CHEMICAL ASPECTS OF THE PD/H – H_2O SYSTEM IN ITS NUCLEAR ACTIVE STATE

BY

DR. STANISLAW SZPAK, WITH DR. FRANK GORDON

OCTOBER 2011

SPAWAR

SECOND DRAFT ARCHIVE REPORT

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Contents

Preface

An occasional gift to one who is interested in documenting research in a field is the emergence of a previously unpublished work. For me this has certainly been the case for Dr. Stanislaw Szpak's book on low energy nuclear reactions (LENR). Thanks go to Dr. Frank Gordon for making the book available for archiving. And I am grateful to Dr. David Nagel for coming up with the idea and suggesting it to Dr. Gordon. Dr. Nagel and Dr. Pam Mossier-Boss have reviewed all or portions of this document.

I am pleased also for the opportunity to provide a summary of Dr. Szpak's LENR research as context for his book. The following have been particularly helpful for preparing the summary.

- Carat, Ruby, 2015. Following Nature's Documents: Stan Szpak LENR Co-Deposition. A Cold Fusion Now Short Film, Featuring Dr. Stan Szpak, Dr. Frank Gordon, and Dr. Melvin Miles. Transcript. https://www.youtube.com/watech?v=OxBJjWzlKl0.
- Clairemont Mortuary, 2016. Obituary for Stanislaw John Szpak. https://www.clairemontmortuary.com/obituaries/Stanislaw-Szpak-41114/#!/Obituary.
- Frazier, C., 2013. Dr. Pam Boss Receives Preparata Medal. Infinite Energy, Issue 111, September/October.
- Gluck, P., 2016. Oct 13, 2016 Sad News, Some LENR Info. Post on Ego Out Blogsite. http://egooutpeters.blogspot.com/2016/10/oct-13-2016-sad-news-some-lenr-info.html.
- Gordon, F., 2016. In Memory of Dr. Stan Szpak. Infinine Energy, Issue 130, November/December.
- Krivit, S., 2016. U.S. Navy LENR Researcher Stanislaw Szpak Dies. New Energy Times. October 13. https://news.newenergytimes.net/2016/10/13/u-s-navy-lenr-researcher-stanislaw-szpak-dies/.
- Krivit, S., date unknown., Szpak Theory Portal: The Stan Szpak Theory of Cold Fusion. New Energy Times. http://newenergytimes.com/v2/sr/Theories/SzpakTheory.shtml.
- Krivit., S.?, date unknown. Space and Naval Warfare Systems Center (SPAWAR) Pacific, Refereed Low-Energy Nuclear Reaction (LENR) Journal Papers. Bibliography Compiled by and copyright New Energy Times. http://newenergytimes.com/v2/reports/SSC-Pacific-Refereed-Journal-Articles.pdf.
- Mosier-Boss, P.A. and L. G. Forsley, 2015. "Condensed Matter Nuclear Reactions". https://www.academia.edu/17964553/Condensed_Matter_Nuclear_Science_October_2015.

This archiving project has been performed under the umbrella of the LENR Research Documentation Initiative (LRDI), previously at the Energy Institute of The University of Texas at Austin and now at LENRGY[1]. Gratitude is expressed to the Anthropocene Institute[2] for financial support of the LRDI.

Thomas Grimshaw, President
LENRGY, LLC
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[1] http://www.lenrgyllc.com/.

[2] https://www.anthropoceneinstitute.com/.

1 Introduction

Cold fusion was announced on March 23, 1989, by Martin Fleischmann and. Stanley Pons. The immense potential energy benefits of cold fusion (also referred to as Low Energy Nuclear Reactions, LENR) were immediately recognized. Humankind's need for a source of cheap, clean, inexhaustible, and safe energy seemed to be permanently satisfied. However, LENR was rejected by mainstream science within a year or so, and it remains highly marginalized to this day. On the other hand, the phenomenon has continued to be rigorously pursued by many investigators in several countries. The mounting evidence for the reality of LENR shows that its potential benefits may yet be realized.

One of the first organizations to verify the claims of Fleischmann and Pons was the U.S. Navy Space and Naval Warfare (SPAWAR) Systems Command[3] in San Diego, California. This work was under the leadership of Dr. Stanislaw Szpak, an experienced electrochemist who was well aware of Fleischmann's research and strong reputation in the field. Dr. Szpak's investigations took place at SPAWAR in Dr. Frank Gordon's department and included Dr. Pamela Mosier-Boss and Jerry Smith[4]. Larry Forsley subsequently worked closely with the LENR research team at SPAWAR.

The SPAWAR team has been among the most active in conducting LENR research and advancing understanding of the phenomenon. One of Dr. Szpak's accomplishments was a book on the topic that has not been published. The objective of this report is to provide a brief review of his LENR research and to present his book for archiving to ensure it is not lost to the field.

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[3] Now named Naval Information Warfare (NAVWAR) Systems Command.

[4] Krivit, S., 2016. U.S. Navy LENR Researcher Stanislaw Szpak Dies. New Energy Times. October 13.

2 Summary of LENR Contributions

Dr. Szpak (Figure 2-1) was friends with Martin Fleischmann and apparently knew of his and Stanley Pons' work with electrolytic cells prior to the March 1989 press conference[5]. When the announcement was made, he and Dr. Pam Mosier-Boss almost immediately began LENR experiments in their lab at SPAWAR.

Figure 2-1.
Dr. Stanislaw Szpak in 2015
Source: "Cold Fusion Now" as Referenced in Gordon's Memorial Article

Dr. Frank Gordon has published a description of Dr. Szpak in a memorial article[6], which is excerpted below.

> Dr. Stanislaw (Stan) Szpak, electrochemist and cold fusion scientist, passed away on October 12, 2016 in San Diego, California, one month short of his 96th birthday.
>
> Stan was born on November 17, 1920 in Schenectady, New York. His family returned to Poland when he was two years old. He attended a Polish university, receiving degrees in Chemical Engineering. It was during his time in college that he woke up one morning to the sound of airplanes passing over his house on their way to bomb a small airport a few miles away, which was the start of the German invasion of Poland during World War II. Stan did not register as required by the occupiers and instead became part of the resistance, using his technical skills to make soap and tan leather, which his family used to barter for other goods. During this time he accidentally triggered a land mine that caused him to lose sight in his left eye and suffer some hearing loss.

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[5] Frazier, C., 2013. Pam Boss Receives Preparata Medal. Infinite Energy, Issue 111, September/October.

[6] Gordon, F., 2016. In Memory of Dr. Stan Szpak. Infinine Energy, Issue 130, November/December.

> When the war ended and Poland was under Russian influence, Stan – who was a U.S. citizen by virtue of his birth in the U.S. – applied to return to the U.S. and was successful in 1948. He got a job at the General Electric electroplating facility in Schenectady. After a few years, he resigned to attend the University of Pennsylvania, where in 1961 he earned a Ph.D. in Chemical Engineering. The title of his dissertation was "The Role of the Interface in Liquid-Liquid Mass Transfer (Transfer of Acetic Acid Across Water-Benzene Interface)." His advisor was Dr. Norman A. Hixson and others mentioned in the preface include Lawrence Delaney, Hans Lindermann and Robert Thygeson.
>
> Stan relocated to the San Francisco, California area, where he worked for Lockheed. After a few years Stan began a Government career by accepting a civil service position at the Vallejo Naval Shipyard. It was while he was working there that he met and married his wife, Bozica, who had grown up in Serbia during the war before she immigrated to the U.S. In 1972 they relocated to San Diego, California to work for a predecessor to the SPAWAR Systems Center Navy Laboratory.
>
> In 1989 following the announcement by Martin Fleischmann and Stanley Pons that they had produced nuclear reactions in electrochemical cells, Stan switched his focus to understanding that process which at the time was known as cold fusion but is more commonly known today as low energy nuclear reactions (LENR). Among his many accomplishments, Stan is credited with being the first to use co-deposition to prepare the palladium cathode, which has since been successfully used by scientists worldwide.
>
> During his career Stan co-authored numerous technical papers and was also an inventor on several patents. A partial list includes the Journal of Chemical Physics, Electrochimica Acta, Trends in Electrochemistry, Techniques for Characterization of Electrodes and Electrochemical Processes and the Journal of the Electrochemical Society. Many of those papers dealt with processes involved in electro-deposition so it is not surprising that he was the first to apply co-deposition techniques to prepare the LENR cathode.
>
> Following retirement as a government employee, Stan accepted an Emeritus position at the SPAWAR lab, which allowed him to continue to work on LENR.

Among Dr. Szpak's contributions to the LENR field were the introduction of the powerful codeposition experimental method, the initial observation of "hot spots" (demonstrating the scattered locations of LENR in a sample), and a high rate of reproducibility[7]. Dr. Pam Mossier-Boss received the Preparata Medal[8], one of the most prestigious in the LENR field, in 2013 for her work, much of which was in collaboration with Dr. Szpak.

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[7] Carat, Ruby, 2015. Following Nature's Documents: Stan Szpak LENR Co-Deposition. A Cold Fusion Now Short Film, Featuring Dr. Stan Szpak, Dr. Frank Gordon, and Dr. Melvin Miles. Transcript.

[8] Frazier, C., 2013. Pam Boss Receives Preparata Medal. Infinite Energy, Issue 111, September/October.

3 Publications

Dr. Szpak placed a high priority on getting research results published. Besides his unpublished book, he authored or coauthored numerous papers, reports and presentations. About 50 reports and other items were found for him on the LENR-CANR.org online library (Table 3-1). An additional 12 items are on the New Energy Times list of SPAWAR refereed journal article list[9] (Table 3-2).

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[9] Krivit., S.?, date unknown. Space and Naval Warfare Systems Center (SPAWAR) Pacific, Refereed Low-Energy Nuclear Reaction (LENR) Journal Papers. Bibliography Compiled by and copyright New Energy Times.

Table 3-1.
Dr. Spzak's LENR Publications: Search of LENR-CANR.org for "Szpak"

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1991	Szpak, S., et al., Electrochemical charging of Pd rods. J. Electroanal. Chem., 1991. 309: p. 273.
1991	Szpak, S., P.A. Mosier-Boss, and J.J. Smith, On the behavior of Pd deposited in the presence of evolving deuterium. J. Electroanal. Chem., 1991. 302: p. 255.
1991	Szpak, S., P.A. Mosier-Boss, and J.J. Smith. Reliable Procedure for the Initiation of the Fleischmann-Pons Effect. in Second Annual Conference on Cold Fusion, "The Science of Cold Fusion". 1991. Como, Italy: Societa Italiana di Fisica, Bologna, Italy.
1992	Szpak, S., P.A. Mosier-Boss, and S.R. Scharber, Charging of the Pd/(n)H system: role of the interphase. J. Electroanal. Chem., 1992. 337: p. 147.
1992	Szpak, S., P.A. Mosier-Boss, and J.J. Smith. Comments on Methodology of Excess Tritium Determination. in Third International Conference on Cold Fusion, "Frontiers of Cold Fusion". 1992. Nagoya Japan: Universal Academy Press, Inc., Tokyo, Japan.
1994	Szpak, S., P.A. Mosier-Boss, and C.J. Gabriel, Absorption of deuterium in palladium rods: Model vs. experiment. J. Electroanal. Chem., 1994. 365: p. 275.
1994	Szpak, S., P.A. Mosier-Boss, and R.D. Boss, Comments on the analysis of tritium content in electrochemical cells. J. Electroanal. Chem., 1994. 373: p. 1.
1994	Szpak, S., P.A. Mosier-Boss, and J.J. Smith, Deuterium uptake during Pd-D codeposition. J. Electroanal. Chem., 1994. 379: p. 121.
1995	Mosier-Boss, P.A. and S. Szpak, The Metal Hydrogen System: Interphase Participation in H-Transport. 1995, Naval Control, Command and Ocean Surveillance Center, RDT&E Division.
1995	Szpak, S. and P.A. Mosier-Boss, Anomalous Behavior of the Pd/D System. 1995, Office of Naval Research.
1995	Szpak, S. and P.A. Mosier-Boss, Calorimetry of Open Electrolysis Cells. 1995, Naval Control, Command and Ocean Surveillance Center, RDT&E Division.
1995	Szpak, S., et al., Cyclic voltammetry of Pd + D codeposition. J. Electroanal. Chem., 1995. 380: p. 1.
1996	Szpak, S. and P.A. Mosier-Boss, Nuclear and Thermal Events Associated with Pd + D Codeposition. J. New Energy, 1996. 1(3): p. 54.
1996	Szpak, S. and P.A. Mosier-Boss, On the behavior of the cathodically polarized Pd/D system: a response to Vigier's comments. Phys. Lett. A, 1996. 221: p. 141.
1996	Szpak, S., P.A. Mosier-Boss, and J.J. Smith, On the behavior of the cathodically polarized Pd/D system: Search for emanating radiation. Phys. Lett. A, 1996. 210: p. 382.
1998	Evan, D. and S. Szpak, Note on the release of nH1 from cathodically polarized Pd electrodes. 1998.
1998	Szpak, S., et al., On the behavior of the Pd/D system: Evidence for tritium production. Fusion Technol., 1998. 33: p. 38.
1998	Szpak, S. and P.A. Mosier-Boss, On the release of n/1H from cathodically polarized palladium electrodes. Fusion Technol., 1998. 34: p. 273.
1999	Mosier-Boss, P.A. and S. Szpak, The Pd/(n)H system: transport processes and development of thermal instabilities. Nuovo Cimento Soc. Ital. Fis. A, 1999. 112: p. 577.

1999	Szpak, S., P.A. Mosier-Boss, and M. Miles, Calorimetry of the Pd+D codeposition. Fusion Technol., 1999. 36: p. 234.
2002	Miles, M., et al. Thermal Behavior of Polarized Pd/D Electrodes Prepared by Co-deposition. in The 9th International Conference on Cold Fusion, Condensed Matter Nuclear Science. 2002. Beijing, China: Tsinghua University: Tsinghua Univ. Press.
2002	Mosier-Boss, P.A., et al., Thermal and Nuclear Aspects of the Pd/D2O System (1), ed. S. Szpak and P.A. Mosier-Boss. Vol. 1 A Decade of Research at Navy Laboratories. 2002: SPAWAR Systems Center, San Diego, U.S. Navy.
2003	Szpak, S., et al. Polarized D+/Pd-D2O System: Hot Spots and "Mini-Explosions". in Tenth International Conference on Cold Fusion. 2003. Cambridge, MA: LENR-CANR.org.
2003	Szpak, S., et al. Polarized D+/Pd-D2O System: Hot Spots and "Mini-Explosions" (PowerPoint slides). in Tenth International Conference on Cold Fusion. 2003. Cambridge, MA: LENR-CANR.org.
2004	Szpak, S., P.A. Mosier-Boss, and F. Gordon. Precursors And The Fusion Reactions In Polarised Pd/D-D2O System: Effect Of An External Electric Field. in Eleventh International Conference on Condensed Matter Nuclear Science. 2004. Marseille, France.
2004	Szpak, S., P.A. Mosier-Boss, and F. Gordon. Precursors And The Fusion Reactions In Polarised Pd/D-D2O System: Effect Of An External Electric Field (PowerPoint slides). in Eleventh International Conference on Condensed Matter Nuclear Science. 2004. Marseille, France.
2004	Szpak, S., et al., Thermal behavior of polarized Pd/D electrodes prepared by co-deposition. Thermochim. Acta, 2004. 410: p. 101.
2005	Szpak, S., et al., Evidence of nuclear reactions in the Pd lattice. Naturwiss., 2005. 92(8): p. 394-397.
2005	Szpak, S., P.A. Mosier-Boss, and F. Gordon. Experimental Evidence for LENR in a Polarized Pd/D Lattice (PowerPoint slides). in American Physical Society Meeting. 2005. Los Angeles.
2005	Szpak, S., et al., The effect of an external electric field on surface morphology of co-deposited Pd/D films. J. Electroanal. Chem., 2005. 580: p. 284-290.
2006	Szpak, S., P.A. Mosier-Boss, and F. Gordon. Experimental Evidence for LENR in a Polarized Pd/D Lattice. in NDIA 2006. 2006. Washington, DC.
2007	Mosier-Boss, P.A., et al. Pd/D Co-Deposition: Excess Power Generation and Its Origin (paper and PowerPoint slides). in 233rd ACS National Meeting. 2007. Chicago, IL.
2007	Mosier-Boss, P.A., S. Szpak, and F. Gordon. Production of High Energy Particles Using the Pd/D Co-Deposition Process (PowerPoint slides). in APS March Meeting. 2007. Denver, CO.
2007	Mosier-Boss, P.A., et al., Use of CR-39 in Pd/D co-deposition experiments. Eur. Phys. J. Appl. Phys., 2007. 40: p. 293-303.
2007	Szpak, S., P.A. Mosier-Boss, and F. Gordon, Further evidence of nuclear reactions in the Pd lattice: emission of charged particles. Naturwiss., 2007. DOI 10.1007.
2008	Mosier-Boss, P.A., et al., Detection of Energetic Particles and Neutrons Emitted During Pd/D Co-Deposition, in Low-Energy Nuclear Reactions Sourcebook. 2008, American Chemical Society: Washington, DC. p. 311-334.
2008	Mosier-Boss, P.A., et al., Reply to Comment on 'The Use of CR-39 in Pd/D Co-deposition Experiments': A Response to Kowalski. Eur. Phys. J. Appl. Phys., 2008. 44: p. 287-290.
2008	Mosier-Boss, P.A., et al., Triple tracks in CR-39 as the result of PdD Co-deposition: evidence of energetic neutrons. Naturwiss., 2008. doi:10.1007/s00114-008-0449-x(96): p. 135-142.

2008	Szpak, S., et al. LENR Research Using Co-Deposition. in ICCF-14 International Conference on Condensed Matter Nuclear Science. 2008. Washington, DC.
2008	Szpak, S., et al. SPAWAR Systems Center-Pacific Pd:D Co-Deposition Research: Overview of Refereed LENR Publications. in ICCF-14 International Conference on Condensed Matter Nuclear Science. 2008. Washington, DC.
2009	Mosier-Boss, P.A., et al., Characterization of tracks in CR-39 detectors obtained as a result of Pd/D Co-deposition. Eur. Phys. J. Appl. Phys., 2009. 46.
2010	Mosier-Boss, P.A., et al., Comparison of Pd/D co-deposition and DT neutron generated triple tracks observed in CR-39 detectors. Eur. Phys. J. Appl. Phys., 2010. 51.
2012	Fleischmann, M., et al., Experimental Evidence of Nuclear Reactions Generated in a Polarized Pd/D Lattice. 2012, LENR-CANR.org.
2012	Szpak, S. and J. Dea, Evidence for the Induction of Nuclear Activity in Polarized Pd/H-H2O System. J. Condensed Matter Nucl. Sci., 2012. 9: p. 21-29.
2013	Szpak, S. and F. Gordon, The Fleischmann-Pons Effect: Reactions and Processes. J. Condensed Matter Nucl. Sci., 2013. 12: p. 143-157.
2014	Szpak, S. and F. Gordon, Cathode to Electrolyte Transfer of Energy Generated in the Fleischmann-Pons Experiment. J. Condensed Matter Nucl. Sci., 2014. 14.
2014	Szpak, S. and F. Gordon, Forcing the Pd/^1 H - ^1H2O System into a Nuclear Active State. J. Condensed Matter Nucl. Sci., 2014. 13.
2014	Szpak, S. and F. Gordon, On the Mechanism of Tritium Production in Electrochemical Cells. J. Condensed Matter Nucl. Sci., 2014. 14.
2014	Szpak, S., The Pd + D Co-Deposition: Process, Product, Performance. J. Condensed Matter Nucl. Sci., 2014. 14.
2015	Szpak, S., On the Path Leading To The Fleischmann-Pons Effect. J. Condensed Matter Nucl. Sci., 2015. 17.

Table 3-2.
Additional Items on New Energy Times' SPAWAR List of Refereed Publications

Szpak, S.	2002	Szpak, S., and P.A. Mosier-Boss, Thermal And Nuclear Aspects Of The Pd/D2o System Vol. 2: Simulation Of The Electrochemical Cell (Icarus) Calorimetry, TR 1862, Vol. 2, (2002)
Forsley, L.P.	2007	Forsley, Larry P.G., Mosier-Boss, Pamela, Phillips, Gary W., Szpak, Stanislaw, Khim, Jay W. and Gordon, Frank E., "Time Resolved, High Resolution, Gamma-Ray and Integrated Charged and Knock-on Particle Measurements of Pd:D Co-deposition Cells," American Physical Society, Denver, Colorado, March 5, 2007
Gordon, F.	2009	Frank Gordon, Pam Mosier-Boss, Stan Szpak, "20 Years of LENR Research using Co- Deposition," ACS, March, 22, 2009
Gordon, F.	2009	Frank Gordon, Pam Mosier-Boss, Stan Szpak, Melvin Miles, Lawrence Forsely, Mitchell Swartz, "20 Years of LENR Research using Co-Deposition," University of Missouri Colloquium, 2009
Mossier Boss, P.A.	2010	P.A. Mosier-Boss, F.E. Gordon, and L.P.G. Forsley, "Characterization of Energetic Particles Emitted During Pd/D Co-Deposition for Use in a Radioisotope Thermoelectric Generator (RTG)," Low Energy Nuclear Reactions Source Book II , American Chemical Society, (2010).
Mossier Boss, P.A.	2010	P.A. Mosier-Boss, L.P.G. Forsley, and F.E. Gordon, "Comments on Co-deposition Electrolysis Results: A Response to Kowalski," Journal of Condensed Matter Nuclear Science,3 (2010) pp 4–8
Marwan, J.	2010	J. Marwan, M. C. H. McKubre, F. L. Tanzella, P. L. Hagelstein, M. H. Miles, M. R. Swartz, Edmund Storms, Y. Iwamura, P. A. Mosier-Boss and L. P. G. Forsley, "A new look at low- energy nuclear reaction (LENR) research: a response to Shanahan", Journal of Environmental Monitoring,12, 2010, pp 1765-1770
Mossier Boss, P.A.	2011	P. A. Mosier-Boss, JY. Dea, F. E. Gordon, L.P. Forsley, M.H. Miles, "Review of Twenty Years of LENR Research Using Pd/D Co-deposition", Journal of Condensed Matter Nuclear Science 4 (2011) pp 173–187.
Mossier Boss, P.A.	2011	P.A. Mosier-Boss, L.P.G. Forsley, P. Carbonnelle, M.S. Morey, J.R. Tinsley, J. P. Hurley, F.E. Gordon, "Comparison of SEM and Optical Analysis of DT Neutron Tracks in CR- 39 Detectors", Radiation Measurements, 47, pp 57-66. online doi:10.1016/j.radmeas.2011.10.004, (2011) (Pre-print)
Mossier Boss, P.A.	2011	P.A. Mosier-Boss, L.P.G. Forsley, P. Carbonelle, M.S. Morey, J.R. Tinsley, J. P. Hurley, F.E. Gordon, "Comparison of SEM and Optical Analysis of DT Neutron Tracks in CR-39 Detectors", Hard X-Ray, Gamma-Ray, and Neutron Detector Physics XIII , edited by Franks, James, and Burger, Proceedings of the SPIE Vol. 8142, (2011) pp K1 – K8
Forsley, L.P.	2013	L.P. Forsley, P.A. Mosier-Boss, P.J. McDaniel, F.E. Gordon, "Charged Particle Detection in the Pd/D System: CR-39 SSNTD vs. Real-Time Measurement of Charged Particle Stimulated Pd K-Shell X-Rays,"Electrochimica Acta 88, p. 373– 383, (2013)

4 Unpublished Book

Dr. Szpak's book reflects the experiments and insights he gained during his research at SPAWAR. The book is included as an attachment to achieve the archiving objective of this report. Its contents are shown in Table 4-1.The book covers Dr. Szpak's ideas for modeling nuclear activity in the palladium-hydrogen-water system, a description of his understanding of the system, the codeposition experimental method, and the thermal and nuclear effects of the system. The epilog sets forth his concerns and reflections for the LENR field.

Table 4-1.
Contents of Dr. Szpak's Book

Section I: MODELING OF NUCLEAR ACTIVITY IN THE CONDENSED MATTER
- 1.0 Background to cold fusion
- 1.3 The Spring meeting of APS
- 2.0 Early research at SPAWAR and elsewhere
- 3.0 Formulation of research program
- 4.0 Evolution of a model at SPAWAR: history in outline
- 5.0 Construction of the present model
- 6.0 Conclusion
- 7.0 Appendix

Section II: THE Pd_nH − H2O SYSTEM
- 1.0 Elctrochemical cell – system representation
- 2.0 Reaction volume
- 3.0 Associated driving forces
- 4.0 Intermediate reaction products
- 5.0 The interphase
- 6.0 Quantitative approach to selected processes
- 7.0 Cell in external fields

Section III: CO–DEPOSITED CATHODES: PROCEDURE AND PROPERTIES
- 1.0 Massive vs co–deposited cathodes
- 2.0 Electrochemistry of co–deposition
- 3.0 Co–deposition: advantages vs disadvantages

Section IV: The Pd_n/H – H_2O SYSTEM: Thermal Effects
- 1.0 Early observations
- 2.0 Electrochemical calorimetry – general remarks
- 3.0 Basic cell/calorimeter designs
- 4.0 Thermal behavior
- 5.0 Remarks concerning open cell calorimetry
- 6.0 Nature of heat sources
- 7.0 Catastrophic events
- 8.0 Catastrophic thermal event in the Pd/H–H2O system
- 9.0 The recombination theory

Section V: NUCLEAR EFFECTS
- 1.0 Electromagnetic radiation: X–ray, γ – emission
- 2.0 Tritium production
- 3.0 Production of new elements
- 4.0 Emission of charged particles
- 5.0 Electron and its role in nuclear reactions

Section VI: EPILOG
- 1.0 Challenges
- 2.0 Sending a message
- 3.0 Being a messenger
- 4.0 Reflections

Appendix : BYU memorandum circulated in 1989

The appendix includes two items: 1) an undated memorandum entitled, “Piezonuclear Fusion at Brigham Young University”; and 2) a copy of a 1986 paper in the Journal of Physics G: Nuclear Physics entitled, “Piezonuclear Fusion in Isotopic Hydrogen Molecules”. Reference is also made to another document entitled, “Brief History of Cold Fusion at Brigham Young University”, 2004, by J. Balliff, W. Evenson, and S. Jones. This document is available as no. 131 on Ludwik Kowalski’s LENR website[10].

Dr. Pam Mossier-Boss, who worked closely with Dr. Szpak, has reviewed his book and provided several editorial comments. They are listed below.

Page	Line	Comment
8	27	Insert “had”
25	N/A	Better figure needed
48	3	Replace “answer” with “answers”
57	32	Replace “Dr” with “Dr.”
60	9	Replace “eplosions” with “explosions”
63	4	Replace “^{4}He’s” with “helium-4 atoms”
	Last ¶	The experiment was done on a Ni screen not Au. The Pd had melted and some of it solidified on the plastic wall in close proximity to the cathode.
70	6	Replace “particulr” with “particular”
84	4	Replace “substsntial” with “substantial”
90	8	Replace “prof.” with “Prof.”
	Last	Replace “Dr” with “Dr.”
92	3	Replace “he” with “the”
	fn 120	Replace “2007)” with “(2007)”
	fn 121	Replace “Dr” with “Dr.”
	fn 121	Replace “prof.” with “Prof.”
93	1	Replace “H23O” with “H2O”
94	1	Replace “viathe” with “via the”
97	2	Replace “Pd?H” with “Pd/H”
109	2	Replace “drown” with “drawn”
125	11	Replace “prof.” with “Prof.”
	15	Replace “prof.” with “Prof.”

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[10] https://msuweb.montclair.edu/~kowalskil/cf/131history.html.

Attachment.

Chemical Aspects of the Pd/H – H_2O System in Its Nuclear Active State

CHEMICAL ASPECTS OF THE Pd/^{n}H – H_2O SYSTEM IN ITS NUCLEAR ACTIVE STATE

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with

Frank Gordon

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CONTENT

Section VI: EPILOG

SECTION I: MODELING OF NUCLEAR ACTIVITY IN THE CONDENSED MATTER

On the 23rd March 1989 Professors Fleischmann and Pons announced that, at low temperatures and pressures, nuclear events can be initiated and sustained by electrochemically compressing deuterium into the palladium lattice and that these events manifest themselves in the form of excess enthalpy generation. More importantly, this could be demonstrated by a simple experiment which did not require the use of specialized equipment: only a cell/calorimeter, a palladium rod, a platinum counter electrode and a galvanostat were needed. The availability of equipment and the idea of low temperature nuclear reactions created great deal of interest. Unfortunately, at that time, only few of the many attempts to confirm the Fleischmann–Pons results were successful.

1.0 Background to “cold fusion”

Any discussion of the nuclear activity in the Pd lattice, should not be attempted without first describing the reasons that prompted this research. The information assembled here was taken from the writings and lectures given by Prof. Fleischmann, F.R.S. and the memorandum prepared by the faculty of the Brigham Young University[1].

1.1 The Fleischmann–Pons effect

Fleischmann in an article published in the ethics in science journal *Accountability in Research* outlined the thought processes that led him to the discovery of cold fusion. This discovery did not happen spontaneously but rather was the outcome of a long chain of empirical results. As early as the late 40’s, Fleischmann realized that the Pd/^{n}H^{+} (n=1,2,3) system is *the most extraordinary example of an electrolyte*, *i.e.* exhibiting behavior that could not be explained in terms of the Debye–Huckel theory. In the 60’s, he was convinced that the correct approach to the behavior of the Pd/^{n}H system must be in terms of quantum electrodynamics (QED). His underlying goal was to illustrate the need to apply QED reasoning when explaining the behavior of condensed matter as well as demonstrating that such effects can be probed using electrochemical procedures. This led Fleischmann, in 1983, to two questions: (i) *“would the putative*

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[1] Reproduced in Appendix A

reaction of D^+ compressed into a host lattice be different from reactions in a dilute plasma (or reactions of highly excited D in solids)? and (ii) *could such changes in the reactions be observed"?* At that time, he expected the answers: Yes to the first and No to the second. In intervening years, Fleischmann collected enough evidence to change the answer to (ii) from No to Yes. Finally, in March 1989, Fleischmann and Pons presented the evidence of nuclear activities in the Pd/D system. In the lecture given at the First International Conference on Cold Fusion (ICCF – 1), Fleischmann said the following: *"We, (F. and P.), for our part would not have started this investigation, if we have accepted the view that nuclear reaction in a host lattice could not have been affected by coherent processes"*. Indeed, this view forms the core of present theories.

1.2 Research at Brigham Young University

The question that needs an answer: Was the work presented by Fleischmann and Pons unique or was there a similar work done elsewhere? And, if the answer is yes, then was there an unauthorized "sharing" of information? According to the faculty members of the Brigham Young University and, in particular to Prof. S. Jones, the answer is yes to the first and, most emphatically, no to the second. This statement was challenged by University of Utah Prof. S. Pons. In response, the Brigham Young University issued a memorandum *Piezonuclear fusion at Brigham Young University* in which they identified the scientific team (5 from the department of physics and astronomy, 1 from chemistry department, 7 students, 1 faculty member from University of Arizona)[2] and included copies of 6 pages (one notarized) from their logbooks. Reading this memorandum one gets an impression that: (i) students colloquia and term papers are equal to presentations at scientific meetings and publications in journals, (ii) that an "hit and run" (if this does not work, try something else) approach to research is acceptable, (iii) being first is the most important achievement. And there is the copy of notarized page from Czirr's logbook with the notary's statement: *The catalyzed fusion process outlined above was explained and formulated on or prior to 4/7/86 (April). Lee R Phillips, notary, Provo UT, expires 12/23/87.* It would require lot of explanations to comprehend and understand the content therein. As an example, in the upper right corner one reads: Al, Cu, Ni – not hydrite forming; next line: Pt, Pd dissolve much H; next line: Li, Al – hydrite. Here, Al is both – hydrite and not hydrite forming and Pt dissolves much H. That is interesting – in other laboratories Pd does, and Pt does not, absorb hydrogen – facts known for more than a century.

In reading this memorandum one finds two dates that are of interest (introduction of electrochemical procedure): 07 April 1986 and 24 August 1988. In 1986 – *"plans for research were extensively developed"*; August 1988 – *"further experiments planned, research program set out and performed vigorously* from August 1988". Approximately one month later, Prof. Jones is asked to review a proposal on " The behavior of electrochemically compressed hydrogen and deuterium" which was submitted to DoE by professors Pons and Fleischmann in September 1988 and represented work done within the time period 1983 and 1988. Because of the nature of the work (nuclear

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[2] It is interesting that Prof. D. N. Bennion, past president of the Electrochemical Society was not the team member

reactions in a test tube), and because the University of Utah and Brigham Young University are within 30 miles from each other, it should not be surprising if information was transmitted. Besides, it is difficult to accept the notion that after reading Pons and Fleischmann proposal, Jones could not be influenced by the thoughts and methodology contained therein.

1.3 The Spring APS meeting – Baltimore MD, May 1989

The physics community rejected the F–P discovery because they considered it to be an example of pathological science. In fact, during the APS annual meeting in Baltimore, MD (May 1989), a professor of theoretical physics in the known Institution of Higher Learning, declared: *It's all very well to theorize how fusion might take place in a palladium electrode. One could also theorize about how pigs could fly, if they have wings, but pigs don't have wings*[3]. It is true, professor, pigs don't have wings, but nuclear events do occur in a palladium lattice!

2.0 Early research at SPAWAR San Diego and elsewhere

As elsewhere, also in this Laboratory, there was excitement and the desire to further explore the mystery of "cold fusion". Also, as elsewhere, it was assumed that this is as very simple process, in particular, a process not requiring special attention. Such assumption was a mistake. In many cases, the investigators were not prepared for, and should not undertake research of the Fleischmann–Pons effect.

2.1 Amateurs at work

Due to enormous importance of this discovery, the predictable reaction to the Fleischmann and Pons announcement was to duplicate their experiments. In the SPAWAR Laboratory, a couple of physicists rushed to their work station and the same evening started their research. They procured a Pd rod of questionable quality and started their experiment. With this accomplishment, they immediately applied for IR[4] funds. In that, they were successful.

Even a cursory examination of their experimental procedure led me to the conclusion that their effort, in all likelihood, would end in failure. Indeed, that was what has happened. The reasons for their failure was not the errors committed by Fleischmann and Pons, but the lack of basic knowledge of processes involved and their unwillingness to acknowledge it. Instead, following the teachings of the Establishment, one of them wrote [5] *... there is the loss of 6 months to a year of hundreds of scientists productivity which went into the teaching these two miscreants some basic nuclear physics (e.g. no neutrons – no fusion). This was foolishness on my part.* Was it foolishness or was it to tap into available IR funding to do research in an area completely foreign to him? In this connection, we must observe that his statement was made after his IR funding has ended. He continued as follows: *Even now (27 Oct. 90), after the whole thing has*

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[3] statement by S. Koonin, *cf.* S.B. Krivit and H. Winocur, The rebirth of cold fusion, Pacific Oaks Press, Los Angeles, 2004

[4] Independent Research program

[5] the date:27 Oct. 1990; Article:1370 of sci.physics.fusion

been shown to be nonsense, there are still people out there getting money from their managers to work on this. Undoubtedly, he referred to our effort.

2.2 Reformulation of the F–P protocol: The co–deposition

My involvement in the research of the F–P effect dates back to May 1989 when I proposed a variant of the usually employed "massive" electrodes, *viz* working electrodes prepared by the Pd+D co–deposition, a process in which the Pd^{2+} ions are electro–deposited onto an Au substrate in the presence of evolving deuterium. Preliminary data, reported in 1991, indicated that electrodes prepared by co–deposition represent an ideal research tool because (i) they retain all features of "massive" electrodes, *e.g.* positive feedback, life–after–death, (ii) there is no incubation time, (iii) the D/Pd atomic ratios exceeding unity are obtained within seconds, (iv) the intensity of the F–P effect is higher and (v) the reproducibility is 100%. Thus, the use of working electrodes prepared by the co–deposition process removes a number of difficulties associated with massive electrodes. For example, metallurgical aspects, so much in play when massive electrodes are employed, are not existing because in co–deposited films the surface morphology as well as the bulk structure are controlled by the cell current and solution composition.

2.3 The ICCF–1 and ICCF–2 meetings

The first international conference on cold fusion, ICCF–1, was held on 23–31 March 1990 at the University Park Hotel in Salt Lake City, UT. In an opening address, Dr Fritz Will[6] stated that the objective of this conference was to *provide forum for scientists to present results of their research and discuss the findings with others thereby stimulating new ideas and advancing our understanding*. Referring to the difficulties encountered by many, he noted that a multi–million (dollars) expenditure was needed to achieve reproducible performance of the semi–conducting devices. As to the attitude of the Establishment, he pointed out that the Fleischmann and Pons discovery was not the first where the discovery was rejected by the Establishment by citing Galileo and his views on whether the Earth circles the Sun or *vice versa* and that *experimental results cannot be declared wrong by voting* – a clear objection to the statistics of failures[7]. He concluded his opening remarks with an observation that *the freedom of science is just as basic as freedom of speech*.

At the time of the Conference, we had already demonstrated that (i) electrodes prepared by co–deposition yield the F–P effect and (ii) that excess heat is not of purely chemical origin. Moreover, we demonstrated that this technique reproducibly produces excess enthalpy, low intensity X–ray and exhibits sporadic tritium production. Although a preliminary note was already submitted,[8] we were prohibited from presenting our results.[9] By not presenting our results we put ourselves far behind others. Parenthetically, the results of our research were known to Prof. Fleischmann who strongly encouraged their presentation at the ICCF–1.

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[6] at that time Director of National Cold Fusion Institute

[7] Note: only negative results are reproducible – a position taken by, among others, Morrison

[8] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem. **392** 255 (1991)

[9] Office of Naval Research decision communicated to us by Dr. R. Novak.

The ICCF–1 conference was open to media and politicians (Officials of the State of Utah). During intermissions and in the evenings, there were interviews, comments, predictions, etc. An entirely different atmosphere was created for the ICCF–2 conference. The meeting was held in Villa Olma in Como, a small town in northern Italy. The exuberance of the ICCF–1 was absent. Instead, a serious attitude was established. Also absent were comments no neutrons – no fusion, although Morrison still insisted that statistics (ratio of negative to positive results) does not allow him to accept the reality of cold fusion. [10]

It is generally agreed that the turning point regarding the F–P effect was the ICCF–2 meeting in Como, Italy in 1991. One of the highlights of this meeting was the remark made by Prof. H. Gerischer, a noted German electrochemist and a former director of the Max Planck Institute for Physical Chemistry. By his own admission, he came to Como as a skeptical observer, he left convinced that the effect discovered by Fleischmann and Pons is real and worth further study. It was at this meeting that we, for the first time, presented our results. As we have learned subsequently, these results were, in part, instrumental in Gerischer's comments [11].

2.4 Navy reaction to ICCF–2: Attempts at collaborative effort

Shortly after the ICCF–2 meeting, the Office of Naval Research (ONR) proposed a collaborative effort involving SPAWAR, NWC China Lake and NRL to investigate the anomalous effects associated with the prolonged charging of the Pd–D_2O system. This effort's basic premise was *to contribute in collegial fashion to a co–ordinated tri–laboratory experiment*. The proposed procedure was to be as follows: each laboratory will try to duplicate the findings of the other by following the respective experimental protocols as closely as possible. As it turned out, this requirement was a mistake because no one would admit that improvements could not be made – thus invalidating the concept of collaborative effort. Moreover, at that time there seem to be little interest in our technique.

Collaboration may have also an opposite effect – instead of (i) acceleration it may delay the progress, (ii) it may end without ill feelings or (iii) it may produce unexpected consequences. A good example of the latter is the case dealing with responsibilities and credits. The names of parties involved are not important – the lessons learned are. Briefly, in 1994 an experiment involving infrared (IR) imaging of the electrode surface during the reduction of D_2O revealed the existence of "hot spots". Three participants were involved: the first offered laboratory space and the necessary equipment (galvanostat, temperature recorder), the second an IR camera. We provided the working cell and experimental protocol. The presence of "hot spots" as well as the large temperature difference between the electrode surface and the electrolyte, added significantly to our understanding of the nature of the F–P effect. We prepared a brief communication for publication [12] with all participants as co–authors. For reasons not known to us,

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[10] In this connection, Dr Peter Glueck observed: There were 100 shots fired at Caucescu, but only 20 hit him, *ergo* Caucescu is still alive. So much for the statistical considerations.

[11] expressed to us at the informal dinner arranged by Dr Nowak, ONR

[12] S. Szpak, P.A. Mosier–Boss and J.J. Smith, Physics Letters A **221**, 141 (1996)

they did not want to be listed as co–authors.

The importance of "hot spots" was recognized by Fleischmann who cited the results in his lecture [13]. In the audience was present one member of the "collaborative team". He claimed that we had "pirated" his ideas. He called me complaining that because he was not listed as a co–author, his professional standing has suffered. In the course of our conversation I reminded him that he did not want his name to be associated with the publication. That ended this un–pleasant incident. Lesson learned – a clear understanding of responsibilities as well as credits must precede any collaborative effort.

3.0 Formulation of a research program

History teaches that the development of a theory is not an instantaneous event but follows the set of observations made by many in the course of years. If, as originally assumed, the reaction path is the d + d fusion, then any theory must answer the following: (i) how can the Coulomb barrier be penetrated at the observed rates and (ii) how can the process avoid the restrictions imposed by the asymptotic freedom[14]? If, on the other hand, other reaction paths are possible, then the existence of the Coulomb barrier is irrelevant. One such reaction path was examined by Lawrence *et.al.*.[15]

In 1989 not much was known about the F–P effect, except that long time periods were required before the onset of the event. These time periods were identified as due to diffusion controlled charging and a set of unknown processes preceding the onset of the event, the incubation time. While the first was well understood and, therefore, could be dealt with, the second was a complete mystery. Experience teaches that the resolution of complex problems often requires decisive action involving bold simplification, lucid interpretation and useful generalization.

3.1 The first decision

A new phenomenon – the room temperature nuclear reaction in a test tube – was announced. Since this effect was discovered by two distinguished professors of chemistry – one would expect that methods and reasoning of chemistry would be helpful in the interpretation and further development of the understanding of this phenomenon. The starting point was to eliminate the diffusion controlled charging time. For this reason, cathodes prepared by co–deposition were selected. Indeed, cells employing such electrodes yielded excess power shortly after initiation of cell current flow. Moreover, characteristic features of the thermal behavior, *i.e.* positive feed–back and life–after–death, were retained.

3.2 The second decision.

Because of the system's thermal behavior, it became evident that, at least in part, teachings of nuclear physics would have to be invoked. Consequently, a dilemma – what direction to follow – is it chemistry or physics? While the first decision dealt with

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[13] M. Fleischmann, Cold fusion: Past, present and future, ICCF–7, p.230

[14] Asymptotic freedom states that on the length scale associated with nuclear reactions particles must behave as if they were free particles

[15] E.O. Lawrence, E. McMillan and R.L. Thornton, Phys. Rev., **48**, 493 (1935)

practical matter, that of reducing or eliminating the charging time, the second considered the interpretation of observed facts. It is my position that to do so, it is preferable to employ chemical methodology with frequent reference to mechanical analogs when interpreting complex situations. Following this procedure, one can arrive at a realistic model that, in general way, agrees with observation.

3.3 Concept of a model

To describe the transition from the qualitative demonstrations to quantitative analyses one must consider all the facts collected and deductions advanced therefrom. Thus, the problem in constructing a model is two–fold: (i) how can the actual phenomenon, as observed and measured, be presented by a set of simple statements, (ii) how can these statements be reduced to quantitative expressions. In seeking an answer one has two choices: either to start with an abstract treatment, frequently involving advanced mathematics, or to examine the available empirical evidence. The choice that we have made was to follow the latter. This decision was prompted by the quotes:

(i) *My advice to those who wish to learn the art of scientific prophecy is not to rely on abstract reason, but to decipher the secret language of Nature from the Nature's documents, the facts of experience.*[16]

(ii) *It is the qualitative demonstrations which are unambiguous; the quantitative analyses of the experimental results can be subject to debate but, if the quantitative analyses stand in opposition to the qualitative demonstrations, then these methods must be judged to be incorrect.*[17].

(iii) *It should be entirely general line of attack in the study of all types of systems and processes to begin with a thermodynamic analysis and to exhaust the possibilities of thermodynamic reasoning before introducing models and assumptions of a mechanical or molecular nature.*[18]

(iv) *...it has now been verified that there indeed exists new dynamic state of matter induced by a flow of energy far from equilibrium. Such states are governed by a new physical chemistry on a supermolecular level while all laws referring to a molecular level remain essentially unchanged. In all cases considered, the coherent behaviour on the supermolecular level corresponds in fact to amplification of specific molecular properties (such as kinetic constants) in far from thermodynamic equilibrium conditions.*[19]

(v) *.... it should be possible to describe new developments in a thorough–going and*

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[16] This is the closing sentence taken from an address given to the Durham Philosophical Society and the Pure Science Society, Kings College, at Newcastle–upon–Tyne on 21 May 1943 by Max Born at that time professor of Natural Philosophy University of Edinburgh.

[17] A thought expressed by Prof. Fleischmann in his address to the Royal Institute of Chemistry

[18] quote from van Rysselberghe, The thermodynamic structure of electrochemistry, Modern Aspects of Electrochemistry, Plenum Press 1966

[19] the quote from Thermodynamic Theory of Structure, Stability and Fluctuations by P. Glandsdorff and I. Prigogine, Wiley Interscience, 1971

satisfactory manner without the use of advanced mathematics.[20]

(vi) *Insofar as the propositions of mathematics give account of reality, they are not certain; and insofar as they are certain, they do not describe reality*.[21]

These quotes tell us that there are observations, the *Nature's documents*, that point the direction to follow. In the case of conflict between experiment and generally accepted interpretation, the experiment must prevail. Before considering atomistic or mechanical models, one should fully explore the thermodynamic reasoning which, at far from equilibrium, may lead to extraordinary conclusions. Finally, mathematics should not be the primary tool in the treatment of complex situations, because it may lead to models not connected to reality.

A rational approach to the selection of topics to be investigated is *via* the answer to a specific question or a group of questions. Thus, whatever the approach, one is confronted with (a) by what mechanism the energetically low level activities lead to high level response, i.e. by what mechanism chemical reactions/processes transit into nuclear reactions? (b) what is the dynamics of absorbed D^+/D at high D/Pd atomic ratios? (c) can a nuclear event be modeled as a separate entity, *i.e.* without consideration of environment (problem of coherence) (d) is there a clearly defined boundary separating chemistry from physics?

3.4 Modeling of a chemical system

The development of a model representing a phenomenon, whether controversial or not, is not based on a single experiment or even a set of experiments unless there exists a logical connection between them. The preferred course of action is: First the phenomenon is studied to determine what is known and what is not known; Second, to select experiments, the *Nature's documents*, that would answer what is not known. These criteria guided the approach taken in the construction of a model leading to the initiation of conditions for the nuclear reactions in the Pd lattice. It is customary to define chemical aspects as any process or reaction that (i) contains the statement of conservation of mass and charge, (ii) where the initial state is controlled by the experimental protocol and the final state by energy considerations, (iii) where the heat of reaction is on the order of eV and (iv) that is governed by the Arrhenius type rate equation.

The starting point in constructing a model representing the F–P effect, is to model a chemical system: $\Sigma \mathrm{R} \rightarrow (\mathrm{X}_i, \mathrm{Y}_j) \rightarrow \Sigma\mathrm{P} + \mathrm{Q}$. Here, the R's are reactants, the P's products and Q energy produced or consumed. The quantities in the bracket $(\mathrm{X}_i, \mathrm{X}_j)$ stand for a set of intermediate reactions and/or processes whose identification is sought. The transition from R to P may occur *via* a set of consecutive, parallel (reversible, irreversible), coupled and chain reactions in which the direction is determined by the concentrations of substances entering the reaction (reactants). These reactions, if and when identified, provide the basis for an acceptable model. As written, the system consists of unbounded particles (*i.e.* without considerations of their state), in practice

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[20] L. Pauling, The Nature of Chemical Bond, third edition, Cornell University Press, 1960

[21] a quote attributed to Einstein

the state of both, reactants and products is characterized by p, T, c_i.

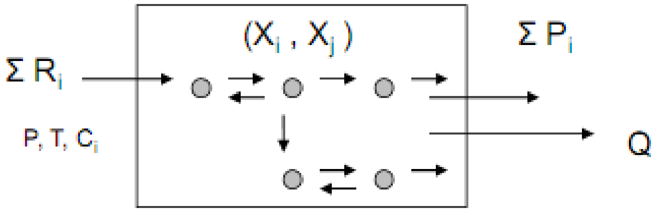

Figure 1:

In this model, the input variables are controlled by experimental protocol while the output variables by the reaction kinetics and energy considerations. At this stage the correct way to proceed is to follow the path: experiment – data collection – data analysis – conclusions – new experiments – etc. That is to say, the development of a model is not the product of a brain wave (storm) but the result of collected empirical evidence. In formulation of such a model one should keep in mind the precise language of thermodynamic reasoning, noting, however, that the *Nature's documents* lead the way.

4.0 Evolution of a model at SPAWAR: history in outline

Without the introduction of time one cannot describe the evolution, a process of change from a primitive to advanced state, which is usually characterized by time periods in which a set of events, often called "milestones", representing significant advancements, takes place. One way to present the history of progress is to define time periods that are separated by: (i) significant observations or (ii) work conditions. With this in mind, the two decades of research activities at SPAWAR into the F–P effect can be divided into five (5) periods:
Time period 1989–1991; end of pathological science
Time period 1991–1993; change in personnel, Dr. Gordon assumes the position of an Executive Director of NISE – WEST.[22]
Time period 1993–1995; change in personnel, my retirement
Time period 1997–2003; production of new elements, particle emission
Time period 2003- 2009; Construction of a model.

4.1 Time period 1989 – 1991.

The F–P discovery divided the scientific community into "believers" and "skeptics". The believers produced experimental facts – the skeptics declared them wrong. In spite of the opposition from the Establishment, the research continued: in this Laboratory the emphasis was on the development of electrodes prepared by co–deposition. Within this time period my interest was directed toward the "getting to know the Pd/D–D_2O system" so that I could construct a realistic model and assess the probability of developing

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[22] NISE – WEST is a Navy organization responsible for In–Service Engineering Activities of electronic and communication system throughout the Pacific.

an energy source. In getting to know the system, we started with the comparison of the original F–P protocol with the adapted procedure that of employing cathodes prepared by co–deposition. This procedure eliminates charging time and provides micro–defects that not only increase the surface area but create conditions favorable for the D^+/Pd lattice interactions, and the [D]/[Pd] atomic ratio greater than one is obtained within seconds.[23] One further comment: The conservation of mass and charge together with the time rate of change of the fractional occupation of sites demands that the interfacial layer specifies the correct boundary conditions, and that the interphase is not a passive but an active element.[24]

The 1991 model: On the basis of available information, one could construct a very primitive model, in which the reactants entering the reaction volume are: D and e^-, the known reaction $D \rightarrow D^+ + e^-$ and the products: excess enthalpy, Q, and X–rays. Here, the kinetics of the reaction(s) preceding the occurrence of the nuclear event, the (X_i,X_j), is unknown except that involves reactants e^- and D^+ in concentrations 100 molar in D^+ and 1000 molar in e^-, respectively.

Publications:
On the behavior of Pd deposited in the presence of evolving deuterium, J. Electroanal. Chem., **202**, 255 (1991)
Electrochemical charging of Pd rods, J. Electroanal. Chem., **309**, 273 (1991).

4.2 Time period 1991 –1993

The review of known facts suggested that, in planning further research activities, the most fruitful approach is to follow the advice given by van Rysselberghe, *i.e.* to exhaust all possibilities of thermodynamic reasoning before introducing specific models – which means that the F–P effect is to be viewed in connection with the system as a whole and that it permits free flow of energy to and from an environment (open system).

4.3 Time period 1993–1995

The 1991 model does not yield any information on events occurring during the incubation period, except for a vague term "self–organization". In order to gain some insight into their respective behavior, and to search for the *Nature's documents* that might be useful in further development of the model, an electrochemical cell should be treated as a system where (i) the properties of an interphase, (ii) the nature of the self–organization and (iii) the system's thermal behavior, should be examined in some detail.

The empirical evidence collected during the 1993 – 1995 period (results of the IR imaging and piezoelectric experiment, *cf.* IV.6.0) led to a model in which temporal and spatial distributions of heat sources[25] suggests that coherent processes are responsible for the self–organization, *i.e.* the formation of new molecular structures in well defined

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[23] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem., **379**, 121 (1994)

[24] S. Szpak, C.J. Gabriel, J.J. Smith and R.J. Nowak, J. Electroanal. Chem., **309**, 273 (1991

[25] assuming reasonable heats of reaction, it would require 10^4 –10^9 single events occurring within a volume of radius 100 Å and within 10^{-4} seconds

reaction volumes. Furthermore, the evidence led to the description of the interphase region as consisting of two inhomogeneous regions on both sides of the contact surface. Its thickness is determined by the relaxation times of processes that affect its structure (the van Rysselberghe concept of an active interphase). *cf.* II.5.0

The 1995 model extended the concept presented in 1993 in that (i) temporal and spatial distribution of hot spots points to self–organization as the characteristic feature of the (X_i,X_j) reaction scheme and (ii) the interphase acts as an interfacial engine, the latter *via* contact with outside world. In particular, change in the controlling parameters, *e.g.* overpotential, η, temperature, T, whether periodic or aperiodic, will produce changes within the interphase that affect the cell performance in either positive or negative direction.

Publications:

Absorption of deuterium in palladium rods: model vs experiment, J. Electroanal. Chem., **365**, 275 (1994)
Comments on the analysis of tritium content in electrochemical cells, J. Electroanal. Chem., **373**, 1 (1994).
Deuterium uptake during Pd–D codeposition, J. Electroanal. Chem., **379** 121 (1994)
Cyclic voltammetry of Pd+D co–deposition, J. Electroanal. Chem., **380**,1 (1995) *On the behavior of cathodically polarized Pd/D system: search for emanating radiation*, Physics Letters A **210**, 383 (1996)
On the behavior of cathodically polarized Pd/D system: a response to Vigier's comments, Physics Letters A **221**, 141 (1996)
On the behavior of the Pd/D system: evidence for tritium production, Fusion Technology, **33**, 38 (998)
On the release of ^2H *from cathodically polarized palladium electrodes*, Fusion Technology, **34** 273 (1998)
Calorimetry of the Pd+D co–deposition, Fusion Technology, **36** 234 (1999)

4.4 Time period 1997 – 2003

After reviewing the progress made elsewhere during the inactive period (1995–1997) I decided to change the course of research from thermal effects to other manifestations of nuclear activity. Up to the 1995, the experimentally controlled variables affected the kinetics of the electrode reactions and only peripherally the structure of the interphase region. In 1997, in addition to p, c, T and η, a new controlling variable, namely an external field was introduced, ψ_e and ψ_m for the electrostatic and magnetostatic field, respectively. This approach was selected for one reason: an external electrostatic field tends to deform the shape of a solid conductor, *i.e.* it would increase the number of the Pd lattice defects and, presumably, increase thermal output. This was expected – what was not expected was the occurrence of violent localized reactions which changed (deformed) the initial morphology. One example is shown in Fig. 2

A set of SEM's photographs, Fig. 2a –e, shows the effect of the placement of an operating cell in an external electrostatic field. It is seen that the initially almost spherical globules, Fig. 2a, were re–arranged to (i) produce a layered structure, Fig. 2b, chaotic

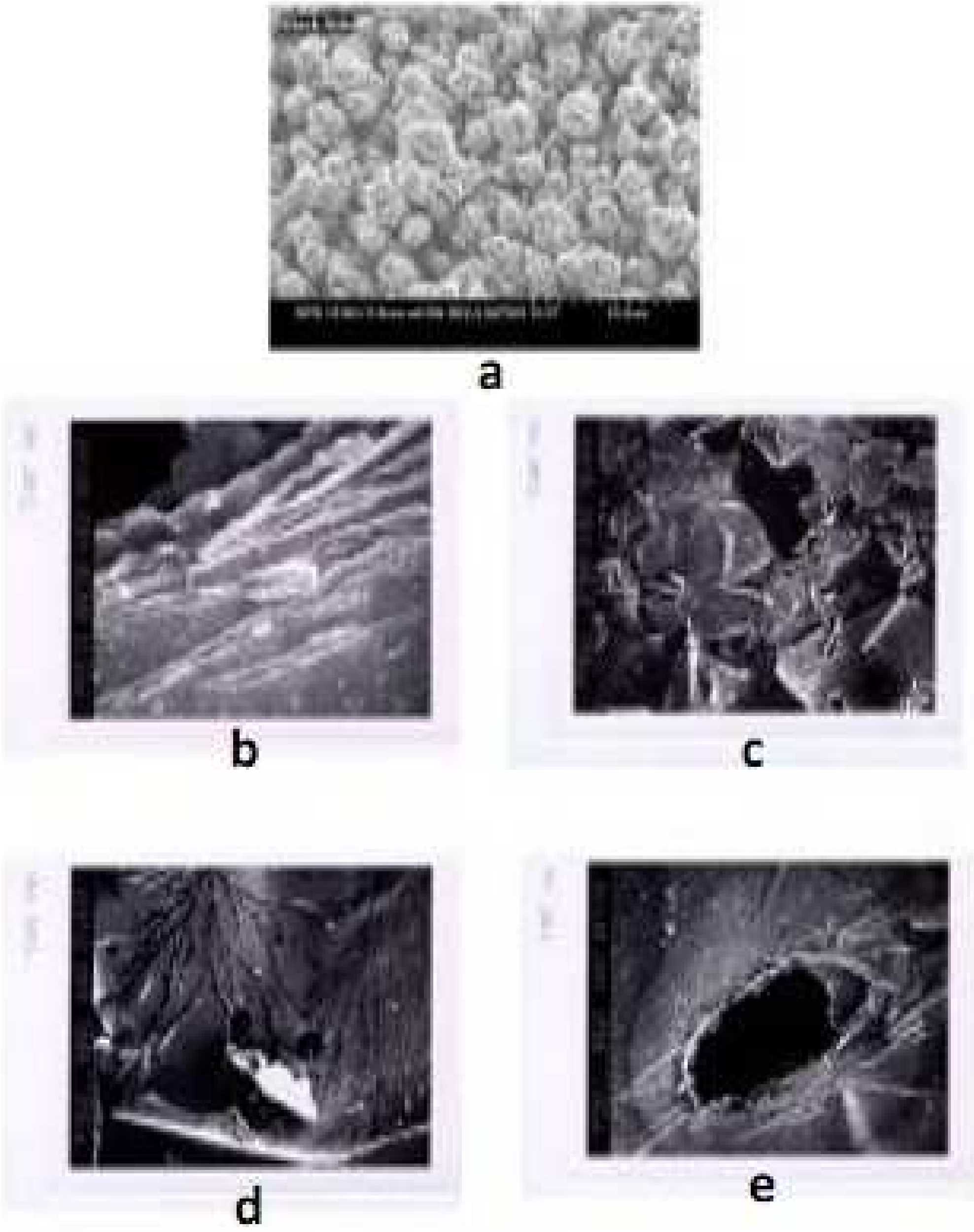

Figure 2:

placement of small thin fragments, Fig. 2c, a bended large thin plate (foil?) with, what appears to be, branches of wires attached to its surface, Fig. 2d, and a violent event resembling explosion, Fig. 2e. Even a cursory examination leads to a conclusion that the energy required to produce such changes is far in excess of that which, under the conditions of experiment, could be extracted from the field at the required rate and intensity. One could offer a number of explanation as to the physical meaning of the observed facts – all of them, most likely, highly speculative. What is clear, however, is that a substantial energy expenditure was needed to create the observed new structures. Thus, the question: Is the energy transferred from the external field sufficient to account for the observed facts and, if not, then what is the its source?

In 1995 the fundamental question concerning the mechanism by which the energetically low level process/reaction can generate energetically high response was recognized but not specified[26]. The simplest mechanism for the transition from eV/at heat of reaction to the MeV response is for the product of the eV/at reaction to become a reactant for the MeV response. The search for the appropriate eV/at reactions begins with the consideration of the state of the system, *i.e.* its distance from equilibrium and the high concentration of both D^+ species and free electrons, e^-, together with one of the observed emission of soft X–rays[27] suggests the possibility of an electron capture as the energetically low level process.[28]

In the Landau and Lifshitz treatise on Theoretical Physics[29] one finds that the capture of an electron by a nucleus $e^- + ^A_Z(X) \rightarrow ^A_{Z-1}(X) + \nu$, can be treated as a chemical reaction. Consequently, for this reaction to occur is the inequality $\mu(e^-) > \mu[^A_Z(X)] - \mu[^A_{Z-1}(X)]$. Applying this argument to the reverse reaction of a neutron decay $p^+ + e^- \rightarrow n$, the production of neutrons will occur when $\mu(e^-) > \mu(n) - \mu(p^+)$.

The chemical representation of neutron production in the Pd lattice, specifically in the interphase region is conditioned on whether or not (i) the absorbed deuterium is in its nuclear state, (ii) there is sufficiently high concentration of free electrons and (iii) the required energies could be generated. The experimental as well as theoretical evidence indicates that, indeed, the necessary conditions exist. First, the absorbed deuterium is in its nuclear state; second, within the cathodically polarized interphase the concentration of free electrons is high and third, the interaction of charged particles with the potential fields on the order of 10^9 Vcm^{-1} can assure production of needed energies.

4.5 Time period 2003 – 2009

As a general observation, any system placed in an external field (magnetic or electric) increases its energy density in amount equal to the energy transfered into the volume

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[26] The approach taken by theorists was to find how the Coulomb barrier can be circumvented so that the reaction d + d would occur at reasonable rates. Theorists assumed that the d + d fusion to be independent, *i.e.* they excluded from consideration the coherent behavior. Such assumption is contradicted by the existence of the incubation time. There are other indications that the "hot fusion" concepts cannot be applied when discussing nuclear events occurring within the confines of the Pd lattice

[27] S. Szpak, P.A. Mosier–Boss and J.J. Smith, Physics Letters A **210**, 383 (1996)

[28] The electron capture by deuteron was discussed by us at the ICCF 11 – in 2004

[29] Vol. V, Statistical Physics, Pergamon Press, 19980, p.319

through its surface. Just how the system, as a whole, responds to the external field and what processes/reactions are affected is illustrated in Fig. 3 a –c. In the absence of external fields, D^+ ions are brought to the charge transfer layer by diffusion. Following their reduction, deuterium atoms are adsorbed at the contact surface (open circles – at low and high energy sites) and penetrate very fast into the subsurface layer where (i) molecular–ions are formed (solid/open circles), (ii) ionization occurs (open circle), (iii) interaction with the Pd lattice defects that leads to cluster formation and (iv) various reactions (e.g. self–organization, nuclear events) could be found.

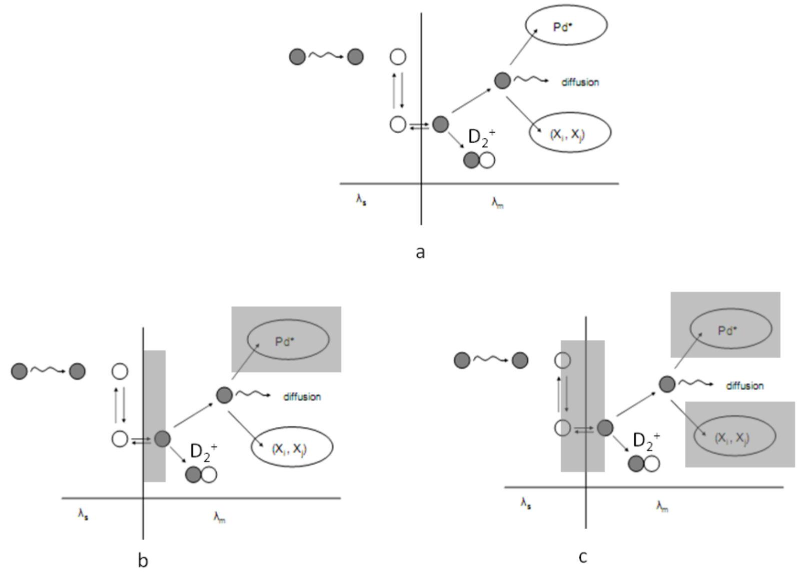

Figure 3:

An external electric field affects the behavior of the polarized electrode through the activities in the electrolyte, at the contact surface and indirectly within the structure and thickness of the interphase *via* relaxation effects, as high–lighted in Fig 3b. An external electrostatic field affects the electrolyte by interacting with ions which are accelerated with net force arising from applied field and opposed by the forces associated with its structure (ions are solvated). The contact surface, a conductor, when placed in an electric field suffers a "negative pressure" and cannot exist in stable equilibrium and it does not directly interact with the interphase region except through the Gauss' theorem, *cf.* II.7.3.

The interaction of the magnetic field with the polarized Pd–D_2O system is far more complex. Here, both the Lorentz as well as the gradient forces are engaged. The Lorentz forces, acting at the solution side of the contact surface, substantially change the hydrodynamics (outside the Prandtl layer) and through it, the distribution of the charge transfer current density. The gradient forces, operating within the interphase, affect shape, stability and distribution of aggregates within the clusters found around the mobile Pd defects and, additionally, though non–specific electronic interactions, with kinetics of the various reactions. These activities are highlighted in Fig. 3c.

In general, an external electrostatic field affects the potential while the magnetic field the kinetic energy of a system. The common feature is their interaction with the contact surface and thus also with the interior *via* the Gauss' theorem. This combined interplay between the surface and interior result in behavior resembling an "interfacial engine"[30] The action of the interfacial engine, and therefore also its output, can be magnified by varying the intensity of the external fields. To affect the processes preceding the nuclear event, slow changes would be required – their frequency would depend on the relaxation of relevant processes.

Publications

Evidence of nuclear activities in the Pd lattice, Naturwissenschaften, **92**, 394 (2005)
Further evidence of nuclear reactions in the Pd-D lattice:emission of charged particle, Naturwissenschaften, **94**, 511 (2007)

5.0 Construction of the present model.

In the construction of a model representing nuclear activities in the Pd lattice we relied on the set of observations logically linking one to another. The model described here applies to cathodes prepared by the co–deposition process and, most likely, also to massive electrodes. In general, there are two ways to view modeling: The first, for the paradigm/model driven research, three conditions must be evaluated, *viz (i) is it applicable (ii) does it violate the second law of thermodynamics, (iii) is it physically and mathematically complete and (iv) is it reducible to mathematical expressions that are useful to experimenters.*[31] The second, to visualize something that cannot be directly observed. Now, because of the very complex nature of this phenomenon, its description cannot be, at the present time, reduced to mathematical expressions that are useful to experimenters. However, the conditions: is it applicable and does it violate the second law of thermodynamics, must be retained. This we have done by constructing a model based on two key observations, *viz* discrete heat sources and production of new elements, interpreted *via* two concepts: (i) self–organization and (ii) electron capture by a proton/deuteron.

5.1 Key observations

(i) *Hot spots*. The characteristic feature of the thermal behavior is the presence of discrete, short lived heat sources distributed in time and space. These fast reactions, releasing thermal energy in a very short time, resemble mini-explosions and lead to the following conclusions: (a) transition from non–reactive to reactive state occurs

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[30] A term used by Sterling and Scriven, [A.I.Ch.J. **5**, 514 (1959) some 60 years ago, to describe the interfacial instability during the mass transport across the liquid–liquid interface.

[31] Chubb in *Thermal and nuclear aspects of the Pd/D$_2$O system*, SPAWAR Systems Center San Diego, TR 1862 vol. I, Feb 2002

at discrete sites, (b) heat producing reaction are fast, (c) a large number of reacting particles is confined in a small volume, (d) statistics of the explosion pattern suggests a structured chaos rather than random distribution. These reactions occur within a volume having radius of few hundreds Angstroms with the reaction time in nano–seconds involving milligrams of total mass.

(ii) New elements. While the observations of hot spots is the starting point in understanding of the chemical aspects, the production of new elements and the associated electromagnetic radiation and particle emission, provides information on the type of nuclear events that take place within the confines of the Pd lattice, *cf.* V.3.0.

5.2 Key concepts

Key observations, *i.e.* the hot spots and production of new elements do not lead directly to the construction of realistic model. Their content and meaning must be related to the processes. To do so, we use two concepts: (i) self–organization and (ii) electron capture by a nucleus treated sas a chemical reaction.

(i) Self–organization implies that there exists a volume element within the system having dimensions much larger than the characteristic molecular dimensions but smaller than the total volume of the system. Within this volume fluctuations behave coherently thus modifying its microscopic behavior. At far from equilibrium, new structures, involving coherent behavior are formed and can be maintained only through a sufficient flow of energy. The interaction between entities constituting the system takes the form of a struggle – some are eliminated others are formed, *i.e.* there exists a state of dynamic equilibrium that allows for the existence of different entities.

(ii) A simple nuclear process, treated as a chemical reaction is not a new concept – it is discussed in the notes on Lectures in Nuclear Physics, given by Fermi in 1949, and in the treatise on the Course of theoretical physics by Landau and Lifshitz. In 2004, I applied this concept to the $e^- + p^+ \rightarrow n$ reaction which, in my opinion, is the key element that triggers the nuclear event in the Pd lattice.[32]

5.3 The three–stage model

The basic question relevant to the construction of model is: what are the processes that led to localized heat sources and to the formation of stable new elements (*i.e.* nuclei containing large number of neutrons and protons and the associate orbital electrons)? It appears, on the basis of experimental evidence, that a three–stage model, shown in Fig. 4, adequately describes the behavior of the negatively polarized Pd/D–D_2O system. It summarizes the following points: simultaneous reactions occur within the same volume, indicated by circle within a circle, located in the interphase region, denoted $\lambda_1 + \lambda_2$.

(i) Reactions/processes in stage I

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[32] Somewhat later, Widom and Larsen presented an alternate treatment based on the concept of weak force interaction leading to production of "ultra–low momentum neutrons" that trigger the nuclear events in condensed matter.

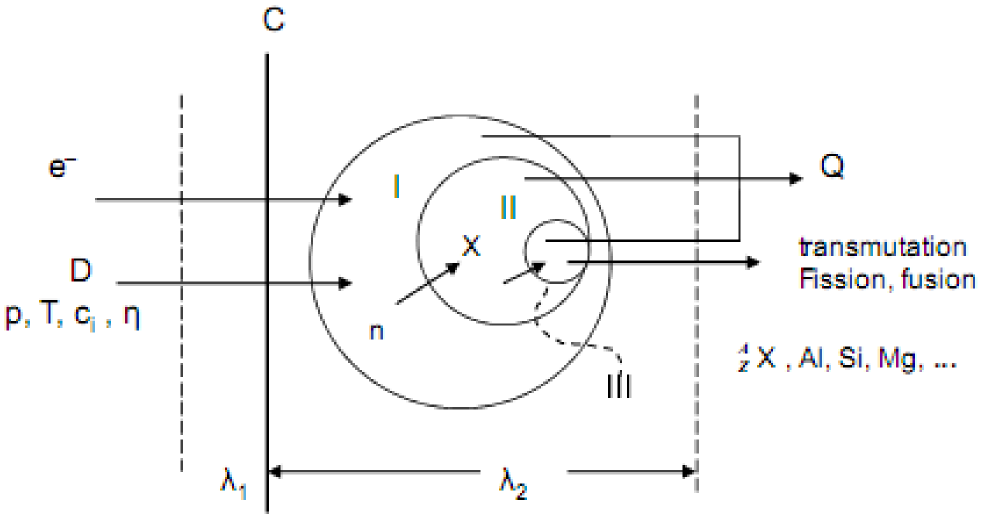

Figure 4:

Reactions within the first step are: (i) ionization of absorbed deuterium $D \rightarrow D^{+} + e^{-}$, (ii) production of molecule–ions $D + D^{+} \rightarrow (D_2^{+} \cdot e^{-})^{+}$, (iii) self–organization leading to the formation of clusters of aggregates $Pd^{*} + nD^{+} \rightarrow Pd^{*}...D_n^{+}$, (iv) production of neutrons $e^{-} + D^{+} \rightarrow 2n$, (v) production of hybrid molecule–ion $n + D_2^{+} \rightarrow (D^{+}T^{+} \cdot e^{-})^{+}$. Reactions (i) – (iii) are typical chemical reactions while (iv) and (v) are chemical reactions within the context of nuclear chemistry.

(ii) Reaction/processes in stage II

Reactants entering stage II are the products of self–organization. These reactants can interact with electrons (electron capture) or with neutrons. In either case, the dynamic equilibrium of stable aggregates generated by self–organization, is disturbed and the affected aggregates can either collapse or explode, depending on the type of interaction with environment[33]. The collapsing cluster/aggregate concept is not singular to our point of view. Similar notion, in concept but not in detail, was postulated by others, as presented at the ICCF 10. For example, Adamenko postulated the *creation and evolution of self–organized and self–supported collapse of electronic–nuclear plasma of initial solid–state density under the action of coherent electronic driver up to a state of large non–stationary electronic–molecular clusters with density close to that of nuclear substance*. There is (i) self–organization, (ii) dense plasma corresponding to clusters interacting with the Pd lattice defects, *e.g.* mobile dislocations, (iii) “of solid–state density” corresponding to the action of the FΔΦ and asymmetric stress field and

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[33] Any mechanical system in dynamic equilibrium can be made to collapse/explode if appropriate change in the force distribution is made.

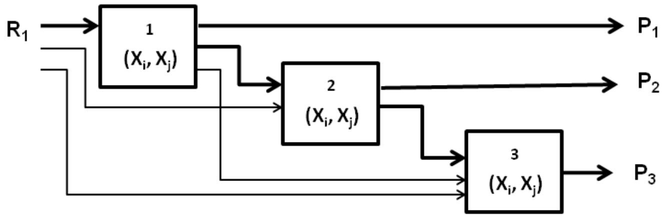

Figure 5:

(iv) “coherent electronic driver..., ” is equivalent to the formation of new structures on the “super–molecular level.

As an another example is the Kozima point of view. He postulated: (i) existence of CF – matter consisting of neutrons, protons and electrons, a neutron drop, denoted $^A_Z\Delta$, (ii) the CF matter is located within the surface boundary layer, (iii) the reaction path follows the scheme $^A_Z(X) +^a_z \delta \rightarrow^{A+a}_{Z+z} (X)$. His model is, in fact, similar to the proposed model except thatmymodel is based on a set of reproducible empirical evidence while his, as presented, has evolved from *ad hoc* made assumptions[34].

(iii) Reactions/processes in stage III

While the processes in stages I and II can be treated in terms of chemical concepts, those in stage III involve system stabilization *via* the transition from unstable nuclei to stable ones by various decay processes. A very general picture is as follows: The first step, in stage I, is the production of neutrons *via* the electron capture process: e$^-$ + D$^+$ → 2n. Stage II is the seat for nuclear reaction of the type $n + \Sigma^A_Z X_i \rightarrow \Sigma^{A+1}_Z X_j$. Here the reaction product, entering stage III, is a set of new unstable nuclei $^{A+1}_Z X^*_j$ which undergo process of “stabilization” by fusion, fission, particle emission and electromagnetic radiation: $\Sigma^{A+1}_Z X^*_j - [p^+, \alpha^{2+}, n, \gamma, X] \rightarrow \Sigma^{A_1}_{Z+1} X_j$.

5.4 Alternate representation

Figure 4 displays processes taking place in each stage. In an alternate representation, illustrated in Fig. 5, we view the operating system in terms of a flow diagram and assign specific function to each stage.

Functions assigned to individual stages are as follows:
Stage 1 – system is in dynamic equilibrium established through the order–disorder ac-

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[34] An alternate concept, that of a sequential addition of proton and neutrons, has been proposed. This concept appears not to be realistic because no method was provided for stopping the progress of addition to account for the presence of multiple elements. In order to select the most likely model one must view it in terms of available empirical evidence, *viz* low intensity of radiation (low energy X–rays), and the composition of new elements. The absence of significant X–ray emission would suggest that the specie undergoing reaction must have “bound” electrons (electrons in orbits). The multiple new elements indicate that the Pd..D^+_n clusters contain distribution of molecular species

tivity at the contact surface. Its principal activity, denoted (X_i,X_j), is to provide, *via* self–organization, reactants that produce excess power and new elements (transmutation). These are essentially chemical processes and could be either exo – or endothermic

Stage 2 – principal seat of excess power production *via* the unspecified set of nuclear processes .

Stage 3 – production of new elements *via* the various decay paths leading to the formation of stable elements such as Ca, Al, Si, etc. Although the transition from unstable to stable nuclei is highly exothermic, its contribution to the cell power output is minimal.

6.0 Concluding remarks

Early in my work, I recognized the importance of chemical aspects when examining the nature of the phenomenon of "cold fusion". This approach led me to the construction of, what I believe to be, a realistic model representing the behavior of the negatively polarized Pd/D–D_2O system in which the Pd/D films are prepared by co–deposition. Briefly, in this model, processes in subsystems I and II are chemical (including nuclear chemistry) in nature while those in subsystem III can only be treated by methods of physics. The common approach – modeling of the nuclear activity arising from electrochemical compression of absorbed deuterium requires the knowledge of input variables, the reactants and products as well as the energy change of the whole system.

SECTION II: THE Pd/$^nH--^nH_2$O SYSTEM

It is generally agreed that the correct approach to study the F–P effect is to consider its interdisciplinary character. This is certainly true when “massive” cathodes are used. It is my view that, when co–deposited cathodes are employed, one should use the system approach to identify the problems and offer solutions. In a typical run, reaction products are: excess power, electromagnetic radiation, particle emission, and transmutations, the latter refers to the production of new elements such as T, Al, Si, Zn, ... The poor reproducibility associated with the original protocol and the lack of repeatability in cells employing co–deposited cathodes indicates a complex reaction paths that are influenced by the magnitude of external constraints such as temperature, changes in overpotential or external field. Knowing the reaction paths involved, it should be possible to manipulate the external parameters in order to affect the composition of reaction products in such a way that the desired product is obtained.

This section provides essential information needed to resolve the complexity of the system and to suggest further research. In particular, it contains recognition that the nuclear active state is due to a set of processes and reactions including the interphase transport and the effect of external electric and magnetic fields.

1.0 Electrochemical cell – system representation

Starting with the proposition that an operating electrochemical cell is a system displaced from equilibrium, with the distance from equilibrium measured by the cell potential, one can examine all factors leading to he construction of a model representing nuclear active state of the Pd/H–H_2O system. To provide a rational interpretation one must define the system and its initial conditions, *i.e.* variables that keep the system either in equilibrium or non–equilibrium state as well as the interaction between individual system’s elements. This is illustrated in Fig. 6. The system consists of three sub–systems: cathode (negative electrode), solution and anode (positive electrode). Of these three sub–systems, the negative electrode plays dominant role with minor contribution from the solution phase. Consequently, this sub–system (the cathode) is further broken into elements, *viz* the reaction volume, the interphase and the contact with outside world. It is here, where the concepts and definitions must be clearly presented and, it is here, where the complex interplay involving kinetic and thermodynamic considerations must be recognized.

Each element is further broken into sub—elements. Within each sub–element the processes are interacting with each other to a various degree. This interaction may involve activity (i) within the same sub–element, *e.g.* excess heat affects its generation rate through positive feed–back, or (ii) between two sub–elements, *e.g.* Pd lattice defects and reaction kinetics, or (iii) the natural fluctuations and reaction kinetics. But the really important interaction, leading to a set of nuclear events, is the contact with the outside world and the reaction volume *via* the interphase, specifically *via* its contact surface and the electric double layer. While the element *reaction volume* is the location of the nuclear events, the contact surface plus the electric double layer is the most important because it serves as a link between the reaction space the outside world, Putting it differently, it transmits the action of external forces that lead to various couplings, self–organization and, in the end, to the initiation of nuclear event.

Reiterating, Figure II.1 (i) illustrates the difficulty in resolving the complexity of an operating electrochemical cell, (ii) implies that statistical experiments design is not practical and (iii) a search for, and identification of, key observations is the way to proceed. But above all, it stresses the importance of the connection between the outside world and the reaction volume *via* the energy and/or matter exchange. Proper understanding of the relationship between activities of connecting elements is the primary objective, when assessing systems limitations as well as to predict its behavior. To meet this objective, we take as the starting point the content and meaning of a chemical reaction. In particular, we selected the order indicated by systems elements shown in Fig. 6, *i.e.*: (i) reaction volume, (ii) interphase, its structure and function (response to transport and fluctuations), (iii) effect of external fields: electrostatic and magnetostatic (formation of new structures, domains).

2.0 Reaction volume

Within the reaction volume element there are five sub–elements of special interest, *viz* reaction –its content and meaning, reaction kinetics, associated driving forces, types of reaction and reaction products.

2.1 Reaction – its content and meaning

A chemical reaction is usually described by either (i) aA + bB $\rightarrow$ cC + dD + Q or (ii) A + a $\rightarrow$ B + b + Q where $Q>0$ denotes an exothermic and $Q<0$ an endothermic reaction. The first is the usual representation of a chemical reaction indicating that a moles of A react with b moles of B to yield c moles of C and d moles of D. The second is that of a nuclear reaction of the type A(a,b)B + Q. As written, in both cases only limited information is provided, *viz* conservation of matter and charge. In both cases the initial and final states are not specified, *i.e.* the system consists of unbounded particles in the sense that there is a continuous range of possible energies. In reality, the initial state in both cases is controlled by experimental protocol and the final state by energy considerations.

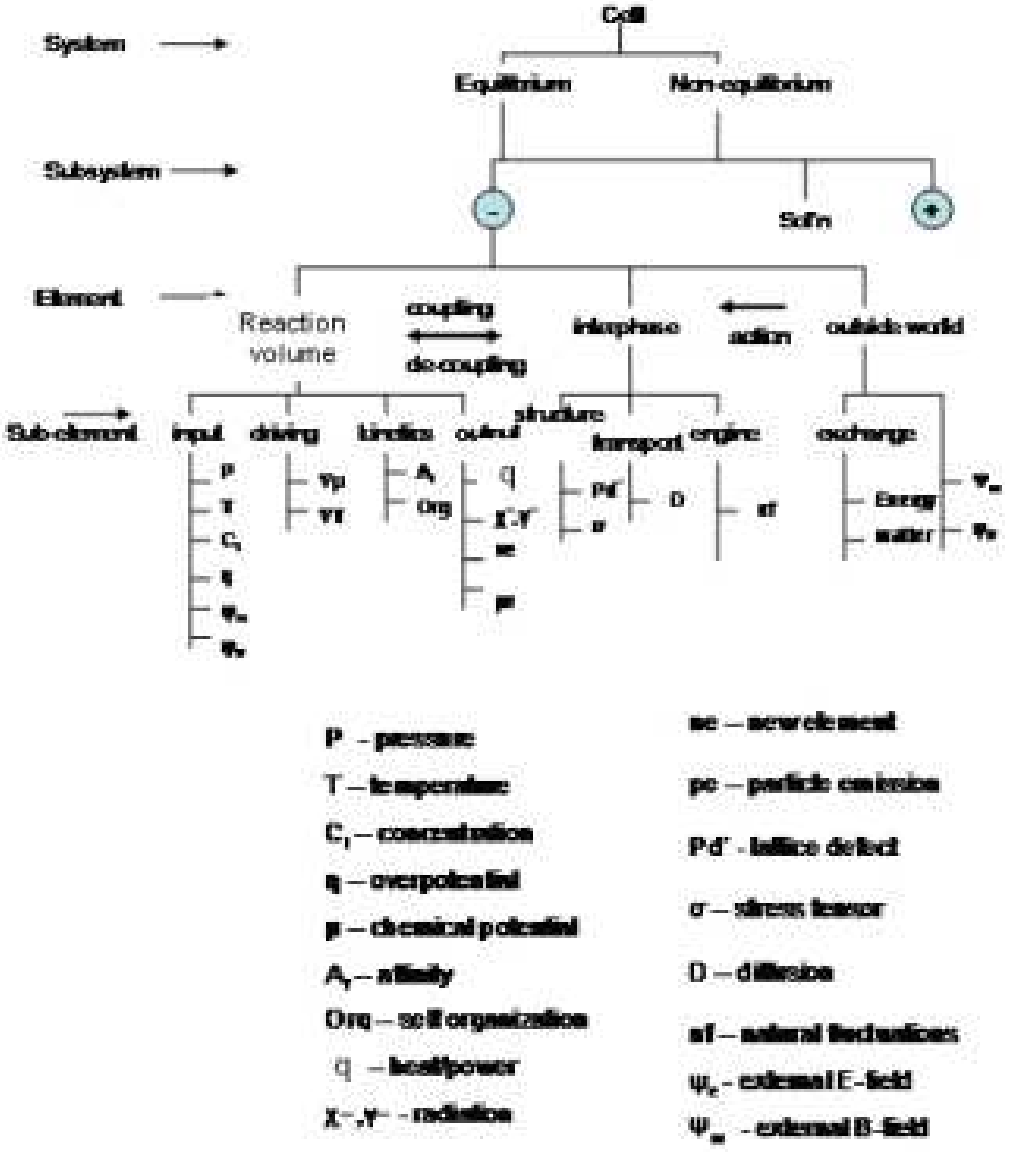
System
Subsystem
Element
Sub-element
Cell
Equilibrium
Non-equilibrium
-
Sol'n
+
Reaction volume
coupling
de-coupling
interphase
action
outside world
input
driving
kinetics
output
structure
transport
engine
exchange
P
T
C_i
η
Ψ_m
Ψ_e
$\nabla\mu$
∇T
A_r
Org
q
χ^-,γ^-
ne
pe
Pd'
σ
D
nf
Energy
matter
Ψ_m
Ψ_e
P - pressure
T – temperature
C_i – concentration
η – overpotential
μ – chemical potential
A_r – affinity
Org – self organization
q – heat/power
χ^-,γ^- - radiation
ne – new element
pe – particle emission
Pd' - lattice defect
σ – stress tensor
D – diffusion
nf – natural fluctuations
ψ_e - external E-field
Ψ_m - external B-field

Figure 6:

I should like to observe that chemical reactions rarely occur in an ideal environment, *i.e.* at constant temperature in time and space and at constant concentration in space. This consideration leads to recognition that to uncover the true mechanism one must consider (i) conditions of an experiment, (ii) problems arising from energy (heat), matter and charge transfer, and (iii) description of laws that govern transport of matter and energy under the conditions of an experiment.

2.2 Reaction kinetics

Reactions are classified as either simple, v = kc_r, *i.e.* if the reaction velocity depends only on concentration of a reactant, or complex, if v = k_1c_r - k_1 c_p, where it depends on both concentrations of reactants and products. The reaction velocity may also be expressed in terms of their affinities, *i.e.*

$$v = v_0[exp(A_f/RT) - exp(A_r/RT)] \tag{1}$$

where A_f is the affinity in forward direction and A_r in reverse, and where v_0 is the exchange velocity. Affinity is defined as $A = -\Sigma_i \nu_i \mu_i$, *i.e.* by the sum of chemical potentials of reactants and products[35]. Here, the affinity plays the same role as $\nabla\mu$ in transport processes or ∇T in heat conduction.

2.3 Coupled reactions.

In complex systems, involving number of processes taking place simultaneously, chemical coupling may occur. Chemical coupling means that one reaction affects the velocity of another. Conditions for the existence of such coupling were discussed by van Rysselberghe,[36] who showed that this is possible if (i) there is another reaction, the coupling reaction (dQ' = $A_2 \times v_2 > 0$) occurring in the direction dictated by its affinity and (ii) if the sum $A_1 \times v_1 + A_2 \times v_2 > 0$, *i.e.* when the coupling reaction proceeds at sufficient rate and where the needed energy is supplied by the coupling reaction.

One example of coupled reactions, also referred to as induced reactions, is the initiation of nuclear activity in the Pd/H–H_2O system when it is placed in an external magnetic field, *cf.* IV.7.1. Once initiated, the nuclear activity is maintained by two simultaneous reactions, that of neutron and deuterium production, with the first proceeding in the non–spontaneous direction and requiring energy input, the second exothermic. Here, we have a situation where the electron capture reaction produces a substance that is used up in another reaction with the liberation of energy which, in turn, accelerates the first reaction. Such dependence is referred to as coupling, more accurately the first reaction is coupled the second coupling.

3.0 Associated driving forces

Chemical and electrochemical potentials are the forces that drive the transport of particular species within the electrode structure as well as reactions that occur therein.

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[35] I. Prigogine and R. Defay, *Chemical Thermodynamics*, Longmans Green Co, London – New York – Toronto, 1954

[36] P. van Rysselberghe, Bull. Ac. Roy. Bel.,**22**, 1330 (1936); ibid. **23**, 416 (1937)

Because of the very complex nature of the Pd/D–D_2O system, specifically of the interphase region, the applicable driving force (chemical and electrochemical potential) takes on different form.

Formally, the chemical potential is defined as the partial molar quantity, $\mu = \partial Z/\partial n_i)_{n_i \neq n_j}$ of thermodynamic quantities such as: internal energy, U, enthalpy, H, Helmholtz free energy, F, and Gibbs free energy, G. Note that each of these quantities exhibits the property of additivity, *i.e.* each depends on the number of particles in the system under consideration. The additivity property means that, if the number of particles is changed by a given factor, then the quantity itself is changed by the same factor. Stating it differently, it means that the quantity in question is an Euler homogeneous function of first order with respect to the number of particles.

3.1 One component system

The chemical potential, μ, is obtained by differentiating any one of the thermodynamic quantities with respect to the number of particles, remembering however, that they are expressed in a different set of variables as indicated by the subscripts. Thus,

$$\mu = \frac{\partial U}{\partial n}|_{S,V} = \frac{\partial H}{\partial n}|_{S,p} = \frac{\partial F}{\partial n}|_{T,V} = \frac{\partial G}{\partial n})|_{T,p}.$$

Note that for a single particle the chemical potential is just its negative binding energy.

3.2 Many component system

In systems with more than one component, the chemical potential of the i–th specie is

$$\mu_i = \frac{\partial U}{\partial n_i}|_{S,V,n_i \neq n_j} = etc$$

3.3 Effect of external and internal fields

The chemical potential in a system containing charged particles in mechanical and thermal equilibrium is given by $\mu_i = \frac{\partial \Delta G}{\partial n_j}|_{p,T,n_j \neq n_i}$. When this system is placed in an external field, ψ (e.g. gravitational field, electrostatic field, magnetostatic field, etc.), the electrochemical potential is given by $\mu_i = \frac{\partial \Delta G}{\partial n_j}|_{p,T,n_j \neq n_i,\psi}$. If the particles interact with an internal or external field, then any change in their number must include energy change (or reversible work) and the chemical potential takes on the form $\overline{\mu} = \mu + u(x,y,z)$ where $u(x,y,z)$ is the interaction energy. Examples of an internal field are: (a) stress field when the addition or subtraction of a particle deforms the body (b) an electrostatic field. In the first case $\overline{\mu} = \mu + u(\sigma, \tau)$, in the second case $\overline{\mu} = \mu + e\Phi$. The latter is the electrochemical potential.[37]

3.4 Chemical potential of i–th species in thin films

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[37] Note that when placed in an external electrostatic field, the potential energy of charged particles becomes a function of position, and the constancy of the chemical potential requires non–uniform distribution of particles.

Of interest in the interpretation of the nuclear active state are the driving forces operating within the interphase which, in turn, are viewed as an assembly of non–autonomous thin layers. A sufficiently large phase, in contact with another, and where the molecular interaction between them is negligible, is referred to as autonomous phase. In such phase, the chemical potential of the i–th species is $\mu_i = \partial F/\partial F|_{T,V}$. But, a thin layer, being influenced by composition of adjacent phases, exhibits the cross–chemical potential which is defined as

$$\frac{\partial F^{\alpha,\beta}}{\partial n_{i,\alpha}} = \frac{A}{V^\alpha}\varepsilon^{\alpha,\beta}_{i,\alpha}$$

so that the complete chemical potential of the i–th component, in the α layer is:

$$\overline{\mu}^\alpha_i = \mu^\alpha_i + \frac{A}{V^\alpha}\varepsilon^{\alpha,\beta}_{i,\alpha}$$

If the α–layer is sandwiched between $(\alpha-1)$ and $(\alpha+1)$, then

$$\overline{\mu}^\alpha_i = \mu^\alpha_i + \frac{A}{V^\alpha}\left[\varepsilon^{\alpha,(\alpha-1)}_{i,\alpha} + \varepsilon^{\alpha,(\alpha+1)}_{i,\alpha}\right]$$

In these expressions, superscripts indicate the layer in question while the subscripts the component.[38]

The effect of the surface discontinuity, *i.e.* the junction of two layers, α,β, on the chemical potential takes the form: $\mu^* = \mu^\alpha + \frac{S}{V}\varepsilon_{i,\alpha}$, where the cross chemical potential term, $\varepsilon_{i,\alpha}$, is the measure of the effect of change in the i–th component on the fee energy of contact. Conversely, the change in the free energy of contact surface will affect the chemical potential of the i–th species in the α layer, and therefore, its gradient and the transport rate.

4.0 Intermediate reaction products

In section I we noted that hot spots provide some information. Here we consider only the time/space distribution which we have identified as a structured chaos, *cf.* IV.6.0. It follows that the intermediate reaction products arising from self–organization are found in the stage I of the three – stage model, *cf.* I.5.4, Fig. 5, but their stability is subject to conditions prevailing in the stage II in response to the action of external factors. The situation is quite accurately described by four statements:[39] (i) a complex interplay of kinetic and thermodynamic quantities determines system's stability, (ii) an open system must be maintained, if self–organization is to occur, *i.e.* spontaneous self–organization can occur if the system is able to exchange part of the energy or matter with the outside world (iii) the formation of new structures is always the result of an instability which may be due to either internal or external fluctuations to the system, (iv) fluctuation,

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[38] Definitions and symbols: 1. A non–autonomous character of a layer indicates that there is a significant number of molecules interacting with molecules in adjacent layers. 2. The measure of the interaction between adjacent layers is the free energy of contact, alternatively of the discontinuity surface. For further discussion consult *Surface Tension and Absorption* by R. Defay and I. Prigogine, Longmans, Green and Co, London, 1966

[39] P. Glansdorff and I. Prigogine Thermodynamic theory of structure, stability and fluctuations, Wiley Interscience, 1971; G. Nicolis, Self–organization in non–equilibrium systems, Wiley Interscience 1977

either internal or external to the system, is always followed by the response which may bring the system to its original conditions or may produce a new structure.

As the departure from equilibrium is increased, the system becomes unstable and evolves to form new structures exhibiting coherent behavior. In particular, non–equilibrium open physico–chemical systems undergo "self–organization" processes which, in turn, yield structures of spatial domains characterized by generation of bursts of chemical activity. Incidentally, we define equilibrium as a state generated by the balance between operating forces. Mathematically it is expressed by a minimum of the free energy (thermodynamic interpretation), or by the probability of the system being at its maximum (statistical interpretation), or by the equality of forward and reverse velocities (kinetic interpretation).

4.1 Molecules, aggregates, clusters and domains

A molecule is defined as an assembly of two or more atoms bound together to form a structure with sufficient stability to consider it as an identifiable specie[40]. As used here, an aggregate is an assembly of molecules, cluster is an assembly of aggregates held together by forces operating in chemical systems. A domain denotes a volume containing one or more clusters interacting with lattice defects.

In the Pd/H–H_2O system the absorbed hydrogen, and its isotopes, interacting with the lattice defects, generates various molecular assemblies. Molecular assemblies are products of self–organization and involve a network of Pd defects, hydrogen isotopes and electrons. By inference, hot spots suggest that the molecular assemblies contain thousands of atoms. Thus, it might be useful to list factors that might be helpful in the interpretation of observed facts, e.g. type of forces and molecular stability.

4.1.1 Types of forces The acting forces in chemical systems are:

(i) electro–valence – forces governed by the Coulomb's law.
(ii) co–valence – attractive strong heteropolar forces between uncharged atoms.
(iii) metallic forces – in which atoms are positively charged with free electrons located between them.
(iv) van der Waals forces. These forces arise partly from polarization and partly from resonance. They produce weak attraction at larger interatomic distances. They are always present – form molecular aggregates and decompose at higher temperatures. Van der Waals forces, by themselves may not lead to the formation of aggregates of molecules in definitive ratios. However, if there are steric considerations, then the formation of aggregates with definitive number of atoms is possible.
(v) hydrogen bonding – exists in solid state and forms molecular aggregates.
(vi) exchange forces. Exchange forces represent a continuous transfer of energy between the coupled particles. Mechanical analog of exchange forces: Two equal and coupled pendulums interact with one another whereby the motion of the first pendulum is gradually transferred to the second pendulum and when the second attains maximum amplitude, the first comes to rest.

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[40] L. Pauling, The Nature of Chemical Bond, Cornell University Press, Ithaca, New York, 1960

(vi) magnetic field induced orientation of non–spherical aggregates arises from the magnetic torque acting on the anisotropic molecules that constitute these assemblies.

4.2 Molecular stability

Conditions for molecular stability are the same as for equilibrium, *i.e.* balance of forces. The Feynman theorem[41] states that, at the equilibrium configuration of a molecule, the resultant force acting on each nucleus vanishes. This means that when in an equilibrium configuration the repulsion of a nucleus by the other nuclei is balanced by the attraction due to electrons. The force acting on each nucleus is calculated using classical electrostatic theory. This balance can be disturbed either gradually or rapidly as in e.g. hot spots, *cf.* IV.6.0. In the latter case, the molecular assembly can collapse or explode depending on whether the attractive or repulsive forces are affected.

The forces that keep molecules and aggregates together are, by analogy, the same as forces between molecules in liquid. Just as in the drop of liquid, here also, spherical symmetry is assumed (spherical symmetry arises from the action of molecular forces and the surface tension). Mechanical stability of the molecule ion [(D^+. e^-) –D^+] as well as the stability of domains of complexes, Pd...[D^+...]$_n$ can be examined using the liquid drop analogy, *i.e. via* the energy considerations and, in particular, by the change in the potential energy associated with the deformation of the spherical drop. This energy consists of two parts: (i) binding energy (*i.e.* energy needed to take the molecular complex apart) and (ii) surface (capillary) energy and, in the case of a domain, also its electrostatic energy. The stability conditions for both molecules, aggregates, clusters and domains are derived from the energy considerations associated with the shape change resulting from the motion that occur within the molecular assembly. In all cases, the "capillary" energy plays important role. Such shape changes can arise from fluctuations which, in turn, modify the magnitude of the potential and kinetic energies of the molecular assembly or the Pd...[D^+...] domain. The interaction with an environment, the Pd lattice defects and/or electrons, occurs in a coherent way. In a special case, shape change might be explosive resulting in molecular collapse.

A complete energy balance must, in addition to the internal energy of system's components, include (a) kinetic energy, (b) mechanical work and (c) heat. Of these, the kinetic energy is of interest when describing molecular stability. Now, change in the kinetic energy, $E_k = mv^2/2$, is equal to work done by force F on the corresponding trajectory, r. If this force depends only on position, then equation dE = F dr can be integrated and the integral $(-\int F dr)$ is the potential energy. The sum of kinetic and potential energies is constant during motion.

Another useful property in the interpretation of the polarized Pd/H–H_2O system, *cf.* IV.8.0, is: A system enclosed by rigid walls that do not permit flow of matter, energy and electric current but allow for the presence of time–invariant (stationary) external fields, is in equilibrium when the condition $\delta U + \delta E_{pot} = 0$ is fulfilled. Here U denotes the internal energy and E_{pot} is the potential energy determined by the stationary conservative forces. Thus, for any virtual displacement in the external potential field,

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[41] R.P. Feynman, Phys. Rev., **56**, 340 (1939)

individually U and E(pot) can change by the distribution of matter but their sum must remain constant.

4.2.1 Specific models

Of the various model available, we selected two, *viz* the Bohr's compound nucleus and the Teller and Sahlin model that might be helpful in interpretation of nuclear events.

Bohr's compound nucleus.

A liquid drop model represents a nucleus in which surface energy is attributed to unbalanced forces that act on a particle at the surface. As another particle approaches the surface and comes in contact with the surface, it is coupled, *i.e.* the energy of the approaching particle is shared with other nucleon and its identity is lost. At some later time, a nucleon can find itself due to statistical fluctuations at the surface and escape. Because of the long period of time, the decaying nucleus forgets how it was formed.

The Teller and Sahlin model

In Teller and Sahlin view,[42] molecule structural units (atoms, electrons) are held together by weak electromagnetic forces with a r^{-2} dependence. A molecule or an aggregate may be viewed as a mixture of particles having very different properties in which slowly oscillating nuclei are immersed in a sea of electrons. This very nature of particles presents both difficulties and advantages in the treatment, because, if a physical system which has variables that change slowly and those that change rapidly, then the behavior of fast variables is not significantly affected by the rate of change of slow particles (decoupling).

4.3 Growth of aggregates

Transmutations or generation of new elements, *cf.* V.4.0, suggest that a cluster contains aggregates of different composition. The reason for the difference in the composition of an aggregate can be given by considering its rate of growth. Here, we assume that the growth of molecular aggregates follows a model: a j–macromolecule is formed either by the acquisition of a monomer by a (j-1) macromolecule or the loss of a monomer by the (j+1) macromolecule. Thus, the time rate change in the concentration of j–macromolecules, N_j is

$$\frac{dN_j}{dt} = \alpha_{j+1}N_{j+1} + \beta_{j-1}N_1N_{j-1} - \alpha_jN_j - \beta_jN_1N_j \tag{2}$$

where α_j is the reaction rate constant for the detachment of a monomer from a j–macromolecule and β is the rate constant for the acquisition of a monomer from the parent phase where the concentration of monomers is N_j.

In addition to Eq. (2), an equation giving the monomer concentration, N_1, is required. For this purpose, we use the relationship: $N_1 - Gt - \Sigma_{j=2}jN_j$, whereby N_1 is expressed as the difference between the number of monomers that have been supplied at the constant rate G and the number of monomers that have been bound in j–macromolecules,

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[42] E. Teller and H.L. Sahlin in *Physical Chemistry – An Advanced Treatise*, vol. V, Academic Press

up to time t. The difficulty in the application of these equations to real systems lies in the determination of the α and β rate constants whose numerical values are controlled by internal and external fluctuation[43], *cf.* Fig. 6.

5.0 The interphase

An interphase identifies an inhomogeneous region separating two homogeneous phases. In the present case, it identifies the inhomogeneous region separating metallic electrode from an electrolyte. To illustrate the inhomogeneity one might construct a test particle to probe the changes in the medium as it approaches the contact surface, *i.e.* surface separating two homogeneous phases. Under equilibrium conditions and far from the contact surface the medium is homogeneous, *i.e.* a test particle sees no difference that would depend on the viewing direction or time. However, as the test particle approaches the contact surface, the situation changes, the test particle sees non–uniformity in force distribution, *i.e.* the homogeneity is lost.

While the processes in the electrolyte phase up to and including adsorption are well known, those occurring on or after crossing the contact surface remain unclear. When the system is in equilibrium, its structure can be defined in terms of physical properties. When the system is not in equilibrium, it is often convenient to discuss its structure in terms of occurring processes, *i.e.* the interphase can be viewed as an assembly of a set of layers whose structure is determined by the operating processes[44] while the interface is the contact surface (*i.e.* the surface of discontinuity) separating the electrolyte and electrode phases. Thus, in a number of situations, it is convenient to represent this region as consisting of a number of thin layers that are homogeneous where, to assure their homogeneity, an average value of a particular variable is taken. The imposition of homogeneity of each layer results in its non–autonomous character which arises from the interaction of molecules in adjacent layers. Consequently, changes in any part cause changes throughout the whole region. To follow these changes, it is convenient to adapt the multi–layer concept, *i.e.* the interphase is divided into a set of non–autonomous layers. The effect of change in one layer on any other can be examined by energy considerations, specifically in regard to a phase transition assumed to precede the initiation of an exothermic reaction. An abrupt change in any of the surface variables will produce a relaxation spectrum which leads to the development of gradients.

5.1 Crossing the interphase

Transport of deuterium across the interphase is due to coupling processes at the contact surface followed by the transport into the electrode interior. The solvated D^+ ions are driven toward the negative electrode contact surface at the rate determined by the cell current. The electro–deposited deuterium is removed from the contact surface by gas evolution (the Heyrovsky – Horiuti and the Tafel paths) and by absorption. The adsorbed/absorbed deuterium is distributed as follows: (i) The subsurface, D_s is formed

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[43] Detailed analysis and methods of solution can be found in S. Szpak and C.J. Gabriel, J. Electrochem. Soc., **126**, 1914 (1979).

[44] P. van Rysselberghe, in*Modern Aspects of Electrochemistry*, vol, IV, Plenum Press 1966

just below the top–most layer of Pd atoms and provides a link between the chemisorbed surface atoms, D_a, and dissolved in metal, D_m, (ii) two energetically different D_a exist, (iii) with chemisorption there is associated surface reconstruction, but only D_s is responsible for its maintenance, (iv) there is an energy barrier separating D_a and D_s which affects transport in both directions. Chemisorbed D is responsible for surface reconstruction while absorbed D maintain state of reconstruction[45]

6.0 Quantitative approach to selected processes

Standard approach to examine any process is *via* the use of differential equations because they provide a procedure for the description of a process/reaction in terms of small increments that appear as derivatives, *i.e.* as the limits of the quotients of the increments of the variables that describe the process/reaction over the increment in time or space in which it takes place. But the differential equation does not completely describe a given problem. For a full description one needs to specify the initial and boundary conditions, *i.e.* to specify as many conditions as the number of arbitrary functions or constants involved in the integration process.

The events of interest take place within the confines of the interphase, i.e, within the very inhomogeneous thin layer are described by the partial differential equation containing four independent variables - three space coordinates and the time, *i.e.* of the form $\frac{\partial X}{\partial t} = A\nabla^2 X$, where A is a constant. Whether or not this equation can be used to describe events (*e.g.* transport) within the interphase depends on the physical meaning of the ∇^2 operator. Hopf[46] showed that the quantity $\nabla^2\phi$ is a measure of the difference of the scalar ϕ_0 taken at a point zero and its average value of ϕ_{ave} in the infinitesimal distance from it. Consequently, it is used when it is assumed that the process description deals with physical properties that are averaged over elements of volume and ignores the microscopic variations resulting from molecular structure.

6.1 Defects/interactions[47]

Defects in metals that affect hydrogen transport are: (i) point defects, (ii) solute defect complexes, (iii) dislocations, (iv) internal boundaries and (v) isolated metal clusters.
(i) The simplest point defect is the vacancy, *i.e.* an empty lattice space. A strong interaction (attraction) exists between the interstitial hydrogen and the open volume character of the defect. In other words, the interstitial hydrogen is driven into the vacancy by the presence of an open volume. The strength of binding energy is similar to that of chemisorbed hydrogen.
(ii) The interaction with the solute and solute–defect complexes is weaker than with vacancies. Here, the strength of interaction is influenced by elastic distortion and by electronic differences in H/D –bonding between the host and impurity atoms.
(iii) The open volume effects produced by the lattice strain are less than those generated by vacancies thus resulting in weaker interactions. As the H/D atom approaches the dislocation, the binding energy changes. Mobility of interstitial H/D species is re-

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[45] RJ Behm, J. Chem. Physics, **78**,7486 (1983)

[46] L. Hopf, *Introduction to Differential Equations of Physics*, Dover Publications, New York, 1948, p.63

[47] It is assumed that the D–interaction with defects differs from the H– interaction in intensity but not in kind

duced by attractive interaction with dislocations.
(iv) The interaction between internal boundaries and interstitial hydrogen are of special interest. In fully metallic boundaries, the binding energies are much less than for vacancy trapping. The smallness of interaction is related to the absence of open–volume defects on these boundaries. However, if the boundary contains a non–metallic phase which tends to form a covalent bond to the ^{n}H (n =,2,3) atom the situation is different; the binding energy is substantially higher. In addition, the boundary plays another role; it provides paths for accelerated diffusion, which is ascribed to a reduced vacancy formation in the excess volume of the boundary.
(v) The dislocation velocity is enhanced by the presence of absorbed hydrogen. The effect was observed (by transmission electron microscopy) for edge, screw and mixed dislocations.[48]

6.2 Forms of diffusion equation

Diffusion is normally described as a spontaneous and irreversible process of equalization of concentration. It was first observed by Bertholet in 1803 and, somewhat later in 1855, formulated by Fick. Since then, the diffusion has become the topic of interest in both science ad engineering, because the diffusion controlled transport can be used to represent basic ideas concerning a process or it can be a convenient tool to explore properties of a medium in which it occurs.

Transport of absorbed deuterium generated at the contact surface of the negatively polarized Pd–D_2O system is governed by the diffusion equation. Depending upon conditions of the transport path, different forms of the diffusion equation are used.

The quantitative analysis of the transport of species across the solid side part of the interphase must take in account their interaction with lattice defects. For this reason, motion of a particle, whether charged or not, is complicated by its interaction with Pd lattice defects as well as being affected by gradient of the stress field. Thus, the use of a simple form of the diffusion equation is inappropriate.

(i) Ideal system (non interacting particles)

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \tag{3}$$

This simple form of the diffusion equation was used to describe electrochemical charging of Pd rods. The results of the diffusion controlled modeling of the charging process of massive electrodes were in agreement with observation.[49]

(ii) Transport of interacting particles. Transport of absorbed deuterium is described by the diffusion equation of the Smoluchowski type

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} + \beta\frac{\partial[cf(x)]}{\partial x} \tag{4}$$

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[48] H. K. Birnbaum, E. Sirois and P. Sofronis, in Metal – Hydrogen Systems, Proc. Third International Symposium, Uppsala, June 1992 vol. II.

[49] S. Szpak, C.J. Gabriel, J.J. Smith and R.J. Nowak, J. Electroanal. Chem., **309**, 273 (1991)

where β is the mobility ($\beta = v/f$) and $f(x)$ is the force that either accelerates or slows the motion of absorbed deuterium.

The nature of the $f(x)$ function depends on the interaction between absorbed deuterium and the Pd lattice defects. In many cases to simplify the quantitative treatment and still reflect the reality, it is sufficient to consider f(x) = constant. In that case, the applicable equation is

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} + v\frac{\partial c}{\partial x}. \tag{5}$$

The term $v\frac{\partial c}{\partial x}$, by setting $\frac{\partial c}{\partial t} = 0$, yields information concerning distribution of diffusing particles and by virtue of $f = gradE$ also the effect of local field on transport.

(iii) *Diffusion with reaction (sink)*

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} + \beta\frac{\partial[cf(x)}{\partial x} - k(x)c \tag{6}$$

This form of the diffusion equation is used when the diffusing particles are immobilized by e.g. interaction with lattice defects.[50]

(iv) *Diffusion with rapid increase in concentration.* In general, models may, or may not, reflect the real situation. Rapid change in the chemical/electrochemical potential may affect the kinetics of the various processes and thus invalidate the usual treatment by solving the parabolic equation governing transport. In many practical instances, the diffusion flux $j = -D\partial c/\partial x$ may not accurately describe a real situation. Suppose that at some point within the diffusion field there is a sudden increase in the concentration of diffusing particles. Under these conditions, the correct expression for flux is $j + \tau\partial j/\partial t = -D\partial c/\partial x$ where τ is the relaxation time. Using the equality $\partial c/\partial t = -\partial j/\partial x$ (conservation of matter) the diffusion equation takes on the form

$$\frac{\partial c}{\partial t} + \tau\frac{\partial^2 c}{\partial t^2} = D\frac{\partial^2 c}{\partial x^2} \tag{7}$$

i.e. the diffusion process is described by hyperbolic rather than parabolic equation. This equation implies tat a strong discontinuity in concentration propagates with certain velocity (of the diffusion field) resulting in maximum transport.

7.0 Cell in external fields

The usual input variables of an operating electrochemical cell are pressure, temperature, concentration of reactants and cell current or voltage. Of the input variables, usually the effect of temperature and overpotential on the cell performance, is examined. In particular, the temperature, as expected, produces measurable results, *viz* as the temperature increases so does the cell thermal output (positive feed–back), *cf.* IV.4.0. A question that naturally arises: Do other input variables, such as external electro – or

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[50] A.M. Berezhkovskii *et.al.*, J. Chem. Physics, **109**, 4182 (1998)

magnetostatic field, affect the cell performance? Here, we examine the effect of external electro– and magneto–static fields on the behavior of the Pd/$^nH -^n H_2O$ (n=1,2) system during electrolysis.

To facilitate the identification of dominant processes, it is desirable to assemble background information on processes responsible for change in both surface morphology and the bulk structure. Those are: (i) thermodynamic considerations, (ii) field interaction with conductors, liquid dielectrics and (iii) the laws governing the bulk response to the surface forces.

7.1 Thermodynamic considerations

An increase in the energy of the system placed in an external electromagnetic field is equal to the energy flowing through the boundary surface of the volume less the work done by the field plus the reversible work of electrification and magnetization of regions that can be polarized (conservation of energy). The energy transferred from the field into molecules by stationary fields of less than 10^4 Vcm^{-1} or 0.1 T is small when compared with the energies of chemical bond. Consequently, from the thermodynamic point of view, an external field is regarded as a new variable and its effect is included in the $\Sigma_i l_i \delta L_i$ term of the infinitesimal change in the internal energy

$$\delta U = T\delta S + \Sigma_i l_i \delta L_i + \Sigma_j \mu_j \delta m_j \tag{8}$$

where $\Sigma l_i L_i = dW$ and where l_i and L_i are the work coefficients and work co–ordinates (other symbols have their usual meaning). In a homogeneous and isotropic medium, this term reads: $dW = -pdV + \mathbf{E}\delta(\mathbf{P}V)$ where the work coefficient is the electric field strength, **E**, and the conjugate work co–ordinate, $\mathbf{P}V$, is the product of the electric polarization, **P**, and volume V. For the system in a magnetic field, the corresponding term is: $dW = -pdV + \mathbf{H}(\mathbf{I}V))$ the work coefficient is the product of magnetic polarization, $\mathbf{I}V$ and the conjugate work coordinate is the magnetic field strength, **H** (for the electrified interface additional terms must be added). The thermodynamics of solid systems is analogous to that of liquid with six (6) quantities $V_0\sigma_i$[51] replacing volume V.

7.2 System in an external field.

The variety of morphologies resulting from the exposure to an external field, *cf.* Fig. 2, strongly suggests that they arise from the co–operative or competitive interaction between relevant processes and their driving forces. An operating cell is viewed as a system consisting of three subsystems, *viz* bulk Pd/D, the interphase and the electrolyte. The electrolyte, an ionic conductor is treated as a dielectric with added mobile charges, the interphase is an assembly of non–homogeneous layers and the bulk is a conductor. Just before the application of an external field all intensive state variables are constant in time. Consequently, to provide rational interpretation, one must (i) consider the interaction of the field with the system, and in particular, with its components: conductor (Pd/D film), liquid dielectric (electrolyte) as well as the relationship between the

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[51] the quantity $V_0\sigma_i$ is a product of reference volume and the strain component, the associated intensive parameter is the stress component), *i.e.* internal forces are associated with the strain just as the pressure is associated with the volume in liquid systems.

surface forces and bulk response, (ii) define the system and its conditions just before placement in an external field, and (iii) examine the effect of the field on the behavior of individual components of the cell.

(i) Electrostatic field, ψ_e and a conductor

The electrostatic field affects each individual component, *viz* electrolyte, interphase, in a different way. A conductor, when placed in an electric field: (i) reduces the field energy, (ii) an uncharged conductor, located outside of the field, is drawn toward the field, (iii) it cannot remain in stable equilibrium and (iv) a "negative force" acts on the surface causing appropriate response in the bulk phase[52]. A conductor placed in an external field does not suffer changes in thermodynamic quantities because electric field does not penetrate into its interior, *i.e.* changes, in its physico–chemical properties, are limited to the surface.

(ii) Electric field, ψ_e, and the electrolyte.

While a conductor, when placed in an electric field always suffers "negative" pressure, a dielectric may either expand or contract, *i.e.* it may experience either positive or negative pressure, depending on condition of an experiment. The situation is more complex for a liquid dielectric containing charges: the behavior of ions must be included. In particular, a single ion when in contact with water is solvated, *i.e.* the central ion is surrounded by an appositively charged ionic cloud. When subjected to an external electric field, it is accelerated with a net force arising from the applied field and opposed by the force associated with the structure of moving ion, *i.e.* central ion is accelerated in one direction while its ionic cloud in another, augmented by the force associated with relaxation of the ionic cloud.

(iii) Magnetic field, ψ_m, in general

In standard textbooks, one reads that the equations describing electric and magnetic fields are the same, *ergo* their action should be identical. This, however, is not true and generally: electric field affects the system's potential energy while magnetic field, its kinetic energy[53] – this is especially true for molecular assemblies.

The interaction of a magnetic field with electrochemical systems can be roughly divided into three main areas, *viz* (i) magneto–hydrodynamic effects, i. e. those affecting mass transport *via* the reduction of the diffusion layer thickness, (ii) magneto–dynamical effects, i.e, those involving shape change of micro–globules as well as macro–molecules, and (iii) non–specific interactions of electronic nature, *i.e.* those affecting the structure of the interphase, the electrokinetics and electro–catalysis. These effects are attributed to the action of forces generated by the gradients of magnetic energy density, namely the paramagnetic gradient force, $F_1 = (\chi/\mu\mu_0)B^2\nabla c$ and the gradient field force, $F_2 = (\chi c/\mu\mu_0)B\nabla B$, *i.e.* by forces arising from non–homogeneity

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[52] L.D. Landau and E.M. Lifshitz, Electrodynamics of Continuous Media, Pergamon Press, 1960, pp. 7, 31

[53] F. Noether, in P. Frank and R. von Mises, Die Differential und Integralgleichungen der Physik, vol. II, p. 724, Dover Publications, 1961

of the magnetic field. In an operating cell there is an additional force, the Lorentz force, $F_3 = q[E + v \times B]$.

Expected effects associated with the magnetic field are:
(i) When an electrode is exposed to magnetic field, there is shift in the rest potential if (a) material is ferromagnetic or (b) corrosion takes place – in the latter case, it is attributed to the action of Lorentz forces.
(ii) When the interphase is placed in an external magnetic field, the magnetic "order" of one layer affect the "order" of another being in contact with the first. This is of particular interest when the Pd/D film is in contact with Ni, a ferromagnetic substance.
(iii) When the "host/guest" complexes (where the Pd lattice defect acts as the host for the guest D_n^+, e^- complexes) are exposed to magnetic field, deformation of their structure is very likely to occur due to magnetic torque.

7.3 Shape change

Any solid undergoes shape change when the internal forces exceed the elastic limits. In general, three types of forces can be identified as acting during the deformation of a solid. These are (i) internal forces, *i.e.* forces that obey Newton' law, (ii) applied (or external) forces and (iii) capillary forces (forces that act between the internal and surface molecules, or between solid boundary and the molecules of surrounding liquid). By definition, when the surface forces are not uniformly distributed, they act as external forces. If, in fact, the latter are involved in producing shape changes, Fig. 2, then their action can be magnified by an external electrostatic field, *i.e.* by placing an operating cell in an electrostatic field.
The relationship between surface forces and the bulk response is given by the Gauss theorem

$$\int div\mathbf{A}dV = \oint A_n ds \tag{9}$$

In this equation, the left side term is the algebraic sum of all sources/sinks distributed over the whole volume – the right side defines the outflow, if positive and inflow, if negative. Furthermore, it indicates that forces acting on any finite body can be reduced to forces applied to the surface and *vice versa*. It follows that the shape change at constant volume is the result of motion due to forces acting on the surface. Consequently, the deformation is determined by the distribution of surface forces while the rate of deformation, by their magnitude.

Shape change affects transport of dislocations and boundary sliding. The intensity of these processes would strongly depend on the type of interaction existing between vacancy and dislocation. In general, vacancy may be instrumental in creating dislocations or may have an opposite effect. Consequently, it determines the distribution of deuterium present within the interphase.

SECTION III: CO–DEPOSITED CATHODES: PROCEDURE AND PROPERTIES

In all our work we employed cathodes prepared by co–deposition. We have chosen this type of cathodes to reduce the diffusion controlled charging time. As it turned out, this electrode structure is an ideal tool to explore the nature of the nuclear active state of the Pd/$^{n}H - H_2O$ (n=1,2,3) system. To illustrate the correctness of the choice that we have made, we structured this section as follows: First, we describe the behavior of massive electrodes and compare it with that of co–deposited electrodes. Next, we discuss the electrochemistry of the diffusion controlled co–deposition and conclude with the assessment of practicality.

1.0 Massive *vs* co–deposited cathodes

Experimental protocol devised by Fleischmann and Pons was as follows: electrolyze, under galvanostatic control, heavy water containing Li^{+} and OD^{-} ions in a cell in which massive palladium served as the cathode and platinum as an anode. Characteristic features associated with their experimental protocol are: (i) The electrode charging with the electrochemically generated deuterium is diffusion controlled, (ii) an undetermined time period, the incubation time, is needed to initiate the F–P effect. During the incubation time, atomic/molecular interactions between the D^{+} species and the Pd lattice and/or the Pd lattice defects occur, *i.e.* the "self–organization"[54] takes place; (iii) the reproducibility and intensity of the F–P effect depends on the metallurgy of massive palladium and the charging procedure.

1.1 Charging of massive electrodes

An early research indicated that successful production of excess enthalpy depends on, among other factors, the metallurgy of massive palladium and the rate of charging. Charging/discharging of the Pd/D electrode is a very complicated process. Hence, the development of a general model for the transport of absorbed deuterium is not a trivial

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[54] self–organization refers to a set of processes that occur during the incubation time. These processes are responsible for the change from an inert to active state needed for the initiation of the excess power, the F–P effect.

undertaking. In our work, we considered the simplest model, *i.e.* a model in which the flux is determined by the concentration gradient alone and where the relevant processes are: (i) $D_2O + e^- \rightarrow D(a) + OD^-$ with k_1, k_{-1} and η, (ii) $D_2O + D(a) + e^- \rightarrow D(s) + OD^-$ with k_2, k_{-2} and η and/or (iii) $D(a) + D(a) \rightarrow D_2(s)$ with k_3, k_{-3} where k_i and k_{-i} are the rate constants for the forward and reverse reactions and η is the operating overpotential. The absorption of D into the Pd lattice is given by (iv) $D(a) \rightarrow D(l)$ with k_4, k_{-4}

(i) Hydrogen absorption rate

This simple model states that the time rate of absorbed deuterium is governed by both the flux across the electrode/electrolyte contact surface and diffusion in the bulk. This, in fact, specifies the boundary conditions for numerical solution of transport equation which requires three types of input parameters, *viz* those associated with the electrode/electrolyte system, those characterizing the initial conditions and those that examine the effect of rate constants on charging process itself. The electrode charging and the effect of rate constants can be examined within well defined time periods of which the first time interval, $0 < t < \tau_1$ represents the charging of the double layer, the second $\tau_1 < t < \tau_2$ covers the period for the attainment of quasi–steady state and the third $t > \tau_2$ is the time in which the electrode begins to accept the electrochemically generated deuterium, Fig. 7, curve Q. It is noteworthy to indicate that during the time of the attainment of the quasi–steady state of the surface coverage, the amount of absorbed deuterium is insignificant – in all case examined it was on the order of 0.0001% above its equilibrium value.

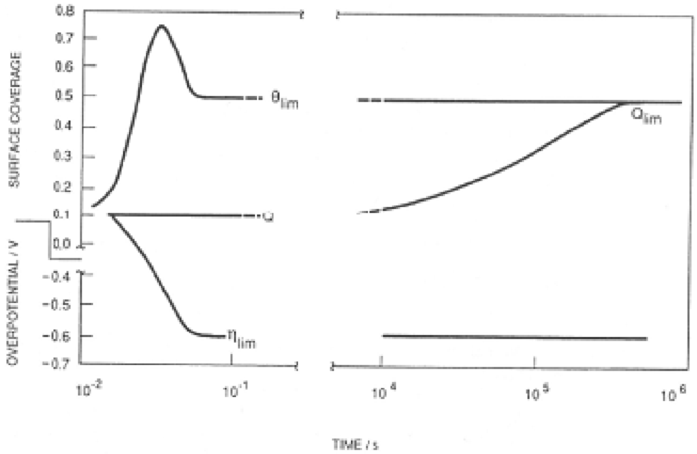

Figure 7:

At first sight this model ascribes a dominant role to the rate of absorption, *i.e.* the higher the rate, the faster the electrode charging. However, due to coupling between surface processes, no such conclusion should be drawn, unless the interphase controls the overall event. As a rule, an increase in the rate of absorption shifts the attainment of the quasi-steady state to somewhat longer times while the decrease in the absorption rate shows greater tendency to develop a maximum in the $\theta(t)$ curves. The effect of the choice of the rate constants and the charging current is discussed in great detail by Szpak et al[55].

(ii) Predictive capabilities

This simplified model predicts rather well the experimentally observed behavior of the charging of palladium rods.[56] The data published by Riley *et al*[57] show a number of characteristic features, among them: (i) the existence of apparent threshold value for the cathodic current density, *i.e.* a value above which the initial charging rate does not increase; (ii) slower rate for unloading than for loading which implies that the rate of transport across the interphase depends on direction – an unlikely situation for diffusion controlled transport; (iii) initial charging rate depends, for the most part, inversely on the radius of the Pd electrode and (iv) an unexpected dependence of the asymptotic electrode loading on cathodic current density, *i.e.* appearance of a maximum.

In a subsequent paper[58] we compared Riley's *et. al.* data with predictive capabilities of this simple model and found that, indeed, there is: (i) saturation of the initial charging rate; (ii) non–monotonic dependence of the asymptotic charging on current density (iii) asymmetry in loading and deloading. These features imply that: (i) the cell current density controls the mode of charging/discharging; (ii) directional transport across the interphase and (iii) the importance of geometrical considerations.

1.2 The search for simplicity and accuracy

To understand the causes of the observed poor reproducibility, all of the system's variables, that might affect it, must be considered. Fleischmann *et. al.* in their paper *"Calorimetry of the Pd–D_2O system: The search for simplicity and accuracy"*[59] formulated a general equation governing thermal behavior of this system. It can be expressed as a function of a set of time/space dependent variables and their differentials,

$$Q_f = f_0 \times f_1\ (\psi_1, \psi_2, \psi_3). \tag{10}$$

Equation (10) states that the thermal behavior of the Pd/D_2O system can be adequately represented by the product of two sets of parameters: One dealing with the metallurgical aspects of the cathode material and electrolyte composition, f_0, and the other containing variables characterizing the processes/reactions associated with the cell operation, $f_1(\psi_1, \psi_2, \psi_3)$. The first set, $\psi_1(E, dE, T, dT)$, contains the system variables, the second set, $\psi_2(\theta, d\theta, \eta, d\eta, \phi, d\phi)$, the surface (interphase) variables and the third, $\psi_3[X, dX, (\beta \to \gamma), d(\beta \to \gamma)]$, – the bulk variables. Here, E_c – cell voltage, η –cathodic overpotential (in particular its Volta component on the solution side), θ– surface coverage, X – deuterium content (expressed as the D/Pd atomic ratio), and the

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[55] S. Szpak, C.J. Gabriel, J.J. Smith and R.J. Nowak, J. Electroanal. Chem., **309**, 293 (1991)

[56] S. Szpak, P.A. Mosier–Boss, C.J. Gabriel and J.J. Smith, J. Electroanal. Chem., **365**, 275 (1994).

[57] A.M. Riley, J.D Seader, D.W. Pershing and C. Wallling, J. Electrochem. Soc., **139**, 1342 (1992)

[58] S. Szpak, P.A. Mosier–Boss, C.J. Gabriel and J.J. Smith, J. Electroanal. Chem., **365**, 275 (1994)

[59] ICCF–4

formation of the $\gamma-$ phase denoted by $(\beta \rightarrow \gamma)$. A word of caution, these variables are not independent of each other and, consequently, there is a high probability of the development of instabilities and oscillations.[60] Evidently, in order to specify conditions of reproducibility, a large number of experiments would have to be performed which, in turn, requires commitment in personnel and resources.

It became obvious, *cf.* Eq. (10), that (i) the statistical design of experiments (high/low variables) is not the preferred way to proceed and (ii) that a new approach in the manufacturing of cathodes is required. Thus, it was necessary to review the facts about the Pd/D–D_2O system. Clearly, (i) for a successful experiment high D/Pd atomic ratios are required (preferably D/Pd = 1 or higher), (ii) electrochemical charging, being diffusion controlled, takes several days to obtain the desired D/Pd ratio (depending on the electrode geometry and size as well as mode of charging), (iii) there are even longer "incubation" times, *i.e.* times during which the needed "structure" is formed, and (iv) poor reproducibility that may arise from difficult metallurgical problems.

2.0 Co–deposition

In the course of search for more reproducible and effective cathode materials three avenues were considered: (i) palladium alloys, (ii) co-deposition and (iii) other geometrical forms, *e.g.* thin films. Relevant questions with respect to (i): Would the addition of an alloying element change the solubility of deuterium (hydrogen) in palladium? With respect to (ii) and (iii): Is the electrode structure (morphology) an important factor? Since the electrode morphology is important, the electrodes prepared by co–deposition were selected for further study.

The co–deposition is a process whereby the palladium is co–deposited from a Pd^{2+} salt solution onto a substrate that does not absorb deuterium (such as Au, Cu). The applied current or potential is so adjusted as to deposit palladium in the presence of evolving deuterium. Consequently, charging by diffusion is eliminated, the incubation time is non–existing since the "self–organization" is an integral part of the co–deposition. The structure of the electro–deposited palladium is controlled by the solution composition and the cell current profile.

In the early stages of the investigation of the F–P effect we decided to use cathodes prepared by co–deposition. In the first publication[61] (submitted in November 1990, published in 1991) we wrote: *This note reports on an alternative experimental approach to produce conditions favorable to the observation of this extraordinary behavior by exploiting the Pd/D co–deposition. This approach, because an ever expanding electrode surface is created, assures the existence of non–steady state conditions as well as simplifies the cell geometry by eliminating the need for uniform current distribution and, more importantly, eliminates long charging times effectively.*

2.1 Electrochemistry of co–deposition

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[60] In this formulation, for a given experimental run, the first set, f_0 can be viewed as being constant. This assumption is probably valid for the reduction of D_2O, but not in acid solutions where the D^+ ions affect the chemical potential.

[61] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem., **302**, 255 (1991)

The very brief discussion of the charge transfer kinetics presented here covers (i) the Butler–Volmer and van Rysselberghe approach, (ii) the special case of diffusion controlled co–deposition and (iii) the art of co–deposition.

2.2 Charge transfer kinetics

It is customary to use the Butler–Volmer equation

$$j = j_0 e^{-\alpha F \eta / RT} \tag{11}$$

to examine kinetics of the charge transfer reaction. In this equation, j_0 is the exchange current density and η is the overpotential. Applying this equation to the reduction of heavy water ($D_2O + e^- \rightarrow D + OD^-$) with electrons flowing to the working electrode (cathode) from the power supply, we have

$$j/F = k_{-1} c_{OD^-} \theta e^{\alpha F \eta / RT} - k_1 c_{D_2O} (1-\theta) e^{-(1-\alpha) F \eta / RT} \tag{12}$$

with

$$j_0/F = k_{-1} c_{OD^-} \theta e^{\alpha F \Phi / RT} = k_1 C_{D_2O} (1-\theta) e^{-(1-\alpha) F \Phi / RT} \tag{13}$$

where Φ is the Nernst potential, k_{-1} and k_1 are the rate constants for the cathodic and anodic reactions. Here, j_0 is the exchange current density (equality of forward and reverse reactions at equilibrium).

The correct use of the Butler–Volmer equation requires (i) specification of the planes between which the overpotential is measured, (ii) concentrations that are used (interfacial or bulk concentrations) for the forward and reverse reactions and (iii) understanding of the physical significance of the α factor. An extensive discussion of the charge transfer kinetics can be found in Bockris and Reddy treatise.[62]

An alternate approach to the kinetics of charge transfer reaction, based on the thermodynamic structure of electrochemistry, was developed by van Rysselberghe. The essential difference between these two approaches is not in the form of appropriate rate equations but in the identification of respective driving forces, *e.g.* overpotential *vs* reaction affinity and exchange current density *vs* reaction exchange velocity. In particular, kinetics of charge transfer reaction following the conventional Butler – Volmer formulation differs from that based on the thermodynamic structure of electrochemistry, in that: the first, is based on the *a priori* introduction of a model of mechanistic and/or molecular nature while the second, uses thermodynamic reasoning. Which approach to follow depends on the problem to be examined and the preference of an investigator. A detailed discussion of this subject can be found in publications of van Rysselberhe[63] augmented by reading the *Chemical Thermodynamics* by R. Defay and I. Prigogine[64].

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[62] J. O'M. Bockris and A. K. .N. Reddy, Modern Electrochemistry, Plenum Press, New York, 1974

[63] cf. Chapter I in Modern Aspects of Electrochemistry, J. O'M. Bockris, ed Vol. 6, Plenum Press, New York, 1966

[64] I. Prigogine and R. Defay, Chemical Thermodynamics, Longmans Green and Co, London – New York – Toronto 1954

Incidentally, the kinetics of deuterium evolution is of no special interest when the Pd+D co–deposited electrodes are employed. It is of interest, however, when massive electrodes are used because it determines the conditions for their effective charging.[65]

2.3 The art of Pd+D co–deposition

The Pd+D co–deposition may be viewed as a special case of production of alloys by electrochemical processing – it involves simultaneous deposition of more than one component. This is done by the reduction of ions present in the electrolyte[66]. Briefly, for the production of Pd/D alloy the relevant reactions are: $Pd^{2+} + 2e^- \rightarrow Pd$ and $D_2O + e^- \rightarrow D + OD^-$. In practice, however, these reactions depend on the electrolyte composition. For the co–deposition from a solution containing $PdCl_4^{2-}$ complexes, Naohara[67] found that the reduction of Pd^{2+} proceeds *via* the reduction of an adsorbed $PdCl_4^{2-}$ complex resulting in a layer–by–layer growth of the Pd film. However, the orderly growth of deposited palladium is disturbed by the adsorbed deuterium generated by the reduction of heavy water. Ohmori et al[68], using a scanning tunneling microscope, proposed a model where the H^+ ions are adsorbed and reduced at the surface. A part of the adsorbed hydrogen enters the Pd lattice and accumulates around lattice defects. Through this process, the surface is transformed into a modular–like structure.

The structure, and therefore the performance characteristics, of the co–deposited electrodes depends on the way the were prepared. To obtain the desired structure it is convenient to use Fig. 8 as a guide.

Figure 8a illustrates the procedure when (i) reduction of $PdCl_4^{2-}$ and D_2O are independent of each other, (ii) electrolyte volume and electrode surface remain constant, (iii) reduction of Pd ions is diffusion controlled[69] and (iv) no other charge transfer reactions occur. Under these conditions the electrode rest potential, $\Phi(0)$, is determined by solution composition. At t = 0, a constant cell current, I_1, is applied. In practice it is much smaller than the limiting current, $I_1 << j_{1,lim}$.ote that in Fig. 8 j* denotes the limiting current. This is done to assure an adherent Pd deposit. With the passage of time, the Pd^{2+} ions are depleted, the electrode potential, driven by the cell current, becomes more negative. At $t = t_1$ the cell current is increased to I_2, *i.e.* to a value very close to the limiting current. This is done to assure a long co–deposition period. When the applied current density, I_2, by reducing the concentration of Pd^{2+} ions, becomes the limiting current (for that concentration), the electrode potential is driven into the region of heavy water instability and at $t = t_2$, the reduction of D^+ ions commences[70] and the co–deposition begins. If the co–deposition is galvanostaticaly controlled and if the cell current exceeds the limiting current, then, at constant volume and surface

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[65] S. Szpak, C. J. Gabriel, J.J. Smith and R.J. Nowak, J. Electroanal. Chem., **309**, 273 (1991)

[66] A detailed discussion can be found in J. O'M. Bockris and A. K. N. Reddy, Modern Electrochemistry, Plenum Press, 1974, Vol. II, p. 1223

[67] H. Naohara, S. Ye and K. Uosaki, J. Phys. Chem B **102**, 4366 (1998)

[68] T. Ohmori, K. Sohamaki, K. Hashimoto and A. Fujishima, Chemistry Letters, 1991, p. 93, The Chemical Society of Japan

[69] The diffusion limited current density is estimated by substituting δ = 0.05 cm which yields $j^* = j_{lim} = 0.025$ zc Acm^{-2}, where z is the number of positive charges and c in g–ion/l.

[70] The reduction of heavy water is controlled by the overpotential and has the form of $j = j_0 \exp(\alpha\eta F/RT)$.

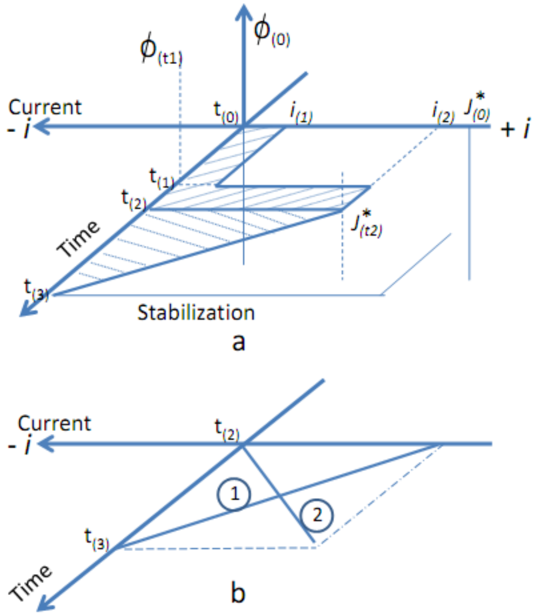

Figure 8:

area, the Pd^{2+} ions concentration decreases linearly with time. The co–deposition is completed at $t = t_3$.

During the co–deposition period the $I = j_{1,lim} + j_2$ and the j_2 component also changes linearly with time resulting in the time dependent composition of the deposit as displayed in the I/t plane, the line 1 for palladium and the line 2 for deuterium. The desired D/Pd atomic ratio can be obtained inspecting Fig. 8b.

In our publications we described the co–deposition as simultaneous reduction of Pd^{2+} and D^+ ions. Such description does not identify the many problems that non–chemists may face. It is well known that electrodeposition of metals in the presence of evolving hydrogen yields poorly adhering powdery deposits. Reiterating, inspection of Figs. 8a and 8b suggests that the first step is to select electrolyte composition[71]. To assure good

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[71] Electrolyte composition: 0.03 M $PdCl_2$ + 0.3 M LiCl in D_2O was employed in all experimental work. Other formulations can be used provided that the Pd+D co–deposition occurs and that the mechanical integrity of the deposit is maintained. Incidentally, chemical properties of D_2O are identical with those of H_2O. The difference, more pronounced at lower temperatures, is in the magnitude of equilibrium constants and reaction velocities. In general, the D_2O reactions are slower, solubility of salts is less and ionic mobility is about 1.5 times slower than in H_2O. In a mixture: light/heavy water reaction: $H_2O + D_2O \rightarrow 2HDO$ + 0.03 kcal, occurs. Other useful data: (i) ionic product $[D_3O^+]\,[OD^-] = 1.6 \times 10^{-15}$ (*i.e.* about eight times less than ionic product of light water, (ii) maximum density at 11.6 C, (iii) freezing point at 3.82 C, boiling

bonding of the electrodeposited palladium, one starts with the current density that is a small fraction of the limiting current for the selected electrolyte composition. When the Pd deposit becomes *ca* 10^{-5} cm thick, the current density is increased to speed up the co–deposition process, *cf.* V.3.3.1 for the procedure used in this laboratory.

3.0 Co–deposition: advantages vs disadvantages

The suitability of the electrode material is judged by its advantages as well as disadvantages. The first and foremost is the immediate initiation of the excess enthalpy generation and the 100% reproducibility. Other advantages are (i) excess enthalpy generation as well as its rate exceeds those observed on massive or thin film electrodes, (ii) excess enthalpy is produced also at low current densities, *e.g.* 6 – 10 mAcm^{-2}, and (iii) greater flexibility in fabricating working electrodes, *e.g.* fabrication of cell elements based on fluidized bed concepts.

The principal drawback is due to a "cauliflower–like" structure of the co–deposited films. Such structures are mechanically weak – they can be damaged by a vigorous gas evolution when high cell currents are used. Experience shows that the damage is insignificant at current densities as high as 400 mAcm^{-2}. Thus, the answer is yes because it assures not only a reproducible excess enthalpy production but also exhibits other characteristic features such as positive feed–back as well as heat–after–death.

In spite of these characteristics there was reluctance in using co–deposited cathodes before answering two questions: First, are the cathodes prepared by co–deposition equivalent (in performance) to those originally proposed by Fleischmann and Pons? Second, would they aid or make it more difficult to understand the nature of the phenomenon of excess enthalpy production? To illustrate the equivalence it is useful to start with examination of the thermal behavior of cells. In both cases, excess enthalpy is generated upon electrolysis of heavy water; in both cases a positive feed–back and "life-after–death" [72] was observed.

The problem of whether the interpretation and inquiry into the nature of the Fleischmann–Pons effect be more difficult in co–deposited cathodes is examined *via* the electrochemical charging process. To understand the difference, we considered a simple model in which the transport is controlled by diffusion and the boundary conditions are determined by the kinetics of the charge transfer reactions coupled with the adsorption/desorption exchange processes. In massive electrodes the boundary conditions usually assumed are: the electrode surface is homogeneous with respect to adsorbed deuterium which is in equilibrium with absorbed deuterium. In real systems the situation is quite different, *viz* one cannot assume homogeneity with respect to transport. In reality, the distribution of both, the charge transfer current density and the solution composition are not uniform.

The question whether or not it obscures the understanding of the nature of the F–P ef-

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point at 101.42 C, (iv) potential difference: H/H$^+$ and D/D$^+$ is 0.003 V. Production costs: to obtain 10 mL of 98 % D_2O 1000 L of water is to be electrolyzed; for the separation factor 5 and operating at 3.6 V, 1000 kWh is needed.

[72] term used to indicate thermal activities at zero cell current

fect – the answer is “probably not”. The absence of the incubation time, the elimination of diffusion controlled charging and the 100% reproducibility, eliminates the need for the “statistical experiment design” and thereby excluding a great number of factors that appear to be essential in interpretation. Furthermore,the structure of the co–deposited Pd+D films is controlled by the rate of co–deposition and solution composition while the production of controlled massive palladium is much more difficult.

SECTION IV: THE Pd–^{n}H–H$_2$O SYSTEM; THERMAL EFFECTS

The enormously large quantities of excess enthalpy generation in the Pd/D–D$_2$ system, the F–P effect, raised questions concerning its origin. Specifically, questions that were posed, and needed answer, were: (i) Is the enthalpy generation of nuclear origin? (ii) Where is the heat source located – at the surface, in the sub–surface region, in the bulk? (iii) Is the heat source uniform throughout the whole electrode volume or distributed, *e.g.* at discrete sites? (iv) Is the excess enthalpy generation reproducible?

The discussion of thermal effects observed in polarized Pd/D–D$_2$O system, presented here, is structured as follows: (i) the development of calorimetric equation for cells with adiabatic walls, (ii) excess enthalpy generation, (iii) thermal run–a–way, (iv) fluidized bed design and concludes with (v) the "recombination" theory. It covers the work done in the SPAWAR Laboratory with limited discussion of thermal effects observed in the Dewar–type calorimeters.

1.0 Early observations

The working electrode employed by Fleischmann and Pons was massive palladium rod or wire. In the search for more effective cathode materials, we selected cathodes prepared by the co–deposition. With this approach a new set of questions has arisen that needed an answer, *viz*: (i) Is the co–deposited film a suitable cathode material? (ii) If yes, then what is the magnitude of excess enthalpy as compared to that reported for "massive" electrodes? (iii) Are the characteristic features (e.g. positive feedback, heat–after–death) preserved? (iv) Is the excess enthalpy generated uniformly through the electrode volume or is it produced at discrete sites? To provide an answer to (i), the following procedure was devised:[73] A copper–constantin thermocouple, shielded by a glass tubing, was cemented to a copper foil. A second, identical thermocouple was immersed in the electrolyte. If, during the co–deposition at cell currents assuring a visible gas evolution an excess enthalpy is generated, then the temperature recorded by the thermocouple cemented to the copper foil should exceed that immersed in the electrolyte. Indeed, this was the case in all runs, with the temperature difference being

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[73] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem., **302**, 255 (1991)

2–4 C. Answers to (ii) – (iv) are yes[74]

2.0 Electrochemical calorimetry – general remarks

Standard procedure for measuring thermal effects is to use calorimetry. Calorimetry of electrochemical cells – a subject neglected prior to March 1989 – is based on conservation laws. Its correct use requires the knowledge of all processes involved, the sequence of events, the construction of an apparatus and the experimental procedure employed. That is to say, the development of the calorimetric equation of an operating electrochemical cell employs conservation of energy and adjusts the applicable walls and constraints in the manner that they are consistent with the cell design and the relevant experimental procedure. Consequently, a successful calorimetry must consider not only heat generation rate but also possible heat transfer paths, *i.e.* the establishment of an accurate model of an experiment is important. For this reason, a brief description of the relevant features involving modeling of electrochemical cells is given.

2.1 Model of an electrochemical cell in an outline

An electrochemical cell, used in the study of the F– P effect, is an example of the driven or substance producing cell in which energy is generated by nuclear processes. The passage of cell current, I, results in D_2 gas evolution at the negative electrode (cathode) and a gaseous O_2 at the positive electrode (anode). The respective charge transfer processes are:
at the cathode: $2\ D_2O + 2\ e^- \rightarrow D_2(g) + 2\ OD^-$
at the anode: $2D_2O \rightarrow O_2(g) + 4\ D^+ + 4e^-$
and within the electrolyte: $D^+ + OD^- \rightarrow D_2O$.
The evolving gases saturate the electrolyte and the escaping gas bubbles, that are traveling through the electrolyte, contain water vapor in the amount depending on local conditions (ambient pressure, temperature, etc.).

From the thermodynamic point of view an electrochemical cell is an open system consisting of subsystems representing the cell working elements, e.g. an electrolyte, positive and negative electrodes, and auxiliary items such as thermistors, calibrated resistive heaters, etc. In its simplest arrangement, an electrochemical cell is a three phase, multi–component assembly wherein the charge transfer reactions and the associated transport processes occur.

The subsystems and the associated processes are:
(i) the negative electrode (subsystem 1) acts as an electron source. The charge transfer reaction is the reduction of D_2O.
(ii) the electrolyte (subsystem 2) where the recombination $D^+ + OD^- \rightarrow D_2O$ occurs
(iii) the positive electrode (subsystem 3) acts as an electron sink. The associated charge transfer reaction is the oxidation of D_2O
(iv) the head space (subsystem 4) contains the vapor phase in contact with an electrolyte. It is assumed that at all times, it contains only D_2O, D_2 and O_2 molecules.
(v) The operating cell is immersed in a bath (subsystem b) which is in contact with

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[74] S. Szpak, P.A. Mosier–Boss, M.H. Miles and M. Fleischmann, Thermochim. Acta, **410**, 101 (2004)

environment.

Initially, the system is in thermal, mechanical and chemical equilibrium. Upon initiation of the current flow, significant changes in temperature and concentration take place, *viz* the development of gradients which, in turn, initiates transport processes in the electrolyte phase as well as across the electrolyte/gas interface. The increase in temperature arises from irreversible processes: the Joule heating, the electrodic processes and the exothermic absorption of deuterium by the palladium electrode as well as due to the heat exchange with the environment. In the absence of the electrolyte stirring, temperature and concentration gradients promote the development of convective flow. If the convective flow is sufficiently intensive, then it results is uniform composition and temperature throughout the electrolyte phase.

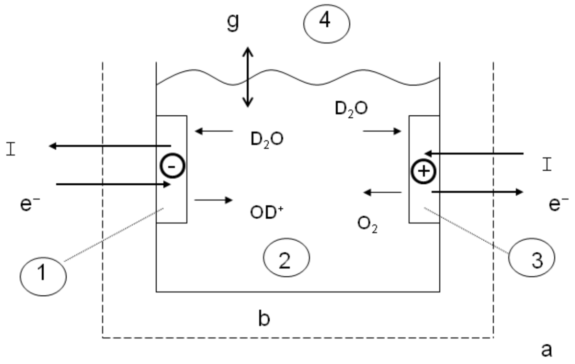

Figure 9:

During the cell operation there is an exchange of enthalpies between the cell elements and the electrolyte as well as between the electrolyte and other items such as walls, thermistors etc. Other condition that must be considered (to a lesser degree) is the magnitude of the cell current since it could have an effect on the current efficiency and the expression for the heat transfer coefficient. It is noted that some of the fluxes may have to be modified due to the particular experimental protocol as well as calorimeter construction.

2.2 Development of calorimetric equation

Equilibrium state in any system is completely characterized by the internal energy, U, the volume, V, the temperature, T, and the mole number, n_j, of chemical components, U = U(T,V,n_j). If constant pressure processes are examined, then the use of the enthalpy function is more convenient. In the enthalpy representation, the pressure is substituted for volume, H(T,p,n_j)= U + pV. Both quantities, U and H, are unambiguously defined in closed and open systems because the changes in state variables are independent of whether the change in composition, dn_j, is due to an exchange or chemical reaction.

A statement of energy conservation in open systems involves the following steps: Within the time period, dt, the infinitesimal change in its internal energy is

$$dU = dQ - dW - pdV - \Sigma f_k p \frac{\partial V^{(a)}}{\partial n} dt - \Sigma f_k \frac{\partial U^{(a)}}{\partial n} dt \tag{14}$$

where dQ is the heat transferred into the system, dW is the work done by the system on surroundings exclusive of the expansion work, pdV, the first sum is the work done on the surroundings by the removal of material and finally, the last term is the energy carried out by the removal of material. The first two terms on the right hand side are the non–exact differentials since neither Q nor W are state variables. The f_k indicates the flow (flux) outward, if positive, of the k–th component.

An equivalent equation in the enthalpy representation is derived by noting that the last two terms can be combined to read $\Sigma f_k (p \frac{\partial V^{(a)}}{\partial n_k} dt + \Sigma \frac{\partial U^{(a)}}{\partial n_k} dt)$ so that

$$dH = dQ - dW - \Sigma f_k \frac{\partial H^{(a)}}{\partial n_k} dt \tag{15}$$

The calorimetric equation is the time rate of temperature change of the system

$$mc_p \frac{dT}{dt} = \frac{dQ}{dt} - \frac{dW}{dt} - \Sigma f_k (\frac{\partial H^{(a)}}{\partial n} + \frac{\partial H}{\partial n} \frac{dn}{dt}) \tag{16}$$

The term dW/dt contains only electrical work done and ignores other mechanical work, the pdV work having been included in the derivation of equations. The enthalpy flux terms $\Sigma f_k \frac{\partial H}{\partial n}$ for charged species is assumed to take place at constant potential since all the electrical work is included in the dW/dt term. The heat transfer between systems at different temperatures is accounted for by the temperature dependence of the molar enthalpies $\partial H^a / \partial n_k$ and $\partial H / \partial n_k$ and should not be introduced separately. Moreover, in operating electrochemical cells, the work is always positive because the term IE_c must be positive irrespective of whether the cell operates in the driving or driven mode. Equation (16) may be modified to reflect cell design and experimental protocol and often written as

$$C \frac{dT}{dt} = Q - \Sigma_k J_k \tag{17}$$

where $C (= mc_p)$ is the heat capacity of the electrolyte, $Q (= Q^* + I[E_c - E_{th}])$ is the rate of change of the heat content and $J_k (= k \Delta f)$ is the flux in and out calculated using the generalized Ohm's law.

The initial conditions needed for solving the calorimetric equation are: uniform temperature throughout the system, zero cell current and vapor composition containing O_2, D_2 and D_2O molecules only.

3.0 Basic cell/calorimeter designs

Two open cell/calorimeters designs employing co–deposited cathodes were used in examining thermal behavior of the Pd/D–D_2O system, *viz* the Dewar type and cells with an adiabatic enclosure. The discussion of performance characteristics of the Dewar type cell/calorimeter can be found in Technical Report SSC/SD – TR – 1862, vol. 2.

3.1 Cells with adiabatic enclosure

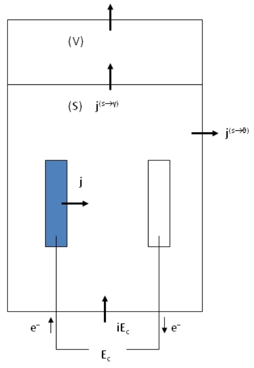

Figure 10:

To develop the calorimetric equation for the cell/calorimeter with adiabatic enclosure we consider a system consisting of an electrochemical cell containing a known amount of electrolyte and totally immersed in a water bath, Fig. 10. Initially, this system is in equilibrium and, for the duration of an experiment, the bath is in contact with an infinite heat sink ($T^{(e)}$ = const.). Applying the conservation of energy in the condensed form, the time rate of the temperature change in the cell, after its activation, is

$$C_1 \frac{dT_1}{dt} = Q_1 - J_q^{(1\to2)} - J_q^{(1\to e)} \tag{18}$$

and in the water bath

$$C_2 \frac{dT_2}{dt} = Q_2 + J_q^{(1\to2)} - J_q^{(2\to e)} \tag{19}$$

where $C_1 = \Sigma c_{1,i}$ is the heat capacity of the electrolyte and includes all cell components (cell constant), C_2 is the heat capacity of the bath fluid, $Q_1 = [J^* + I(E_c - E_{th})]$ denotes the rate of heat production in the electrolyte phase, Q_2 is the heat supplied to the bath in order to maintain $T^{(1)} = T^{(2)}$, and the J's are the heat fluxes exchanged between systems elements, *viz* the electrolyte, bath and environment.

Equation (18) is the energy balance expressed in terms of heat generation due to the irreversibility of the charge transfer processes, the rate of heat exchange between the cell and the water bath, and the rate of heat loss to the environment. Since the term Q_1 is always positive, it follows that $T^{(1)} > T^{(2)}$ resulting in an outflow of the heat generated within the cell.

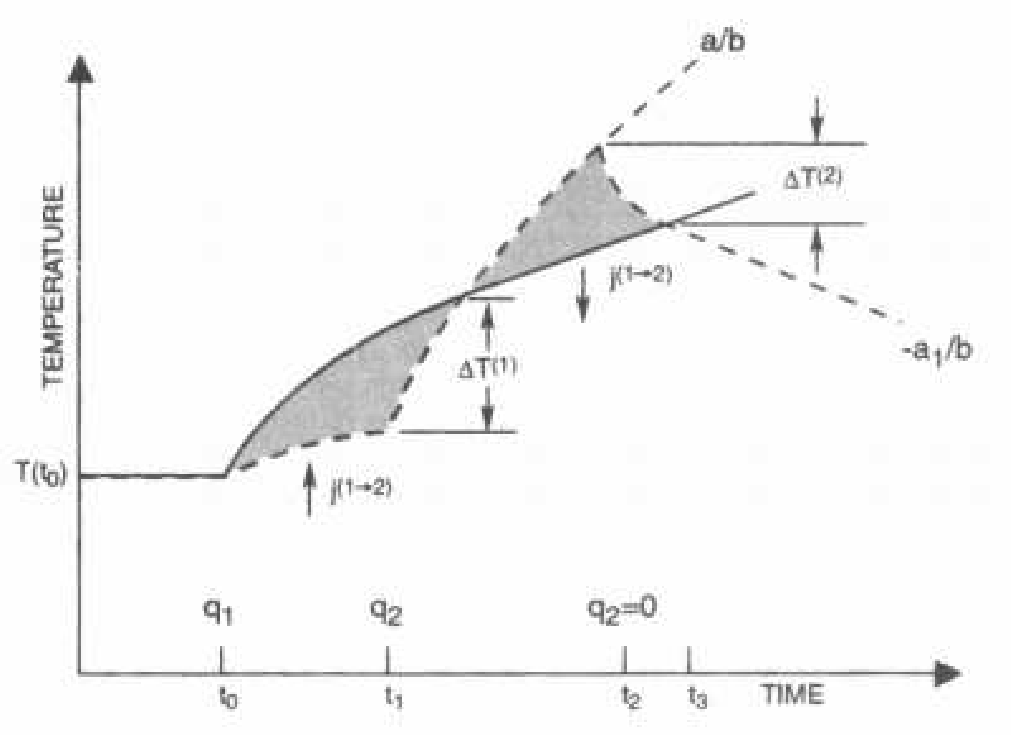

Figure 11:

The construction of an adiabatic wall requires that $J_q^{(1\to2)} = J_q^{(2\to1)}$ at all times. In practice, $\Delta T \neq 0$; consequently, the maintenance of an adiabatic wall requires that $\int J_q^{(1\to2)} dt = \int J_q^{(2\to1)} dt$, *i.e.* that ΔT oscillates about its zero value. To maintain an adiabatic wall, the positive ΔT due to $q_1 > 0$ must be countered by q_2 to reverse the direction of heat flow. One way to construct and maintain an adiabatic wall is as follows: At $t < t_0$, the system is in an equilibrium. At t_0 the flow of the cell current is initiated causing the system's temperature to rise as shown schematically in Fig. 11, by solid $T^{(1)}$ and dashed $T^{(2)}$ lines. At t_1, the difference ΔT reaches an *a priori* specified value, and the heat source q_2 in the bath is activated. If $q_1 > q_2$, then the temperature $T^{(2)}$ raises faster than $T^{(1)}$, in time at t_2 reducing ΔT to zero. To maintain an adiabatic wall, it is necessary to transfer the same amount of heat in both directions.

4.0 Thermal behavior

In the introductory remarks a number of questions was raised, *viz* (i) Is the excess enthalpy generation of nuclear origin as initially suggested by Fleischmann and Pons and (ii) where are the heat sources located. The answer to the first question, based on the emission of electromagnetic radiation, neutrons and charged particles (*cf.* V.3.0 and V.4.0), is yes. The answer to the second question cannot be given with any degree of assurance, *i.e.* putting it differently, there is no known experimental procedure that could provide an unambiguous answer[75]. The remaining questions can be answered and the answers constitute the bulk of this section.

4.1 Excess enthalpy generation

Thermal behavior of cells employing cathodes prepared by co–deposition is not different from that observed in cells using massive Pd cathodes[76]. Whether or not a particular cell generates excess enthalpy is determined by mass and energy balance. Two examples of an excess enthalpy generation are presented: (a) in a Dewar–type cell and (b) in a cell with adiabatic walls.

4.1.1 Case a – Dewar type cell[77]

The evaluation of the thermal behavior is based on the examination of the $E(t)$ and $T(t)$ data in response to the input enthalpies $I[E_c - E_{th}]$ and $Q(t)$. Figure 12a shows the $E(t)$ and $T(t)$ data due to the enthalpy inputs, $I[E_c - E_{th}]$ at times indicated by arrows facing upwards and by resistive heater with $Q = 0.2500$ W by arrows facing down. Briefly, three distinctly different time periods can be identified.

The first period includes the co–deposition at 0.006 A, and charging at 0.1 increased to 0.2A and followed by a stepwise reduction to 0.05 and 0.02 A. The second time interval is the period of low rate charging for ca 72 hrs. The third time interval is characterized by drastic changes in cell operation – the cell current was increased/decreased in larger steps and, in addition, resistive heater was engaged at times indicated.

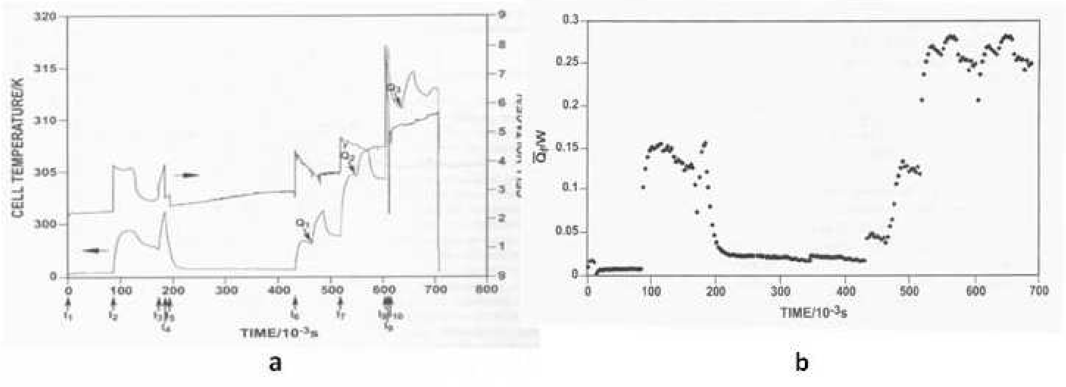

Figure 12:

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[75] Experimental evidence points to the first few atomic layers, *i.e.* within the metal side of the interphase region as the most likely seat of the heat generating reactions

[76] Of special interest, however, is the excess enthalpy generation during the co–deposition process, *i.e.* at low current densities. If, in fact, an excess enthalpy is generated during co–deposition, then that would have a profound influence on the understanding of its origin. Indeed, making reasonable kinetic and thermodynamic assumptions, one could conclude that an excess enthalpy production during co–deposition might occur.

[77] S. Szpak, P.A. Mosier–Boss, M.H. Miles and M. Fleischmann, Thermochim. Acta, **410**, 101 (2004)

The $T(t)$ data are normal, in the sense that an increase in the enthalpy input causes an increase in the cell temperature and *vice versa*. However, the examination of the $E(t)$ data reveals that there exists time periods when the cell temperature increases with the decrease in the enthalpy inputs. As in the case of cells employing conventional electrodes, in this instance energy conservation requires the presence of additional (unidentified) heat sources.

Figure 12b shows the excess enthalpy generation for a typical run. The data show that the excess enthalpy is somewhat higher than that observed on massive electrodes. An excess of *ca* 40 –50% at cell temperatures of 30 – 40 C, is not unusual. As a rule, the efficiency increases with an increase in temperature. Although, at this time, we do not have data, we have evidence of an increased activity as the temperature approaches the boiling point.

One interesting and potentially significant observation is the excess enthalpy generation at low cell currents. As illustrated in Fig. 12b, at current densities of 6 mAcm^{-2} there is generation of excess enthalpy. At the present time, we cannot state whether or not the observed excess heat is due solely to exothermic absorption because not much is known about the current efficiencies of the various operating reaction paths.

4.1.2 Case b – Cell with adiabatic wall[78]

The applicable equation is

$$mc_p\frac{dT}{dt} = I(E_c - E_{th}) + Q(t). \tag{20}$$

This equation represents the enthalpy balance for the case where the only reaction is the deuterium evolution. Thus, it applies to “massive” electrodes when a complete saturation with deuterium has been achieved, *i.e.* when the enthalpy of the evolved deuterium and oxygen can be recovered by the recombination reaction. This applies also to electrodes prepared by the co–deposition technique after the completion of the co–deposition.

An example of an excess enthalpy generation during the co–deposition as well as the electrolysis of D_2O taking place in a cell with an adiabatic enclosure is shown in Fig. 13.

The excess enthalpy produced/consumed during the time interval Δt, *i.e.* the term $Q(t)$ can be evaluated from the plot $0.239 mc_p \Delta T - \int_{\Delta t} I[E_c - E_{th}]dt$ vs $\int_{\Delta t} IE_c dt$. A plot for a typical run is shown in Fig. 13. Evidently, at first, $Q(t)$ is negative and becomes less negative with an increase in the co–deposition time. During the co–deposition, a part of electrochemically generated deuterium is absorbed, $i_{2,1}$ and part appears as evolving gas, $i_{2,2}$. In the balance equation, the first is connected with heat source/sink and only the second can be recovered by the recombination reaction. Consequently, the term $\int_{\Delta t} I[E_c - E_{th}]dt$ should read $\int_{\Delta t}[IE_c - i_{2,2}E_{th}]dt$ so that the $Q(t)$ term is evaluated by plotting $0.239 mc_p \Delta T - \int_{\Delta t}[IE_c - i_{2,2}E_{th}]dt$ vs $\int_{\Delta t} IE_c dt$. The negative value of $Q(t)$

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[78] S. Szpak, P.A. Mosier–Boss and M.H. Miles, Fusion Technology, **36** 234 (1999)

indicates that at the beginning of the run the cell acts as a refrigirator, *i.e.* in an apparent violation of the second law of thermodynamics.

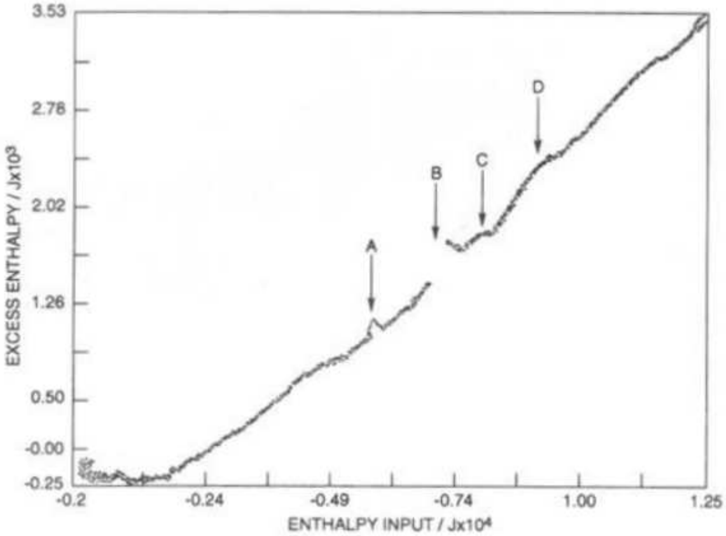

Figure 13:

In the presence of the F–P effect, the shape of both, the experimental and corrected $Q(t)$ vs $\int_{\Delta t} IE_c dt$ represent a sum of at least two terms, $q_1(t)$ and $q_2(t)$, the first denoting the endothermic absorption (acting as a sink) and the second, the exothermic F–P effect (acting as a heat source). Initially the increase in the $Q(t)$ suggests that the principal process is the endothermic absorption. The development of a shallow minimum means that shortly after the initiation of the current flow, a positive term (excess enthalpy production) became active. At first, the contribution is small. However, with the passage of time, it appears that the rates of both processes were in constant ratio. With the further passage of time, the exothermic F–P effect becomes dominant.

5.0 Remarks concerning open cell calorimetry

The solution of Eq. (20) requires specification of initial conditions and evaluation of all other terms consistent with the mode of operation and the cell design. The initial conditions are the equilibrium conditions, *i.e.* the temperature of the whole system is that of the environment and the composition of the gas phase is $D_2 : O_2 = 2 : 1$ with the D_2O vapor in equilibrium with the electrolyte. The rate of the heat transfer out of the cell depends on cell geometry, construction of the enclosure and mode of transport. The simplest case is that of an adiabatic wall. If, however, the enclosure is a diathermal wall, then the heat transfer may occur either *via* radiation with some convective contributions or *via* convection with minor radiative contributions.

The criticism of the open cell calorimetry is centered around the assumption of a steady state heat transport to the environment and the selected calibration procedure. To assure correct interpretation of the thermal behavior of the electrolyte phase it is necessary to know the rate controlling process, its temperature dependence and relaxation time[79].

Characteristic features of thermal output of electrochemical cells are
(i) Thermal energy generated by electrochemical cells based on the Pd/D–D_2O system, employing co–deposited electrodes, is delivered at temperatures not exceeding 100C. The delivery at higher temperatures is possible but would require substantial redesign of cells.
(ii) The delivery at low temperatures should not be considered as an impediment since substantial energy usage is at temperatures less than 70C.
(iii) Advantages of electrodes prepared by co–deposition are: (a) 100% reproducibility, (b) on average, higher efficiencies than those reported for massive or thin film geometries, (c) greater flexibility in cell design (e.g. fluidized bed)
(iv) The disadvantage is its poor mechanical strength. This, however, may be corrected by co–deposition at low current densities, alternatively from other Pd–complexes dissolved in D_2O.

6.0 Nature of heat source

To confirm the existence of the F–P effect one needs only to establish an excess enthalpy production. This is done by calorimetry, a method that not only measures the excess enthalpy generation but it can also provide additional information by examining the relaxation times associated with perturbations in the enthalpy inputs (*i.e.* change in cell current or activation of the resistive heater). But calorimetry, being an integrating procedure, cannot do much more. There are other tools, however, that can add to a better understanding the nature of the polarized Pd/D–D_2O system. One such tool is the infra–red (IR) imaging of the surface of an active electrode; the other, prompted by the first, is the use of a pressure sensitive substrate, onto which the Pd/D films are co–deposited, to demonstrate mechanical changes that do occur when an instantaneous and high intensity exothermic reaction takes place.

6.1 Infrared imaging. Early in the 90's, Prof. Simnad (UCSD) and Dr Evans (General Atomic International) viewed the surface of a Ni electrode during electrolysis of light water containing carbonates using an IR camera. They observed the most unusual pattern of temperature distribution, in particular, formation of the space/time dependent temperature zones. When the surface of the polarized Pd/D–D_2O system (electrode) was viewed (during co–deposition and electrolysis of heavy water) the formation of short lived "hot spots" was observed.
Experimentally the IR camera A, Fig. 14a, views the surface of an active negative electrode B, placed next to the thin Mylar sheet C, affixed to the wall of a rectangular

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[79] The concept of the relaxation time is well defined, it is expressed in terms of a variable $\xi(t)$ which is the measure of the approach to equilibrium. It follows the exponential law $\xi(t) = \xi_{eq} + Ce^{-t/\tau}$ where C is the integration constant and τ is the relaxation time.

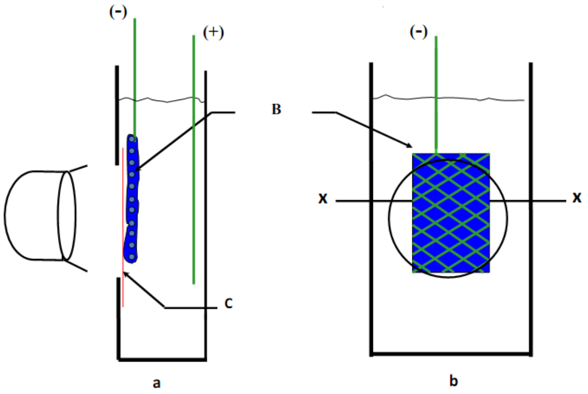

Figure 14:

cell made of clear plastic. Two conditions must be met for a successful monitoring of the thermal behavior, *viz* (i) the amount of the D_2O between the electrode surface and the IR camera must be minimal (in order to avoid the attenuation of the signal), and (ii) the electrode surface facing the IR camera must be open (in a sense that the electrodic processes are accessible to viewing by the IR camera). These conditions are met by co–depositing the Pd/D film on an open substrate, *e.g.* on a Ni screen placed in close proximity to the thin wall (made of Mylar). The IR camera can be operated in two modes: (i) to monitor temperature distribution on the electrode surface and (ii) to measure the cell temperature across the cell (*i.e.* along the X–X line, Fig. 14b.

Results of IR viewing the surface of an active cathode are summarized in Figs. 15a – 15c. The negatively polarized Pd/D–D_2O system shows the development of short lived “hot spots”. These thermal activities, illustrated in Fig. 15a - 1,2,3,4, were observed at indicated times early during the Pd+D co–deposition. Temperature profile recorded across the electrode surface when the IR camera was operated in the second mode is shown in Fig. 15b. The temperature/time profile at the electrode surface and in the solution is illustrated in Fig. 15c.

The distribution of hot spots indicates that (i) the rate of excess enthalpy generation is not uniform,(ii) thermal activities occur at low charge transfer current densities as well as during deuterium evolution, (iii) rough estimates of their intensity can be obtained. Of these statements only (ii) can be regarded as certain – others are subject to interpretation. Thus (i) is not random distribution but a structured chaos and (iii) any estimate would require a set of assumptions that may, or may not, be realistic.
The chaotic distribution of hot spots, Figs. 15a 1 – 4, elevates the surface temperature, Fig. 15b section A-A, above that of a solution, Fig. 15b section B – B. The cell temperature profile was taken periodically during electrolysis. The difference between surface temperature and that of solution increases with time, being initially at *ca* 2C

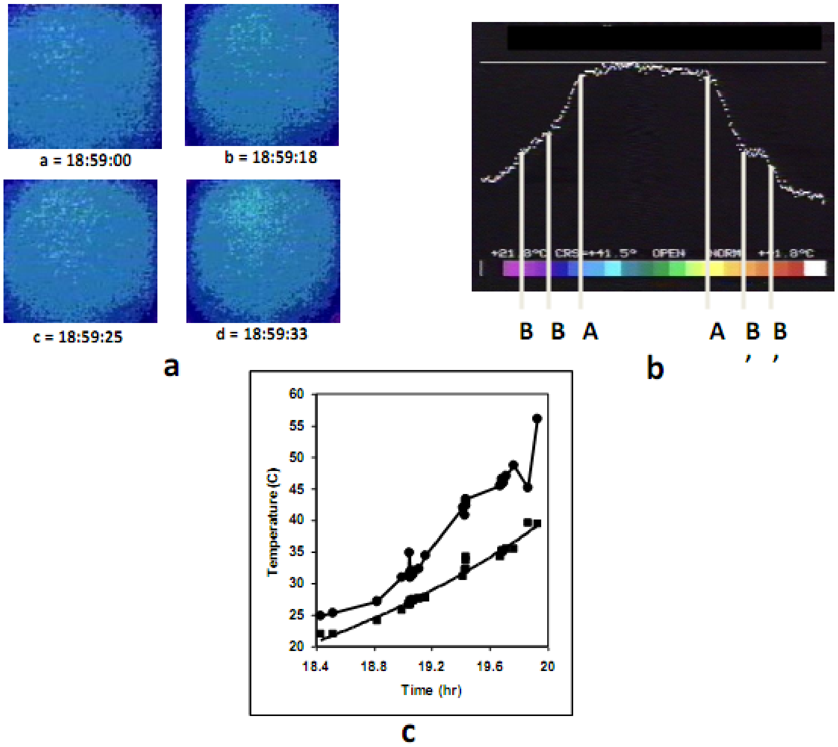

Figure 15:

and reaching a value as high as 17C, two hours later. The increase in the surface temperature, curve a, Fig. 15.c, is irregular, indicating bursts in heat generation. In contrast, the solution temperature, curve b, Fig. 15c, increases smoothly which is an expected behavior because of the large weight (volume) of solution. The temperature difference between the surface and solution, displayed at any instant by the IR camera is substantially higher higher than those measured by thermocouple[80].

6.2 Mini–explosions. The hot spot represents an act of a mini–explosion. In particular, a fast exothermic reaction causes deformation of the electrode structure and the rise in temperature, thus sending pressure and temperature gradients (spikes) away from the source. Such gradients can be detected and, in fact, have been detected when co–deposited films were placed in contact with a piezoelectric substrate. This led us, in 1998, to devise an experimental procedure that would display changes in the electrode structure associated with the formation of hot spots.

An experimental arrangement to record the occurrence of mini–explosions is shown

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[80] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem., **302**, 255 (1991)

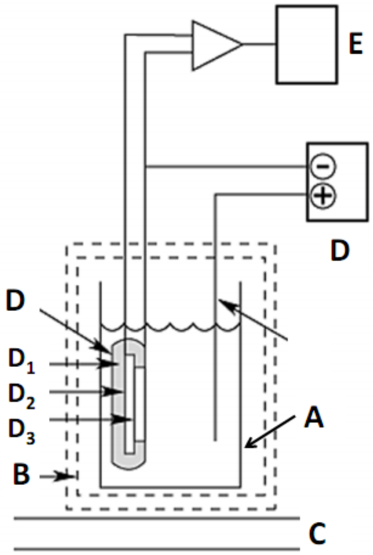

Figure 16:

in Fig. 16. An electrochemical cell, **A**, is placed in a Faraday cage, **B**, (to prevent external noise) and the whole assembly is placed on a shock absorbing pad, **C**. The key part for the successful display of mini–explosions is the construction of the cathode, **D**. Here, a thin circular slice, in the form of a disk (r=1.143 $\times 10^{-2}$ m, l=2.0$\times 10^{-3}$m of the piezoelectric material (lead–zirconium–titanate, source: Piezokinetics, Bellafonte, PA) with a conductor (e.g. Ag) deposited on parallel surfaces was connected to an oscilloscope, **E** and a power source, **F**, in a manner indicated[81]. The Pd/D film was deposited onto one side a piezoelectric substrate.

6.2.2 Mini–eplosions. The characteristic feature of a piezoelectric material is the one–to–one correspondence of direct and reverse effect, *e.g.* compression develops potential shift and shift in the potential produces compression. Thus, upon compression by arriving pressure wave shifts the potential in one direction and upon expansion in an opposite direction. Now, the act of releasing thermal energy in a very short time resembles a mini–explosion which, in turn, deforms the electrode structure and sends pressure and temperature waves. Such waves, impinging onto a pressure sensitive surface, can be displayed if the surface is that of a piezoelectric material.

An ideal display of a mini–explosion, as seen by the sensing device, is illustrated in Fig. 17a where an instantaneous heat source, Q, sends a pressure wave toward the

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[81] The Pd/D co–deposition was carried out at constant cell current from a solution containing 0.03 M $PdCl_2$ and 0.3 M LiCl dissolved in D_2O

sensor's surface causing potential shift downwards, Fig. 17b. The associated heat wave, traveling much slower, arrives at the sensing surface after the system has relaxed to its initial position, and the the temperature change causes an expansion that shifts the potential in an opposite direction.

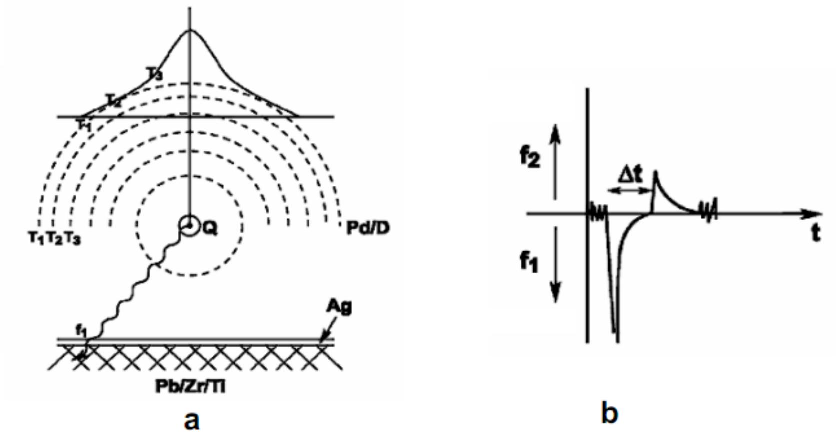

Figure 17:

Shortly into the co–deposition, the pattern associated with a mini–explosion was observed and recorded, Fig. 18a, where a single event followed by a burst, Fig. 18b are shown. As a rule, a single event occurs rarely – a burst of events is the common occurrence. One such burst, in an extended form is shown in Fig. 18b. A single event, isolated from a burst is shown in Fig. 18a. Here, we see clearly a single spike which, in the negative direction corresponds to the pressure pulse. Using a simple model, e.g. that of a spherical reaction space, Fig. 17a, one could, from the magnitude of the voltage spike and the Δt, reach some conclusion concerning the position and strength of the heat source.[82]

It is noteworthy that, (i) as the cell temperature approaches the boiling point, the mini–explosions are far more intensive than those seen at lower temperatures, (ii) singular events can be seen also following the termination of an experiment, Fig. 18c.

6.3 Significance of hot spots and mini–explosions

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[82] For the applicable mathematics see H.S. Carslaw and J.C. Jaeger Conduction of heat in solids, Oxford (at the Clarendon press) 198, p. 216

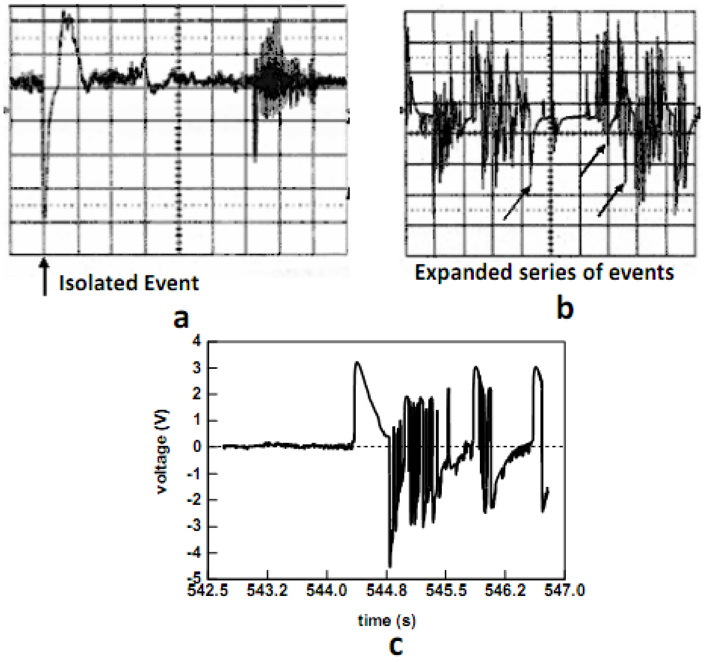

Figure 18:

The observed hot spots are the *Nature's documents* that not only point out the direction to follow but offer some insight into the nature of heat generating reactions. The physical significance and conclusions that can be drawn depend on how this observation was made. Thus, hot spots, as seen by the IR imaging, provide information on their number, intensity and location, or in general, indicating that there are certain location where the concentration of reactant(s) is very large. An assumption can be made that in order to observe hot spots on the electrode surface it is necessary to confine a large number of reacting particles within a small volume. This requirement represents a situation where a microscopically large, but macroscopically small, domains attain a state that can function collectively in absorbing large quantities of deuterium in lattice defects. Excess enthalpy is produced at discrete sites where the transition from stable to unstable regime, alternatively a transition from non–reactive to reactive sites takes place. Restating, one can regard the formation of these domains as the key observation that guides the development of a realistic model representing the F– P effect.

Along the same line of thinking, in a private communication[83], Dr Lowell Wood estimated that *The energy involved in their formation* – (their refers to hot spots) *which is on the order of 0.1 kJ/g or 1 – 10 ergs – must have been developed on at most this time scale; this corresponds to the order of* 10^5 ^{4} *He's formed from D + D fusion over a time–scale of less than 10 nanoseconds,* i.e. *a power level of at least 10 W. In this case, the dimension of the volume of the event was a few hundred Angstroms, and as I noted last Friday, seemingly had something of an elongated nature along the axes of these axisymmetric explosions features; the power level would be at least 100 times higher,* i.e. *at least 1 kW. Since the masses involved are nanograms to femtograms, the specific power generation rates are those seen hitherto only in nuclear explosions.*

In an attempt to arrive at the physical significance of mini–explosions one must define the system in which they take place, *i.e.* one must consider the conditions at the negatively polarized Pd/D–D_2O system. The operating cathode is (i) an open system, *i.e.* a system in which both energy and matter can be transferred between the outside world and the reaction site and (ii) the system is not in equilibrium. Consequently, methods of non–equilibrium thermodynamics are an indispensable tool to examine the nature of the F–P effect. The mini–explosions were recorded as a function of time, thus yielding information on the statistics of occurrence from which one can speculate on the kinetics of formation of precursors[84].

In conclusion, hot spots represent heat sources which are, what appears to be, randomly distributed in time and space, yield substantial amount of information about the reactions and/or processes that constitute the F–P effect. Those are estimation of (i) size of the reaction volume and (ii) number of singular events. From the statistics one can determine whether or not it is random or chaotic, *i.e.* what is the nature of the processes responsible for self–organization.

7.0 Catastrophic events

Three times in this laboratory and, at least once elsewhere[85], thermal run–a–ways were observed. The description in all cases reported is sketchy because these events occurred when no one was present and when no appropriate instrumentation was in place to record relevant data. Nevertheless some pertinent features are consistent. These are: (i) temperature rise is fast, (ii) electrolyte evaporation is rapid but not "explosive" (*i.e.* no spilling), (iii) there is no excessive production of nuclear ash (*i.e.* no increase in X– and $\gamma --$ radiation).

First case: Thermal run—a–way has occurred in the course of collecting data on electromagnetic radiation[86], that resulted in (a) deformation of cell walls and lifting of the cell cover and (b) deposition of Pd on cell wall, illustrated in Fig. 19. In this case, the Pd/D film was co–deposited onto an Au foil (1 × 0.5 cm) and operated at current density of 100 $mAcm^{-2}$. Evidently, the temperature of the cathode exceeded the melting

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[83] Email dated 17 Oct. 2000 to Dr FE Gordon

[84] An extensive discussion of domains formation and their stability can be found in IR Epstein and JA Pojman, *An introduction to non–linear chemical kinetics*, Oxford University Press, New York, 1998

[85] Explosive thermal run–a–way in Prof. Pons laboratory at the University of Utah

[86] S. Szpak and P.A. Mosier–Boss, Physics Letters A, **219**, 382 (1996)

point of palladium and the glob of molten palladium and gold was propelled toward the cell walls where it solidified.

Figure 19:

Second case: During routine examination of the cell performance using as the cathode a Pd/D film co–deposited onto a Ni screen (area 2×1 cm^2). In this case, the event was less damaging – the cell walls were deformed, the cathode temperature was less than that necessary to melt palladium. The SEM of the electrode material is shown in Fig. 20.

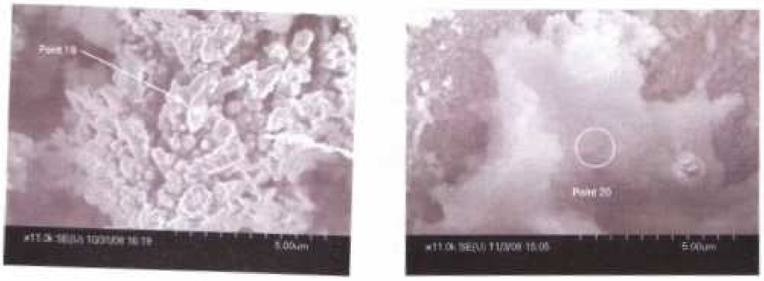

Figure 20:

This moderate thermal run–a–way did not damage the electrode as a whole but radically changed the structure of the deposit. Even a cursory inspection of Fig. 20a shows that (i) a change from globular structure to sub–micron fragments, mosaic in appearance, has occurred and that (ii) there are areas, indicated by arrows, which are significantly different, Fig. 20b. These areas are uniformly distributed throughout the electrode. The EDX analysis of these areas showed the presence of multiple new elements, namely Al, Mg, Si and Ca, thus indicating that the thermal run–a–way resulted from nuclear reaction(s) and not because of unspecified chemical reactions.

Third case: Thermal run–a–way occurred in a cell designed for the investigation of the diffusion of absorbed deuterium assisted by an electrostatic field. A palladium wire, coated with a thin layer of electro–deposited copper and an insulating layer, except for a 1 cm long section, Fig. 21a, was immersed in the 0.3 M LiCl in D_2O solution. Here, thermal run–a–way occurred during D_2O electrolysis.

This case is of particular interest because, in a qualitative way, it describes the events associated with the explosive thermal run–a–way seen in the nuclear active state of the Pd/H–H_2O system. Figure 21a shows a small section of the Pd wire in which the Pd/D layer is separated from the pure palladium. Upon closer inspection of both, the Pd/D layer and the Pd, one sees a concentration of dark spots, indicated by arrows, their number becoming less as the distance from the Pd/D+Cu boundary increases. The EDX analysis of these spots revealed the presence of a new element, namely calcium. One example is shown in Fig. 21b.

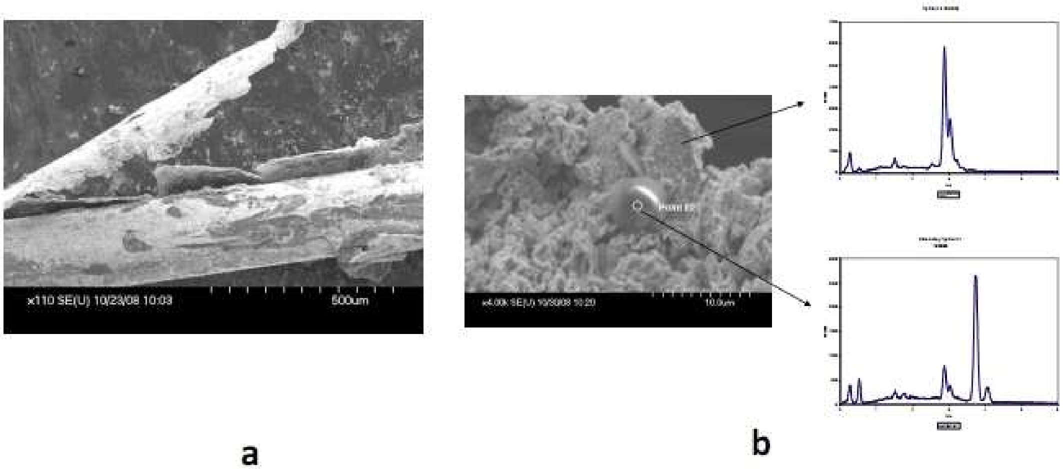

Figure 21:

To reiterate, the SEM and EDX examination of the electrode material, taken from the second and third event, showed the presence of localized, distinctly different morphology. When analyzed, either a single new element, Ca, or multiple, e.g. Al, Si, etc. were found. In the second case, these distinct structures were uniformly distributed, while in the third case, their number decreased with the distance from the reaction point. As illustrated in Fig. 21b, the reaction produces not only localized melting of palladium but also shows mechanical damage due to the action of forces generated by the action of a nuclear event. These three cases show that the thermal event may be of varying intensity – from catastrophic to mild. Although rare the phenomenon of thermal run–a–way must be considered and resolved before the F–P effect could be reduced to practice.

7.1 Catastrophic thermal event in the Pd/H–H_2O system

Several catastrophic thermal events were observed in the course of investigation of the effect of external magnetic field on the behavior of cells using light water based electrolyte, . In one case, after three days of electrolysis with cell current varying between -300 mAcm^{-2} and 5 mAcm^{-2}, a catastrophic thermal event has occurred that resulted in cell deformation, loss of electrolyte due to evaporation and leaking through a punctured cell bottom. The damage, about 1/3 of total area, viewed from the outside and inside the cel is consistent with placing a very hot object in contact with plastic material, Fig. 22. This observation led us to conclusion that even using H_2O based electrolyte, the system can be put into nuclear active state. A communication was submitted to Journal of Radioanalytical and Nuclear Chemistry. The editor decided not to publish the submitted material (the copy of submitted communication can be found in the section VI).

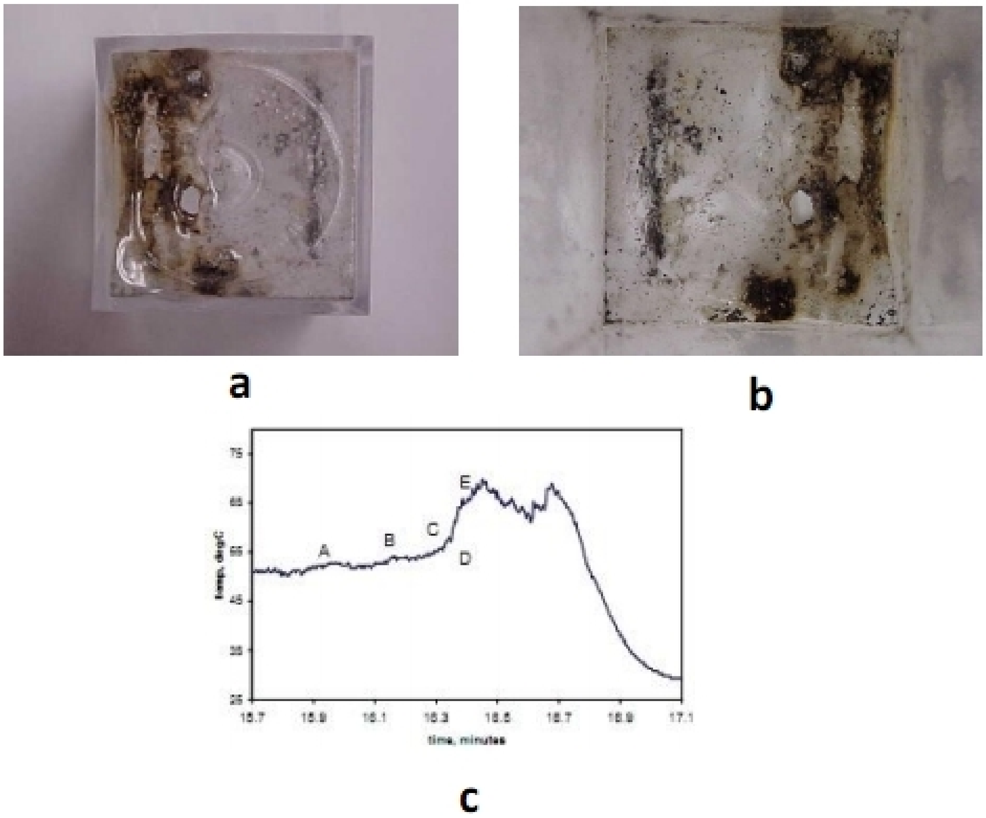

Figure 22:

Viewing the observed damage, *i.e.* shape change and wall puncture, it is reasonable to assume that a very hot object contacted the cell bottom. The black powder, firmly attached to the cell bottom, indicates that the Pd/H film exploded and hot fragments were deposited onto cell bottom. The location of the black residue around relatively clear area suggest that the explosive fragmentation occurred after large segment of the Pd/H deposit hit the surface. Furthermore, the wall deformation implies that sufficiently high temperature softened the acrylic plastic. These facts suggest that it is reasonable to assume a non–chemical energy source and that it remains active even after detachment from the platinum substrate.

8.0 Where are we and where are we going?

The description of thermal events in cells employing co–deposited electrodes seem to indicate that now is the time to think about transition to engineering type of activities, *i.e.* to formulate program leading to the development of practical, energy producing cells.

Fluidized bed electrodes. Almost a decade ago, Fleischmann suggested[87] that *the time is ripe to investigate fluidised bed electrodes*. This remark was based on the unique feature of electrodes prepared by co–deposition, *viz* that an excess power is generated at low current densities. An example of the construction of a cell employing fluidized bed electrode is shown in Fig. 23.

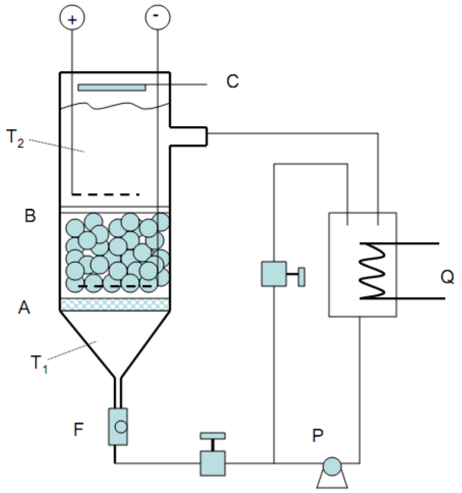

Figure 23:

The functional elements are: A – thin structured layer to assure uniform flow velocity throughout the fluidized bed: B – thin layer preventing contact between fluidized bed particles and anode; C – recombiner (for reaction $2D_2 + O_2 \rightarrow 2\ D_2O$); T_1 and T_2 are the temperature measurements ports. The flow pattern that leads to the transfer of power from the cell to perform useful work is shown by light lines.

The use of fluidized bed electrodes offers flexibility in cell construction which depends

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[87] Fleischmann to Szpak, letter dated 11 Jan, 01

on the positioning of current feeder electrodes and the direction of current and fluid flow. Moreover, because of the porous structure of fluidized beds, the electrochemical reaction would be confined to special zones of the beds as determined by the direction of current and fluid flows[88]. This confinement may add to a better understanding of the Pd/D–D_2O system.

They are, of course, limitations, *e.g.* the interference due to excessive gas generation. This, however, can be, to a degree, controlled by the correct placement of current feeders as well as the adjustment of the velocity of flow and current density. Although there are no data as to the efficiency of such designs, nevertheless one could state, with reasonable certainty, that devices using low grade heat can be easily realized.

9.0 The "recombination theory".

Except for Joule heating, the exothermic absorption (the endothermic absorption occurs during the co–deposition) and the F–P effect, no other heat sources are activated during the co–deposition process. The frequently cited $D_2 + O_2$ recombination reaction, as being responsible for excess enthalpy generation, is not supported by experiment (recombination of evolving gases yielded volumes that were better than 1.0% of those calculated assuming 100.0% Faradaic efficiency[89], or theoretical considerations[90]. And yet the notion that recombination is responsible for the excess enthalpy generation persists. For example, Shanahan[91] observed that the short–lived hot spots support the recombination theory. In his view, to quote: *"The infrared photography of Szpak* et.al. *is supportive evidence of this, if one considers the oxidation in subsurface bubbles to be rapid, which should be true of $D_2 + O_2$ flames"*. Shanahan, in his publication *Comments on "Thermal behavior of polarized Pd/D electrodes prepared by co-deposition*[92] writes *this author has consistently proposed that entrained bubbles are the source of the effect*. Such interpretation is, indeed, difficult to understand. Moreover, just because he *consistently proposed, etc* it does not make it so.

What is most annoying is the fact that it is impossible to dispute and provide a correct interpretation because the editor of this journal, L.E. Hansen (Brigham Young University), has rejected the submitted rebuttal: *This paper is being rejected without review for the following reasons...* among them *I was an author on what I believe is the first paper to provide experimental evidence of the recombination reaction and suggest it as an explanation of "excess heat"*. With this in mind, one might ask: should the journal editor reject submitted material just because his own work has not been cited or agreed with? The politics of publications is taken up again in the concluding remarks, *cf.* VI.

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[88] An example of the effect of the direction of flows was discussed, among others, by Szpak *et.al.* in Electrochim. Acta, **11**, 934 (1966)

[89] W. N. Hansen and M.E. Melich, Trans. Fusion Technology **26**,355 (1994)

[90] F. Will, J.Electroanal. Chem..,**426** 177 (1997)

[91] K.L. Shanahan, e–mail to M.A. Imam, 20 June 2002

[92] Thermochim. Acta, 428 (2005) 207

SECTION V: NUCLEAR ASPECTS

Although still controversial, the excess enthalpy generation during electrochemical compression of deuterium in the Pd lattice, is generally accepted. This, however, cannot be said about the material presented in this section, *i.e.* that the excess enthalpy production must originate from nuclear reactions, and that the nuclear ash consists of electromagnetic radiation, transmutation and particle emission. Because of the lack of theoretical guidance, we present the experimental evidence only and avoid interpretation of the mechanism giving rise to radiation, transmutation and particle emission except when nuclear reactions are written in the form of equations that correspond to those employed in chemical kinetics.

In hot fusion nuclear events occur at isolated locations where the temperature is on the order of thousands K. In "cold fusion", an event takes place in the reaction volume where (i) the concentration of D^+ is about 100 molar and that of electrons ten times higher, (ii) where the driving forces are chemical potentials, and (iii) where coherent processes determine the composition of the "nuclear ash". Hence, one of the still unanswered questions: Is it chemistry or physics? One answer can be found in a treatise on inorganic chemistry[93] in which we find that *The essential goal of nuclear physics is to interpret the nature of nuclear forces. One important approach to this objective is the study of transmutations occurring in nuclear reactions, and the magnitude of energy liberated in such processes. Originally this subject could be investigated only by purely physical methods, and as such could be considered a section of pure physics*. Here, we cast the problem in chemical terms and accept the fact that when an explanation is offered for an observation, it is, as a rule, rejected by those who suffer from the pathological disbelief syndrome. In their response they invent reasons that often violate well known principles. An example – detection of deuterium by mass spectroscopy (mass number 2) is not deuterium but unspecified impurity, *cf.* VI.

Topics discussed in this Section include: (i) electromagnetic radiation, *e.g.* X– and γ–radiation, (ii) tritium and helium production[94], (iii) production of new elements, (iv)

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[93] H. Remy, Treatise on inorganic chemistry, vol. II, p. 563, Elsevier, 1966

[94] Our search for the nuclear ash was limited to electromagnetic radiation and tritium production; helium production was studied by Dr M.H. Miles at NWC – China Lake, Ca

particle emission, (v) induction of nuclear active state in the Pd/H–H_2O system and (vi) the role of an electron in nuclear reactions.

1.0 Electromagnetic radiation: X-ray, $\gamma-$ emission

In general, the energy deposited within the Pd lattice should result in emission of X–rays arising from the disturbance of the electronic structure of the Pd/D–D_2O system. In particulr, soft X– ray emission (5 –55 keV) can occur through any number of processes involving (i) the excitation of the solid through processes involving individual atoms, (ii) through coherent many–body electron effects, or (iii) it may indicate the formation of tight orbits where the corresponding quantum jumps should yield soft X–rays.[95]

The material presented here is a summary of a two years long study of spectral regions 15 — 3000 keV and 7 – 40 keV on two vastly different surfaces of the Pd/D films (morphology effect) with and without electrolyte modification (addition of small amounts of thiourea and Be^{2+} ions).

1.1 Qualitative demonstration. Qualitative demonstration of X–ray emission is usually done by recording images on photographic film. In the simplest arrangement, a photographic film, enclosed in a light–tight and solution–impermeable envelope, is placed in close proximity to the screen electrode. One example[96] – after exposure for 24 hrs (or more), in an operating cell (at cell current that assures visible evolution of "electrolytic gases", D_2 and O_2) the developed film showed areas exposed to the radiative flux and those partially blocked by the Ni screen on to which the Pd+D film was deposited.

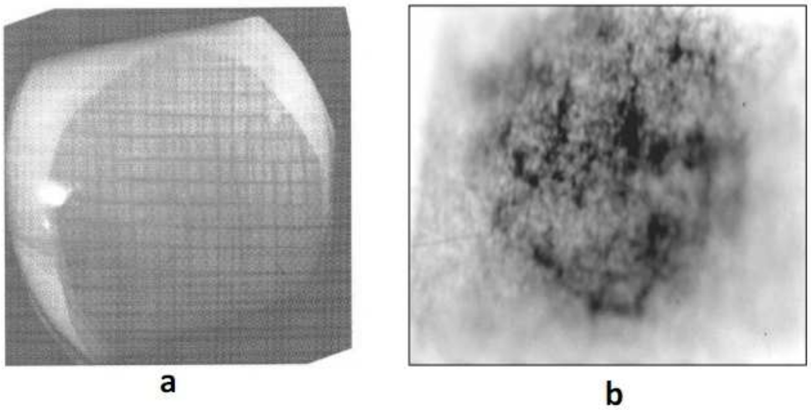

Figure 24:

What appears to be a double or triple exposure is most likely due to a slight displacement of the film, Fig. 24a. We should like to point that the emission of the electromagnetic radiation persisted after termination of cell current flow, Fig. 24b.

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[95] J.P. Vigier, Physics Letters A **221**, 138 (1996)

[96] S. Szpak, P.A. Mosier–Boss and J.J. Smith, J. Electroanal. Chem., **392**, 255 (1991)

1.2 Quantitative evaluation. Quantitative evaluation of electromagnetic radiation is not a simple matter. It requires careful consideration of (i) background radiation, (ii) sensitivity of detecting devices and (iii) appropriate cell design. In practice, the background radiation must be reduced as much as possible. This is usually done by placing the equipment in well–shielded caves (by *e.g.* 5.0 cm thick lead bricks) which reduced the background radiation to acceptable levels. In our work, for data collection, the p–type high purity Ge and Si(Li)detectors were employed and the spectral data analyzed using standard procedure. Movable cathode was designed to maximize the collection of electromagnetic flux[97].

(i) Spectral region 15 — 3000 keV. This spectral region was selected to provide an overview of the electromagnetic radiation with particular attention to the surface morphology and the effect of surface active agent(s), e.g. thiourea $CS(NH_2)_2$. Inspection of Fig. 25 shows that both surface morphology and presence of certain additives, e.g. Be^{2+} ions, affect the intensity of electromagnetic radiation. On the co–deposited surface an increase in the count rate (ca 4%) occurred shortly after the initiation of the Pd/D co–deposition while several days of charging were required to observe the same effect on a smooth surface, Figs. 25a and 25b respectively. The effect of the an additive, *e.g.* thiourea, was the same, namely the reactivation of the X–ray producing processes, Fig. 25a, or their initiation, Fig, 25b.

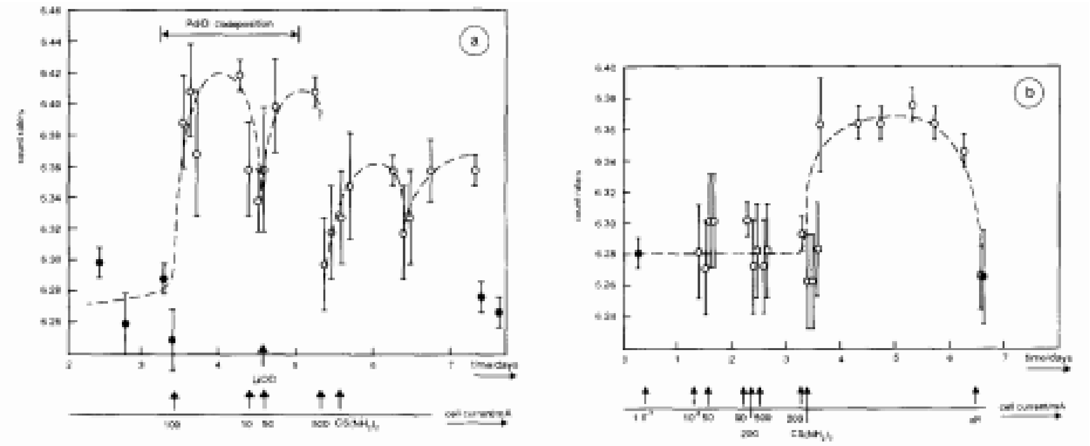

Figure 25:

The electrode response to the additives suggests a substantial change in the dynamic equilibrium within the interphase region.

(ii) Spectral region 7 – 40 keV. The emission of low energy radiation is of special interest. The emission of soft X–rays indicate the electron capture by a nucleus and, constitute the key reaction for the initiation and sustainment of nuclear active state in the $Pd/H^n–H_2O$ system (n= 1,2). The low energy distribution of the electromagnetic

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[97] S. Szpak, P.A. Mosier–Boss and J.J. Smith, Physics Letters A **210**. 382–390 (1996)

emission, shown in Fig. 26, reveals two features, *viz* the presence of weak peaks, one at *ca* 20 keV and another between 8 – 12 keV. Our interpretation assigns the 20 keV peak to the Pd K_α which is superimposed upon the bremstrahlung arising from the oscillating plasma of the polarized Pd/D–D_2O system. The 8 – 12 peak is most likely associated with the Pt which has the L line emission within the 10 – 13 keV range (Pt anode).

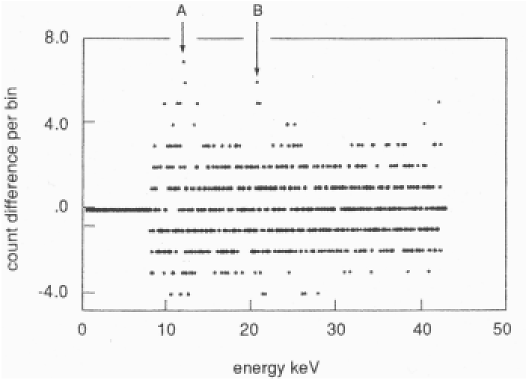

Figure 26:

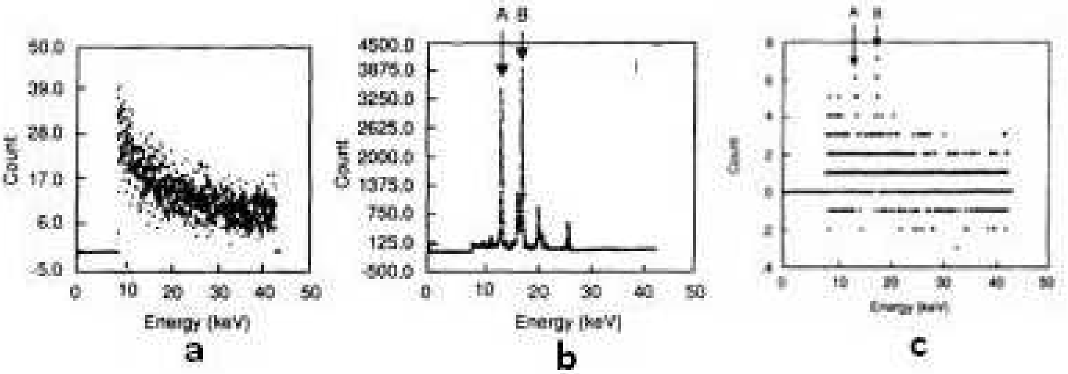

Figure 27:

To demonstrate the correctness of our interpretation we constructed a model which consisted of a simulated bremstrahlung by recording thorium oxide spectrum and an americium spectrum to represent the contribution of line source. Addition of these spectra, Fig 27a, and the subtraction of the background, Fig 27b, resulted in the spectrum shown in Fig. 27c, *i.e.* the spectrum that exhibits a structure very similar to that shown in Fig. 26. While such procedure is not a definitive proof of the offered interpretation of the meaning of Fig. 27a, it demonstrates that a spectrum consisting of a few energy peaks superimposed on a bremstrahlung background is a reasonable interpretation.

In conclusion several points can be made, among them: (i) The cathodically polarized system emits X– rays with a broad energy distribution and with an occasional emergence of recognizable peaks, (ii) the emission of the X–rays appears to be of varying intensity, (iii) surface morphology influences radiation: the co–deposited Pd/D electrodes emit electromagnetic radiation during co–deposition while electrodes with smooth surface require much longer times, (iv) addition of certain ions, *e.g.* Be^{2+} ions or thiourea into electrolyte, significantly affects the intensity and the "incubation" time of the emitted radiation.

2.0 Tritium production

Initially, in support of conclusions reached by Fleischmann and Pons, the search for the nuclear ash was concentrated on the production of tritium. This was based on the belief that the governing reaction is $d+d \rightarrow He^* \rightarrow t+p$ in which the highly excited He^* atom decays into tritium and proton. Usually, in the search for tritium production open cells were used and its presence was determined by the analysis of the electrolyte phase. This procedure does not yield useful information because it requires relatively high tritium production rate and the knowledge of the separation factor, usually taken to be 2.0. Reliable determination of tritium content and its rate of production in electrochemical cells is done in either closed cells[98], or in closed cell systems[99]. The closed cell system, was used in this Laboratory[100]. The advantage in the use of closed cell system rather than closed cell is that it allows for tracing the rate of tritium production with time.

2.1 Experimental protocol

(i) Cell design and sampling schedule. The closed cell system used in the the search for tritium production is shown in Fig. 28a with the sampling schedule in Fig. 28b. The cell with graduated walls to provide check on the volume of electrolyte, was connected to another cell containing a catalyst with sufficiently large surface area to assure complete recombination of evolving gases[101]. A known volume of electrolyte with known tritium content is electrolyzed at constant current for a period of $\Delta t = t_2 - t_1$. During this time period the electrolyte volume is reduced by $V_e(t_2) = IM_w\Delta t/2F\rho$. At time t_2 a sample is withdrawn, further reducing the electrolyte volume by V_s. Immediately after sampling, the electrolyte volume is restored by addition of electrolyte in the amount of $V_a(t_3) = 2V_s + V_e$. After a few minutes to allow for mixing, a second sample, $V_s(t_4)$, is removed for tritium analysis. Removal of a second sample, following the electrolyte addition, ensures that the electrolyte volume and tritium content are accurately known at the beginning of the next time interval. The sampling and addition procedures are to

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[98] F.G. Will *et.al.*, Studies of electrolytic and gas phase loading of palladium with deuterium, in "The science of Cold Fusion", ICCF–2, Como, Italy, 1991

[99] S. Szpak *et.al.*, J. Electroanal. Chem, **373**,1 1994

[100] Szpak *et.al.* Fusion Technology, **33**,38 (1998); Szpak *et.al.*, J. Electroanal. Chem., **373**, 1 (1994)

[101] The design of gas combining cell must assure that samples are representative of the total D_2O and DTO contents, *i.e.* the area of the catalytic surface must be sufficiently large to yield 100% efficiency.

be carried out without the interruption of the cell current flow.

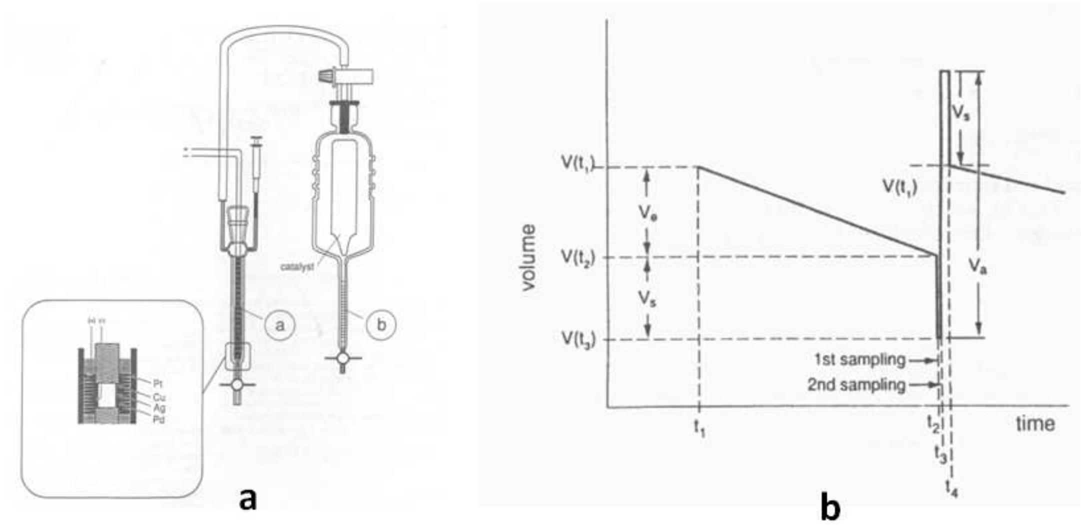

Figure 28:

(i) Tritium analysis. The determination of the tritium generation rate is a two step procedure, consisting of transfer of tritium from the electrode to the liquid phase (an electrolyte) followed by scintillation count. The method employed here was: one (1) mL of sample was added to 10 mL of Fisher Scientific ScintiVerse E Universal LSC cocktail in a borosilicate vial. The prepared solution was counted for 600 minutes in a Beckman LS 600 LL scintillation counter. A word of caution: to eliminate interference from chemiluminescence a sample was distilled to dryness and the distillate analyzed for tritium content. Incidentally, the greatest source of error is the determination of tritium content, particularly at low tritium concentrations. When operating open cells with multiple samplings, care must be taken to assure good control of the electrolyte volume.

2.2 Computational

In the closed cell system the fluxes associated with the electrolysis of heavy water in cells employing deuterium absorbing cathode, illustrated in Fig. 28a, are summarized in Fig. 29. The gaseous products, generated during electrolysis , *i.e.* D_2 and/or DT evolving at the cathode, C, and O_2 at the anode, A, are catalytically recombined in a separate vessel, R. Because of the absorption of hydrogen isotopes by the cathode material, the hydrogen flux produced by electrolysis is divided into three streams: entering the electrolyte by the Heyrovsky–Horiuti path or other exchange processes, entering the gaseous phase by the Tafel path and entering the bulk electrode by absorption. If tritium is produced/consumed during the experiment, then either the magnitude of these fluxes or other appropriate fluxes should be considered. The mass volume of tritium in the electrolyte, Fig. 29, where r denotes rates of mass change with subscripts 1, 2, 3 and 4 indicating electrolyte addition, removal by sampling, evaporation and electrolysis, respectively; f is the tritium mass fraction with subscripts g and l indicating gas and liquid phases. Cell components are shown by heavy lines.

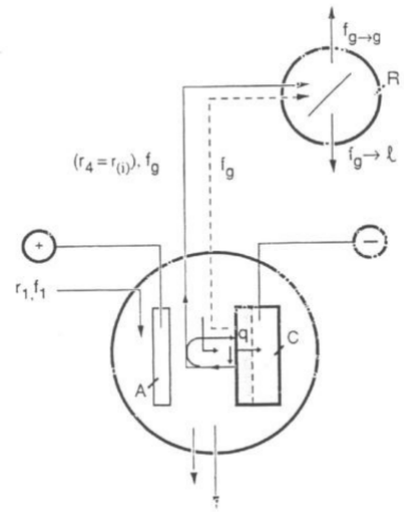

Figure 29:

The time dependence of tritium concentration in the electrolyte phase is given by solving the equation

$$\frac{d(fm)}{dt} = \Sigma_n f_n r_n kq \tag{21}$$

subject to experimental conditions specified by $\frac{dm}{dt} = \Sigma_n r_n$ where f denotes the tritium mass fraction, m is the mass of electrolyte and n identifies the relevant process. In particular, r_1 is the rate of D_2O addition to replenish that removed by electrolysis, r_2, 5_3 and $r_4$are the rates of removal by sampling, evaporation and electrolysis respectively and $kq, (k < 1)$ is the fraction of tritium produced or consumed in the electrode interior, that has been transferred to the electrolyte phase irrespective of the transport mode. As written, equation (21) implies that if tritium is generated within the electrode it is transferred to the electrolyte phase or if is consumed, it is removed from the electrolyte phase. Tritium balance on the recombiner and electrode are formulated in an analogous manner with the inclusion of the relevant fluxes identified in Fig. 29.

Equation (21) is an equation of the type

$$\frac{dy}{dt} + P(t)y = Q(t) \tag{22}$$

which has a solution

$$y = y(0)exp\left(-\int_0^t Pdt\right) + exp\left(-\int_0^t Pdt\right)\int_0^t Qexp\left(\int_0^t Pdt\right)dt \tag{23}$$

For an open system with intermittent sampling, an initial m(0) is electrolyzed at constant cell current, i, the rates r_1 and r_2 are zero and $r(i)$ is constant (Faraday law). With r_1 and r_2 being zero, *i.e.* with $P = (s-1)r(i)/[m(0)-r(i)t]$ and $Q = kq(t)/[m(0)-r(i)t]$, where s is the isotopic separation constant, the solution is

$$f(t) = [m(0)-r(i)t]^{s-1} \times \left[\frac{f(0)}{m(0)^{s-1}} + \int_0^t \frac{kq(t)}{[m(0)-r(i)t]} dt\right] \tag{24}$$

In practice, for constant tritium generation rate, the integral in Eq. (24) can be treated as a weighting factor, *i.e.* Eq. (24) becomes Eq. (25).

$$f(t) = f(0)\left[\frac{m(0)-r(i)t}{m(0)}\right]^{s-1} + \frac{kq}{(s-1)r(i)}\left[1-\frac{m(0)-r(i)t}{m(0)}\right]^{s-1} \tag{25}$$

For a closed cell system, the balance on the recombiner must be included. The tritium mass balance for recombiner working with the efficiency ε_r is given in terms of the tritium mass fraction

$$f_g = \varepsilon_r f_{g\to l} + (1-\varepsilon_r) f_{g\to g} \tag{26}$$

where subscript $g \to l$ refers to mass fraction found in the liquid collected during catalytic conversion and $g \to g$ indicates the lost fraction due to inefficiency of the catalytic converter.

2.3 More about the isotopic separation factor, s

The hydrogen isotopes evolve at different rates during electrolysis. Consequently it is convenient to introduce a quantity called isotopic separation factor, s, which is proportional to the relative rates of hydrogen isotopes evolution and which can be used in the quantitative analysis of “electrolytic gases”. The separation factor used in the analysis, *cf.* Eq. (24), is defined as s= $(\frac{c_T}{c_{D\,g}})/(\frac{c_T}{c_{D\,l}})$ and it should be inverted to conform to values usually cited in the literature.

Irrespective of the rate determining step, species undergoing electro-reduction, $c_m^{(r)}$, are in equilibrium with those in the bulk (b) electrolyte (superscript r refers to the location where the reactive species affect the rate through their electrochemical potentials). The set of restrictive conditions that assures constancy of the isotopic separation factor are: (i) evolution reaction of hydrogen isotopes is independent of each other, (ii) sufficiently high overpotentials are applied so that the reverse reaction can be neglected and (iii) the system operates in a stationary state, *i.e.* equilibria between various species are established resulting in d θ/dt = 0.[102] For the deuterium absorbing electrode material, the concept of interphase must be extended to include the metal side.

Figure 30 shows the structure of the innerphase where r is the reaction site, a adsorption site, e charge transfer site, n indicates the nucleation and gas evolution plane. The sites are located where the reactive species affect the rate through their electrochemical potentials. The associated fluxes are: in Fig. 30a the i - i_D + i_T denotes the cell

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[102] J. O’M. Bockris and S. Srinivasan, J. Electrochem. Soc., **111**, 844 (1964)

current producing gaseous deuterium and tritium. If tritium is produced within the bulk electrode and transferred to the electrolyte phase, then it must be first brought to the adsorption plane (a– plane). The constancy of the s– factor requires an equilibrium condition between species located within the interphase while the applicability of Eq. (25) demands that all tritium enter the electrolyte phase, *i.e.* follow the path, Fig. 30a. If, however, the conditions are such that transport to the electrolyte phase is prevented, path Fig. 30b becomes operative, resulting in substantial enrichment of the gas phase.

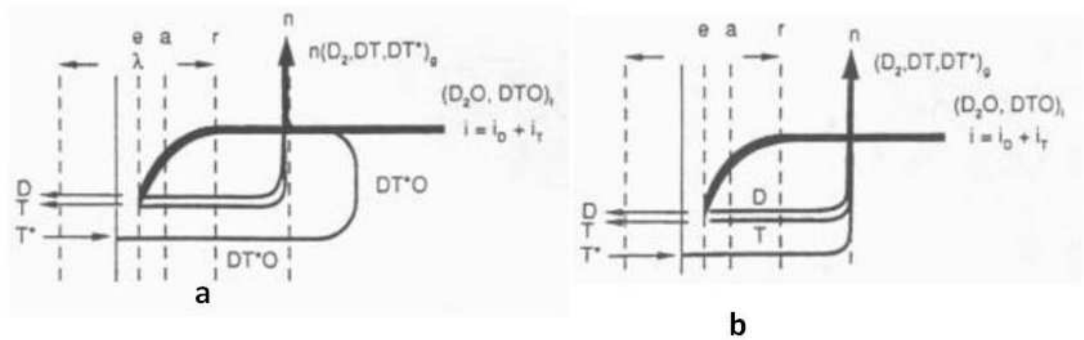

Figure 30:

2.4 Tritium production rates

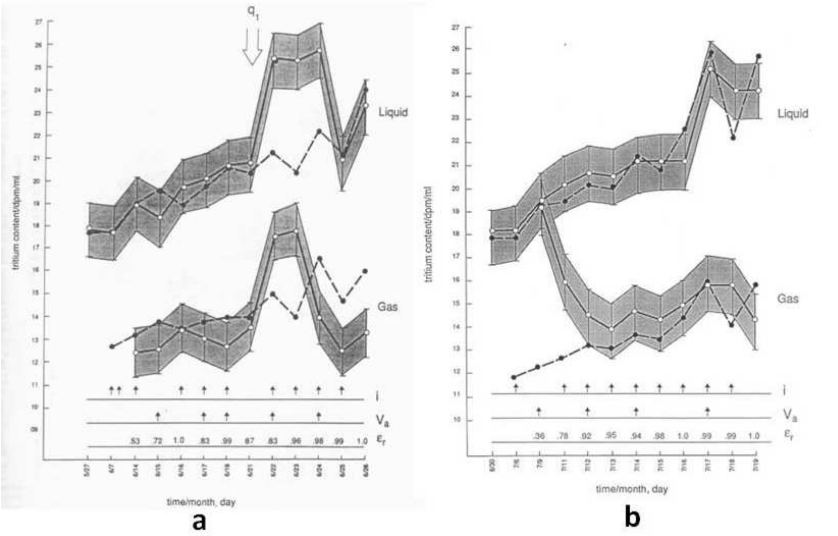

Figure 31:

A qualitative assessment of tritium distribution between the electrolyte and gas phases resulting from a prolonged evolution of deuterium generated by electrolysis of D_2O is shown in Figs. 31a and 31b. The electrolysis was carried out under varying cell current profiles. For convenience, changes in cell current, i, times of sampling, (V_a), and recombiner efficiency, ε_r, are indicated. Two different scenarios can be seen from the data, *viz* sporadic tritium production with periods of inactivity, Fig. 31a, and sporadic tritium production in which selective enhancement of the phase tritium level occurs, Fig. 31b.

The first scenario, Fig. 31a, shows an active period of approximately three days duration, starting 30 days after the initiation of charging. The constancy of the isotopic separation factor indicates that the reaction path for the cathodic charge transfer did not change and that tritium generated during the active period first entered the electrolyte phase resulting in isotopic distribution controlled by the electrodic reaction, *i.e.* the distribution of tritium between the phases follows transport*via*route A, Fig. 30a.

The second scenario – the data summarized in Fig. 31b show an active period in which tritium enrichment is limited to the gaseous phase only. This enrichment occurred shortly after the completion of co–deposition process and was subsequently followed by a period of inactivity. This selective enrichment of the gaseous phase suggests that path B, Fig. 30b, is the method of transport.[103]

3.0 Production of new elements

Low temperature nuclear reactions were first reported nearly a century ago. In a brief communication Oliphant *et.al.*[104] reported that bombardment of perdeutero inorganic compounds by deuterons produced tritium and hydrogen. One year later, Lawrence *et.al.*[105], in an apparent extension of Oliphant *et.al.* work, showed that there exists a class of barrierless nuclear reactions of the type $^A_Z(X) +^2 H \rightarrow^{A+1}_Z (X) + p^+$. The occurrence of "room temperature" nuclear reactions was not challenged for the next 50 years. The situation has drastically changed following the Fleischmann and Pons announcement on 23 March 1989.

The production of light elements such as tritium and helium was expected. However, the absence of a direct correspondence between the amount of tritium and helium produced and the excess power generated in cells suggested that other, as yet, unknown set of nuclear processes takes place. To test this hypothesis, a search for other, much heavier elements, was undertaken. The first attempt to do so was that by Rollison and O'Grady[106] who, through surface analysis, showed the presence of a number of heavy elements and concluded that their concentration at the surface was due to segregation associated with transport of impurities rather than to an undetermined nuclear process. The search for new elements, *i.e.* elements not originally present in the system, continued. A review of this effort was provided by, among others, Miley and Shrostra[107]. In most cases, however, the experimental methodology was different from that associated

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[103] The sporadic nature of the burst–like behavior would not be detectable using a closed cell system

[104] M. Oliphant, P. Hartreck and Lord Rutherford, Nature **133**,413 (1934)

[105] E.O. Lawrence, E. McMillan and R.L. Thornton, Phys. Rev, **48**, 493 (1935)

[106] D.R. Rollison and W.E. O'Grady, Anal. Chem., **63**, 1697 (1991)

[107] G.H. Miley and P.J. Shrostra, Trans. Am. Nucl. Soc., **88**, 627 (2003)

with Fleischmann and Pons experimental protocol.

If not segregation then what? In our work we emphasized the importance of key observations in planning research activities. One of the key observation is the presence of localized heat sources, the hot spots, because they imply the existence of domains containing large number of D, D^+ and electrons interacting with the network of Pd lattice defects. Their physical state, of what could be described as an "oscillating plasma", is governed by two factors: (a) the $F\Delta\Phi$ and (b) the σV terms. The first term, acting through the electrochemical potential of absorbed deuterium produces high pressure[108] which, under non–equilibrium conditions, results in formation of new structures (molecular assemblies). The second term, σV, arising from the asymmetry of the stress field, adds little to the compression but affects the molecular structures within the oscillating plasma through interaction with the network of lattice defects.

3.1 Experimental

In general, before the nature of any event, occurring within a system, can be examined, it must be first created in a reproducible way, under well defined conditions as to its environment, location and the distance from the initial equilibrium. This applies to the evidence presented in this section. To avoid irrelevant comments offered by *pathological disbelievers*, all experiments were conducted employing standard procedure. The presentation is structured as follows: (i) experimental protocol, (ii) evidence, and (iii) interpretation.

(i) Experimental protocol. Typical electrochemical cell used in this set of experiments is shown in Fig. 32. It consist of (i) a rectangular cell (A) made of clear plastic with copper plates (4) attached to parallel walls, (ii) negative electrode (1) – an Au foil anchored to a polyethylene base (3), (iii) positive electrode (2) – a Pt screen to allow for the escape of gaseous reaction products, (iv) an electrolyte – 20 mL of the solution having composition 0.03 M $PdCl_2$ + 0.3 M LiCl in D_2O. The cell assembly is connected to a galvanostat (not shown), the copper plates (4) to a regulated high voltage source. The cathode assembly is placed perpendicular to an external field.
The standard experimental protocol covers three time periods, *viz*— (a) preparation of the Pd/D electrode, (b) "stabilization" of the system – Pd/D + D_2 + D_2O and (c) exposure to an external field. The Pd+D co–deposition takes place at current density 1.0 $mAcm^{-2}$ for the first 24 hrs followed at 3.0 $mAcm^{-2}$ for the time necessary to reduce all Pd^{2+} ions. The stabilization process refers to the time needed to assure uniform distribution of absorbed deuterium throughout the electrode volume. This is done by rising the current density to 30 – 50 $mAcm^{-2}$ for 2 –3 hours. After the stabilization process is completed, the cell is placed in an external electrostatic or magnetostatic field and operated at current densities of 100 $mAcm^{-2}$ or higher for 24 hours or more, the applied field was $3000Vcm^{-1}$ for electric and 0.1 T for the magnetic field.

(ii) The evidence. The placement of an operating cell in an external electric or magnetic field results in dramatic morphological changes. A set of SEM's photographs, *cf.* Figs. 2a –e, shows the effect, namely that the almost spherical globules, Fig. 2a, were

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[108] Fleischmann in ICCF–1

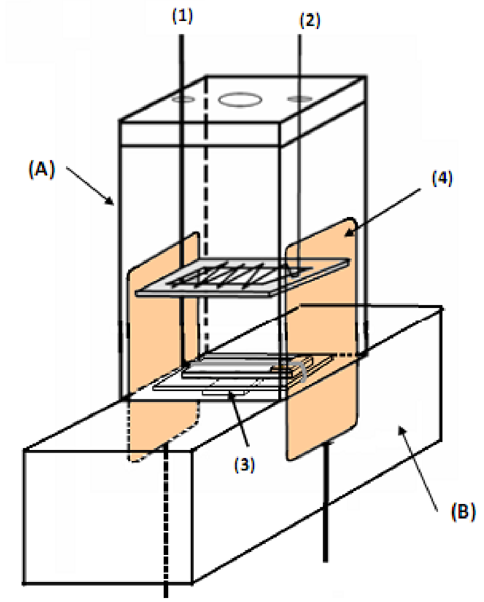

Figure 32:

re–arranged to (i) produce a layered structure, Fig. 2b, chaotic placement of small thin fragments, Fig. 2c, a bended large thin plate (foil?) with, what appears to be, branches of wires attached to its surface, Fig. 2d, and a violent event resembling explosion, Fig. 2e. Even a cursory examination leads to a conclusion that the energy required to produce such changes is far in excess of that which, under the conditions of experiment, could be extracted from the external field at the required rate and intensity. One could offer a number of explanation as to the physical meaning of the observed facts – all of them, most likely, highly speculative. What is clear, however, is that a substantial energy expenditure was needed to create the observed new structures. Thus, the question: Is the energy transferred from the external field sufficient to account for the observed facts and, if not, then what is the its source?

(iii) Selected examples of the external field effect

Upon placement of an operating cell in an external electric field the structure of the deposit undergoes massive change. At first, by visual inspection, one could notice swelling of the Pd/D deposit followed by it leaning toward the negative plate of the condenser, Fig. 32. If the substrate (e.g. an Au foil) is not firmly attached to a supporting structure (e.g. a plastic strip), then, upon placement in an external electric field, there is lifting and bending of the Au/Pd/D deposit, indicating competition between forces of electrical and mechanical nature. During the electrolysis of heavy water for an extended period, e.g. 24 to 48 hrs, the electrode structure, as seen by the SEM, undergoes substantial changes. The examination of the structural changes, recorded under varying field strengths and cell currents, shows changes that can be explained by current distribution within the porous Pd/D deposits and those that cannot. The first group is shown in Figs. 33b – e, the second in Figs. 34b –e.

(iv) Shape changes shown in Fig. 33

The structural changes shown in Figs. 33b – e can be explained in terms of known effects of current distribution in porous structure of the co–deposited Pd/D film. In particular, Fig. 34b shows the effect of disintegration of the "cauliflower –like" deposit into much smaller particles dispersed in an electrolyte. This can happen for a number of reasons, among them vigorous gas evolution arising from an uneven distribution of the current density. If a current passes through such dispersion, then, depending on local conditions, each particle may act as a bipolar electrode, *i.e.* when the current flows through the particle rather than the electrolyte. This happens when the potential drop in the electrolyte is greater than the combined anodic and cathodic overpotentials of the relevant charge transfer reactions.

The charge transfer reactions, whether cathodic or anodic, do change local composition of the electrolyte, thus producing concentration gradients and electrolyte flow that are amplified in porous media. This, in turn, affects the shape of deposits: It may result in (i) non–specific arrangement, Fig. 33b, (ii) the sponge–like structure, shown in Fig. 33c, (ii) dendritic growth[109], shown in Fig. 33d, or (iii) it may produce fine wires[110], illustrated in Fig. 33e.

In the discussion and interpretation of the observed morphological and structural changes at least three factors should be considered: (i) the external field, (ii) the cell current distribution and (iii) the presence of gaseous deuterium within the confines of the electrode structure. Now, since the depth of current penetration (for a given electrode kinetics, current density, etc.) into an electrode depends on the pore size, a different response to the action of an external field is expected at different sites. At sites of a relatively large pore size, a micro–globule is acted upon by two factors: *viz* the electric field and the convective flow arising from mixing by the evolving deuterium.

(v) Shape changes shown in Fig. 34

The production of new elements in cells using co–deposited cathodes is insignificant. However, if these cells are placed in an external electrostatic or magneto-static field, then the rate of new elements production is markedly enhanced. Measurable quantities of new elements can be found at locations exhibiting distinct features, e.g. boulders, or violent reaction, (formation of craters, *cf.* Fig. 2). We noted that at these distinct sites substantial energy expenditure is required to produce the observed morphologies and, at these sites one would expect to find the nuclear ash if the required energy is of nuclear origin. Indeed, at these sites composition is different from that found in

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[109] An extensive discussion of conditions promoting dendritic growth can be found in J. O'M. Bockris nd A.K.N. Reddy, Modern Electrochemistry, Plenum Press, New York (fifth printing) 1974, vol. II, pp. 1218 – 1221

[110] J. C. Bradley, J. Crawford, M. McGee and S.G. Stevens, J. Electrochem. Soc., **145**,L45 (1998)

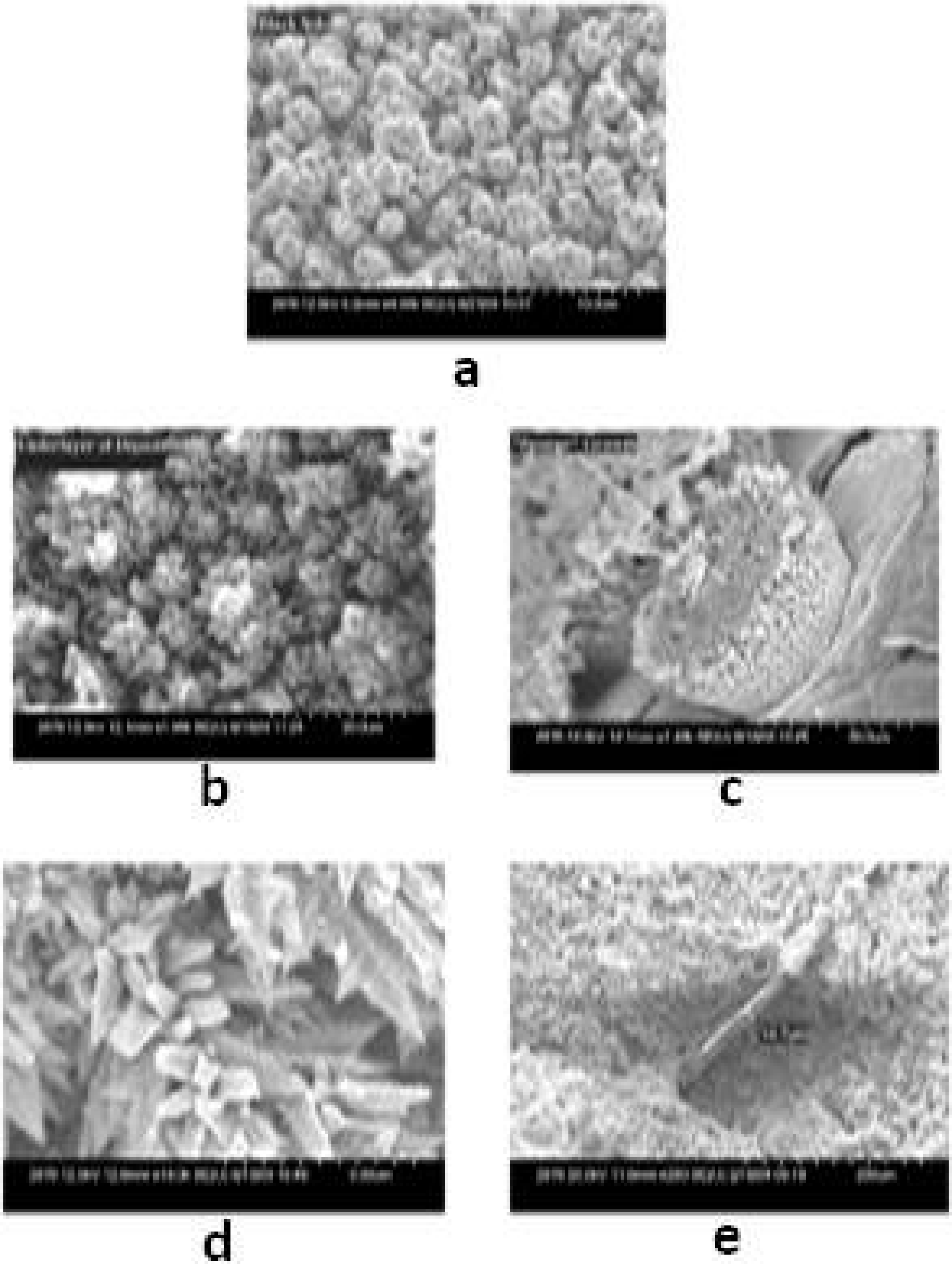

Figure 33:

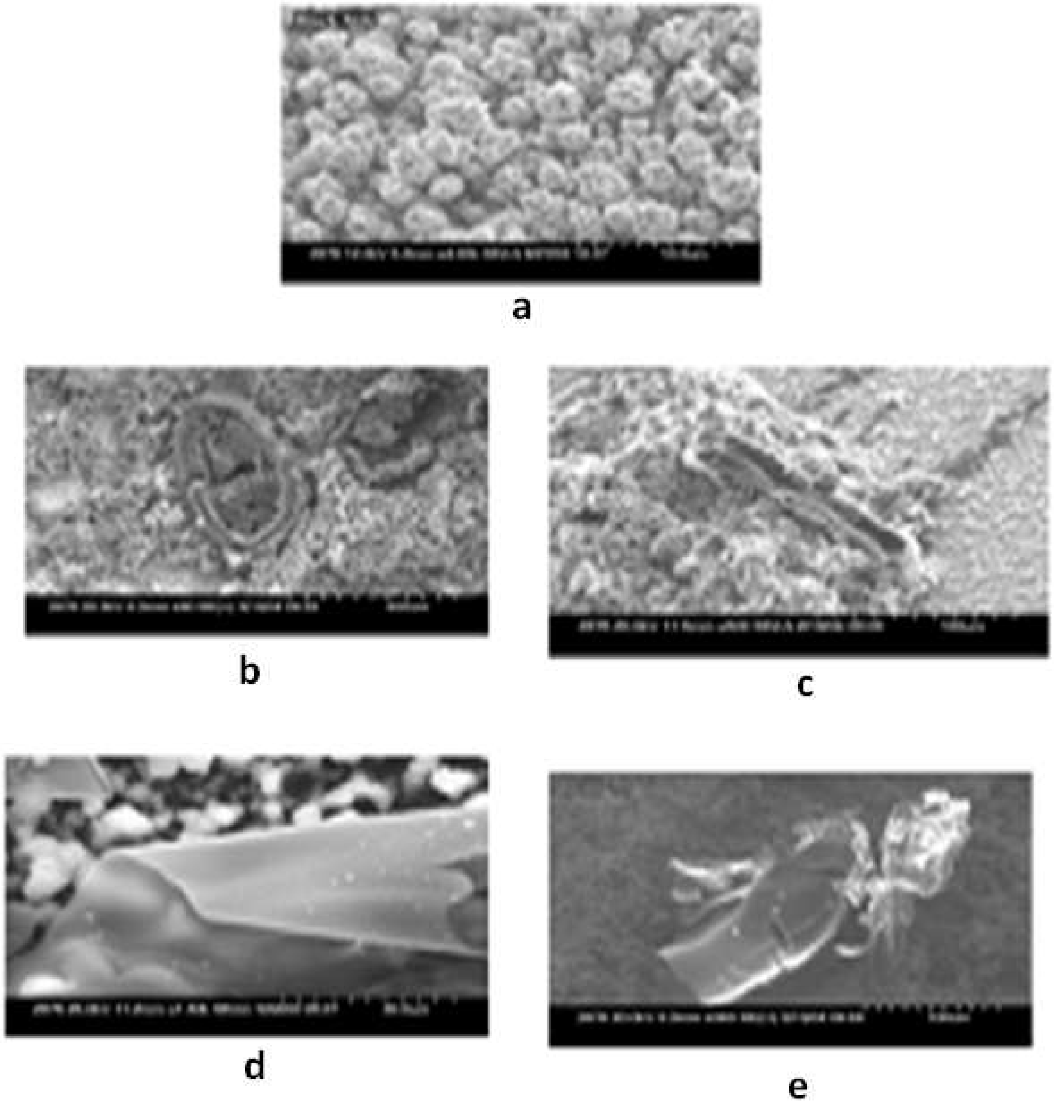

Figure 34:

neighborhood, *i.e.* the presence of new elements[111].

The set of SEM photographs, assembled in Figs. 34b – 34e, shows sites usually seen when a molten metal solidifies under liquid. These shapes are difficult to explain, except that their formation would require substsntial energy expenditure. Briefly, even a cursory examination of Figs. 34b –34e suggests a plausible explanation, namely that, at certain sites, temperatures exceeding Pd/D melting point result in formation of molten blobs which subsequently solidify under liquid while subject to directional forces of electronic nature (field strength on the order of several hundreds Vcm^{-1} is available). The situation is so complex that even a qualitative discussion is not possible at the present time, nevertheless, some speculative explanations can be offered. Thus, (i) Fig. 34b represents, what appears to be, a small explosion in which molten metal solidifies in the form of more or less circular rings, (ii) Fig. 34c where the solidification forms a linear lump. Quite different conditions are associated with forms illustrated in Figs. 34d and 34e. In the first case the folded thin fragment suggests that it was separated from main body, raised and bended. In the second case, it appears that the nuclear reaction took place at one site and its product deposited at another site.

(vi) Effect of an external electric field. A number of selected examples of new elements and their location, generated during electrolysis of D_2O on cathodes prepared by co–deposition is assembled in Fig. 35.

The selected examples show that the number of new elements varies from a single element to multiple elements[112] and that the number does not depend on the strength of applied field. The selected examples illustrate the effect of an external electric field. Evidently there are no recognizable distinct morphological features that would indicate the number or the identity of new element(s). For example, the EDX analysis of, what appears to be a violent explosion, Fig. 35a, one finds two new elements, Mg and Al. Other characteristic morphologies features, *viz* that of a boulder, Fig. 35b and a folded layer, Fig. 35c, contain multiple new elements, *i.e.* Mg, Al, Si and Ca, Fig. 35b, and 35c. These two examples indicate that different morphologies may have the same chemical composition which, in turn, implies that the observed morphology is determined by local forces that guide the development of a particular shape. Rarely, except in thermal run–a–way (*cf.* IV. and V.), one sees cases containing a single new element. One such case is shown in Fig. 35d where the deposited fragment contains Al only.

(vii) Effect of magnetic and magnetic plus electric fields

An external electrostatic field changes conditions at the contact surface, and only *via* the relaxation mechanisms, extends them into the interphase. On the other hand, an external magnetostatic field affects not only the conditions at the interphase (*via* Lorentz forces) but penetrates the interphase where the gradient forces and to a lesser degree the Lorentz forces are active. Hence, it is not surprising that magnetic field would affect both the Pd/D structure and the reaction products in different way. This is illustrated

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[111] S Szpak, P.A. Mosier–Boss, C. Young and F.E. Gordon, Naturwissenschaften, **92**, 384 (2005)

[112] Oxygen, chlorine and Pd are not new elements since they are cell components

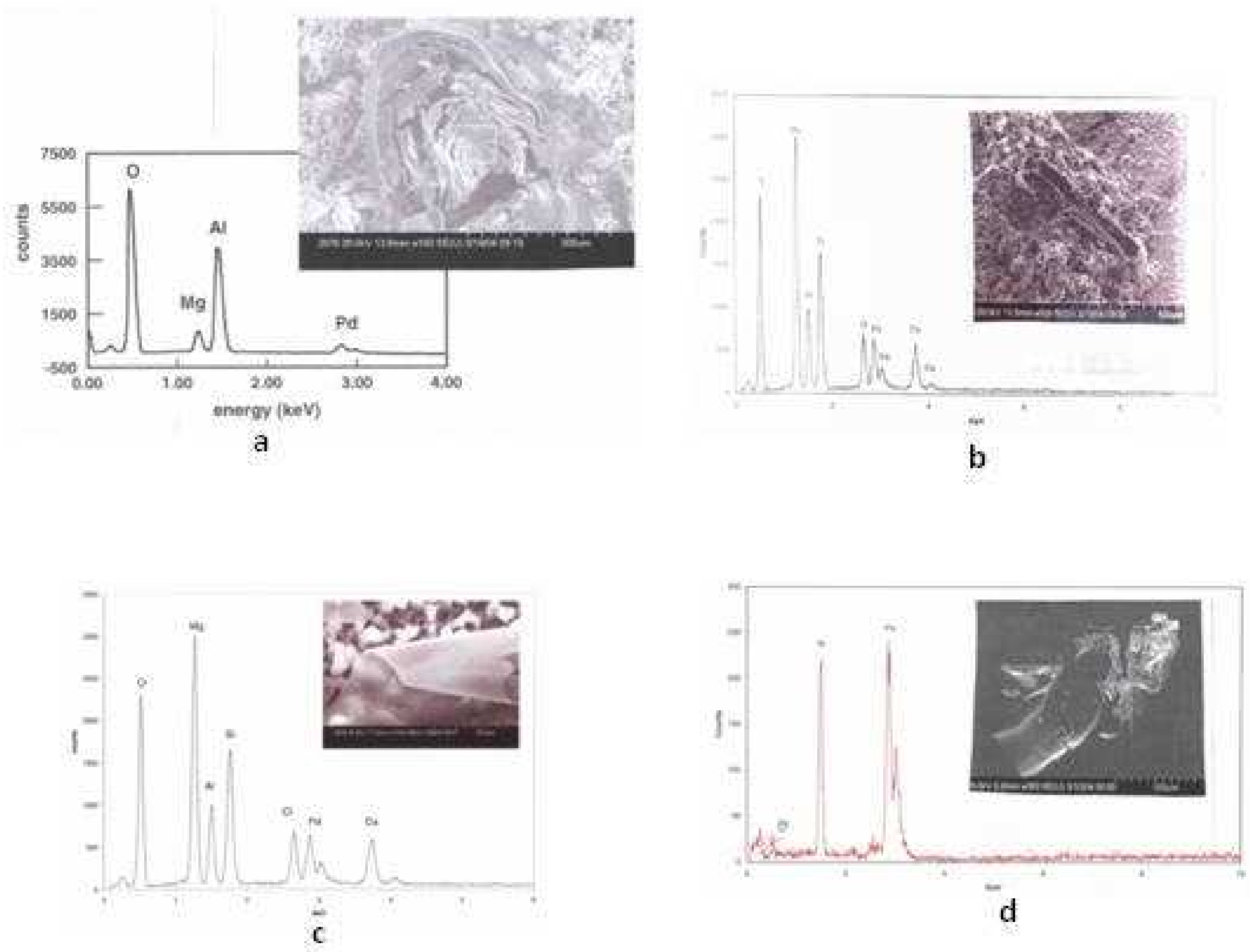
7500
5500
3500
1500
-500
counts
O
Mg
Al
Pd
0.00
1.00
2.00
3.00
4.00
energy (keV)
a
b
O
Mg
Al
Si
Cl
Pd
Ca
c
d

Figure 35:

in Fig. 36a –c, where (i) Fig. 36a shows change in the electrode structure, (ii) Fig. 36b – the EDX results (composition) taken on the same "pancake" and (iii) Fig. 36c – the effect of combined fields, *i.e.* where the cell was exposed to external electric and magnetic fields.

(viii) *Structural change*. Figure 36a shows that the shape change of the ' co–deposited film when placing an operating cell in an external magnetic field is somewhat different from that associated with the electric field – the distinct morphological features (boulders, craters) are replaced by "pancake–like" structures. To emphasize the difference we selected examples that show production of new elements (transmutation) not seen in electric field. The EDX analysis of selected sites shows production of Fe, Cr in addition to Al. Elements such as Mg, Si were found in other samples. As a rule, the number of new elements found is larger than that observed in an electric field.

(ix) *Distribution of new elements – EDX results*. Even a cursory examination of the distribution of new elements differs from that observed on sites generated by an external electric field. As illustrated in Figs. 36b and 36c there is no change in either composition or concentration of new elements sampled in close proximity to each other , Fig. 36b or at some distance, Fig. 36c.

(x) *(c) Effect of combined fields*. The effect of combined electric and magnetic fields, illustrated in Fig. 36c. On the basis of a limited number of observations, it appears that when the co–deposited cathode is exposed to a combined action of electric and magnetic external fields, the magnetic field seems to dominate, *i.e.* the elements usually seen in the case of electric field are eliminated or suppressed.

(xi) Single element, Ca. The transmutation to a singe element, namely calcium, is seen in all cases of thermal run–a–ways. It takes place on isolated spots in the form of spherical blobs, Fig. 37a. or within an area where the Pd/D film is separated from the substrate, Fig. 37b. We have selected this case to demonstrate that nuclear reaction is localized by analyzing the area in the proximity of the spherical blob of Ca.

3.2 Interpretation.

Before offering my interpretation of the transmutation, I considered (i) is the transmutation real or an illusion and (ii) if real, is the production of new elements a key observation, *i.e.* an observation that will point the direction to follow in planing further research?

(i) *Reality or illusion?*. In the introduction to this section we noted that while excess power generation is generally accepted that the same cannot be said about the production of new elements. Thus, we need to answer he question: Is the reported production of new elements real or just a contamination? Arguments against the latter are: (a) Total amount of impurities in 20 mL of electrolyte is about 0.5 mg, *i.e.* too small to be detected, (b) Impurities would be co–deposited during the early stages and uniformly distributed throughout the electrode volume. (c) The segregation associated with the surface transport processes is not likely to occur in the cauliflower–like structure. (d) All cell components were analyzed for the presence of "new elements" – none was

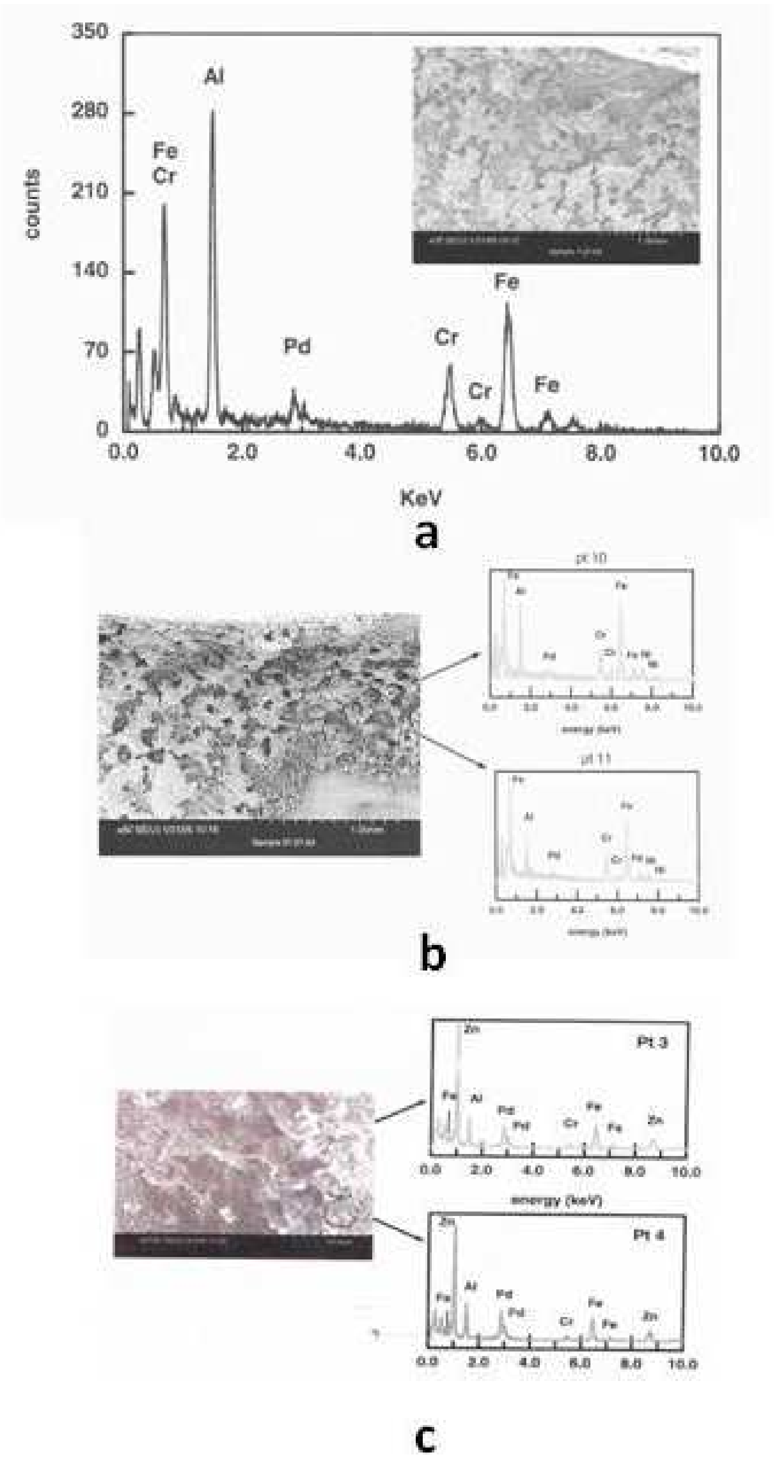
350
280
210
140
70
0
counts
Al
Fe
Cr
Pd
Cr
Cr
Fe
Fe
0.0
2.0
4.0
6.0
8.0
10.0
KeV
a
pt 10
pt 11
energy (keV)
b
Zn
Pt 3
Pt 4
Fe
Al
Pd
Cr
Fe
Zn
energy (keV)
c

Figure 36:

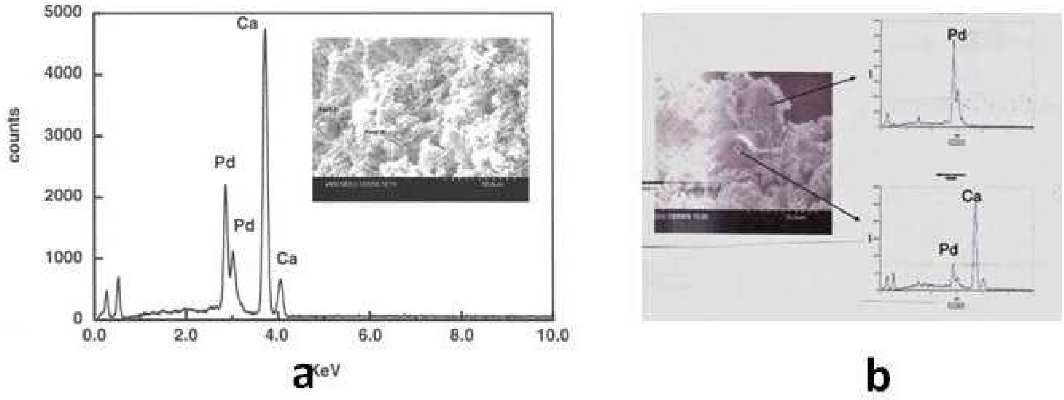

Figure 37:

found[113].

(ii) *Transmutation – a key observation*. In the construction of a model (*cf.* section I) we relied on two observations, *viz* hot spots and production of new elements, and two concepts – self–organization and electron capture by a nucleus. Transmutation, if viewed as a separate problem is interesting but it does not add much to the understanding of the F–P effect. However, when viewed in connection with hot spots, it yields information on the state within the domains based on facts and conclusions derived therefrom. The conclusions arising from localized heat sources were discussed in section IV – here, we analyze (a) the facts leading to conclusions associated with transmutation (production of new elements) *via* (b) arguments considered by some investigators to be speculative.[114]

(a)*Facts:* Exposure to external electric and/or magnetic fields exhibits localized and morphologically distinct features well defined in electric field, *cf.* Fig. 2, less in magnetic, *i.e.* in electric field the variety of shapes – in magnetic flattening of globules with protruding spikes along the peripheries, *cf.* 37a. In particular, (a) Transmutation is found at isolated locations, clearly identifiable in the case of electric field, not so in magnetic field. (b) Transmutation yields single and multiple elements. (c) New elements are stable elements. (d) The effect of an external electric field differs from magnetic (magnetic field produces Fe, Cr, Zn in addition to those seen in electric field). (e) Nuclear reactions are fast (within nanosec range). (f) Fast reactions, expected in case

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[113] Some of the reaction products can be determined quite accurately, e.g. excess enthalpy production and to a degree X–ray and proton emission. The amount of "new elements" can be determined as a fraction (%) of surface but not as a fraction of volume. The isotopic abundance can be determined. Since quantitative evaluation would be expensive, it would appear that at the beginning a qualitative treatment would suffice.

[114] From a chemist's perspective the arguments that follow are consistent with the state of the system, imposed constraints and the nature of electron – nucleus reaction rather than arbitrarily assumed set of approximations. From the physicist's point of view these arguments are pure speculations.

of nuclear reactions, lead to hypothetical situations.

(ii) *Hypothetical:* Transmutation (production of new elements) does not occur in vacuum rather it is just the final step in a series of events of which the initial state is of special interest. In section IV, we described the hot spots as displaying a nuclear character and containing large number of single events and noted that they are fast reactions which, in turn, implies rapid collapse of existing molecular structures. This leads to a set of topics that, we argue, leads to a most probable transmutation mechanism. They are (a) cluster formation and their composition, (b) molecular collapse, (c) transmutation to stable reaction products.

(a) Cluster formation. In II.4.1 we defined cluster is an assembly of aggregates where aggregates represent an assembly of molecules and where localized implies the formation of domains arising from an interaction between the D^+, e^- particles and mobile palladium lattice defects. A cluster can be viewed as two interacting systems that, under non–equilibrium conditions and allowing for energy exchange with the environment, yield an assembly of new molecular structures of aggregates bound together by the van der Waals forces. Here, the effect of an external field is that of the steric factor which affects the structures and composition of aggregates and through it transmutation to multiple stable elements. Moreover, it accounts for the observed difference associated with either electric or magnetic fields.

Equation 2 suggests that the composition of an aggregate is governed by the appropriate rate constants subject to internal and external fluctuations that, in turn, are affected by the steric factor associated with the action of external fields. The situation is further complicated by the interaction with mobile Pd lattice defects.

(b) Cluster stability. In II.4.3 we noted the stability conditions for any system are the same as its equilibrium conditions, *viz* the balance of forces. Now, if a cluster is viewed as an analog of mechanical system, *i.e.* a number of molecular aggregates bound together, then molecular stability requires balance of attractive and repulsive forces. Imbalance within a cluster, aggregate or molecule causes their collapse, *viz* implosion if attractive forces are greater then repulsive and explosion if the repulsive forces are greater. Molecular collapse may involve a cluster or an aggregate or even a single molecule leading to the production of multiple new elements.

(c) Transmutation. It is my position that the generation of "new elements", e.g. Al, Mg, Si, etc. denoted as $\Sigma_{Z_1}^{A_1} X_j$, is, most likely, the product of a series of reactions of the type

$$n + Pd^* \ldots (D_2^+)_i \rightarrow ??? \rightarrow \Sigma_Z^A X_j^* - [p^+, \alpha^{2+}, n, X-, \gamma-] \rightarrow \Sigma_{Z_1}^{A_1} X_j$$

where ??? denotes an unknown process (or a set of processes) preceding the formation of an unstable nuclei, ΣX_j^*. When viewed in connection with hot spots, it suggests that the composition within the cluster is not uniform, *i.e.* that the cluster contains aggregates of species with different number of the Pd ...D_n complexes.

A note of caution – events (processes, reactions) associated with the transmutation constitute only a very small fraction of nuclear reactions. Such conclusion is based on the analysis of reaction products *viz* helium.

4.0 Emission of charged particles

The detection and characterization of particles emitted from polarized Pd/D films, when such films were placed in an external field, yields significant information about the chemistry and physics of the nuclear processes occurring within these films. One way to display the particle emission is to use the CR –39 chips. The discussion of the usefulness of this technique to our work can be traced to Summer of 2003 when Mr Lewis Larsen (Lattice Energy LLC)[115] described the work done in prof. Miley laboratory where CR–39 chips were used to detect and characterize particles emitted from the polarized Pd/D system.

The CR–39 detector is very reliable if correctly used. When a charged particle enters and travels through the CR–39 detector, it leaves a trail of broken molecular chains and free radicals along its track. The damage done to the CR–39 material by the emitted particles can be exposed by etching in concentrated NaOH solution. The condition of etching, e.g. time, temperature and concentration depends on the type of information sougth.[116].

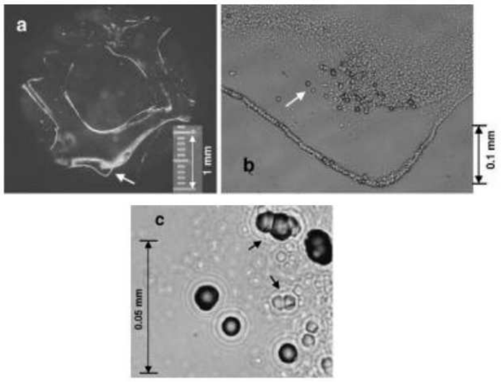

Figure 38:

4.1 Experimental. Our interest in particle emission dates back to 2005 when CR–39 chips were used to detect the emission of particles during the heavy water electrolysis. If CR–39 chips are used to display particle emission, then an appropriate procedure must be developed that would exclude misinterpretation. One such procedure involves Pd+D co-deposition on an open metallic substrate, *e.g.* a screen or a single wire, placed in contact with the CR–39 chip. If the emission of particles from the Pd/D film occurs then they could be detected only along the electrode edge, as illustrated in Fig. 38a.

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[115] At this meeting Mr Lewis was shown a set of SEM photographs, *cf.* Fig. 2, illustrating structural changes caused by placing an operating cell in an external electric field. He admitted that he has not seen anything like that and proposed a joint effort in further work. The "joint effort" ended with him obtaining samples.

[116] The detailed discussion of strengths and weaknesses of the CR–39 sensors can be found in publications by Dr Mosier–Boss[117]

The bright line along the peripheries of a single eyelet is, in fact, an overlap of hundreds of impingement tracks, as displayed in Fig. 38b. In particular, Fig. 38b is an expanded area indicated by arrow in Fig. 38a. Clearly, the bright line represents a set of impingement tracks next to each other while an arrow points to an area of numerous tracks. The images in Fig. 38c show double and triple tracks implying that there are reactions emitting two or three particles having approximately the same mass and energy.

In an attempt to understand the behavior of an operating system, we were concerned with when and how the energetic particles are emitted. Figure 39 shows clusters of tracks recorded after one hour of exposure indicating that coherent domains, arising from self–organization, are formed shortly after placement in an external field. Moreover, the grouping of tracks, being consistent with the hot spots, implies the reactions leading to either transmutation or particle emission, occur together with those producing excess power.

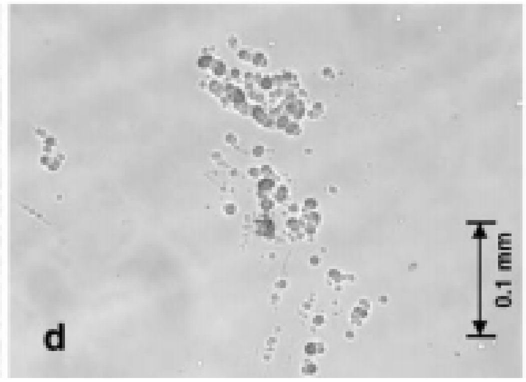

Figure 39:

4.2 Concluding remarks. With the publication of two papers *Evidence of nuclear reactions in the Pd lattice*[118] and *Further evidence of nuclear reactions in the Pd/D lattice: emission of charged particles*[119] the climate in the SPAWAR laboratory has changed. With these publications and considering the advances that we have made, notably in the importance of the electron capture, being the first step in the initiation of the F–P effect, it became obvious that further support was warranted. It was at this point that I decided to change the direction of my research and concentrate on the role of an electron – hydrogen reaction in condensed matter while Drs Gordon and Mosier–Boss on the important aspects of particle emission. Their research covered three topics : (i) CR–39 solid state nuclear detector, (ii) semi–quantitative analysis of emitted particles and (iii) particle distribution.

5.0 The electron and its role in nuclear reactions

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[118] S. Szpak *et.al.*, Naturwissenschaften **92**,394 (2005)
[119] S. Szpak *et.al.*, Naturwissenschaften, **94**,511 (2007)

The Pd/D–D_2O system is an assembly of deuterons, electrons and the Pd lattice defects. When polarized it becomes nuclear active (it yields the F–P effect). It was assumed that the nuclear state is due to d + d fusion and that electrons are involved only in th screening process. The evidence that the electron capture by a deuteron may occur (emission of soft X–rays) was generally ignored. With the passage of time it became obvious that the electrons play a decisive role in the initiation and maintenance of the nuclear activity in the Pd/D – D_2O system.

In September 2006, in the paper submitted to Naturwissenschaften,[120] we suggested that reaction $e^- + d^+ \rightarrow n_2 \rightarrow 2n$, plays significant role in the initiation of the F–P effect. In June 2007, we submitted a manuscript "On the evidence for and origin of nuclear activities in polarized Pd/D–D_2O system" to Zeitschrift fuer Physikalische Chemie.[121] In this paper we suggested a set of nuclear reactions, where the reactants within the reaction volume are: D^+, and neutrons generated by the electron capture and where the reaction products are: excess enthalpy, $\gamma-$ radiation, tritium and helium. The corresponding nuclear reactions are:
tritium production: n + $D^+ \rightarrow {}^3_1H^+ + \nu$
helium–3 production (by β^- decay): ${}^3_1H^+ - e^- \rightarrow {}^3_2He^{2+} + \nu$
helium – 4 production: n + ${}^3_2He^{2+} \rightarrow {}^4_2He^{2+} + \nu$
helium –4 production*via*step ${}^3_1He^+(n,e^-)\,{}^4_2He^{2+}$.

In 2007, the available evidence pointed out that the representation and treatment of the electron capture as a chemical reaction is a probable mechanism. The convincing evidence would be the $e^- + p^+ \rightarrow n$ reaction. This could be done by replacing heavy water by light water because the needed reactants, *viz* protons, electrons and Pd lattice defects, are present. If such reaction was possible and if it was followed by n + $p^+ \rightarrow d^+$, then, after prolonged electrolisis, the system would approach that proposed by Fleischman and Pons. But such behavior was never observed – thus, can the Pd/H–H_2O system be put into its nuclear active state?

5.1 Conditions for electron capture by nucleus. The condition for the electron capture by a proton to occur is the inequality $\mu(e^-) > \mu(n) - \mu(p_l^+) > 0$. Now, all protons located in the reaction volume interact with the Pd lattice to a various degree. The chemical potential of interacting protons, p_l^+ and p^+ species is of the form $\mu(p_l^+) = u(p^+) + u(r)$, where u(r) denotes the degree of interaction. To evaluate the u(r) function we use the relation $\mu(p) = -\varepsilon(p)$, *i.e.* the chemical potential of a particle is its negative binding energy. In this representation, the u(r) function indicates that part of the interacting site is incorporated into the proton itself, *i.e.* it represents the degree of overlap which, in turn, determines whether or not the electron capture by proton can occur. To force the system into its nuclear active state or for an electron capture by a proton to occur, the quantity $\mu(n) - \mu(p^+ +_l)$ must be positive. If the binding energy of the interacting proton is larger than that of a neutron, then the proton will not accept an electron, irrespective of its energy. Now, since the electron capture reaction was

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[120] S. Szpak *et.al.*, **94**, 511 2007)

[121] This paper was send for review on 14 June 2007 by Mr Martin Radke who advised us to check the review status with the Editor–in–Chief, Prof. Dr Baumgaertel. Our attempts to contact prof. Baumgaertel were not successful.

never observed in the Pd/H–H$_2$3O it means that $mu(n) - \mu(p_l^+) < 0$.

5.2 Forcing the Pd/H–H$_2$O system into nuclear active state. The problem is how to do it. Would the placement of an operating cell in an external magnetic field force the Pd/H–H$_2$O into its nuclear active state? To answer, one must consider the magnetic properties of the [Pd*.. ..H]$_n$ domains and their interaction with the magnetic field. [122]

Discussing the effect of magnetic field, (*cf.* II.3.2) we noted that, in addition to the Lorentz forces, the gradient forces are involved. The open question, what system property is affected? In seeking an answer we used the Pd/H–H$_2$O system and examined the open circuit rate of hydrogen desorption.[123] The data presented in Fig. 40 show a typical potential/time curves for the desorption of hydrogen. In particular, they show that in absence of an external magnetic field, curve a, the electrode potential/time relation, $\Phi(t)$, shows three distinct time periods indicating three different desorption rates. At first, there is a linear increase of the electrode potential with the slope of 2.2×10^{-5} Vs^{-1} followed by the transition to much higher rate , segment BC, and asymptotically approaching its equilibrium potential. Upon application of an external field, point P curve b, the linear segment of the $\Phi(0,t)$ is much slower and is followed by the transition, segment B'C', to its equilibrium potential. Such behavior shows that proton transport controls the rate of hydrogen desorption while, when in magnetic field, the contact surface is involved. Or, putting it differently, magnetic field weakens the proton/Pd defect interaction.

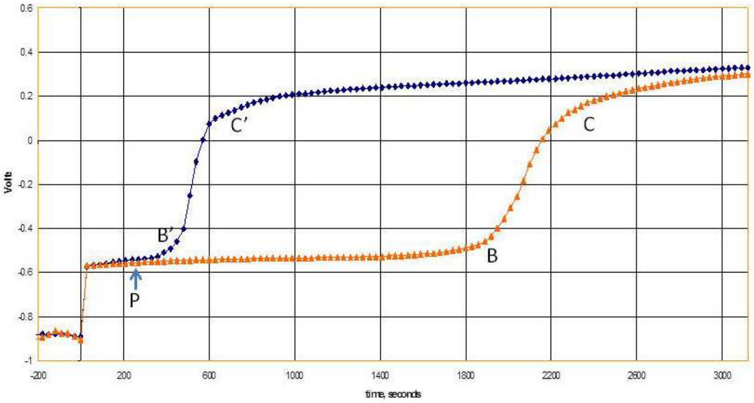

Figure 40:

5.3 Concluding remarks. An external magnetic field, as well as the modulated cell cur-

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[122] The measure of the interaction of the magnetic field with a substance is given by its susceptibility, χ, *i.e.* by its receptiveness for lines of magnetic force. A substance with $\chi > 0$ is called paramagnetic and palladium shows very high susceptibility ($\chi = 5.72 \times 10^6$) which decreases linearly with increase in hydrogen concentration reaching zero at H/Pd atomic ratio of 0.65. B. Svensson, Annalen der Physik, **18**, 299 (1933).

[123] S. Szpak and J. Dea, Unpublished data (An attempt to publish was unsuccessful because, paraphrasing the editors comment “just another cold fusion paper in disguise”.

rent, affect the system's dynamic equilibrium by the change in surface forces and,*via*the Gauss theorem, the distribution of stress fields within the interphase, thus creating a new state of dynamic equilibrium that results in an increase of active protons, *i.e.* protons that can participate in the electron capture. Experience shows that external magnetic field is more effective and that, when combined with the modulated cell current, results in nuclear activity whose rate is only slightly less than that observed in heavy water.

(i) The $e^- + p^+ \rightarrow n$ reaction. Although the reaction that drives the Pd/H–H_2O system into a nuclear active state is the electron capture, the addition of a neutron to proton is far more relevant from both theoretical and practical point of view. The radiative capture of a neutron by proton is the most common nuclear reaction in which the compound nucleus emits only a 2.2 MeV γ photon to produce a stable isotope of the same element; it transmutes hydrogen into deuterium and deuterium into tritium. It is noteworthy that no hard radiation was observed in this system or in the related Pd/D–D_2O system. This is an expected behavior in view of the theoretical arguments presented by Widom and Larsen. They concluded that the polarized Pd/^{n}H–^nH_2O (n=1,2) system has built–in $\gamma-$ shields that prevent the emission of the 2.2 MeV γ radiation. In particular, the shielding is the transfer of energy from $\gamma-$ photons to electrons.

SECTION VI: EPILOGUE

Two dates are important: 23 March 1989 and 1 May 1989. On the 23-rd March a new phenomenon of potentially great importance was disclosed – on the 1-st of May it was declared an illusion by uninformed professors in a well known institution of higher learning. On the 23 March, professors Fleischmann and Pons presented evidence of a new phenomenon, that of a nuclear process occurring in the test tube at room temperature. On the 1-st of May, professors Koonin and Lewis, in an unfriendly, or better, unprofessional manner,[124] expressed their views without first examining the claims made, because these claims did not fit into their understanding of the underlying physics. By doing so, they effectively slowed research leading to the full understanding of this phenomenon.

Today, 20 years later, we can say with certainty that the effect is real and that the position taken by the American Physical Society during their May 1989 Baltimore MD meeting was wrong. Moreover, latest results show that the *grandeur* predicted by Preparata is near. Indeed, on the basis of a sequence of observations and guided by the *Nature's documents*, the hot spots and production of new elements, we arrived at what we believe to be a realistic model (*cf.* Section I).

1.0 Challenges

There are two sets of challenges, *viz* technological issues and information dissemination. The first involves the formulation of research programs concerned with the control of the output, *e.g* excess power, deuterium, tritium and neutron production and the second, to bring the Fleischmann–Pons discovery back into the main stream science. This, of course, would involve information dissemination *via* publications in journals and presentations at meetings.

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[124] S. Koonin –"My conclusion is that we are suffering from the incompetence and perhaps delusion of Drs. Pons and Fleischmann"; N. Lewis "And what we see in our laboratory is, no evidence for any unusual nuclear or chemical reaction" expressed at the APS Spring meeting, Baltimore MD; D. Goodstein "All three scientists from Caltech executed between them a perfect blocked shot that casts cold fusion right out of the area of main stream science" quotes taken out from the lecture given by Prof. B. D. Josephson at the Nobel Laureates' meeting 30 June 2004

1.1 Technological issues. In the area of thermal behavior, the transition to engineering activities is long overdue. In the area of nuclear output, it is too early to pass sound judgment. The transition from research to engineering would require additional work involving cell design, in particular the co–deposition process and the transfer of power from cell to the work station. Further development of the co–deposition is needed to increase their effectiveness. Also electrolyte flow pattern would have to be examined in great detail to assure cell's maximum performance. Briefly. the proposed model, *cf.* I.6.0, which represents rather well the system's behavior, is incomplete inasmuch as it does not identify the exact nature of processes/reactions involved. However, it appears that in order to control the cell's output the response of the interphase to external stimuli must be given priority. Two research programs, that appear to be feasible, are (a) design of cell employing fluidized bed cathodes and (b) how to control reaction rates at point close to the thermal run–a–way. The latter applies to the behavior of the Pd/H–H_2O system.

1.2 Information dissemination. While the scientific side of the F–P effect was the subject of this work, not much has been said about how to force its return to the main–stream science. Traditionally this would be done *via* publications in journals and presentations at meetings. The route to follow is outlined in an article *The politics of publications*[125] where one finds that publication involves interaction between authors, editors and reviewers – each has a specific responsibility. Authors submit results of their work, editors accept or reject submitted material. Reviewers, in the course of review, often face conflict of interest, e.g. they may delay response to assure priority in publication of their results on the same subject.

This is a normal procedure. A completely different set of rules seems to apply to submitted material dealing with the F–P effect. The F–P effect is, at the present time, dismissed in spite of overwhelming evidence in its favor. In reality, it is not the question of truth of the claims made therein, rather their denunciation takes over by reviewers, who often have no familiarity with the subject matter. When in the denunciation mode, they use "made–up" arguments instead of scientific reasoning. Frequently used expression: *Extraordinary claims require extraordinary evidence*, means that an acceptable proof can be stretched without limits, effectively blocking the publication of submitted material by requiring never–ending demands of additional evidence, often irrelevant.

In writing a paper, one is faced with two choices – send the message or be a messenger. In the first case one communicates results of one's research; in the second case, one wishes to be an active partner in particular research activities. In what follows, we describe in some detail our experience in trying to send a massage extending the F–P effect to the Pd/H–H_2O system and participate in the discussion concerning the "recombination theory". Both communications were rejected.

2.0 Sending a message

The message: The Pd/H–H_2O system can be put in a nuclear active state by (a) placing an operating cell in external magnetic field, (b) using modulated cell current, or (c)

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[125] P.A. Lawrence, Nature, **422**, 259 (2003)

both for increased effectiveness. A paper *On the induction of nuclear activity in polarized PD?H–H_2O* (reproduced below) was submitted to *Journal of Radioanalytical & Nuclear Chemistry* manuscript identification: JRNC1259.

ON THE INDUCTION OF NUCLEAR ACTIVITY IN POLARIZED Pd/H–H_2O SYSTEM

Stanislaw Szpak · Jack Dea

Abstract. In cells employing cathodes prepared by the co–deposition process, the polarized Pd/D–D_2O system becomes nuclear active when the concentration of deuterium, expressed as D/Pd atomic ratio, is equal to or greater than one. In contrast, to activate the polarized Pd/H–H_2O system, action of an external magnetic field, modulation of cell current or both, are required. Evidence for the nuclear active state in the Pd/H–H_2O system, namely impingement tracks, deuterium production and catastrophic thermal event, is presented and discussed.

Key words Palladium Magnetic field Nuclear activity Protons Deuterons Neutrons

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1.0 Introduction

On the 23rd March 1989 Fleischmann and Pons announced, in a press conference, that nuclear events can be initiated and sustained at low temperatures and pressures by electrochemically compressing deuterium in the palladium lattice. The discovery of a nuclear active state in polarized Pd/D–D_2O system did not happen spontaneously, rather it was the outcome of a long chain of theoretical considerations and empirical results. An account of these activities can be found in publications by Fleischmann[1] and his collaborators[2]. Briefly, these activities led to the formulation of three fundamental questions: (i) Would the reaction involving deuterons compressed into a Pd lattice be different from corresponding reactions in dilute plasma, (ii) could such reactions be observed and (iii) by what course of action a low energy chemical process can generate high energy response? Evidently, in 1989, the answer to (i) and (ii) was yes because at that time, Fleischmann and Pons started their research in “cold fusion”. They selected calorimetry as the principal tool to measure the extent of nuclear activities and a solid palladium wire as the cathode material in which the electrochemically generated deuterium was compressed.

The compression of deuterium by a slow diffusion controlled process can be replaced by a fast charging associated with the co–deposition process, a process in which the Pd^{2+} ions are electro–deposited onto a substrate (e.g. Au foil) in the presence of evolving deuterium[3]. Electrodes prepared by the co–deposition process retain all features of massive electrodes[4]. Moreover, in electrodes prepared by the Pd+D co–deposition process, the transition from an inactive to active state occurs during co–deposition as well as during electrolysis of D_2O at both, low (0.001 Acm^{-2}) and high (0.4 Acm^{-2}) current densities. By placing an operating cell in an external electrostatic or magnetostatic field, the range of nuclear reactions is extended to include the production of new elements[5] and emission of particles[6].

It is well documented that the Pd/D–D_2O system becomes nuclear active if (i) it is polarized and (ii) atomic ratio $D/Pd > 1.0$ is obtained. A question that arises: Is the induction of a nuclear active state limited to the Pd/D–D_2O system or can it be extended to the Pd/H–H_2O system as well? This communication discusses conditions that must be imposed upon it to induce nuclear activity and covers chemical aspects only.

2.0 Experimental

Three key elements of the experimental protocol, described here, are: (i) cell construction and operation, (ii) detection of neutron and (iii) deuterium production.

2.1 Cell construction and operation

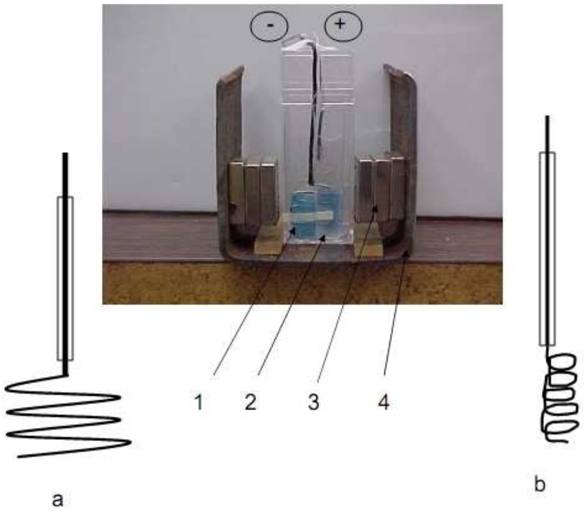


Fig. 1 Electrochemical cell. 1–outside CR–39 detector, 2 – double CR–39 stock located inside the cell, 3 – neodymium magnets, 4 – magnet holder. Cathode and anode identified by - and +; cathode – a designed for neutron detection, b – designed for determination of deuterium content.

A rectangular cell, made of clear plastic, with affixed neutron detectors, (CR–39 chips) and electrodes connected to a galvanostat (Princeton Applied Research, model 363) and cathodes prepared by co–deposition was placed in an external magnetostatic field. Upon placing an operating cell (with i = - 30 $mAcm^{-2}$) in a magnetic field, Fig. 1, the electrochemical compression of absorbed hydrogen was put under computer controlled regime (LabView program) with i = -400 $mAcm^{-2}$ for 90 seconds and i = 5.0 $mAcm^{-2}$ for 5 seconds. The fabrication of cathodes involves co–deposition from a solution of 0.03 M $PdCl_2$ and 0.3 M LiCl dissolved in H_2O at i= -1.0 $mAcm^{-2}$ for the first 24 h followed by i = -3.0 $mAcm^{-2}$ for the time required to reduce all the Pd^{2+} ions; (b) a 3 – 4 h stabilization period at i = - 30 $mAcm^{-2}$, *i.e.* the time needed to assure uniform distribution of absorbed hydrogen throughout the electrode volume.

2.2 Neutron detection

The CR–39 chips served as detectors of nuclear activity. A stock of two CR–39 chips was placed just behind the cathode and a third CR–39 chip outside the cell, as illustrated in Fig. 1. The zig–zag shape of the cathode was used to assure approximately uniform access to CR–39 chips of emitting neutrons. Upon the completion of electrolysis, the CR–39 chips were processed using standard procedure of etching in 6.5 NaOH for 6 hours at 70C.

2.3 Deuterium detection

The mass spectrograph was used to analyze for deuterium. The Pd/H film co–deposited onto a thin coiled palladium wire was employed to assure retention of the electrochemically compressed hydrogen isotopes, Fig. 1b. This electrode design was selected to meet the requirements imposed by mass spectrograph. A note of caution: To minimize the desorption of hydrogen isotopes the following procedure was adapted (a) remove the cell from magnetostatic field, (b) stop the cell current flow, (c) take out the cathode, (d) cut–off the coiled part of the cathode and (e) analyze the coiled part for hydrogen isotopes as soon as possible (e.g. within 15 minutes).

3.0 Results

In what follows, we present direct evidence of nuclear activity in the polarized Pd/H–H_2O system, viz neutron emission, deuterium production and, an indirect evidence, that of the occurrence of catastrophic thermal event.

3.1 Neutron emission

Figure 2 shows typical images of tracks recorded on CR–39 chips. It is seen that the difference is not in the type of tracks but in their number, the latter being highest on the CR–39 detector facing the cathode and the lowest on the chip located outside the cell. In particular, Fig. 2a shows a typical distribution of images of circular and elliptical tracks, Fig. 2b and 2c illustrate the case of ionizing particle entering either perpendicular to the detector's surface or at an oblique angle, and Fig. 2d a double track. The physical meaning of the images recorded by CR–39 detectors is discussed in detail by Mosier–Boss *et.al.*[7].

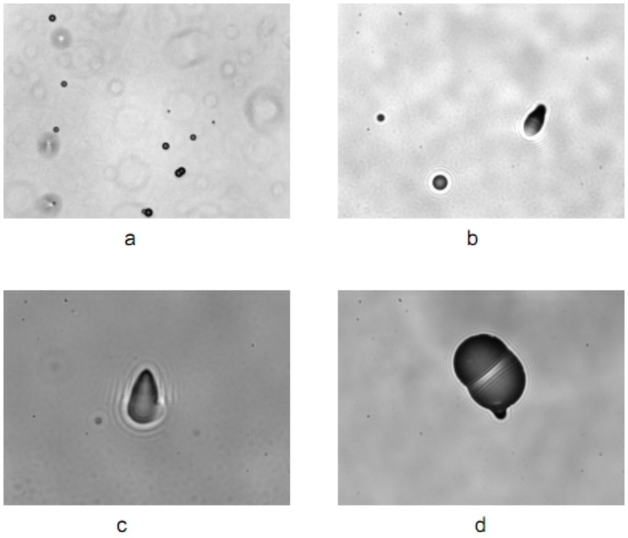


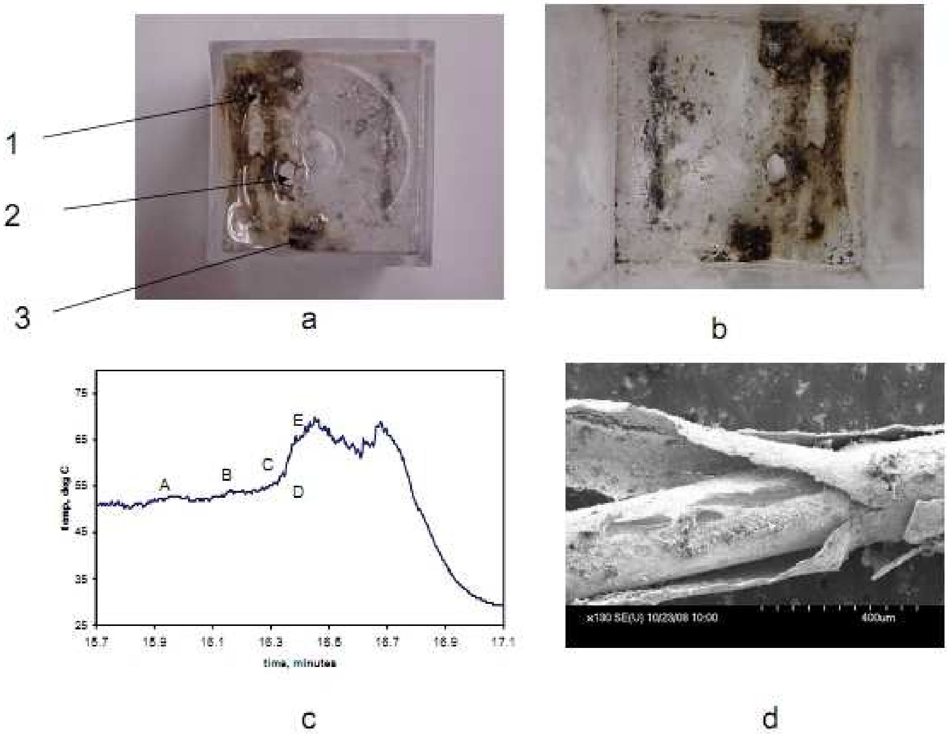
Fig. 2 Images of tracks in CR–39 detectors created in the course of an experiment; a – distribution of tracks at 40× b –illustrates the angle of impingement at 500× c – shows a single track at 1000× d – image of a double track at 500×

3.2 Deuterium production

The mass spectroscopic analysis, performed upon completion of a run, showed the presence of all hydrogen isotopes. Qualitatively, deuterium was the dominant isotope with negligible amounts of tritium. Typically, the D/H atomic ratio greater than one, with a value as high as 5.1, was recorded. Needless to say, that the presence of deuterium in the cathode is of utmost importance because it might provide decisive insight into the mechanism of nuclear reactions in condensed matter. As a rule, mass spectroscopic analysis yields results that are unambiguous. However, if additional identification is required, then this can be done by (i) changing the method of analysis while retaining the sampling procedure or (ii) employing the original procedure (e.g. mass spectroscopy) and analyzing samples with known D/H atomic ratios. Here, the latter was employed in which the desired D/H atomic ratios were obtained by electrolysis of a mixture of H_2O and D_2O in corresponding proportions using thin palladium wires as cathodes.

3.3 Catastrophic thermal event

In the course of investigation several catastrophic thermal events were observed. In one case, after three days of electrolysis with cell current varying between -300 $mAcm^{-2}$ and 5 $mAcm^{-2}$, a catastrophic thermal event has occurred that resulted in cell deformation, loss of electrolyte due to evaporation and leaking through a punctured cell bottom, Fig. 3. The damage, about 1/3 of total area, viewed from the outside, Fig. 3a, and inside the cell, Fig. 3b, is consistent with placing a very hot object in contact with plastic material.



a b

c d

Fig. 3 Damaged cell bottom a – outside view b – inside view Arrows indicate well defined damage areas (1 – thinning, 2 – hole, 3 – embedded particles) c – solution temperature during thermal run–a–way d – separation of Pd/D film from Pt substrate

4.0 Discussion

The induction of a nuclear active state in the polarized Pd/H–H_2O system was demonstrated by neutron and deuterium production. The sequence of events leading to the initiation of the nuclear active state is shown in a diagram, Fig. 4. Here, the events within the metal side of the interphase are: an exchange between the adsorbed and absorbed hydrogen, H_{ad}, H_{ab}, the transition from atomic to nuclear state, denoted $H_{ab} \rightarrow p^+$, followed by $p^+ \rightarrow p_l^+$, the latter identifies a proton interacting strongly with the Pd lattice. When an external magnetic field, ψ, is applied, a new set of processes can be identified, *viz* weakening of the interaction, $p_l^+ \rightarrow p_*^+$, electron capture by proton, $e^- + p_*^+ \rightarrow n$, with neutrons either escaping, n_1, or reacting with proton to yield deuteron, $n + P_*^+ \rightarrow d^+$. The simplest, and the only, reaction that yields neutrons in this system is the electron capture by proton.

Since all processes are influenced by the environment in which they occur, the construction of a workable model requires information on the structure of the reaction volume, the type of reaction and the effect of external factors such as overpotential, magnetic field or modulated cell current.

4.1 Reaction volume

The processes, shown in Fig. 4, occur within the interphase region, a region separating two homogeneous phases, where rates are controlled by the nature of interacting sites (lattice imperfections, defects).

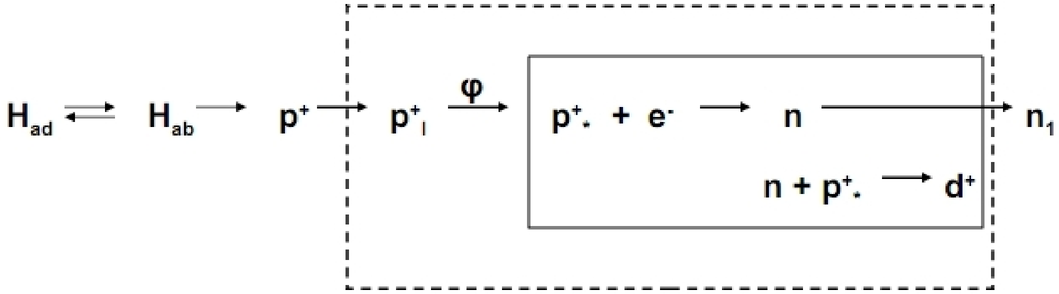


Fig. 4 Schematic diagram showing events within the interphase. Enclosed by broken line are processes affected by an external field (cf. *4.3); by solid line – coupled nuclear reactions (*cf. *4.5 and 4.6.2).*

Within the metal side of the interphase, hydrogen is distributed as follows: the sub–surface hydrogen, in the top–most layer of Pd atoms provides link between the adsorbed and dissolved hydrogen – with the latter interacting with the lattice. With adsorption there is associated surface reconstruction but only the hydrogen in the top–most Pd layer is responsible for its maintenance. Any motion of hydrogen within the interphase generates stresses that, in turn, produce dislocation and other types of interaction sites. That it is to say, within the interphase exists a state of dynamic equilibrium which governs the distribution of hydrogen interacting with the palladium lattice which means that some interaction sites are formed, other disappear thus releasing the interacting protons and making them available for the electron capture reaction (*cf.* 4.4). Corollary – one way to increase the number of non–interacting protons is to continuously change the structure of the interphase*via*changes in the cell operating constrains.

4.2 Electron capture reaction

Electron capture by nucleus is not a new concept. Moreover, nuclear reactions of the type

$$e^- +_Z^A (X) \rightarrow_{Z-1}^A (X) + \nu \tag{27}$$

can be treated as chemical reactions[8]. Consequently, it is not difficult to write down the thermodynamic conditions which govern the chemical equilibrium. Since the neutrinos are not retained by matter, the chemical potential of the neutrinos will not appear in the equation of equilibrium or reaction kinetics. Thus, for the reaction, Eq 1, to occur in the indicated direction, the required condition is the inequality $\mu(e^-) > \mu({}_{Z-1}X) - \mu({}_{Z}X)$. Substituting a proton, p$^+$, for the ${}_{Z-1}(X)$ nucleus, one gets the e$^-$ + p$^+$ → n reaction with conditions for its occurrence: $\mu(e^-) > \mu(n) - \mu(p^+)$.

4.3 External factors

A system described by Eq. 1 is the usual representation of a chemical reaction. As written it provides only limited information, *viz* it is the statement of conservation of mass, energy and charge. Since the initial and final states are not specified, it simply means that the system consists of unbounded particles in the sense that there is a continuous range of possible energies. The unspecified range of possible energies as well as the rate of reactions are controlled by overpotential and external magnetostatic field. The overpotential affects the chemical potential of hydrogen (both in atomic and nuclear state)*via*energy stored in the electric double layer. The effect of magnetostatic field is more complex. Briefly, the interaction of a magnetic field with electrochemical systems can be divided into three main areas: (i) magneto–hydrodynamic effects, *i.e.* those affecting mass transport*via*the reduction of the diffusion layer thickness and through it, the morphology of the surface, (ii) magneto–mechanical effects. *i.e.* those that involve the shape change of micro–globules as well as complex macro–molecules, and (iii) non–specific interactions of electronic nature, *i.e.* those affecting dynamics of the highly concentrated hydrogen in the Pd lattice. These effects are attributed to forces generated by the gradients of magnetic energy density, the paramagnetic gradient and the field gradient forces, *i.e.* by forces that that arise from non–homogeneity of the paramagnetic entity and those associated with non–uniformity of the magnetic field. Briefly, the effect of both, overpotential and magnetic field, is to promote the p$_l^+$ → p$^+$ transition (*cf.* Fig. 4).

4.4 Nuclear reactions in the Pd/H–H$_2$ system

Accepting the electron capture concept and considering condition within the interphase (local electric fields on the order of 10^9 Vcm^{-1} are not unusual), one would expect that, during electrolysis of H$_2$O in cells employing palladium cathodes, nuclear active state should be generated – but none was ever observed. However, if the cell, employing cathode prepared by the co–deposition process, is placed in an external magnetic field or subject to modulated cell current, then neutrons are emitted and deuterium is produced. An external magnetic field as well as the modulated cell current affect the dynamic equilibrium by the change in surface forces and,*via*the Gauss theorem, the distribution of stress fields within the interphase, thus creating a new state of dynamic equilibrium that results in an increase of active protons, *i.e.* protons that can participate in the electron capture. Experience shows that external magnetic field is more effective and that, when combined with the modulated cell current, results in nuclear activity whose rate is only slightly less than that observed in heavy water.

4.4.1 The e$^-$ + p$^+$ → n reaction. The condition for the electron capture to occur is the inequality $\mu(n) - \mu(p^+) > 0$. Now, all protons located in the reaction volume interact with the Pd lattice to a various degree. The chemical potential of interacting protons, p$_l^+$ and p$*+_*$ species is of the form $\mu(p_l^+, p_*^+) = \mu(p^+) + u(r)$ where u(r) denotes the degree of interaction. To evaluate the u(r) function we consider the relation $\mu(p) = -\varepsilon(p)$, *i.e.* the chemical potential of a particle is its negative binding energy. In this representation, the u(r) function indicates that part of the interacting site is incorporated into the proton itself, *i.e.* it represents the degree of overlap which, in turn, determines whether or not the electron capture by proton can occur. If the overlap is greater than that indicated for p_l^+, then the proton will not accept an electron, irrespective of its energy. For an electron capture by a proton to occur, the quantity $\mu(n) - \mu(p_*^+)$ must be positive, *i.e.* the overlap must be less then that for p_l^+.

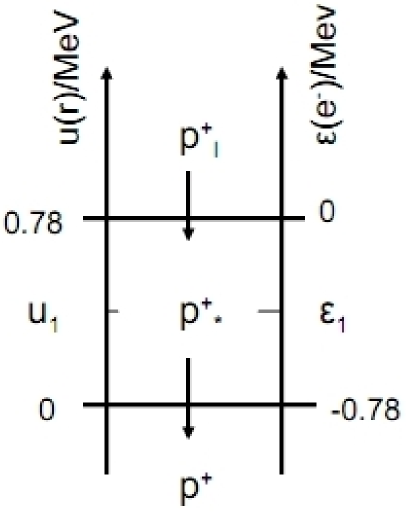


Fig. 5 Diagram illustrating the dominant role of proton/lattice interaction in promoting nuclear activity in the Pd/H–H$_2$O system

The inequality $\mu(e^-) > \mu(n) - \mu(p_*^+) > 0$ implies that there is a set of possible energies of electrons that can react with an interacting proton, p_*^+ which, in turn, is governed by the value of the u(r) function. This is illustrated in Fig. 5, where on the left side the degree of overlap and on the right side the quantity $\mu(p_*^+ - \mu(n)$ [or $\varepsilon(e^-)$] are plotted. Briefly, for u(r) $>$ 0.78 MeV, because the chemical potential of the interacting proton exceeds that of neutron, electron capture cannot take place. Within the range $0 <$ u(r) $<$ 0.78 MeV, for each value of u_1(r) there is associated value of ε_1 which must be exceeded for the reaction to occur. In general, as the u(r) decreases, *i.e.* as the strength of interacting proton decreases, the energy added to activate electron increases reaching the value of - 0.78 MeV, a value corresponding to the rest mass defect, for non–interacting proton.

4.4.2 The $n+p^+ \rightarrow d^+$ reaction. Although the reaction that drives the Pd/H–H$_2$ system into a nuclear active state is the electron capture, the addition of a neutron to proton is far more relevant from both theoretical and practical point of view. The radiative capture of a neutron by proton is the most common nuclear reaction in which the compound nucleus emits only a 2.2 MeV γ photon to produce a stable isotope of the same element, here it transmuts hydrogen into deuterium and deuterium into tritium. It is noteworthy that no hard radiation was observed in this system or in the related Pd/D–D_2O system[9]. This is an expected behavior in view of the theoretical arguments presented by Widom and Larsen[10]. They concluded that the polarized Pd/^{n}H–^{n}H$_2$O (n=1.2) system has built–in γ– shields that prevent the emission of the MeV γ radiation. In particular, the built–in shield is the transfer of energy from γ– photons to electrons.

4.5 Coupled reactions.

The photon to electron transfer of energy provides for the coupling between two reactions: production of neutrons and deuterons. Thermodynamic tells us that, for the electron capture by proton to occur, it must acquire sufficient energy and interact with a proton. However, by arguments presented in 4.4.2 and illustrated in Fig. 5, the production of the p_*^+ specie initiates nuclear activity, indicating that the needed electron energies are readily available. Once initiated, the nuclear activity is maintained by two simultaneous reactions, that of neutron and deuterium production, with the first proceeding in the non–spontaneous direction and requiring energy input, the second exothermic. Here, we have a situation where the electron capture reaction produces a substance that is used up in another reaction with the liberation of energy which, in turn, accelerates the first reaction. Such dependence is referred to as coupling, more accurately the first reaction is coupled the second coupling. Putting it differently, the first reaction, that of electron capture, proceeds in the direction opposite to that dictated by the Clausius un–compensated heat of reaction, dQ' = $\mathbf{A} \times \mathbf{v} < \mathbf{0}$. Conditions for the existence of such coupling was discussed by van Rysselberghe[11] who showed that this is possible if (i) there is another reaction, the coupling reaction (dQ' = $A_2 \times v_2 > 0$) occurring in the direction dictated by its affinity and (ii) if the sum $A_1 \times v_1 + A_2 \times v_2 > 0$, *i.e.* when the coupling reaction proceeds at sufficient rate and where the needed energy is supplied by the coupling reaction.

4.6 Catastrophic thermal event

An indirect evidence of nuclear activity, presented in Fig. 3, covers (i) cell damage assessment, (ii) evolution of nuclear activity and (iii) mechanism of the Pd/H film separation.

4.6.1 Cell damage assessment. Even a cursory examination of Figs. 3a and 3b of the cell damage, *i.e.* shape change and wall puncture, suggests that a very hot object contacted the cell bottom. The black powder, firmly attached to the cell bottom, indicates that the Pd/H film exploded and hot fragments were deposited onto the cell bottom. The location of the black residue around relatively clear area, Fig. 3a, point 1, suggests that explosive fragmentation occurred after large segment of the Pd/H deposit hit the surface, while the wall deformation implies that sufficiently high temperature softened the acrylic plastic. These facts suggest that it is reasonable to assume a non–chemical energy source and that it remains active heat source even after Pd/H film detachment from the platinum substrate.

4.6.2 Evolution of nuclear activity. The evolution of nuclear activity in the Pd/H–H_2O system can be followed by measurement of the heat output. Initially, under the action of an external magnetic field, the transition $p_l^* \rightarrow p_*^+$ takes place and, as indicated (*cf.* 4.4.1), reaction $e^- + p_*^+ \rightarrow n$ occurs. With the passage of time, the second, highly energetic reaction $n + p_*^+ \rightarrow d^+$, tales place to become the primary energy supplier for the first reaction. Here, we have a situation in which the first reaction is accelerated by its product*via*the energy supplied by the chemical change in the substance (here neutrons) which induces the first reaction. Restating: the first reaction is induced by the second reaction*via*the energy transfer. Such kinetics is referred to as induction kinetics[12]. Returning to Fig. 3c, the induction effect became substantial, section ABC, changing to catastrophic, section DE.

The intensity of the heat source can be estimated from the temperature raise of the electrolyte during the last 170 minutes of cell operation, Fig. 3c. In a separate run, a cell, charged with 3.0 ml of 0.03 M $PdCl_2$ and maintained at a constant volume of 5.0 ml, was operated for two days before recording the temperature, with the thermocouple located below the cathode. Within the last 160 minutes the electrolyte temperature remained constant (at 50 C followed by, at first, a slow raise for the next four minutes, Fig. 3c, section ABC, followed by a rapid increase, at 2.6 C/sec., Fig. 3c section DE. During the next 3 – 4 minutes, the electrolyte evaporated and the temperature returned to ambient. For this run, the intensity of the heat source, based on the heat generated by the minute amount of the PD/H film and transferred to the electrolyte, can be estimated to be more than 10 eV/Pd atom, *i.e.* outside the limits of chemical reaction. Furthermore, the temperature raise of the source of ca 250 C/sec. means that substantial amount of the electrolyte was lost by "film boiling".

4.5.3 Mechanism of the Pd/H film separation. A probable mechanism of Pd/H film separation from the Pt substrate can be deduced using photograph, Fig. 3d, representing film separation in another experiment. In this particular case, the cell was charged wit 0.03 M $PdCl_2$ dissolved in heavy water and operated for several days. The set of events associated with the observed damage, illustrated in Fig. 3d, was as follows: (i) a number of localized reactions at the Pt/Pd/D interface forced the separation of the Pd/H deposit (Dea 2009 unpublished). By analogy, the damage, shown in Figs. 3a and 3b, indicates that a Pd/H sleeve, approximately 1 cm long, was separated and propelled away from the cathode to come in contact with the cell bottom. The high temperature and the amount of thermal energy necessary to generate the observed damage implies that nuclear reactions did not terminate upon separation from the cathode but occur during flight as well as after landing at the cell bottom (the "life–after–death situation"[4]).

5.0 Concluding remarks

In conclusion, we note the following:
(i) The negatively polarized Pd/H–H_2 is the simplest system involving nuclear reactions in condensed matter.
(ii) Its initial and final states are well defined, thus simplifying analysis and permitting standard interpretation of observed facts.
(iii) It sheds new insight into the nature of nuclear reaction in condensed matter.

(iv) Chemical approach rather than the ensuing physics is preferred in studying the mechanism of nuclear processes that occur within the confines of the palladium lattice.
(v) The transition $p_l^+ \rightarrow p_*^+$ is the low energy process that generates high energy response.

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2.1 Reviewers comments

Reviewer Nr 2 comments missing

Reviewer Nr 1: In view of the history, ably summarized by the authors, of self–delusion by researchers of nuclear reactions induced by non–nuclear means, the present manuscript would be acceptable only if all questions of experimental conditions were answered. That is far from the case: this manuscript cannot be published without grave harm to the reputation of JRNC.

The authors' qualitative explanations of the observed phenomena are unconvincing, invoking many questionable assumptions such as ignoring the existence of neutrinos, capturing an improbably large fraction of the putative neutrons in hydrogen, and absorbing the 2–MeV capture of gamma rays in situ. But none of this is relevant without hard evidence that there is a phenomenon to explain. They claim to have detected neutrons and deuterium but give no numbers with which the reader can judge the reality of the assertion. No details are given on the mass spectrometry methods, sensitivity, blank experiments, number of experiments. Similarly, the CR–39 observations are worthless without statistically valid comparison with cosmic ray background.

This reviewer must regretfully conclude that the authors have not made him a believer in electronuclear reactions. It would be so exciting if true.

Reviewer Nr 3: It is beyond my comprehension how the fusion H and H (generally limited to the core of the sun) can occur without the availability of necessary energy. The evidence for neutron is not convincing, because in my opinion a CR–39 chip is not a characteristic detector for the neutron. If the authors could demonstrate the formation of a radioactive product*via*the (n,γ) reaction then the readers of this journal would be convinced. Similarly, mass spectroscopic analysis of deuterium may be genuine but the occurrence of impurities must be absolutely eliminated. In short, as a nuclear chemist I am not convinced. My suggestion is that the authors submit this work also to Naturwissenschaften, as they did in many other cases.

2.1.1 Review of reviewers work

Reviewer Nr 1. Evidently, the reviewer Nr 1 suffers from an acute case of the pathological disbelief. All classical symptoms are there: disrespect (self–delusion of researches), explanation and interpretation are rejected because they are not convincing (declaring thermodynamic arguments as rubbish), manuscript should be rejected because it does not answer all questions including those that were not asked (all questions?). And there is a desire to protect good name of the journal (grave harm to the reputation of JRNC). It should be noted that even if our announcement was an example of pathological science – which was not – it would not cause “grave harm”.

Another, rather curious observation – the reviewer did not accept views published in refereed journal. Should all published material discussing the 2.0 MeV gamma rays or the appropriateness of treating the electron capture as a chemical reaction be disregarded? He concluded the review with “It would be so exciting if true”. Although not his responsibility the reviewer could go to the lab, or have someone else to do so, and perform this simple experiment.

Reviewer Nr 3. It is also beyond our comprehension how the fusion H and H can occur without the availability of the necessary energy. Evidently, reviewer Nr 3 did not read the submitted manuscript. The word*fusion* does not appear anywhere in the text, much less fusion of hydrogen atoms. The submitted manuscript dealt with the induction of a nuclear active state in the Pd/H–H_2O system and provided experimental evidence other than the (nγ) reaction. (are we to believe that only (nγ) reactions are of interest to the JRNC readers?).

The reviewer identified himself as a nuclear chemist (still a chemist) but expressed no opinion concerning the underlying chemical reactions. Certainly, as a chemist, he should ask is there any known reaction involving 3×10^{-3} g of Pd in the hydrogen rich environment that could produce the energy needed to cause so much damage?

3.0 Being a messenger

Acting as messengers (*cf.* our paper *Thermochimica Acta* **410***, 101 (2004)* we were drown into the discussion of the recombination theory by being asked to review a paper submitted to Thermochimica Acta by K.L. Shanahan. We declined to review this paper because the evidence obtained in this laboratory and elsewhere was contrary to the claims made and noted that if this paper is published we would respond and offer experimental evidence supporting our position (*cf.* letter to L.D. Hansen dated 5 Oct. 2004).

San Diego, 5 Oct. 2004

Professor Lee D. Hansen
Chemistry and Biochemistry Department
C100 Benson Sience Building
Brigham Young University
Provo, UT 84602–5700

Regretfully, we cannot review the manuscript LDH 963,‘ “Comments on ... ” since our review might be viewed as biased. What we can say is that if one accepts the assumptions made as valid, then there is no reason why this paper should not be published. Obviously, we do not agree with the assumptions made. Should you decide to publish this paper, we will be prepared to defend our position in a separate communication.

There are few points, not affecting the content, that we wish to make:
(i) Referring to the excess enthalpy production as the Fleischmann–Pons–Hawkins effect is inappropriate. The contributions by Hawkins were that of a graduate student performing assigned tasks. Hardly an equal contribution.
(ii) In the introduction we find “... University of Utah chemists, Martin Fleischmann, Stanley Pons and M. Hawkins...”. At that time, Fleischmann and Pons were professors of chemistry and Hawkins a student. Hardly an equal status.
(iii) In conclusion he writes “Unfortunately, they misunderstood the recombination...”. An unfortunate choice of words. We do not believe that anyone could accuse Professor Fleischmann, F.R.S., a member of selected group of eminent scientists, of misunderstanding such a simple chemical process.

It is quite surprising that Dr Shanahan cannot accept the fact that nuclear events do occur in the negatively polarized Pd electrode and are responsible for the F–P effect. Recent report *New physical effects in metal deuterides* by Hagelstein, McKubre, Nagel, Chubb and Hekman prepared for DoE provides sufficient evidence cf the nuclear origin cf the F–P effect. In the lecture given at the meeting of Nobel laureates in Lindau, Josephson discussed the “pathological disbelief” where he assessed the attitudes of the establishment toward the F–P effect. Copy attached.

Dr P.A. Boss

Dr S. Szpak ‘

Upon reading the Shanahan's manuscript, we declined to comment except to say that our data indicate no contribution due to the hydrogen/oxygen re–combination reaction. This was not the only point – although not explicitly stated there were half–truths, disrespect for professors Fleischmann ad Pons, and lack of familiarity with the research done in this country and abroad. This prompted one of us (S. S.) to submit a short response (reproduced below).

Reply to comments on "thermal; behavior of polarized Pd/D electrodes prepared by co–deposition

S. Szpak
SPAWAR Systems Center San Diego, San Diego, CA – retired

K. L. Shanahan has published a paper: "Comments on *"Thermal behavior of polarized Pd/D electrodes prepared by co–deposition"* [Thermochim. Acta, **428** (2995) 207] in which he states that the excess enthalpy generated in electrochemical cells, the F–P effect, is, in fact, due to $2D_2 + O_2 \rightarrow 2D_2O$ recombination reaction, alternatively, to equipment malfunction or incorrect data interpretation and not of nuclear origin. Furthermore, he asserts that *"SMMF fail to differentiate between dissolved and entrained oxygen"*. There are other statements that question our ability to comprehend simple chemical reactions (*cf.* section 2.1O).

The position taken and statements made by Shanahan demand a reply. The considerable delay in response is because we were not aware of Shanahan's paper. It was brought to our attention by the SPAWAR librarian rather than by the journal's editor. It is our experience and, we believe a practice, that in such cases, the journal's editor informs the other party so that a response may be submitted in timely fashion. This has not been done. Another, rather curious observation: Shanahan's paper was received 14 September 2004 and accepted 1 November 2004 while a response to it by Storms required much longer time to process [*cf.* Thermochim. Acta **441** (2006) 207)], received 6 July, revised 25 August 2005, accepted 15 November 2005.

Equipment malfunction – calibration problem

In the introduction in our paper, we wrote "The basis for calorimetric measurements is the conservation of mass and energy and thus it requires the knowledge of processes under consideration, the sequence of events, the construction of apparatus as well as the experimental procedure employed". Thus, any remark, whether critical or not, should be directed toward the case in question and not toward other systems. We note that a whole section (*cf.* section 2.2) was devoted to the discussion of calibration problems not related to the case discussed. Dr Shanahan would be well advised to acquaint himself with the procedure developed by Prof. Fleischmann and described in great detail in technical report SSC/SD – TR – 1862, vol. 2[1].

Recombination theory

In our paper (Thermochim. Acta **410** (2994) 101] there is a quote [from an e–mail Shanahan to Dr Imam (NRL): *"The infrared photography of Szpak* et.al. *is supportive evidence of this, if one considers the oxidation in subsurface bubbles to be rapid, which* *should be true of D_2 + O_2 flames*. Such interpretation is, indeed, difficult to understand and therefore accept. In a paper on *"Comments on ...."* Shanahan modified somewhat his model: there are no flashes but bubbles still exist and *"this burning would occur at the electrode(s), under the electrolyte surface in bubbles"* modeled after *"the classic fire safety triad of fuel, oxidizer, and ignition source"* and *"consistently proposed that entrained bubbles are the source of the effect, and it is intriguing that SMMF fail to differentiate between dissolved and entrained oxygen*. Just because Shanahan consistently advocates his model, it does not make it true.

The picture that emerges is as follows: bubbles containing oxygen collide with bubbles containing deuterium to form bubbles with a volume ratio of 1:2, and are brought to the electrode (cathode) surface by moving liquid. Alternatively, an oxygen bubble is brought the the cathode surface, again by motion of an electrolyte, to combine with the deuterium bubble. He further states that *" mass transport of bubbles to the other electrode (and their ignition!?) should be facile"*. Such statement should be based on the probability of occurrence. Only when the probability is estimated, one can seriously propose a model. This has not been done (to our knowledge).

In the conclusion section (section 3) we find two statements, both incorrect. The first: *"Unfortunately, they have misunderstood the recombination at the electrode, under the surface issue"*. No, we have not misunderstood the issue, there is no issue. In simple terms, during the electrolysis of D_2O in cells employing cathodes prepared by co–deposition there is no recombination, as determined by hundreds of analyses performed over a period of months[2]. The second: *"Thus the conclusion that a nuclear process has been proven is premature"*. Evidently, he has not followed the literature. Excess of power (and/or enthalpy) was confirmed by many, transmutation to other elements were known since the early days and recently confirmed by Szpak *et.al.* in two brief communications[3,4].

It is interesting to note that Shanahan does not want to accept the fact that Fleischmann and Pons are the discoverers of the effect and that both were professors of chemistry at the University of Utah and not just *chemists"*, thus the F–P effect and not the F–P–H effect. Furthermore, the origin of cold fusion is described in the lecture given by Prof. Fleischmann – *"Background to cold fusion: The genesis of a concept"*[5].

One final remark – why does one write a paper? Firstly, to report a new observation, experimental or theoretical. This is done in brief communication, letters to the editor, and the like. Secondly, to inform and offer an interpretation, draw conclusion, *i.e.* in general, to advance science. This usually requires a full size paper. However, when reading the literature, one frequently finds papers that repeat previously offered arguments, often irrelevant. This is done to satisfy the *publish or perish* arguments, *i.e.* to advance, with little effort, one's professional standing. The reader of this note, and the papers by Shanahan and by Szpak *et.al.*, should have no difficulty in reaching a conclusion as to their values.

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3. S. Szpak, P.A. Mosier–Boss, C. Young and F.E. Gordon, Naturwissenschaften **92**, 394 (2005)
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5. M. Fleischmann, ICCF – 10

Since this short rebuttal was ignored by the editor of Thermochimica Acta, we decided to prepare a full size paper that will conclusively show errors propagated by re–combination theorists, including Shamahan. This paper is reproduced below.

THE FLEISCHMANN–PONS EFFECT: IS THE EXCESS HEAT RESULT OF D_2 + O_2 RECOMBINATION?

S. Szpak, J. Dea and F. E. Gordon

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Abstract

The claim that the observed excess heat generated in electrochemical cells, the Fleischmann – Pons effect, is due to deuterium oxygen–recombination reaction is not supported by experiment or existing models of a motion of dispersed gas bubbles in a liquid.

Almost two decades ago, professors Fleischmann and Pons, first in a press conference and somewhat later in a publication[1], disclosed that an unlimited supply of “clean” energy can be obtained by electrochemical compression of deuterium in the Pd lattice. Furthermore, they stated that the “excess enthalpy” is due to, as yet, unidentified set of nuclear reactions. Needless to say, this discovery was of immense interest. However, because of low rates of heat generation and the assertion of its nuclear origin gave rise to opposition which, at times exceeded the accepted norms.

Recently Shanahan, critical of an earlier paper[2], has published: *Comments on “Thermal behavior of polarized Pd/D electrodes prepared by co–deposition”*[3] in which he states that the excess enthalpy generated in electrochemical cells, the F–P effect, is, in fact, due to (i) either equipment malfunction or incorrect data interpretation, (ii) D_2 + O_2 recombination and (iii) not of nuclear origin. Leaving aside its nuclear origin, we shall briefly discuss equipment malfunction and the recombination reaction.

Equipment malfunction – calibration problem

In the introduction to ref. [2], one finds: *“The basis for calorimetric measurements is the conservation of mass and energy and thus it requires the knowledge of processes under consideration, the sequence of events, the construction of apparatus as well as the experimental procedure employed”*. Corollary, any remark, whether critical or not, should be directed toward the case in question and not toward other systems. We note that a whole section (*cf.* [3] section 2.2) was devoted to the discussion of calibration problems not relevant to the case discussed. The correct procedure developed by Prof. Fleischmann is described in great detail in technical report, SSC/SD –TR – 1862, vol. 2.

Recombination theory.

In ref. [2] there is a quote [from an e–mail Shanahan to Dr Imam (NRL)]: *"The infrared photography of Szpak* et.al. *is supportive evidence of this, if one considers the oxidation in sub–surfaces bubbles to be rapid, which should be true of* $D_2 + O_2$ *flames"*. We observed that such interpretation is, indeed, difficult to understand and therefore accept. In his paper[3], Shanahan modified somewhat his model: there are no flames but the bubbles still exist and *"this burning would occur at the electrode(s), under the electrolyte surface in bubbles"*, modeled after *"the classic fire safety triad of fuel, oxidizer, and ignition source"* and *" consistently proposed that entrained bubbles are the source of the effect, and it is intriguing that SMMF fail to differentiate between dissolved and entrained oxygen"*. Just because one (including Shanahan) consistently advocates a model, it does not make it true.

After careful reading and interpreting *entrained* to mean *to draw and transport gas by the flow of a fluid*, (*cf.* Merriam – Webster dictionary) the picture that emerges is as follows: oxygen bubbles generated at the anode collide with bubbles containing deuterium are brought to the electrode (cathode) surface by a moving liquid. Alternatively, an oxygen bubble is brought to the cathode surface, again by motion of an electrolyte, to combine and react with the evolving deuterium bubble. He further states that *"mass transport of bubbles to the other electrode should be facile"*. Such a model should be based on the probability of occurrence. Only when the probability is estimated, one can seriously propose a model. The simple picture of a gas bubble being carried out from one electrode to the other electrode does not reflect the reality. Motion of dispersed gas bubbles in an electrolyte, especially close the the Prandtl layer, is very complex, *cf.* ref. [4].

Experimental evidence

Today, the generation of excess heat is widely accepted, but its origin is subject of extensive theoretical investigations – so far without definitive conclusions. The notion that the effect is due to the $D_2 + O_2$ recombination reaction still persists. Those who favor it, offer no model except the work of Shanahan, who based his model on the presence of hot spots. It is the purpose of this communication to show that the Shanahn's model is not supported by experiment and that hot spots offer some insight into the nature of the F–P effect.

The evolution of thermal activities in the negatively polarized Pd/D_2O system was examined using an experimental arrangement shown in Figs. 1a and 1b. Figure 1a illustrates the position of an infrared camera facing the negative electrode. To record the evolution of thermal behavior it is necessary to place the negative electrode, e.g. a Ni screen, upon which a deuterium saturated Pd film was deposited, very close to the thin cell wall (Mylar film). The infrared camera was operated in two modes: the first, to measure the cell temperature across the cell (along the X–X line), Fig. 1b, and the second, to display the presence of hot spots.

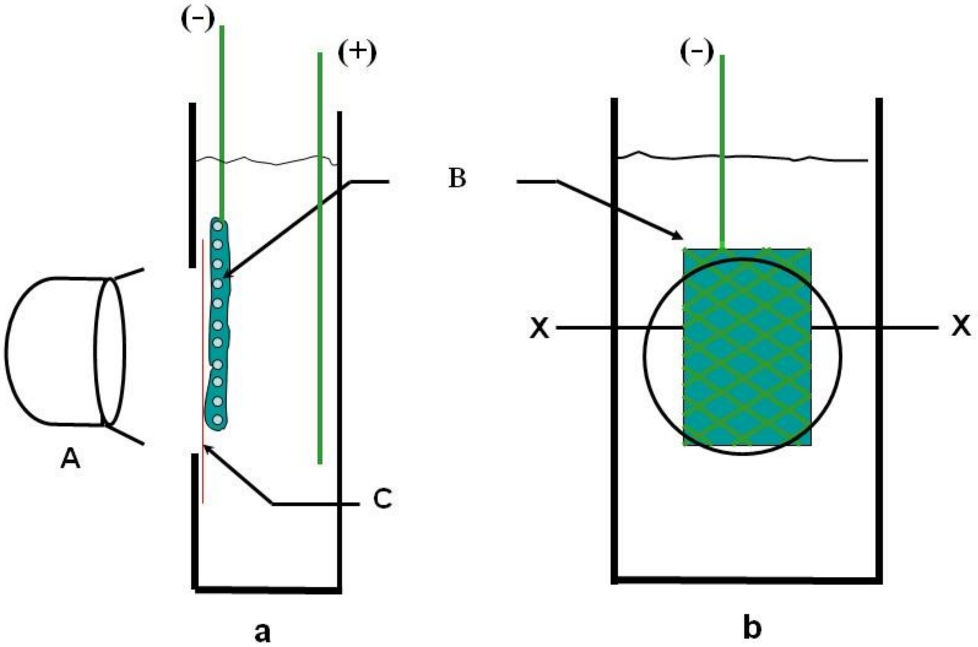
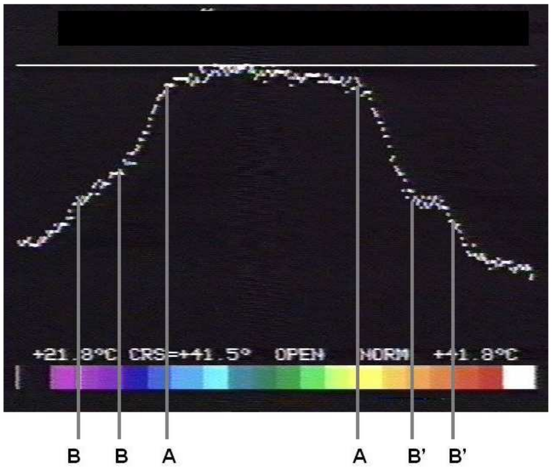


Figure 1. Experimental arrangement for infrared imaging. a (side view) – position of the IR camera; b (front view) – placement of the negative electrode (Ni screen): A – infrared camera, B – Ni screen, C Mylar film.

Cell temperature profile

A typical electrode/solution temperature profile is shown in Fig. 2, where the A–A section indicates the electrode surface temperature while sections B–B and B'–B' the solution temperature. The cell temperature profile, plotted in Fig. 3, was taken periodically during the electrolysis. Evidently, the difference between the electrode surface temperature and that in solution, ΔT, increases with time, being initially at *ca* 2^0C and reaching a value as high as *ca* 17^0C two hours later. It is noteworthy that the increase in the surface temperature, curve a, is irregular indicating bursts in excess enthalpy generation. In contrast, the solution temperature, curve b, increases smoothly which is an expected behavior because the weight (volume) of solution substantially exceeds that of the Pd/D electrode assembly. Incidently, temperature difference arises also from coupling of interfacial processes[5]. However, this contribution is negligible for a system operating at the conditions of this experiment.



[redacted]

Figure 2 Cell temperature profiles. A –A electrode surface temperature; B – B solution temperature.

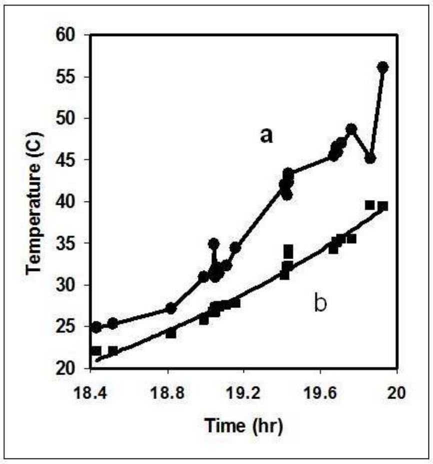
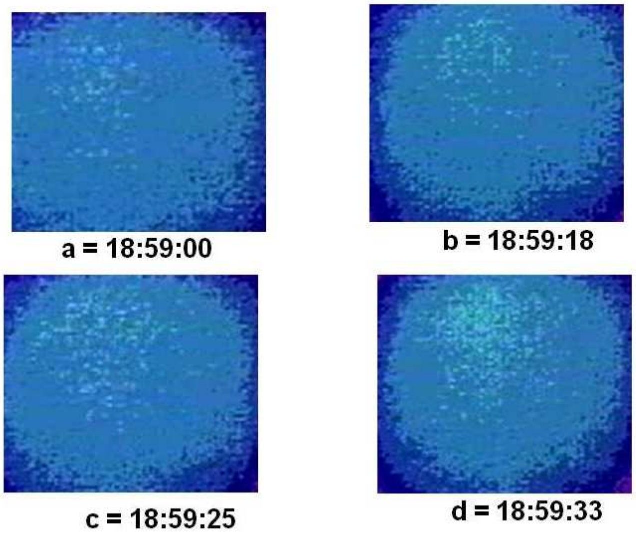


Figure 3. Evolution of the temperature difference, ΔT, as a function of time. a – electrode surface temperature; b – solution temperature.

Development of hot spots

Thermal activity in the form of “hot spots”, Figs. 4a – 4d, were detected early during the Pd/D co–deposition process. Judging from the number of hot spots, we note that (i) the rate of heat generation is not uniform, (ii) thermal activities occur at low cell temperature and at low cell currents. These observation seem to contradict the commonly held view that long incubation times are required and current densities in excess of 100 mA/cm^2 are always needed to initiate and maintain the F–P effect when solid Pd – rods, foils or wires are used.


a = 18:59:00

b = 18:59:18

c = 18:59:25

d = 18:59:33

Figure 4. Time/space dependency of "hot spots", their intensity and frequency during co–deposition. Temperature scale included. "Hot spots" images at times indicated.

The intensity of thermal activity increases with an increase in both cell temperature and cell current. An example is shown in Fig. 5. Here, the temperature of hot spots cannot be estimated because it exceeded the camera range. This conclusion is supported by the temperature profile across the electrode surface as shown in Fig. 5b.

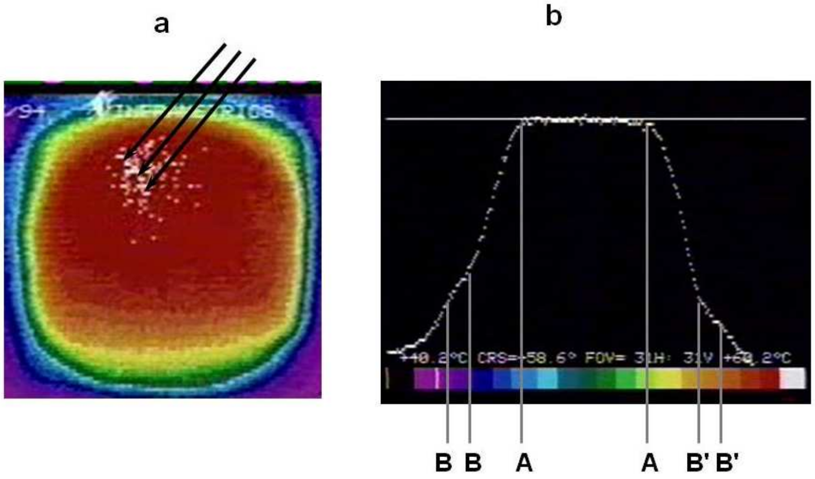


Figure 5. Time/space dependency of "hot spots". a – at elevated temperature; arrows indicated spots whose temperature was outside of the camera range; b – distribu-

tion of cell temperature. A–A electrode surface temperature, B–B solution temperature.

Propagation of pressure gradients

A consequence of localized heat sources is lattice distortion and the development and propagation of stresses within the Pd/D lattice. The display of mechanical distortion can be followed by co–depositing the Pd/D films onto pressure sensitive substrates, e.g. piezoelectric ceramic material[6]. Piezoelectricity is characterized by a one–to–one correspondence of direct and inverse effects, *i.e.* the internal stresses resulting from the electric field are proportional to the field itself and the deformation is accompanied by the appearance of a field strength proportional to the deformation. Because of high sensitivity, its use as a sensing device must be done under strictly controlled conditions. To eliminate external factors that might interfere with the interpretation of the sensor's response the electrochemical cell was shielded (Faraday cage) and the whole assembly placed on a shock absorbing material, as illustrated in Fig. 6.

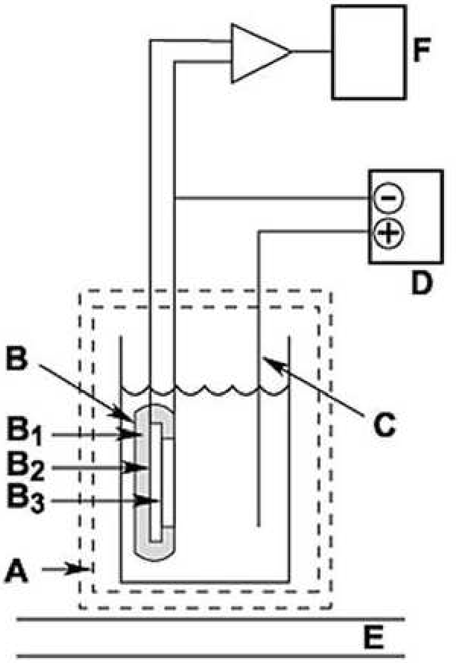


Figure 6. Experimental arrangement for recording mini–explosions. A – Faraday cage, B – negative electrode assembly (B_1 – insulating material, B_2 – piezoelectric substrate, B_3 – Pd/D film), C – positive electrode, D – potentiostat/galvanostat, E – shock absorbing material, F – oscilloscope (LeCroy digital).

An ideal response of a piezoelectric sensing device to thermal mini–explosion is illustrated in Fig. 7. In general, there are two types of forces that interact with the piezoelectric material, *viz* those that cause the contraction of the piezoelectric material and those that result in its expansion.

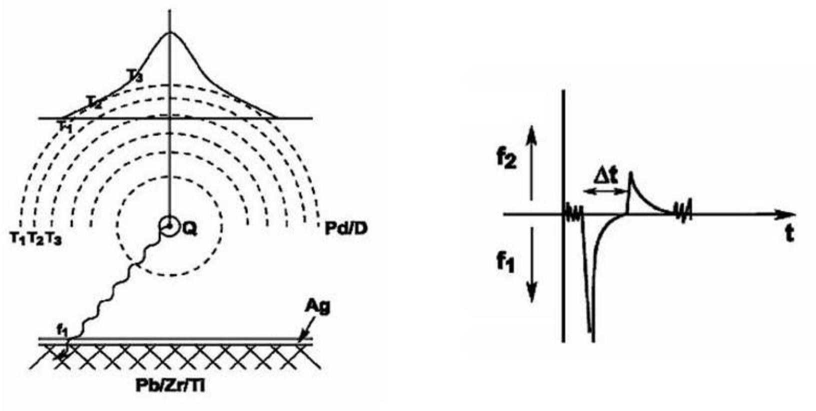


Figure 7. Ideal representation of a mini–explosion. left – location of the instantaneous heat source and associated effects; right – a response of a piezoelectric sensor (f_1 – compression, f_2 – expansion).

An example of the first kind is the pressure change due to *eg* appearance of localized heat source within the Pd/D film; of the second kind – the change in temperature of the piezoelectric material.

A single isolated event is shown in Fig. 8a. Here, we see clearly a single voltage spike which, in the negative direction, corresponds to the pressure pulse. After a brief period of time, $\Delta t = 0.06$ sec., we note the arrival of the temperature front (voltage spike in the opposite direction), followed by the system relaxation. Using a simple model, e.g. that of a spherical reaction site, one could, from the magnitude of the voltage spikes and the Δt, reach some conclusion concerning the position and strength of the heat source. Such singular events are seen in the early periods of the co–deposition. Interestingly, these voltage bursts persisted for hours following the termination of current flow.

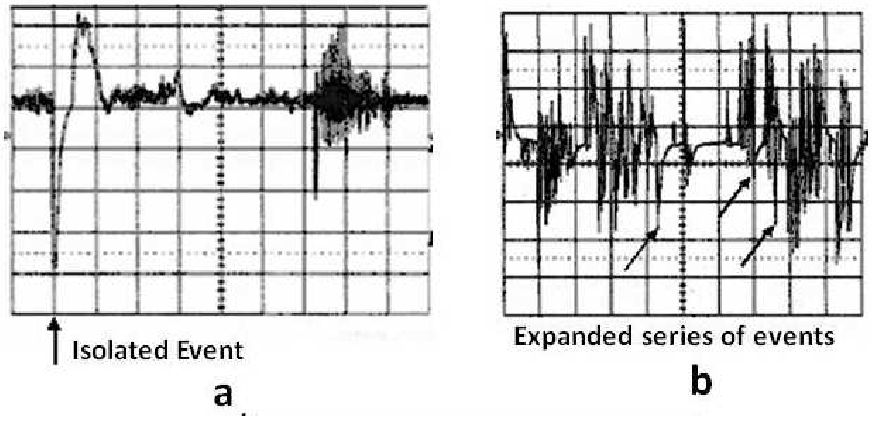


Figure 8. Typical response of a piezoelectric sensor. Left – A an isolated event. Right – B an expanded set of events, arrows indicate recognizable pattern.

A typical voltage–time behavior indicating a burst of events within the Pd/D film is shown in Fig. 8b. These bursts were observed at constant current densities as low as *ca* 4.0 μAcm^{-2}. An expanded trace of such bursts shows a series of of voltage spikes, indicated by arrows. Their frequency and intensity increased with an increase in the cell current and cell temperature. At temperature near the boiling point, thermal activities were very intense as indicated by the magnitude of the voltage spikes, illustrated in Fig. 9.

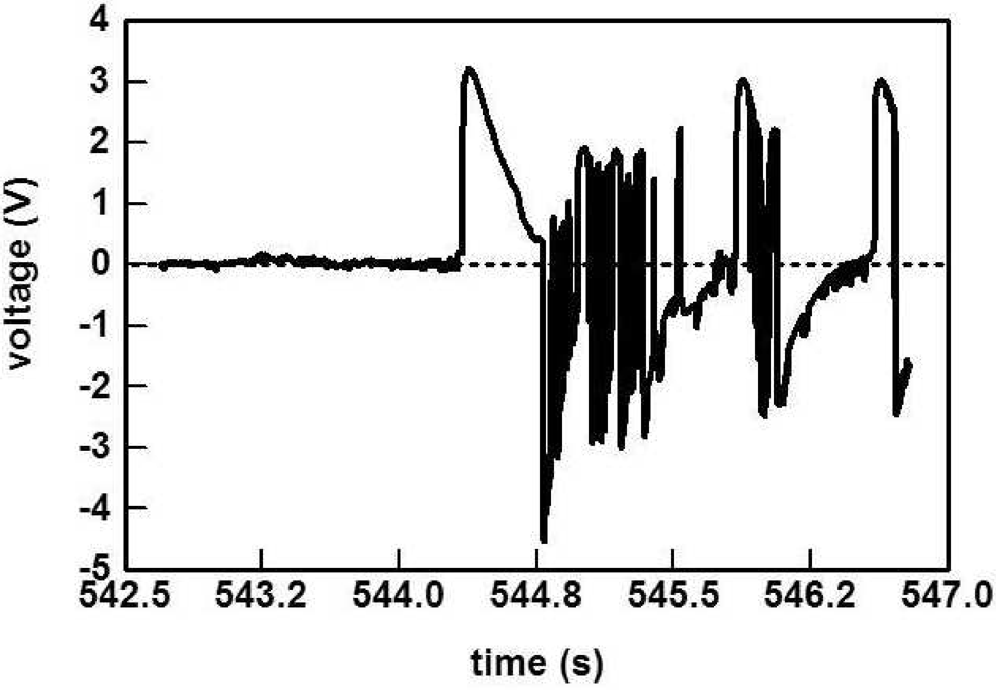


Figure 9. Response of a piezoelectric sensor to events occurring at the boiling point. The high voltage spikes (no amplification) indicate very strong pressure and temperature gradients activating the sensor's response.
The meaning of experimental evidence

In this communication, we presented empirical evidence interpretation of which leads to different conclusion than that reached by Shanahan. The temperature difference between electrode and electrolyte, shown in Fig. 2, does not support the recombination model. Indeed, if the hot spots were due to deuterium oxygen recombination, then the electrolyte temperature should be higher than that of the electrode surface. Such conclusion is reached because the recombination reaction would occur in the gas phase and the heat would be transferred to the solution rather than to the electrode.

Hot spots

Some insight into the nature of the heat sources can be obtained from the analysis of the presented evidence. We refer to Figs. 4,5,8 and 9, *i.e.* we refer to the activities recorded by the IR camera and the piezoelectric sensing device, namely incubation time, size, distribution and the effect of temperature. Thus: (i) Incubation time – the discrete heat sources were noted shortly after the initiation of current flow; (ii) Size – the active site must contain a large number of single events to produce a visible image. Unfortunately, our experimental set–up was not designed to yield quantitative assessment. However, based on other considerations, Chubb[7] estimated the number of single events to be between *ca* $10^4 - 10^9$; (iii) Distribution – the time/space distribution of hot spots as well as their varying intensity with time, Figs. 4a–4d, exclude the existence of fixed location of the active sites. Thus, the direct influence of structural aspects of the electrode material is doubtful. Rather, the distribution and the varying intensity arises from the coupling of the various processes occurring on both sides of the contact surface in response to fluctuations; (iv) Effect of temperature – both, the frequency and intensity are a strong function of temperature. In particular, both increase with an increase in temperature, exhibiting the so–called positive feedback, *cf* Figs. 3 and 5. This is, perhaps, the most direct indication of the influence of the chemical environment on the origin of the hot spots.

Formation of active domains

The appearance of discrete reaction sites, hot spots, implies the existence of a transition from non–reactive to reactive sites (domains). We regard the formation of these domains as being the last step prior to the initiation of the F–P effect. The lack of kinetic data does not permit us to carry any quantitative assessment. However, the thermodynamic reasoning can offer, in a qualitative way, some useful information needed to formulate a phenomenological model.

The formation of active domains represents the instability of the system. It is known that electrochemical (chemical) instabilities, arising from either internal or external fluctuations, lead to self–organization if the system is able to exchange part of the energy or matter with the environment. The self–organization is interpreted as coupling of various processes and/or reactions which implies complex interplay of kinetic and thermodynamic quantities.

Concluding remarks

In the conclusion section of ref. [3] (section 3) one finds two statements, both incorrect. The first: *" Unfortunately, they have misunderstood the recombination at the electrode, under the surface issue"*. No, they have not misunderstood the issue, there is no issue. In simple terms, during the electrolysis of D_2O in cells employing cathodes prepared by co–deposition there is no recombination, as determined by hundreds of analyses performed over a period of months[2].

One of us (S. S.) notes that expressions like *they have misunderstood the recombina-*

tion... and *...failed to differentiate between...* suggest incompetence and should have no place in scientific papers. I do not wish to be disrespectful, but Dr Shanahan's contributions (to science) are no match for those of Prof. Fleischmann, F. R. S., co–author of [2]. Also, it is interesting to note that Shanahan does not want to accept the fact that Fleischmann and Pons are the discoverers of the effect and that both were professors of chemistry at the University of Utah and not just *chemists*, thus the F–P effect and not the F–P–H effect.

One final remark – why does one write a paper? Firstly, to report a new observation, experimental or theoretical. This is done in brief communications, letters to the editor, and the like. Secondly, to inform and offer an interpretation, draw conclusion, *i.e.* in general, to advance science. This usually requires a full size paper. However, while reading the literature, one frequently finds papers that repeat previously offered arguments, often irrelevant. This is done to satisfy the *publish or perish* arguments, *i.e.* to advance, with little effort, one's professional standing. Would the latter be fair assessment of ref. [3]?

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[6] J. Dea, P. A. Mosier–Boss, S. Szpak, *Thermal and pressure gradients in polarized Pd/D system*, American Physical Society Spring meeting, Indianapolis, IN, April 2002
[7] S. R. Chubb, private communication

The submission of this manuscript was confirmed as rejected by the editor L. E. Hansen (copy below)is paper was
Ms. Ref. No.: TCA-D-08-00083 Title: THE FLEISCHMANN-PONS EFFECT: IS THE EXCESS HEAT RESULT OF D2 + O2 RECOMBINATION? Thermochimica Acta

Dear Mr. Jack Dea,

This paper is being rejected without review for the following reasons:
1. The first four sentences of the introduction are a highly biased statement of the situation vis-a-vis "cold fusion". The "F-P effect" is in fact due only to a fundamental error in the way the heat was measured.
2. The technical report cited as giving "the correct procedure" is not readily available, but the procedure given in other sources is incorrectly based on the assumption that heat transfer is mainly by radiation and not by conduction. The fault with most "cold fusion" calorimetry is failure to measure the temperature difference across the major heat transfer path. This failure to abide by the fundamental principle of heat conduction calorimetry has produced most of the reports of "excess heat".

3. Shanahan did not insist on bubbles. Dissolved gases can also be ”entrained”.
4. The statement, ”Today, the generation of excess heat is widely accepted,” is false. Only a few die-hards still believe in it.
5. I was an author on what I believe is the first paper to provide experimental evidence of the recombination reaction, and suggest it as an explanation for some (not all) observations of ”excess heat”. A paper published a short time later showed that recombination was important at low current densities, but became negligible at high current densities. You have failed to cite either of these papers in the present paper.
6. Current density is not given for the experiments reported.
7. The results are readily explained by known effects. Working electrodes should be at a higher temperature than the surroundings depending on current density, heat of the reaction, and irreversible effects. Because the reaction takes place at nucleation sites and because the PdD has a much larger specific volume than Pd metal, hot spots and distortion of the Pd are expected to occur as deuterium dissolves in the metal.
8. ”The electrolyte temperature should be higher than that of the electrode surface” only if the reaction occurs in the electrolyte. If the reaction occurs in the metal, whether it is recombination or formation of PdD, then the electrode will be hotter than the solution. Furthermore, the hot spots may simply be due to current focusing, *i.e.* spots of high current density through the solution interface, similar to the formation of dendrites during electrolytic deposition of metals.
9. The last two paragraphs of the conclusion are not conclusions about science, but personal attacks on Shanahan and have no place in a science journal.

Thank you for giving us the opportunity to consider your work.

Yours sincerely,

L.D. Hansen
Editor
Thermochimica Acta

Upon reviewing the content of the rejection letter, we concluded that any further correspondence would be pointless. Instead we submitted this manuscript with the copy of the rejection letter to professor Vyazovkin, also an editor of Thermochimica Acta, for reconsideration because we felt that the Shanahan’ misconceptions should be answered.

San Diego, 17 March 2008

Professor S. Vyazovkin
Department of Chemistry
University of Alabama at Birmingham
901 S. 14th Street
Birmingham, AL 35294

Dear Professor Vyazovkin,

Recently a manuscript: “The F–P effect: Is the excess heat result of R_2 + O_2 recombination?”, by S. Szpak *et.al.*, was rejected without review but with comments by the editor prof. L.D. Hansen, that show either total unfamiliarity with the subject matter or an extreme bias.

In this letter we ask you to pass judgment on the position taken by L.D. Hansen. To facilitate this task we provide you with the necessary background information that prompted the submission of this communication. One of us (S. S.) was asked to review Shanahan’s paper “Comments .....” [Themochimica Acta, **428** (2005) 207]. He declined to do so because the arguments presented were contrary to experimental evidence accumulated in the SPAWAR Laboratory. Furthermore, he noted that (i) there were objectionable sentences that casted doubt on our ability to comprehend simple chemical processes/reactions and (ii) that, should Shanahan’s paper be published – we would respond. The considerable delay in response is because we were not aware of Shanahan’s paper. It was brought to our attention by the SPAWAR librarian rather than by the journal’s editor. It is our experience and, we believe a practice, that in such cases, the journal’s editor informs the other party so that a response may be submitted in timely fashion. The was not done.

In the rejection letter, addressed to Dr Dea, prof. Hansen listed nine (9) points that justified his decision. Of these nine points, two are particularly troubling. These are point Nr 5 and point Nr 9. Point Nr 5 implies that “unless you cite my papers (and presumably agree with me), I will reject your paper. Point Nr 9 shows double standards – it is OK to show disrespect by stating “fail to differentiate ...etc but it is not OK to response by comparing relative value of Shanahan’s contributions with those of prof. Fleishmann!

We shall now respond to each point listed in the rejection letter.

Point Nr 1. The first four sentences describe what has happened on 23 March 1989. It is just abbreviated statements of facts. It does not involve judgment – hence L.D. Hansen either uses different definition of “bias” (a highly personal and unreasoned distortion of judgment (cf Webster – Meriam dictionary) or did not read carefully the submitted communication.

Point Nr 2. Technical report, TR 1862, is available, cf online:
http//www.spawar.navy.mil/sti/publications/pubs/trindex.hmtl
or by contacting SPAWAR San Diego. Our rejected paper is not about calorimetry, it is about incorrect interpretation of the hot spots, Fig. 4, as indicating recombination reaction. Furthermore, L. D. Hansen is either unaware of the detailed discussion of prof. Fleischmann’s cell/calorimeter by prof. W. Hansen (University of Utah) in 1991 and somewhat later by W. Hansen and M. Melich (Naval Postgraduate School, Monterey, CA) who concluded that, when correctly used, the Fleischmann procedure is very accurate, or dismisses W. Hansen and M. Melich’s work as rubbish.

Point Nr 3. Quote, ”Shanahan did not insist on bubbles” – untrue – Shanahan did insist:

"this burning would occur at the electrode(s), under the electrolyte surface in bubbles". Whether or not "dissolved gases can also be entrained" is immaterial since it depends on the definition of entrained. Shanahan, by "fail to differentiate between dissolved and entrained oxygen" uses the generally accepted definition.

Point Nr 4. L.D. Hansen statement that "only a few die–hards believe in it" is, of course, wrong. Obviously, he does not follow the expanding field. Research into the F–P effect is conducted in a number of countries, e.g. Japan, China, France, Italy, Russia, Israel, USA, to name just a few. Excess heat is always generated if a correct procedure is followed.

Point Nr 5. A sentence in the rejection letter reads" I was an author on what I believe is the first paper to provide experimental evidence of the recombination reaction, etc. You have failed to cite either of these papers". Well, the purpose of our communication was the discussion of the incorrect interpretation of our data by Shanahan and not to review the "recombination theories". Now comes the troubling part – should a journal's editor reject submitted material just because the editor's work was not referenced?

Point Nr 6. The specification of the current density is not necessary in reaching reported conclusions.

Point Nr 7. Comments offered in point Nr 7 do not apply to electrodes prepared by the co–deposition technique. Evidently, prof. Hansen is not familiar with this technique. There is strong indication that the co–deposition is an endothermic process which would result that the electrode would be cooler provided that there is no other exothermic reaction.

Point Nr 8. Our communication was concerned with the Shanahan's model of recombination, *i.e.* gas bubbles containing D_2 and O_2 are in contact with the electrode surface and surrounded by a liquid (an electrolyte). It seem logical to assume that recombination would create a void that would be filled with a liquid – hence, heat generated by the recombination would be transferred to liquid and not to solid. Other points: (i) it is not clear how gas bubble containing deuterium and oxygen could be found in the metal. (ii) Equally puzzling is the assertion that the hot spots are due to current focusing – the results in Fig. 8 do not support this view.

Point Nr 9. Two points: (i) There is a sentence "I do not wish to be disrespectful .." which implies no intention to offend. (ii) The only response to Shanahan's "..intriguing that SMMF fail to ...,etc was to compare Shanahan's contribution with those of prof. Fleischmann who was one of the co–authors. It is difficult to see how such a comparison might be taken as attack on Shanahan.

To summarize – we are convinced that the reasons for rejection of our manuscript are not because of evidence presented, rather because of bias of the editor accompanied by unfamiliarity with the subject. Again, we wish to point out that this communication was to respond to critical remarks made by Shanahan and present evidence in support of our point of view. We addressed this letter to you, who as an editor of Thermochimica Acta will be in position to resolve this troubling situation.

Sincerely,

J. Dea, PhD

Enclosures:
Rejected manuscript
Rejection letter

This material was e–mailed on 17 March 2008 but, as of today (December 2010), no answer was provided. Repeated inquiries were ignored by the editorial office of Thermochimca Acta.

4.0 Reflections

On 23 March 1989, as on any other evening, I watched evening news. What was different this time was prof. Fleischmann, in company of a younger gentleman, speaking about a scientifically exciting discovery – nuclear reaction(s) producing enormous amount of heat in a test tube. The announcement itself was not a great surprise to me since, for some time, September or October of 1988, I have known that prof. Fleischmann was pursuing a new approach to energy production. What I did not know that it involved nuclear reaction(s).

Natural consequence of the press conference was the desire to examine the claims made by Fleischmann and Pons. Attempts to do so took place in well known institutions (Caltech, MIT, U of Texas), government laboratories as well as in private garages. Results vary - a new "social class" is born, *viz* believers and skeptics. Their number grew rapidly. What, at first, appeared to be a simple experiment was, in fact, a complex behavior not easily interpreted. The rather simple question "would the putative reaction of D^+ compressed into a host lattice be different from reactions in dilute plasma (or reactions of highly excited D in solids" and the work of Fleischmann and Pons opened a new area of research that of nuclear events in condensed matter.

In this laboratory, work started in the evening of announcement by personnel not familiar with even the rudimentary electrochemical procedures, thus their effort would, most likely, end in failure. Being somewhat familiar with electrochemistry, I asked myself: should I get involved and, if yes, what can I contribute? I knew that I would not be just another researcher seeking to confirm Fleischmann and Pons results. To resolve this dilemma, I decided to follow the development without making comments or taking a position, but at the same time, because of the ensuing controversy, following closely the developing attitude of laboratory management. At first the management was neither for nor against conducting research in "cold fusion". Somewhat later, after formulating a new experimental protocol (co–deposition), Dr F. E. Gordon, at that time department head (Code ...), , became an enthusiastic supporter and later became an active researcher in this fascinating research.

Review the "who is who" in this country that was involved in "cold fusion" research led to the question: should the direction of research be placed in a physicist's or chemist's hand? If in the latter, then what would be the role of a chemist? The phenomenon of nuclear reactions in condensed matter at room temperature created chaos – it just would not fit into te teachings of classical physics or chemistry. The situation was similar to that experienced, at the end of 19–th century, by Becquerel who studied newly discovered radiation emitted by various materials without definite conclusions. The situation was corrected through identification and isolation of new elements (polonium and radium) using chemical procedures (co–preciptation), by Mme Curie–Sklodowska . Another example of chemical intervention to clear up the nature of reactions arising from the neutron capture by uranium (Fermi, 1934, 1935). It was through the research by Hahn and Strassmann (1939) who showed that lanthanum and barium are the reaction products, i.e that by accepting neutron, the uranium was split into two nuclei with about the same size yielding energy in amount 200×10^6 eV. In 1898 Mme Curie–Sklodowska showed that the difficulties experienced by Becquerel are removed by knowing the system. In 1939 The work of Hahn and Strassmann identified the type of reaction, thus giving physicists clear line of research. In 1989, Fleischmann and Pons work opened a new field for it to be explored. Here by analogy, one expects that the effort of some gifted physicist will provide the acceptable theory. But, in my judgment this will not happen before a procedure is devised in which theoretical predictions do not collide with observations. This is the chemist's job.

APPENDIX A

Shortly after the 23 March press conference a memorandum entitled *Piezonuclear fusion at Brigham Young University* prepared by nameless author(s) was circulated. This memorandum attempts to convince the reader that the so called *cold fusion* was the product of research conducted at BYU long before the Fleischmann–Pons announcement. This memorandum re–appeared in 2004 under the title *Brief history of cold fusion at Brigham Young University*, authored by J. Ballif, W. Evenson and S. Jones and posted by L. Kowalski[126] with somewhat exaggerated comment by referring to it as a historic document.

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[126] http.//pages.csam.mountclair.edu/ kowalski/cf/131/historyhtml

PIEZONUCLEAR FUSION AT BRIGHAM YOUNG UNIVERSITY

I. Scientific Team

A. Brigham Young University Faculty Members - Department of Physics and Astronomy

Steven E. Jones (resume attached)
E. Paul Palmer
J. Bart Czirr
Daniel L. Decker
Gary L. Jensen

B. Brigham Young University Faculty Members - Department of Chemistry

James M. Thorne

C. Brigham Young University students

Stuart F. Taylor
Rod Price
J. W. Wang
David Mince
Eugene Sheeley
Paul Dahl
Paul Banks

D. University of Arizona Faculty Members - Department of Physics

Johann Rafelski

II. History

A. Scientific paper published March 1986 (submitted June 1985)

The roots of our work regarding piezonuclear fusion are described in a scientific paper published in the **Journal of Physics G: Nuclear physics, 12:** 213-221 (attached). This paper was received by the journal on 12 June 1985 and published in March 1986.

1. Theory proposed which explains Brigham Young University experiments

The detailed mathematical framework given in the paper was worked out primarily by Clinton Van Siclen, the first author on the paper. The paper discusses fusion at room temperature and how this might be enhanced by increasing the density of hydrogen isotopes. The paper discusses the transition of hydrogen to the metallic state under high pressures and other technical points. One very significant concept explored in this paper is that a hypothetical particle "with mass twice that of the electron" could lead to room temperature fusion at a rate of approximately one fusion per minute per kilogram of deuterium. This is close to the actual rates observed in later experiments at Brigham Young University by Jones and colleagues, and the theoretical framework given in this early publication continues three years later to be a useful explanation for the piezonuclear effect. Indeed, this paper is often referred to in our paper on the Brigham Young University experiments since it provides a theoretical, mathematical foundation for piezonuclear fusion.

2. "Piezonuclear fusion" coined by Steven Jones of Brigham Young University to describe cold fusion

In addition to initiating the study, Steven Jones (one of the authors) coined the term "piezonuclear fusion" in analogy to the term "thermonuclear fusion," to indicate that the proposed approach is to induce fusion by "squeezing" the hydrogen nuclei together at near room temperatures rather than by heating them to very high temperatures. (The prefix "piezo-" comes from a Greek word meaning to squeeze or compress.)

B. Brigham Young University Physics Colloquium 12 March 1986

1. E. Paul Palmer suggested geophysical applications

The paper was published in March 1986, and on March 12, 1986 some of the concepts in the paper were described by Dr. Jones at a Colloquium of the BYU Physics Department. Paul Palmer was present and associated these ideas with geological data on heat and helium-3 which are correlated in volcanoes and other thermal regions of the earth. Both heat and helium-3 are released in fusion reactions (proton-deuteron and deuteron-deuteron reactions). Dr. Palmer suggested that rock or ions in the earth might help to catalyze the fusion reaction. This creative leap is recorded in Dr. Palmer's logbook, dated March 13, 1986 in some detail (attached).

C. Report to DOE 13 May 1986

Our work on cold piezonuclear fusion was reported to the Department of Energy in the 1985-86 Annual Performance Report, dated 13 May 1986, along with three related documents: the Van Siclen/Jones paper on piezonuclear fusion, a note entitled "Experiments in cold fusion" dated 28 March 1986 by Paul Palmer; and "Comments on Catalyzed Fusion," a note by Steven Jones dated 1 April 1986. It was at this time that Prof. Jones received permission from the DOE funding agent R. Gajewski to pursue research on this aspect of cold nuclear fusion under an already existing DOE grant to Brigham Young University for muon-catalyzed fusion research.

D. Brigham Young University's experimental program

1. Planning began in March 1986

As a result of discussions generated by the Physics Department colloquium on March 12, 1986, an experimental program was worked out to test these new ideas. An important discussion meeting was held at BYU on April 7, 1986, involving Profs. Czirr, Jones, Palmer of BYU and Johann Rafelski of the University of Arizona along with student researchers. Plans for the research were extensively developed at the meeting. Prof. Rafelski had been very active in theoretical work on piezonuclear fusion since late 1985 and strongly urged the active pursuit of this experimental effort at BYU.

2. Use of Pd, Li, Al, Cu, Ni, Pt under non-equilibrium conditions, was outlined 7 April 1986 - notarized lab notebook page

Prof. Jones's brief notes from the April 7, 1986, meeting are attached showing that the metals aluminum, copper, nickel, platinum, palladium (because it "dissolve(s) much hydrogen"), and lithium were discussed as prime candidates for the process. The importance of non-equilibrium conditions was discussed; in particular, "shocked hydrides" and "electric discharge" were considered. These notes were notarized that day by Lee R. Phillips, a notary and BYU attorney, showing the importance attached to these ideas by the physicists present.

3. Geophysical evidence for piezonuclear fusion was sought in the scientific literature as early as April 1986

On April 13, 1986, Prof. Palmer noted in his logbook a number of fusion reactions to be studied, including the deuteron + lithium-6 reaction. (On March 18, he had noted the high amounts of sodium and lithium in magmas; these later became ingredients in our electrolyte solution.) On April 16, he records the finding of a paper by the Russian physicists B. A. Mamyrin, L.V. Khabarin, and V. S. Yudenich [**Dokl. Adad. Nauk. SSSR, 237**:1054 (1978)] in which they report excess helium-3 found in various metals. This paper was encouraging to us, but we were surprised that no follow-up work was recorded in the literature.

4. Electrochemical cell built and measurements taken beginning May 1986

On May 22, 1986, our first electrochemical cell for "electrolytic infusion of hydrogen into metals" was built (see attachment from Prof. Palmer's logbook) and on May 23, D_2O (heavy water) was added. We looked first for gamma rays from proton-deuteron fusion using a sodium-iodide detector and found on May 27 that the foreground rate when the cell was operating was slightly higher than the background rate when the cell was not operating, but the result was not statistically significant. In June, we developed another means of loading hydrogen isotopes into metals, using pressurized gases, and added a crude neutron counter.

5. Work on a highly sensitive, energy resolving neutron detector was begun in 1986, since neutrons of the correct energy are a sure indicator of nuclear fusion

Throughout the summer, the neutron detector was worked on while different electrolytes were tried, including the addition of NaOH or H_2SO_4 to D_2O and the addition of "impurity salts" of various metals. We also tried loading the cathode with deuterium before beginning the electrolysis (see, e.g., 10 September 1986 entry). By September 3, 1986, we saw a foreground minus background rate of about 5×10^{-3} in the neutron counter, but the result was neither statistically significant nor consistently repeatable. However, this rate proved to be consistent with the rate obtained in later work when the neutron counter system had been dramatically improved. As the school term began anew, we concluded that in order to make progress in our work we had first to improve the neutron detector. Bart Czirr and Gary Jensen continued this work. Some of the effort went into trying to find a suitable hydrogen-rich, inorganic scintillator. While this work continued, Dr. Jones pursued muon-catalyzed fusion research.

6. Student papers presented on piezonuclear fusion experiments in March and April 1988

In January, 1988, Prof. Jones organized a student research class along with Prof. Palmer and Prof. Larry Rees. Piezonuclear fusion was one of the principal research topics, pursued by students Paul Dahl and Paul Banks. Both wrote term papers on the topic. Paul Dahl presented an oral paper at the Spring Research Conference of the BYU College of Physical Sciences and Mathematics 1988 on 12 March 1988; his paper was entitled "An Experimental Investigation of Piezo-nuclear Fusion." On March 25, 1988, we prepared some deuterated metal samples which were sent to Harmon Craig of the University of California. at San Diego for helium and tritium analysis. These samples were later sent to Al Nier of the University of Minnesota, but analysis has not been completed as of 30 March 1989.

7. Further experiments planned, research program set out and pursued vigorously from August 1988

In August, 1988, the decision was taken by the scientific team to vigorously pursue experimental piezonuclear fusion research. In particular, Prof. Jones logbook records that a fusion group meeting took place on August 24, 1988, and that this matter was aired. We decided that both gamma and neutron detectors would be used. Since energy applications for muon-catalyzed fusion then appeared remote, we decided to place particular emphasis on our piezonuclear fusion research program. This decision followed work in early August by Prof. Jones in which he outlined a paper on the subject. His intent was to include a discussion of piezonuclear fusion research in his paper for publication in the proceedings of a muon-catalyzed fusion workshop previously held in Florida in May 1988, as recorded in his logbook entry dated 9 August 1988. (Prof. Jones is an editor of this Proceedings, which is publication no. 181 of the American Institute of Physics.) However, during a visit to Provo on August 15-16, 1988, Prof. Rafelski dissuaded Jones from including this work in his paper but rather encouraged more experimental studies. Dr. Jones also discussed the BYU work on this fusion process and his intentions to emphasize piezonuclear fusion studies with Dr. Alan Anderson and Dane Chapman in August and early September, 1988. Our group has vigorously pursued its experimental piezonuclear fusion research at BYU since August 24, 1988.

8. Publishable results obtained in 1988-1989

We started with gamma-ray studies since the sodium-iodide detector is easiest to set up. As before, we saw only non-significant hints of gamma production in our 3 inch sodium iodide counter, so we decided to concentrate on using the neutron spectrometer, which was fully conditioned for use in late 1988. Our first studies with this spectrometer were done using titanium, palladium, tantalum, nickel, aluminum, iron, and lanthanum. We also used several methods of loading deuterium into metals, including the original electrochemical method. Thus, we performed anew the experiment which we had started with in May 1986, namely electrolytic infusion of deuterium into metals, but with a much-improved neutron detector. Of these experiments, Paul Palmer records: "Steve and Bart have set up experiments exactly as we did a year or so ago and looked for fusion-generated neutrons in Bart's liquid-scintillator, low-resolution spectrometer.....As in the previous work, the results were tantalizingly positive." Within a few weeks, the results had reached a statistical significance of over five standard deviations. We also found correlations between tritium detected in Hawaii and volcanic eruptions there, in agreement with expectations that piezonuclear fusion occurs in the earth. We decided in early February to publish our results.

E. Discussions with scientists at other institutions (1986-1988)

Our work in this field has been communicated to a number of scientists outside of BYU in the 1986-1988 period; to name a few: Harmon Craig (University of California, San Diego geophysicist), Al Nier (University of Minnesota), Alan Anderson (Idaho Research Software), Gus Caffrey (Idaho National Engineering Laboratory), James Cohen, Mel Leon, Jim Bradbury, Richard Maltrud, Mike Paciotti (all Los Alamos National Laboratory), Russell Kulsrud (Princeton Plasma Physics Laboratory), Archie Harms (McMaster University), and Mike Danos (National Bureau of Standards).

III. Scientific Contacts with University of Utah Researchers

A. Steven Jones reviewed proposal by Pons and Fleischmann at request of DOE, 20 September 1988

According to Prof. Jones' logbook, he reviewed a proposal by Profs. Stanley Pons and Martin Fleischmann on September 20, 1988, entitled "The Behavior of Electrochemically Compressed Hydrogen and Deuterium." It is not possible from the title to determine that the proposal relates to nuclear fusion. The proposal was sent to Prof. Jones by Ryszard Gajewski, director of the Division of Advanced Energy Projects of the Department of Energy. Dr. Gajewski has funded Jones' work on cold nuclear fusion since 1982, and his specific work on electro-fusion since May 1986. For his part, Dr. Jones has reviewed about eight or ten proposals relating to cold nuclear fusion, his primary field of research. The cover letter with the proposal says nothing about declining to review the proposal if the reviewer was doing related work. Indeed, most of the proposals which Dr. Jones is asked by the DOE to review relate closely to his active research on cold nuclear fusion, including muon-catalyzed fusion. The cover letter did specify that the reviewer agrees to "use the information contained in the proposal for evaluation purposes only." This Jones accepted and acknowledges that he has abided by this agreement. The development of the project at BYU outlined above, including the use of electrochemical cells since May 1986, shows that Brigham Young University was conducting research in electrolytic piezonuclear fusion long before the review of the University of Utah's proposal. When that proposal becomes available to the public it will become obvious, if it is not already clear, that Brigham Young University researchers have not used information from that document. We adhered to our program in a straightforward way.

B. Jones offered to cooperate with Pons and Fleischmann

Prof. Jones recommended that the University of Utah's proposal be approved, despite his unresolved reservations about the theoretical underpinnings. He also suggested to R. Gajewski that he inform Pons and Fleischmann that Jones had been doing related work on piezonuclear fusion since at least 1986 and that perhaps a cooperative effort between the nearby universities (BYU and University of Utah) would be desirable. Jones pointed out that the techniques of the two efforts (e.g. neutron detection at BYU) were complementary and that the research effort could be benefitted by cooperation.

1. Pons responded with telephone call December 1988

Dr. Gajewski did inform Pons of the proposed cooperation, who in turn called Jones in (about) December 1988 to discuss the matter.

2. Jones offered use of neutron detector

In ensuing contacts, Pons requested written information regarding the neutron spectrometer which had been developed at BYU. Jones mailed him this information and offered to allow Pons to use the operating neutron spectrometer at BYU. Pons seemed pleased with the offer.

3. Pons and Fleischmann visited Brigham Young University laboratory, 23 February 1989

Finally, on February 23, Pons and Fleischmann came to BYU to visit Jones and his colleagues in the BYU Underground Laboratory. Pons and Fleischmann were shown the neutron spectrometer and the energy spectra which it produced, including calibration and actual data distributions. In particular, we openly pointed out the fusion neutron signal observed in our data. We also discussed some of our geological evidence for piezonuclear fusion. In the exchange of information, Fleischmann showed us one of their electrochemical cells, although we understood that this particular one was one that did not work. We invited them to bring their (working) apparatus to BYU to verify its operation with our neutron spectrometer. They agreed, and a date of February 26 was set for the test.

4. Researchers agreed to work toward simultaneous publication

Over lunch at BYU that day (February 23, 1989), Jones told Pons and Fleischmann that the BYU group was preparing to publish their data and offered to let them publish simultaneously. Dr. Jones reports that when he made the offer to allow the University of Utah researchers to publish simultaneously with the BYU report, he was attempting to establish an open and cooperative relationship.

The University of Utah researchers did not come back to the BYU laboratory to test their equipment on February 26 as agreed. Rather, they explained that morning that a graduate student had had to travel to a funeral and suggested that they would plan to come at the end of the week. But they did not come then either. Subsequently, a meeting was proposed by University of Utah President Chase Peterson for March 6, 1989 to be held at BYU with the chief scientists and Presidents of the two universities present.

IV. Contacts Between University Administrators

A. Telephone discussion between President of University of Utah and Provost of Brigham Young University, 3 March 1989

On Friday, March 3rd, the President of the University of Utah called the Provost of Brigham Young University. He made some observations about the significance of cold fusion research going on at both universities, and some of the complexities surrounding the projects. He then asked for a meeting with top university administrators and chief scientists involved in the projects as soon as possible. A meeting was scheduled for the following Monday.

B. Meeting between Brigham Young University and University of Utah administrators and principal scientists at Brigham Young University, 6 March 1989

On Monday, March 6th, University of Utah president (Chase Peterson), his vice-president for academic affairs (Joseph Taylor), and the two principal scientists (Drs. Pons and Fleischmann) involved in the cold nuclear fusion experiment at the University of Utah arrived at 9:00 A.M. to begin the scheduled meeting. Brigham Young University participants in the meeting were the president of the university (Jeffrey R. Holland), the provost and academic vice-president (Jae R. Ballif), the associate academic vice-president responsible for research (LaMond Tullis), and the principal scientist who directs Brigham Young University's piezonuclear fusion experiments (Dr. Jones).

Before the meeting the president of the University of Utah met separately with the president of BYU and his provost to discuss the agenda. It was agreed that the University of Utah's president could pursue his agenda so long as it included a brief historical summary of research done at BYU.

President Chase Peterson of the University of Utah first explained how wonderful an invention practical cold nuclear fusion would be. He also said that the large monetary proceeds from said invention could be extremely valuable to the University of Utah. Dr. Jones then held up a small flashlight and stated a strong cautionary note that he would be extremely surprised if enough power could be generated by the process to power even a flashlight, and that he could not see how in any case the proceeds from the invention could be vouchsafed for Utah. Jones then reviewed the history of the BYU research on cold nuclear fusion (at the request of Academic Vice President Ballif). He described much of the history given above. In particular, Jones showed a notarized page from his own logbook dated (and notarized) April 7, 1986, demonstrating that the metals palladium, platinum, nickel, lithium, copper, and aluminum were particularly enumerated for the BYU research on piezonuclear fusion on that date. He also showed copies of pages from Paul Palmer's notebook that demonstrate that experimental research using electrolytic infusion of hydrogen into various metals began at BYU on May 26, 1986, 2 1/2 years before we learned of the University of Utah work in this area, with first positive hints of cold nuclear fusion by this process on May 27, 1986. None of these dates were questioned or challenged by Pons or Fleischmann or anyone present, nor did anyone raise any questions about the proposal-review process. They did not allege that the BYU work had pirated any ideas from their own work. After Dr. Jones' review of detailed documents showing the BYU research over the years, Utah President Peterson turned to BYU President Holland and commented on the remarkable coincidence that such similar research had sprung up independently at the two universities.

Then the meeting shifted to a discussion of releasing the information to the public. The University of Utah researchers stated that they would prefer to have up to eighteen months to quietly pursue their research before announcing it. Dr. Jones stated that he had been funded on the research in question since May 1986, that he had positive results, that he felt obliged under the DOE grant to rapidly publish his results, and that the DOE funding agent had encouraged him to go ahead with a publication on the experimental work. In particular, Jones displayed his abstract for an Invited Paper to the Spring Meeting of the American Physical Society to be held in Baltimore, Maryland May 1-4, 1989. Jones' abstract, sent to the American Physical Society on February 2, 1989, states in part: "We have shown that nuclear fusion between hydrogen isotopes can be induced by binding the nuclei closely together for a sufficiently long time, without the need for high-temperature plasmas....We have also accumulated considerable evidence for a new form of cold nuclear fusion which occurs when hydrogen isotopes are loaded into various materials, notably crystalline solids (without muons). Implications of these findings on geophysics and fusion research will be considered."

The University of Utah contingent expressed great concern about Jones' speaking at the May meeting in Baltimore. In particular, University of Utah President Peterson suggested strongly that it would be desirable for Jones not to give the talk. Dr. Jones replied that he was shocked that Pres. Peterson would suggest that he give up an invited talk on the BYU work, and Pres. Peterson said that he would not ask Jones to cancel his talk. Instead, it was agreed that the two groups would submit papers SIMULTANEOUSLY and quickly (after about three or four weeks) in order to have the papers accepted and hopefully published before Jones' scheduled talk on May 4, 1989. It was also agreed by all that no public disclosure of the research would be made by either group prior to the simultaneous submission of the papers. In keeping with this understanding, Jones said that he would cancel a previously scheduled physics department colloquium at BYU, set for March 8 (two days later), and he did so. Jones also cancelled a talk by a graduate student (Stuart Taylor) on the BYU piezonuclear fusion research scheduled at the BYU Spring Research Meeting on March 11, 1989, in order to strictly adhere to this agreement.

In subsequent discussions between Jones, Pons, and Fleischmann, it was agreed that the precise day for the joint submission would be on March 24, 1989. On March 21, Dr. Pons called Dr. Jones and the joint submission date of March 24 was re-confirmed. Dr. Pons indicated that the University of Utah paper was already ready but assured Jones that it would not be submitted earlier than March 24. No mention whatsoever was made of the University of Utah press conference held on March 23, 1989, one day prior to the agreed date for releasing the information jointly, or of the University of Utah paper on cold fusion submitted March 11, 1989, to the Journal of Electroanalytical Chemistry by Pons and Fleischmann.

C. Agreements from 6 March 1989 meeting

After an extended discussion of what might be done to accommodate the interests of the University of Utah delegation, it was agreed by all present that:

1. Simultaneous publication

Scientists at Brigham Young University and the University of Utah would prepare and submit simultaneous publications to the same journal.

2. Publication prior to APS meeting (May 1989) where Jones was scheduled to speak

Every effort would be made to get publication prior to the American Physical Society Meeting, even though this would be difficult in the short time available. It was agreed that publication in the most prestigious physics journals would be pursued first, but if that could not be accomplished in time, the papers would be submitted to another journal. It was agreed that if necessary journals outside the field of physics would be considered including simultaneous publication in a chemistry journal.

3. Exchange of preprints

The scientists would exchange papers after they were completed.

4. No further public comments on results of research until papers submitted

No further public announcements of the results of either teams' research would be made until after the papers were submitted for publication. Brigham Young

University scientists had a department colloquium scheduled later in the week to discuss their research. Jones volunteered to cancel the presentation and did so following our meeting.

V. University of Utah Press Conference and Subsequent Events

On March 22, 1989, we had calls from people at the Department of Energy about a press release that they had announcing a March 23 University of Utah press conference. It stated that net energy-producing cold nuclear fusion had been achieved at the University of Utah, and that a reviewer of the proposal had confirmed the result! We were shocked and disappointed by the announcement and communicated these feelings to Chase Peterson and James Brophy at the University of Utah. For example, on March 22, BYU Professor Grant Mason, Dean of the College of Physical and Mathematical Sciences, spoke to Dr. James Brophy, University of Utah Vice President for Research, and expressed to him that if the press conference were held, we at BYU would interpret this as a violation of the agreements between the two universitites.

The University of Utah press conference was held there on March 23, 1989. No mention was made of the electro-fusion research at BYU. In fact, a question was asked at the University of Utah press conference: "Is this going on any place else, or is this the kind of process that is currently being developed by anyone else?", to which University of Utah Vice President of Research James Brophy replied: "Let's see, I'll answer it and then perhaps you can. We're not aware of any such experiments going on." It was also intimated that the University of Utah paper on their work had already been submitted, although they would not say to which journal. This was also a great shock to us at BYU. After learning of this, we could not see why we should wait until the next day to send our paper to **Nature**, so, after the press conference, we immediately sent our paper to **Nature**. We have received considerable criticism from University of Utah persons for sending our paper in on this day instead of waiting until March 24. However, it was verified to us on March 24 that Pons and Fleischmann had indeed submitted their paper prior to March 23; their preprint entitled "Electrochemically Induced Nuclear Fusion of Deuterium" contains this statement on the title page: "Submitted to Journal of Electroanalytical Chemistry March 11, 1989." Thus, the University of Utah paper was submitted prior to March 24, although we are given to understand that a paper on the subject also was submitted on March 24 to **Nature**.

There remains one final area to be recorded in this effort to lay out the facts. Numerous allegations and insinuations have been made and continue to be made to the effect that Dr. Jones pirated the idea of fusion in an electrochemical cell or some unspecified ideas from the University of Utah work based on his review of their proposal. For example, such insinuations appeared in a front page article of the Deseret News on April 2, 1989. Similar insinuations have been made to officials at the Department of Energy and to scientists at Los Alamos National Laboratory in New Mexico.

On or about February 14, University of Utah attorney Norm Brown spoke to the BYU attorney Lee Phillips and suggested that Jones had pirated ideas from his review of the University of Utah proposal. This was reported to Jones who reported the allegation to the DOE funding agent Dr. Ryszard Gajewski. Dr. Gajewski then questioned Dr. Pons about this. Dr. Pons apologized for the insinuations of the lawyer both to Dr. Gajewski and directly to Dr. Jones on or about February 21, 1989. We know of no further accusations or insinuations of wrongdoing against Dr. Jones until March 10, 1989, when Dr. James Brophy made allegations of this kind to Prof. John Lamb of BYU. During the week of March 14, 1989, Dr. Pons made general accusations against Jones to Dr. Gajewski.

We have provided evidence from logbooks and other sources that demonstrates that these unsubstantiated allegations are false. When the University of Utah proposal becomes available through the Freedom of Information Act, it will be completely clear, if it is not already, that our work was conducted independently of theirs. (According to agreement with the DOE, BYU retained no copy of the proposal following the evaluation.) Meanwhile, we have opened our logbooks and other documents to public view.

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Piezonuclear fusion in isotopic hydrogen molecules

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Abstract. A rough estimate is made of the rate of fusion of the nuclei in a deuterium molecule at room temperature and atmospheric pressure. As the density of the collection of molecules is increased, the fusion rate will likewise increase, in a manner given by an easily derived semi-classical equation. With sufficient experimental data, perhaps obtained through use of a diamond-anvil high-pressure cell, density-dependent expressions for the internuclear potentials of isotopic molecular hydrogen can be constructed and values for the corresponding fusion rates calculated.

1. Introduction

Fusion of isotopic hydrogen nuclei is a very efficient means of producing energy in the high-temperature, high-pressure interiors of stars. In terrestrial conditions, of course, the nuclei acquire an electron and approach one another no closer than is permitted by the molecular Coulomb barrier. The probability of fusion of the nuclei in molecular hydrogen is then proportional to the probability of quantum-mechanical tunnelling through that barrier, or equivalently, the probability of finding the two nuclei at zero separation.

By replacing the electron in a hydrogen molecular ion with a negatively charged muon, the fusion rate is greatly increased. The reaction is facilitated by the reduced equilibrium internuclear separation due to the much larger muon mass. A similar, though not nearly so extreme, distortion of the molecular potential can be achieved by subjecting the molecules to very high pressures. With the possibility of creating pressures of several million atmospheres presented by the diamond-anvil cell (Jayaraman 1984) it is interesting to consider rates for fusion of the nuclei in isotopic molecular hydrogen as a function of pressure.

Separation of the centre-of-mass motion of the nuclei from their relative motion gives Schrödinger's equation for a particle of mass equal to the reduced mass of the two nuclei, and bound in a potential well constructed from the total electronic energy and the nuclear repulsion. A classical particle moving in the molecular potential $V(r)$ depicted in figure 1 will oscillate harmonically about the point r_0 (corresponding to the equilibrium internuclear separation) and between the turning points r_a and r_b. Its quantum-mechanical counterpart, described by the probability amplitude ψ, is not similarly constrained to lie entirely within the classical turning points, but extends into the classically inaccessible regions.

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Mar 13 1986 Source of volcanic heat.

Colloquium yesterday by Steve Jones of BYU physics set me thinking. He talked of muon catalyzed cold fusion — among other things such as quark search and electron-catalyzed fusion of HD molecules. He talked of spontaneous fusion under pressure (low) and catalyzed fusion (high).

$$H + D \rightarrow {}^3He \quad + \sim 6 \text{ MeV (I believe.)}$$

Well, when earth's sedimentary material at a continental margin gets pulled down in a subduction zone at a plate boundary, fusion could take place as the pressure increased. What is the deuterium concentration (I looked on chart of isotopes. It is .015 %). If all fused, and the energy was 5 MeV that would be, per gram of water (with normal concentration of D),

$$\frac{m\,(g)}{M\left(\frac{g}{Mole}\right)}\; A\left(\frac{\text{molecules}}{\text{mole}}\right)\; F\left(\begin{matrix}\text{ratio of D}\\ \text{to H}\end{matrix}\right)\; 2\left(\frac{\text{ratio of H}}{\text{to } H_2O}\right)\; C\left(\begin{matrix}\text{concentration}\\ \text{factor of}\\ \text{D in water}\end{matrix}\right)$$

$$\times\; 5\left(\frac{\text{MeV}}{\begin{matrix}\text{D atom}\\ \text{fusion}\end{matrix}}\right) \times \left(\frac{\text{Joule}}{\text{MeV}}\right)$$

$$= \frac{1\,g}{18\,\frac{g}{\text{mole}}}\; 6\times10^{23}\,\frac{\text{molecule}}{\text{mol}}\; .00015 \times 2 \times 1 \times 5\times10^{6}\,\text{eV} \times 1.6\times10^{-19}\,\frac{j}{eV}$$

$$= 8\times10^{-4}\times10^{23}\times10^{6}\times10^{-19} = 8\times10^{6} \text{ joule.} \quad \cancel{\text{or}}\; \frac{8\times10^{6}}{4.19\,\frac{\text{Joule}}{\text{cal}}} = 1.9\times10^{6}$$

(cal/g of water containing .015 % D)

That is interesting ~ 2 million cal/g

Suppose we take rock up to 3000 °C. That takes ~ 1000 cal. This gram of water could take 2 kg of rock up to high temperature. In many rocks there is certainly of the order of 1 g of water (in water of crystallization as well as interstitial water) per 2 kg of other stuff.

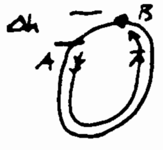
To measure all this, measure the ³He that outgasses from the lava! Simple. These data must be available. The ratio of H to ³He that outgasses would allow computation of the fraction of spontaneous fusions per average water molecule.

Perhaps the rock catalyzes the reaction! Temperature separates H and D from water and even oxygen might catalyze.

(CONT)

Mar 13 1986 (Cont.) Heat from DH fusion.

I'm not sure exactly how to look at the dissipation of heat in a convection cell. Suppose in a cell from a midocean ridge to a trench, the entire energy were dissipated in friction.



Assume energy from A to B around bottom of loop just drives loop and all from B to A is free to be converted to frictional energy at A.
The energy is mgh. For 1 g of rock it is
30 000 ft is ~ 10^4 meter.

$$E = 1(g)\left(10^{-3}\frac{kg}{g}\right)9.8\left(\frac{m}{sec^2}\right)(10^4\,m) \approx 100 \text{ joule} \approx 25 \text{ cal}$$

This is a fair amount $\frac{1}{2}\cancel{m}v^2 = \cancel{m}gh$ $v = \sqrt{2gh} = \sqrt{2(9.8)(10^4)} = 450$ m/sec ≈ 1350 ft/sec

This is 22-bullet speeds. Enough to cause materials to flow but not to melt. This is negligible compared with fusion energy. That is, negligible compared with *total* fusion energy but maybe not compared with slow rates of fusion.

13 March 86 entry read and understood.
Steven E Jones
2 Feb 1990

Mar 14 1986 Catalysis of DT fusion in earth.

I talked to James Baer and Myron Best about the possibility of fusion at plate-subduction regions. They were very excited about the need for local energy sources to supply the heat. We found a paper by Harmon Craig on $^3He/^4He$ isotopic ratio at volcanic regions. It is exciting. Where gas comes from volcanic regions, the $^3He/^4He$ ratio is 8 to 15 times that found in atmosphere and crustal rocks. Craig et. al. attribute this to primordial mantle helium ratios being revealed in plumes. I attribute it to the *generation* of He^3 by DT fusion catalyzed by something or other. I would guess that the something or other are (−) ions of flourine, chlorine, and maybe oxygen. Maybe under pressure, free electrons do it.

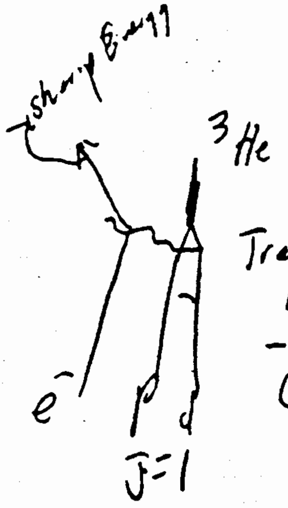
These isotopic ratios support a deep-earth origin of the ^3He but at this time do not indicate the cause of the ratio. Is It primordial or fusion generated? Is there a mantle probe that is not contaminated with possible fusion-generated ^3He? How about meteorites? How about the Soviet deep wells?

What is the solar wind $^3He/^4He$ ratio. Doug Jones gave me a couple of names to look up.

The best test might be a lab experiment - squeeze DH with catalysts in a diamond cell. The trouble with this, if it didn't work, is that it might be because conditions and catalyst were wrong.

Where to proceed? - Search all leads and write a proposal? Or do the work and write a patent and proposal? (cont.)

Jan 4/7/86



Transverse e⁻
borrows E from fusion
– works only for d-p-e
(not d-d) ↓ in gas
but d-d could go in hydride, gas

Al, Cu, Ni – not hydride forming, but H dissolves
Pt, Pd dissolve much H
Li, Al – hydride
H – interstitial)

Partitioning in fusion ⇒ hotter more 4He than 3He
Richness of possibilities particle theory
HDO → OH compounds, hydrous minerals
High pressure H₂O – excellent solvent

→ HDO → what in crust.

→ Travel → term / D.C.
→ Nashville –

1 H-D
2 High-pressure: geochemical
3 Foil – anomalies
d-d: tritium
4 – Shocked hydride – non-equilibrium
or – heat vibration – diamond cell
Pt-H-D + electric discharge
or laser

– Pt + H₂
Pt + D₂
– HD gas

Max Hill – mtg
Diamond anvil: 200–1000 kbar 1/3 mm diam x 1mm thick
Presses: ~60 kbar
Detectors: ~~phot~~ KI – low eff.
liq. scint. – ~20%, t

Czirr

The catalyzed fusion process outlined above was explained and formulated on or prior to 4/7/86 (April)

Lee R Phillips
Notary, Provo UT expires 12/23/87

This says get [illegible]
them.

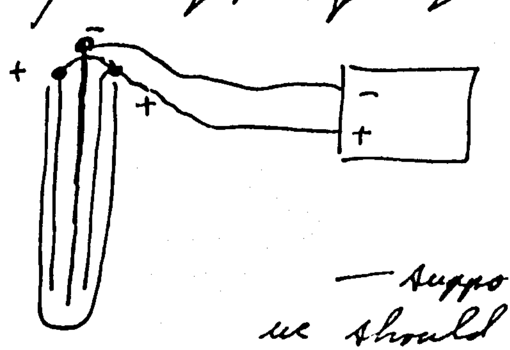
Get common metals and hydride them by chemical means at our instant disposal and test them. (electrolysis and acid bath hydriding)

Get D₂O and hydride metals by common means.

Use vacuum furnace and control hydriding and impurities.

May 22 1986 Electrolytic infusion of hydrogen into metals

We constructed an electrolytic cell in a test tube to try to get hydrogen into metals.



Used a solution of HCl in water. We used copper cathode (−) and nickel anode (+) and ran enough current through to get bubbling at the electrodes.

— supposedly, if Cu behaves like Fe, we should get H in the metal.

After 8-10 hrs, a heavy green coating built up on cathode(−) This flaked off when dry. The cathode was nickel plated underneath in a spotty grey-black silver coating. There was green gelatinous stuff in liquid.

We put this strip which supposedly contained hydrogen and deuterium (in the normal concentration for D of .016 %) around the crystal of a gamma spectrometer. We counted background for 1200 sec and copper strip for about — ~, 2400 sec I believe.

May 22 1986 (cont.) Electrolytic infusion of H.

The results were completely inconclusive. The rate with the hydrated sample was 10% ± 7% greater than background. The count on the background was too short but the equipment was needed for other purposes. As Bart said — you wouldn't bet on cold fusion on the basis of these results — but neither would you bet against it.

We don't know if we had any H in the copper. We are going to weigh it and bake it, but that is of doubtful validity because of oxide formation and surface contamination effects.

May 23, 1986 Electrolytic cell using D_2O.

I rigged up the same cell as before but used 10% D_2O, sulfuric acid, and distilled water with copper anode and cathode. I ran it with 200 mA current and approx 1.5 v across cell. Hydrogen came off the cathode in small bubbles. Ran from ~ 10:00 Am

Bart suggests we use palladium as the metal because we know it has the ability to let H diffuse through it readily. It should work fine.

Ran cell 4½ hr at 200 mA then cut to 100 mA at 3:00 pm

Weighed our first sample and put in furnace to drive off hydrogen and then reweigh — (or maybe we will oxidize and gain weight — who knows.) Turned furnace on at ~ 3:00 pm 500°c

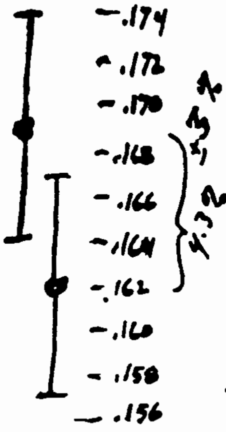
Sample from end outside of electrolytic cell	6.5325 g	before heating
Sample from end in cell — nickel plated	8.4300 g	after heating

May 27, 1986

Rod reported the copper strip gained 50 mg weight — "highly" oxidized and the nickel-plated strip weighed the same. The furnace took so long to get hot that he only took it to 375°C

My Sulfuric acid cell produced a strange strip of copper. It finally ate up the anodes and deposited copper and black stuff (CuO ?) on cathodes. A gamma count gave nothing 1067 background counts in 6600 sec in ~ 3 to 5 MeV and 1149 counts in 6800 sec with Cu strip

May 27, 1986 (Cont.) Electrolysis of Copper in Deuterated Water.



The background rate is .162 ±.005 /sec and the sample rate is .169 ± .005 /sec. At least the trend is in the right direction on paper even though it is not statistically significant.

We do not know if there is hydrogen in the metal. We do not yet know whether the product is a neutron. We do not know whether pressure and/or deformation is required to bring the H-D nuclei closer together for tunneling.

We will look for neutrons next.