CFP 研究の歴史から（6）

― M. Okamoto et al.（1994）によ核変換生成物の局在の最初の確認

常温核融合現象（CFP）が、PdD_x(Li) NiH_x(K, Rb) (x~1) などの CF 試料（CF materials）の中のどの場所で起こるのかという問題は、CFP における核反応の性質がどのようなものなのかに関係した重要な問題であり、最初の Fleischmann et al. (1989) [Fleischmann 1989] の論文でも取り上げられていた問題です。この問題にたいする、最初の具体的な回答を与えたのが Nakada et al. (1993) and Okamoto et al. (1994) の論文でした [Nakada 1993, Okamoto 1994].

1. Okamoto et al. による局在した核反応の確認

1993 年の ICCF3 における彼らの論文から、関連する部分を引用してみましょう：

"*The depth profiles of Li, D and Pd were observed by SIMS technique for several Pd test pieces cut from the Pd electrodes employed in the study of neutron emission from the heavy water electrolysis using Pd-D_2O-LiOD system. The depth profiles can be classified into two types; one is the profile with a structure and the other is the monotonous profile. The former was observed on the Pd test piece from the Pd electrode with neutron emission, while the latter was from the Pd electrode without neutron emission.*" [Nakada 1993 (p. 586)]

ほぼ 1 年後の ICCF4 において、彼らはより精密な実験結果を発表しました：

"*The examples of the depth profiles of D, Li, Pd, Si, and Al are illustrated in Fig. 1. The profiles with bold line represent the depth profiles of each elements obtained from the Pd sample with the nuclear effect of neutron emission and excess heat generation, the medium line for Pd sample with only neutron emission, and the chain-line for Pd sample with no nuclear effect. The curves shown in these figures are normalized to the secondary ion intensity of Pd obtained in each analysis run to carry out the discussion on the concentrations on the same elements. Evidently from the figures, the depth profiles of the elements with no nuclear effects are monotonous as expected from the electrochemical point of view. (2) While, the depth profiles with some nuclear effects have some of irregular structures, especially in the surface within 2 μm for every element. The depth profiles of lithium and deuterium are very similar in each. This fact* *indicates that there is a very strong chemical relation between the lithium behavior and the deuterium behavior as discussed in the previous paper.*" [Okamoto 1994 (p. 14-2)]

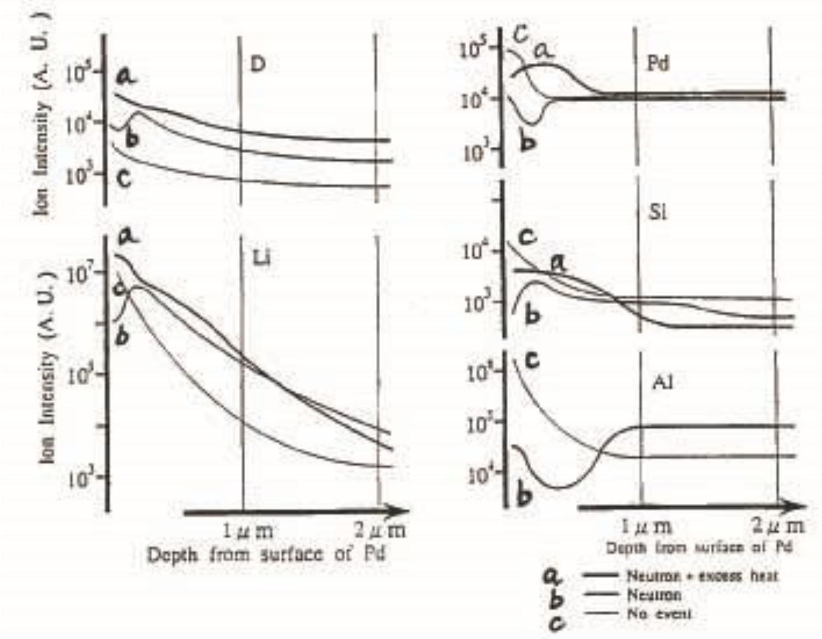

Fig. 1　Examples of depth profiles for each element ([Okamoto 1994 (Fig. 1)]).

この論文（[Okamoto 1994]）を CFRL ウェブサイトの *CFRL News* No. 92 の次に掲示します:

http://www.geocities.jp/hjrfq930/News/news.html

我々は TNCF モデルを使って、Fig. 1 に示された $^{27}_{13}Al$ の減少と $^{28}_{14}Si$ の増加が、次の反応によって説明できることを示しました([Kozima 1996c] [Kozima 1998 (Sec. 11.11e)]);

$^{27}_{13}Al + n \rightarrow {}^{28}_{13}Al^* \rightarrow {}^{28}_{14}Si + e^- + \underline{\nu}_e$,

ここで、 $\underline{\nu}_e$ は電子ニュートリノです。

Dr. Makoto Okamoto のエッセイが、次の CFRL website に掲示してあります:

http://www.geocities.jp/hjrfq930/FTEssay/Essays/Okamoto.htm

Okamoto et al. と同様に Bockris et al. [Bockris 1995] は陰極表面の厚さ数ミクロンの表面層で核変換が起こることを測定しています。

Dr. J.O'M. Bockris のエッセイ "*Cold Fusion 1999*" が次の website に掲示してあります：

http://www.geocities.jp/hjrfq930/FTEssay/Essays/Bockris.htm

2. 多くの研究者が核反応の局在を予想していた

興味のあることは、Okamoto et al. の研究が行われたこの時期に、多くの研究者が表面の重要性に気付いていたことです。

周知のように、Fleischmann et al. は彼らの予想した核反応（d – d fusion reactions and other unidentified reactions）が CF 試料の CF 試料（PdD_x）の容積内（bulk）で起こると結論しています：

“*(a) Excess enthalpy generation is markedly dependent on the applied current density (i.e. magnitude of the shift in the chemical potential) and is proportional to the volume of the electrodes, i.e. we are dealing with a phenomenon in the bulk of the Pd electrodes.*” [Fleischmann 1989 (p. 304)]

しかし、研究が進むにつれて、多くの研究者は試料（CF 物質 CF materials）の表面に実験成功のカギがあることに気付いてきたようで、その最も明確な探求の成果が Okamoto et al. の上記 2 論文であったと言うことができるでしょう。

この時期の多くの研究者の文章に、そのことが記されています。しかしそこには、彼らの予想する核反応（d - d fusion reactions）とは別の、副次的な現象が表面層で起こっていると考えられていた節が読み取れます。例えば、前号（CFRL News No. 91）で取り上げた McKubre et al. では、次のように表現されています：

“*This observation raises the interesting possibility that one or more species, other than deuterium, are required to be present in the cathode in order to observe excess power, species which are not present initially and are thus required to diffuse into the cathode, presumably from the electrolyte. Analyses of used cathodes have revealed the presence of several light elements in the near-surface region (to a depth of several microns); in particular, lithium.*” [McKubre 1993 (p. 18)]

また、Okamoto et al. では、表面層が重陽子の集積に関係した役割をすると考えられています：

“*The process of anomalous accumulation of deuterium was discussed based on the formation of Pd-Li intermetallic compound in the surface area of Pd electrode.*” [Okamoto 1994 (p. 14-7)]

他方、Dash et al. [Dash 1994] は、Pd 陰極表面での Ag and Au の発生を見出し、それが中性子の関係した核変換であろうという予想を述べています：

“Palladium cathodes from two experiments were analyzed microscopically after electrolysis in acidified light and heavy water. For both experiments, more excess heat had occurred in the heavy-water cell than in the light-water cell during electrolysis. For the first experiment, there were localized concentrations of Au detected in areas of the high-current density on both cathodes; greater concentrations of Au were found on the cathodes from the heavy-water cell. For the second experiment, there were concentrations of Ag detected near sites of surface melting on the cathodes from the heavy-water cell. Although occurring in only a small fraction of cathodes, these unexpected elements correlate roughly with measured excess heat and may have arisen through transmutation caused by neutrons from nuclear fusion reactions." [Dash 1994]

さらに、先に取り上げた Bush et al. の Rb→Sr の核変換は（CFRL News No. 89)、Ni メッシュ試料という CF 物質の表面で起こることが明らかであり、試料表面の CFP における重要さを示したものでした：

"Appendix A provides an interpretation of these from the standpoint of hypothetical strontium production. An interesting finding from the standpoint of the CAF Hypothesis [1,3,6] is the fact that, within experimental error, the ratio of the line height for mass number 86 to that for 88 was the same as that for the ratio of the rubidium signals at masses 85 and 87." [Bush 1994 (pp. 346 – 347)]

この実験結果は、TNCF model を用いて説明されました[Kozima 1996a, 1998 (Sec. 11.11b)]。

さらに付け加えるならば､CFRL News No. 88 で取り上げた Morrey et al. の ^{4}He の測定は、TNCF モデルを用いて、CF 物質（この場合は $PdLi_x$）の表面層での Li-*n* 核反応によるものとして説明されたことは前述の通りです[Kozima 1996b, 1997, 1998 (Sec. 11.8a), 1999]

3. 表面層における核反応の性質の探究

このようにして、常温核融合現象における核反応が表面層と深い関係があることが明らかになるにつれて、CF 物質としての体積試料（bulk materials）と層状試料（layer materials）が用いられるようになり、体積試料の場合にもその表面層における核変換が注目されるようになります。T.O. Passell の一連の実験はその中でも優れた成果をあげたものです[Passell 1996, 1998, 2002, 2006]。彼の実験閣下は基本的に TNCF モデルで説明できることを示しました[Kozima 1996d,1998 (Sec. 11.11d)].

このように歴史的な研究の流れで見ると、常温核融合現象における核反応が層状に局在した領域で起こることが明らかになったと言ってよいでしょう。JCF１１で発表した論文［Kozima 2011a、2011b］では、上に概観した論文を含め、Miley et al. [Miley 1996, 1997], Enyo et al. ([Mizuno 1996a, 1996b] and [Ohmori 1996, 1997, 1999], Qiao et al. [Qiao 1997], Iwamura et al. [Iwamura 1998, 2005, 2006a, 2006b, 2006c], および［Celani 2010］など、多くの核変換の実験結果を我々のモデルで説明できることを示しました。

Dr. Michio Enyo のエッセイ“*Open Minded Attitudes to the Science*”が、次の CFRL website に掲示してあります:
http://www.geocities.jp/hjrfq930/FTEssay/Essays/Enyo.htm

最後に、水素吸蔵金属以外の金属を用いた実験について、まとめておきたいと思います。Dash et al. は Pd と硫酸塩（sulfate）を被せた Pt 電極において、Pd 電極と同様な過剰熱が発生することを示しました［Noble 1995］。他方、Ohmori et al. [Ohmori 1996, 1997, 1999］は、Au および W 電極を用いて、表面に核変換によるとしか考えられない異種元素が生ずることを示しました。これらの実験結果は、電極内部よりは表面層が常温核融合現象(CFP)にとって重要であることを示しており、層状試料を用いた Miley et al., Iwamura et al., and Celani et al. の有効性とも関連した現象でもあります。

さらに、Szpak et al. は、電極 - 電解液の界面層の性質が常温核融合現象（CFP）に与える影響に早くから着目して研究を続けてきましたが、PdD_x を Ni, Au や Cu などの表面に co-deposite することで、水素吸蔵を起こさない金属を含めて、陰極表面に PdDx (x 1) 層を持った電極系でのＣＦＰの研究を行っています[Szpak 1992, 1994, 2004, Mosier-Boss 1999]。

References

[Bush 1994] R. Bush and R. Eagleton, “Evidence for Electrolytically Induced Transmutation and Radioactivity Correlated with Excess Heat in Electrolytic Cells with Light Water Rubidium Salt Electrolyte,” *Trans. of Fusion Technology* (*Proc. ICCF4*), **26**, 344 – 354 (1994), ISSN: 0748-1896.

[Bush 1994a] R. Bush and R. Eagleton, “Evidence for Electrolytically Induced Transmutation and Radioactivity Correlated with Excess Heat in Electrolytic Cells with Light Water Rubidium Salt Electrolyte,” *Proc. ICCF4* (Hawaii, USA, Dec. 6 — 9, 1993), Vol.**3**, 2-1 – 2-17 (1994), TR-104188-V3 (EPRI, Palo Alto, California, USA).

[Bockris 1995] J.O'M. Bockris and Z. Minevski, "Two Zones of ‘Impurities’ Observed after Prolonged Electrolysis of Deuterium on Palladium," *Infinite Energy,* **5 & 6**, pp. 67 – 70 (1995 – 96).

[Celani 2010] F. Celani, P. Marini, V. Di Stefano, M. Nakamura, O.M. Calamai, A. Spallone. A. Nuvoli, E. Purchi, V. Andreassi, B. Ortenzi, E. Righi, G. Trenta, A. Mancini, A. Takahashi and A. Kitamura, “First measurement on Nano-coated Ni Wire, at Very High Temperature, under He, Ar, H_2, D_2 Atmosphere and Their Mixtures,” Paper presented at 9th International Workshop on Anomalies in Hydrogen/Deuterium Loaded Metals (Certosa di

Pontignano, Siena-Italy; Sept. 17 – 19, 2010).
[Dash 1994] J. Dash, G. Noble and D. Diman, “Surface Morphology and Microcomposition of Palladium Cathodes after Electrolysis in Acidified Light and Heavy Water; Correlation with Excess Heat,” *Trans. of Fusion Technology* (*Proc. ICCF4*), **26**, 299 – 307 (1994), ISSN: 0748-1896.
[Dash 1994a] J. Dash, G. Noble and D. Diman, “Surface Morphology and Microcomposition of Palladium Cathodes after Electrolysis in Acidified Light and Heavy Water; Correlation with Excess Heat,” *Proc. ICCF4,* Vol. **2**, 25-1 – 25-11 (1994), TR-104188-V2 (EPRI, Palo Alto, California, USA).
[Fleischmann 1989] M. Fleischmann, S. Pons and M. Hawkins, “Electrochemically induced nuclear fusion of deuterium,” *J. E1ectroanal. Chem.,* **261**, 301—308 (1989), ISSN: 1572-6657.
[Iwamura 1998] Y. Iwamura, T. Itoh, N. Gotoh, M. Sakano and I. Toyoda, "Detection of Anomalous Elements, X-ray and Excess Heat induced by Continuous Diffusion of Deuterium through Multi-layer Cathode (Pd/CaO/Pd)," *Proc. ICCF7* (April 20 - 23, 1998, Vancouver, Canada), pp. 167 – 171 (1998). (^{57}Fe/^{56}Fe ratio increased largely.)
[Iwamura 2005] Y. Iwamura, T. Itoh, M. Sakano and S. Sakai, “Observation of Low Energy Nuclear Reactions induced by D_2 Gas Permeation through Pd Complexes,” *Proc. ICCF9* (2002, Beijing, China) pp. 141 – 146 (2005).
[Iwamura 2006a] Iwamura, T. Itoh, M. Sakano, S. Sakai and S. Kuribayashi, “Low Energy Nuclear Transmutation in Condensed Matter induced by D_2 Gas Permeation through Pd Complexes: Correlation between Deuterium Flux and Nuclear Products,” *Proc. ICCF10* (2003, Massachusetts, USA) pp. 435 – 446 (2006), ISBN 981-256-564-7.
[Iwamura 2006b] Y. Iwamura, T. Itoh, M. Sakano, N. Yamazaki, S. Kuribayashi, Y. Terada, T. Ishikawa and J. Kasagi, “Observation of Nuclear Transmutation Reactions induced by D_2 Gas Permeation through Pd Complexes” *Proc. ICCF11,* pp. 339 – 349 (2006), ISBN 981-256-640-6,
[Iwamura 2006c] Y. Iwamura, T. Itoh, M. Sakano, N. Yamazaki, S. Kuribayashi, Y. Terada and T. Ishikawa, “Observation of Surface Distribution of Products by X-ray Fluorescence Spectrometry during D_2 Gas Permeation through Pd Complexes” *Proc. ICCF12* pp. 178 – 187 (2006), ISBN 981-256-901-4.
[Kozima 1996a] H. Kozima, K. Hiroe, M. Nomura, M. Ohta, “Elemental Transmutation in Biological and Chemical Systems,” *Cold Fusion* **16,** 30 – 32

(1996). ISSN 1074-5610. This paper is republished as [Kozima 2015].
[Kozima 1996b] H. Kozima, "Excess Heat and Helium Generation in Cold Fusion Experiments," *Cold Fusion* **17**, 4 – 7 (1996), ISSN 1074-5610.
[Kozima 1996c] H. Kozima, M. Ohta, M. Nomura and K. Hiroe, "Another Evidence of Nuclear Transmutation in Cold fusion Experiment," *Cold Fusion*, 18, 12 – 14 (1996), ISSN: 1074-5610.
[Kozima 1996d] H. Kozima, M. Nomura, K. Hiroe and M. Ohta, "Consistent Explanation of Experimental Data obtained in SRI International and EPRI," *Cold Fusion* **20**, 45 – 48 (1996), ISSN 1074-5610.
[Kozima 1997] H. Kozima, S. Watanabe, K. Hiroe, M. Nomura, M. Ohta and K. Kaki, "Analysis of Cold Fusion Experiments Generating Excess Heat, Tritium and Helium", *J. Electroanal. Chem.* **425**, 173 – 178 (1997), ISSN: 1572-6657 and **445**, 223 (1998) (Errata).
[Kozima 1998] H. Kozima, *Discovery of the Cold Fusion Phenomenon* (Ohtake Shuppan Inc., 1998), ISBN: 4-87186-044-2. For convenience of the readers, the Sections relevant to this analysis are posted at following pages of the CFRL Website:
http://www.geocities.jp/hjrfq930/Books/bookse/bookse01.html
The "References" in this book [Kozima 1998] is posted at the Cold Fusion Research Laboratory (CFRL) Website;
http://www.geocities.jp/hjrfq930/Books/bookse/bookse.html
[Kozima 1999] H. Kozima, K. Arai, M. Fujii, H. Kudoh, K. Yoshimoto and K. Kaki, "Nuclear Reactions in Surface Layers of Deuterium-Loaded Solids" *Fusion Technol.* **36**, 337 – 345 (1999), ISSN 0748-1896.
[Kozima 2002] H. Kozima, K. Yoshimoto, H. Kudoh, M. Fujii and M. Ohta, "Analysis of Zn and Excess Heat Generation in Pd/H_2 (D_2) System by TNCF Model," *J. New Energy*, 6-3, pp. 97 – 102 (2002), ISSN 1086-8259.
[Kozima 2007] H. Kozima, "An Explanation of Nuclear Transmutation in XLPE (Cross-linked Polyethylene) Films with and without Water Trees," *Proc. JCF8*, pp. 44 – 50 (2007).
[Kozima 2011a] H. Kozima, "Localization of Nuclear Reactions in the Cold Fusion Phenomenon," *Proc. JCF11* pp. 59 – 69 (2011), ISSN 2187-2260. And also *Reports of CFRL* (*Cold Fusion Research Laboratory*), **11-2**, pp. 1 – 20 (January, 2011):
http://www.geocities.jp/hjrfq930/Papers/paperr/paperr.html
[Kozima 2011b] H. Kozima and F. Celani, "Brief Explanation of

Experimental Data Set of Excess Heat and Nuclear Transmutation in Multiply Nano-coated Ni Wire," *Proc. JCF11*, pp. 53 – 58 (2011), ISSN 2187-2260. And also *Reports of CFRL* (*Cold Fusion Research Laboratory*), **11-1**, pp. 1 – 8 (January, 2011): http://www.geocities.jp/hjrfq930/Papers/paperr/paperr.html

[Kumazawa 2005] T. Kumazawa, W. Nakagawa and H. Tsurumaru, "A Study on Behavior of Inorganic Impurities in Water Tree," *Electrical Engineering in Japan* **153**, 1 – 13 (2005).

[McKubre 1993] M.C.H. McKubre, S. Crouch-Baker, A.M. Riley, S.I. Smedley and F.L. Tanzella, "Excess Power Observed in Electrochemical Studies of the D/Pd System," *Proc. ICCF3* (Oct. 21 – 25, 1992, Nagoya, Japan), pp. 5 - 19, Universal Academy Press, Inc., Tokyo, 1993, ISBN: 4-946443-12-6.

[Miley 1996] G.H. Miley, G. Narne, M.J. Williams, J.A. Patterson, J. Nix, D. Cravens and H. Hora, "Quantitative Observation of Transmutation Products occurring in Thin-film coated Microspheres during Electrolysis," *Proc. ICCF6* (1996, Hokkaido, Japan) pp. 629 – 644 (1996).

[Miley 1997] G.H. Miley and J.A. Patterson, "Nuclear Transmutations in Thin-film Nickel Coatings undergoing Electrolysis," *J. New Energy* 1-3, pp. 5 – 34 (1996), ISSN 1086-8259.

[Mizuno 1996a] T. Mizuno, T. Ohmori, T. Akimoto, K. Kurosawa, M. Kitaichi, K. Inoda, K. Azumi, S. Shimokawa and M. Enyo, "Isotopic Distribution for the Elements Evolved in Palladium Cathode after Electrolysis in D_2O Solution," *Proc. ICCF6*, 665 – 669 (1996).

[Mizuno 1996b] T. Mizuno, T. Ohmori and M. Enyo, "Anomalous Isotopic Distribution in Palladium Cathode after Electrolysis," *J. New Energy* **1-2**, 37 – 44 (1996), ISSN 1086-8259.

[Mo 1998] D.W. Mo, Q.S. Cai, L.M. Wang, S.Z. Wang, X.Z. Li, "The Evidence of Nuclear Transmutation Phenomenon in Pd-H System using Neutron Activation Analysis (NAA)," *Proc. ICCF7*, pp. 259 – 263 (1998).

[Morrey 1990] J.R. Morrey, M.R. Caffee, H. Farrar, IV, N.J. Hoffman, G.B. Hudson, R.H. Jones, M.D. Kurz, J. Lupton, B.M. Oliver, B.V. Ruiz, J.F. Wacker and A. Van, "Measurements of Helium in Electrolyzed Palladium," *Fusion Technol.* **18**, 659 (1990) , ISSN 0748-1896.

[Mosier-Boss 1999] P.A. Mosier-Boss and S. Szpak, "The Pd/(n)H System: Transport Processes and Development of Thermal Instabilities." *Nuovo Cimento.* A**112,** 577 – 585 (1999), ISSN: 0369-3546.

[Nakada 1993] M. Nakada, Tl. Kusunoki, M. Okamoto and O. Odawara, "A Role of Lithium on Neutron Emission in Heavy Water Electrolysis," *Proc. ICCF3*, 581 – 586 (1993), ISBN: 4-946443-12-6.
[Noble 1995] G. Noble, J. Dash and L. McNasser, "Electrolysis of Heavy Water with a Palladium and Surface Composite," *Proc. ICCF5*, 136 – 139 (1995).
[Ohmori 1996] T. Ohmori, T. Mizuno and M. Enyo, "Isotopic Distributions of Heavy Metal Elements Produced during the Light Water Electrolysis with Gold Cathode," *J. New Energy*, **1-3**, 90 – 99 (1996), ISSN 1086-8259.
[Ohmori 1997] T. Ohmori, M. Enyo, T. Mizuno, Y. Nodasaka and H. Minagawa, "Transmutation in the Electrolysis of Light Water – Excess Energy and Iron Production in a Gold Electrode," *Fusion Technol.*, **31**, 210 – 218 (1997), ISSN 0748-1896.
[Ohmori 1999] T. Ohmori and T. Mizuno, "Nuclear Transmutation Reaction Caused by Light Water Electrolysis on Tungsten Cathode under Incandescent Conditions," *Infinite Energy*, **27**, pp. 34 – 39 (1999).
[Okamoto 1994] M. Okamoto, H. Ogawa, Y. Yoshinaga, T. Kusunoki and O. Odawara, "Behavior of Key Elements in Pd for the Solid State Nuclear Phenomena Occurred in Heavy Water Electrolysis," *Proc. ICCF4* (Hawaii, USA, Dec. 6 — 9, 1993), Vol.**3**, 14-1 – 14-8 (1994), TR-104188-V3 (EPRI, Palo Alto, California, USA).
[Passell 1996] T.O. Passell, "Search for Nuclear Reaction Products in Heat-Producing Palladium," *Proc. ICCF6*, pp. 282 – 290 (1996), (Published by New Energy and Industrial Technology Development Organization (NEDO) and The Institute of Applied Energy (IAE)).
[Passell 1998] T.O. Passell, "Search for Nuclear Reaction Products in Heat-Producing Pd," *Proc. ICCF7*, pp. 309 – 313 (1998), (Published by ENECO).
[Passell 2002] T.O. Passell, "Evidence for Lithium-6 Depletion in Pd exposed to Gaseous deuterium and Hydrogen," *Proc. ICCF9*, pp. 299 – 304 (2002), ISBN 7-302- 06489- X/O ·292.
[Passell 2006] T.O. Passell, "Pd-110/108 Ratio and Trace Element Changes in Particulate Palladium exposed to Deuterium Gas," *Proc. ICCF10*, pp. 399 – 403 (2006), ISBN 981-256-564-7.
[Patterson 1997] J. Patterson, "US Patent #5,607,563" *Elemental Energy* (*Cold Fusion*), 22, pp. 3 – 17 (1997).

[Qiao 1997] G.S. Qiao, X.M. Han, L.C. Kong and X.Z. Li, "Nuclear Transmutation in a Gas-Loading H/Pd System," *J. New Energy,* **2-2**, pp. 48 – 52 (1997), ISSN 1086-8259.

[Szpak 1992] S. Szpak, P.A. Mosier-Boss, S.R. Scharber and J.J. Smith, "Charging of the Pd/^{n}H System: Role of the Interphase," *J. Electroanal. Chem.*, **337,** 147 – 163 (1992).

[Szpak 1994] S. Szpak, P.A. Mosier-Boss and J.J. Smith, "Deuterium Uptake during Pd-D Co-deposition," *J. Electroanal. Chem.*, **379,** 121 – 127 (1992).

[Szpak 2004] S. Szpak, P.A. Mosier-Boss, M.H. Miles and M. Fleischmann, "Thermal Behavior of Polarized Pd/D Electrodes Prepared by Co-deposition," *Thermochimica Acta,* **410,** 101–107 (2004).