

Research Article

# Transmutations and Isotopic Shifts in LENR Experiments An Overview

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## Abstract

This overview presents a brief summary of observations of products of transmutation reactions, which occur in a variety of LENR configurations wherein the “host metal” nuclei react with loaded deuterium or hydrogen, resulting in the formation of new stable elements or isotopes not present prior to an experimental run.

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## 1. Introduction

The term “Transmutations” as used in the LENR field has come to refer to the occurrence of nuclear reactions between the loaded deuterium (or deuterons) or hydrogen (protons) on the one hand, and nuclei of the host metals such as Pd, Ni, Ti or of higher-Z nuclei present in the experimental environment such as those of alloyed elements or impurities in the cathode or even elements present in the electrolyte (in case of electrolysis experiments), on the other. Any nuclear reactions that occur among hydrogenous isotopes such as p or d (or even tritium) present in the reactive zone would generally be classified as “fusion” reactions. As understood in the context of the present discussion the basic difference between transmutations and fusion is that in the former the host metal lattice nuclei participate directly in the nuclear processes taking place while in fusion they merely serve a catalytic role but themselves do not get involved.

Interestingly there are a few instances wherein nuclear reactions do seem to occur even when neither p or d is externally injected into the system nor is any host metal present as such. Examples are the Carbon Arc under water experiments and the phenomenon called “Biological Transmutations”. But in both these cases hydrogen (and deuterium from natural abundance) are already present in the configuration.

When Fleischmann–Pons announced their discovery in 1989, as is well known, it was (and still is) met with considerable skepticism by the Nuclear Physics community. The postulated occurrence of nuclear reactions between two positively charged nuclei having nuclear charge of unity at normal temperatures was dismissed as impossible. Under

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these circumstances any suggestion of the likelihood of a nuclear reaction occurring at room temperature between a deuteron (or proton) and any higher-Z nucleus would be considered as “preposterous”!

It is in the backdrop of this “intense disbelief” of the whole topic of LENR transmutations that we briefly review the experimental evidence gathered by several groups the world over, deploying a variety of different experimental configurations and analytical techniques, over the last couple of decades.

The present overview is a condensed version of a review paper published a year ago [1] jointly with George Miley and Edmund Storms (an electronic version of the preprint of this article which was distributed to participants of ICCF 16 conference is available in [2]). Miley had earlier published periodic status reports on Transmutations [3,4]. Storms had included in his book published in 2007 [5] an exhaustive tabulation of the nuclear products observed by various researchers as reported in a 101 references to LENR Transmutations related publications.

## 2. General Remarks on Experimental Methodology

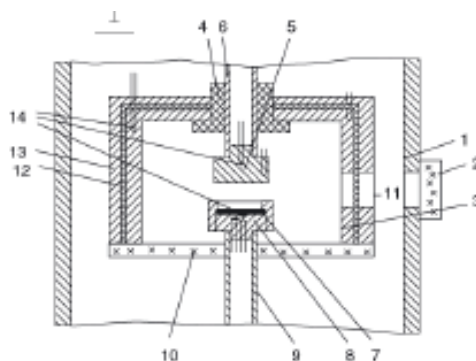
Transmutation experiments generally involve two steps: The experimental run during which a target or test sample is loaded with deuterons or protons by a suitable technique such as electrolysis, gas/plasma loading or other. In the second step, on completion of the experimental run, the test sample is analyzed off line to determine if there is any evidence for transmutation reactions having occurred. This involves measuring the elemental composition and/or isotopic distribution of various components, which were present in the reaction zone. In the case of electrolysis and glow discharge experiments the cathode would be the one mainly investigated. Obviously mere detection of traces of a “new” element which was not present prior to the run does not imply occurrence of transmutations, since in principle cross contamination could have occurred through inadvertent transport of minute quantities of elements from elsewhere in the apparatus. For example during electrolysis trace quantities of impurities could easily have plated out from the electrolytic solutions. In glow discharge experiments plasma etching could have sputtered out some elements and redeposited them on the test sample. It is therefore important to ensure such type of contamination is not the cause of the observations. Having accurate elemental composition of the stock electrolytic solution, especially of trace elements, available prior to the commencement of the experimental runs, would be essential.

However, if the isotopic distribution of the newly found elements or for that matter of any of the materials previously present in the system, are found to be significantly different from their natural abundance values following a run, then it clearly points to anomalous nuclear processes having taken place. Such findings are often referred to as “isotopic anomalies”. Advanced mass spectrometric analytic tools available these days, such as Secondary Ion Mass Spectrometry (SIMS) permit accurate and often *in situ* isotopic distribution measurements. However, mass spectroscopy is known to be subject to errors arising from interference effects caused by molecular ion species having masses close to that of the isotope under measurement. This possibility has to be addressed before concluding that the observed “isotopic anomalies” are real.

Lastly the well-known characteristic of non-uniformity of the LENR phenomenon has to be recognized while interpreting the transmutation results. Ed Storms has propounded the interesting concept of Nuclear Active Environment (NAE), to explain this. Invariably the phenomenon is found to occur in one spot but not in a neighboring one. Also, a systematic variation of the reaction product concentrations is often discerned as one goes from outer layers to deeper layers. Depth profiling of new element concentrations and isotopic ratios has at times influenced acceptance of the genuineness of the transmutation results.

## 3. Russian Glow Discharge Measurements

Karabut and Savvatimova were among the earliest researchers to investigate LENR transmutation phenomena using glow discharge. Figure 1 is a schematic of their glow discharge apparatus, which is basically a double-walled quartz



**Figure 1.** Glow discharge apparatus used by Karabut et al.

vacuum chamber with Mo anode and cathode. The design of the setup permitted use of different cathode material inserts for study [6]. The chamber was evacuated and filled with  $D_2$  gas to a pressure of 3–10 Torr. The region of the cathode bombarded by the plasma ions was typically  $\sim 1 \text{ cm}^2$  in area. Applied voltage varied from 50 V to 1.2 kV; discharge current was  $\sim 100 \text{ mA}$ .

The chamber and electrodes were water cooled to perform calorimetric measurements. The authors have reported observing excess heat consistently with near 100% reproducibility, but not detecting the normal (d–d) fusion reaction products such as neutrons, tritium, or even helium, commensurate with the magnitude of the heat generated. Hence their persistent quest for evidence of transmutation products.

Prior to commencement, the impurity content of virgin Pd cathode material was confirmed to be under 0.01%. Post-discharge Pd cathode buttons were analyzed using: Surface topography by scanning electron microscopy; elemental and isotopic composition using spark mass spectrometry, SIMS, Thermal Ionization Mass Spectrometry (TIMS) and XRF. Autoradiography was used for evidence of any remnant radioactivity. Results have consistently indicated significant deviations from natural abundance values for most elements.

At the Nagoya ICCF 3 meeting (1992), Karabut et al. reported finding as much as 0.1% of Na, Mg, Br, Zn, S, Mo, and Si in the upper crust of the Pd. The top  $1 \mu\text{m}$  layer of the Pd sample was examined at several spots in the front portion, the back portion, and shielded area with a spatial resolution of  $1 \mu\text{m}$  using an X-ray microprobe analyzer. It was found that the content of some elements increased by tens to hundreds of times relative to initial content in virgin Pd.

At ICCF 5 held in Monaco in 1995, they reported finding significant spot to spot variations using an X-ray microprobe analyzer. In some spots, the Ag content was as high as 12–15% and Mo about 5–7%. The concentration of elements such as As, Br, Rb, Sr, Y, and Cd, which were not present in any of the construction materials used in the experimental apparatus, was in the range of 0.1–0.2%. A new result reported at the Monaco meeting was that even with hydrogenous plasma, they observed elements not present in the virgin cathode, but in general the products' yield with deuterium gas was orders of magnitude higher.

At ICCF 9 held in Beijing in 2002, Karabut reported [7] new results obtained by subjecting the discharge device to an "impulsive periodical power source" (pulsed voltage), which led to generation of intense X-ray laser beams. The main impurity nuclides (with more than 1% content altogether) registered in the top 100 nm thick surface layer were  $Li^7$ ,  $C^{12}$ ,  $N^{15}$ ,  $Ne^{20}$ ,  $Si^{29}$ ,  $Ca^{44}$ ,  $Ca^{48}$ ,  $Fe^{56}$ ,  $Fe^{57}$ ,  $Co^{59}$ ,  $Zn^{64}$ ,  $Zn^{66}$ ,  $As^{75}$ ,  $Ag^{107}$ ,  $Ag^{109}$ ,  $Cd^{110}$ ,  $Cd^{111}$ ,  $Cd^{112}$ , and  $Cd^{113}$ . They identified two broad categories of impurity elements: those with masses roughly half of that of Pd (probably caused by deuteron induced fission) and those with masses close to but above that of Pd (possibly caused by

multiple deuteron captures).

At ICCF 12, held in Yokohama in December 2005, Karabut presented further results from discharges carried out with V, Nb, and Ta cathodes and in the inert gases of Xe and Kr besides D<sub>2</sub>. In general, with cathodes other than Pd, “impurity” element yield was significantly lower. In these experiments, Karabut also measured the impurity content yield after peeling off some atomic layers using plasma etching and then again measuring the elemental content using SIMS.

At the same conference in Yokohama, Savvatimova presented [8] a very detailed and exhaustive account of her independently conducted glow discharge results with hydrogen, deuterium, argon, and argon-xenon mixture plasmas. The influence of various experimental parameters such as nature of plasma gas, total dose of bombarding ions, discharge current density (mA/cm<sup>2</sup>), and type of applied voltage (direct or pulsed) on the yield of “additional” elements was studied systematically. This time she also used multilayer cathodes comprising several foils of 100 μm thickness stacked one on top of the other to study differences in product yield characteristics with depth. The greatest changes in “additional” element content and isotope shifts were found in certain “hot spots” (mostly near grain boundaries) where a micro-explosion or plasma micro-discharges had appeared to have taken place. The author makes special mention of elements with mass numbers 59 (Co), 55 (Mn), and 45 (Sc), which were always found in plenty in the post-discharge samples but never in initial samples. One intriguing observation was that the isotopic changes continued to occur for at least three to five months after glow discharge exposure. Several isotopes with masses less than those of W and Ta increased by factors ranging from 5 to 1000 times.

On the whole, Savvatimova found that the more deeply she investigated the LENR glow discharge phenomenon, the more complex it was found to be, as brought out in the 13 tables of results included in her Yokohama paper.

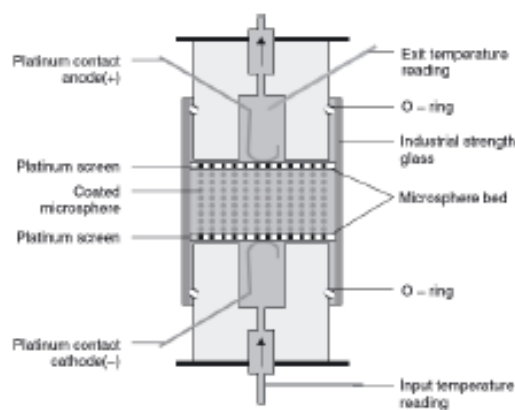
#### 4. Electrolysis Experiments

It was the Miley–Patterson paper first published in 1996 [9] that perhaps really opened the door to acceptance of Transmutation being possibly real even within the CMNS community. Industrial chemist James Patterson had invented a pebble bed cathode, circulating solution electrolytic cell wherein the cathode was made up of a bed of Pd-Ni multilayer thin film coated plastic microspheres (~1 mm dia). There were typically 1000 microspheres in the cell forming a four or five-layer bed constituting the cathode. Li<sub>2</sub>SO<sub>4</sub> solution served as electrolyte as well as coolant. Figure 2 is a schematic of this cell. As this cell showed excess heat with both D<sub>2</sub>O based as well as H<sub>2</sub>O based electrolytic solutions, Patterson entrusted Miley’s group at the University of Illinois to perform elemental analysis of the coating of the post run beads to determine if any nuclear products could be identified.

When Miley found what appeared to be a gamut of new elements he repeated the electrolytic runs in his lab after fabricating his own version of multilayer thin film coated cathodes as well as a fresh electrolytic cell using no metallic components, to preclude the possibility of trace elements from entering the solution and causing contamination.

An advantage of thin film cathodes (coating thicknesses varied in the range of 500–3000 Å) is that high deuterium or hydrogen loadings could be obtained in time durations as short as an hour or two. Also the nuclear products would constitute a larger fraction of the metallic mass, minimizing doubts that the new elements found could be due to impurities deposited from the electrolyte. Miley and his colleagues carried out more than a dozen electrolytic runs with various types of coatings. Following several weeks of electrolysis, beads from the upper layers of the packed bed cathode were retrieved for analysis.

A variety of measurement techniques such as Secondary Ion Mass Spectrometry (SIMS), Energy Dispersive X-ray (EDX) analysis, Auger Electron Spectroscopy (AES), and Neutron Activation Analysis (NAA) were employed. While EDX gave confirmatory data for the higher concentration elements, AES was used for depth profiling of these elements. SIMS was used to obtain an overall picture of the various nuclides present and their relative isotopic ratios while NAA gave a quantitative measure of eight key elements, namely Al, Ag, Cr, Fe, Cu, V, Co, and Zn, present in a gross sample



**Figure 2.** Schematic of Patterson power cell.

containing 10 microspheres. In the case of Cu and Ag, NAA helped establish deviations of isotopic composition from their natural abundance values. NAA has the advantage that it circumvents the molecular ion interference problem. Since NAA typically gives an average value integrated over 10 beads, it averages out the significant bead-to-bead variations in the reaction product yields, which are known to be sensitive to the location of the microspheres in the packed bed.

The results confirmed the presence of a wide range of new elements in the post-run thin films. The reaction products had mass numbers ranging both below and above the atomic mass number of the host metal, spanning across the entire periodic table. Figure 3 is a smoothened out plot of the reaction product yields plotted against the atomic number ( $Z$  value) of the product elements. A characteristic four-humped yield spectrum is evident with humps occurring at  $Z = 6-18$  (peak at Mg–Si),  $Z = 22-30$  (peak at Fe–Zn),  $Z = 44-50$  (peak at Ag–Cd), and  $Z = 75-85$  (peak at Au). In some of the runs, as much as 40% of the initial metal atoms of the thin film coating was transmuted. Miley speculates that each of these groups of elements is derived from one of the main elemental components used in the construction of the cell such as sulphur, nickel, palladium, and platinum (which was the anode material).

SIMS results indicated that the isotopic composition of most of the elements showed substantial deviation from natural abundance, whereas data of the control beads corresponded to natural isotopic ratios only. NAA data for Ag and Cu also confirmed significant deviations from natural abundance. It was however not possible to discern any systematic in the isotopic shift results, since there was considerable scatter in the cathode bed.

Miley's papers have also discussed the differences in yield spectrum between different base metal coatings, differences in product yield between plastic beads and glass microspheres and differences between  $H_2O$  runs and  $D_2O$  runs. The similarity of this four humped yield curve with the well-known double-humped yield curve observed in neutron induced fission has led to speculation that there could be an analogous proton or deuteron induced fission of the compound nucleus formed between a host metal nucleus and one or more protons or deuterons in LENR configurations.

Inspired by Miley's findings reported at the First International Conference on Low Energy Nuclear Reactions held at College Station, Texas, in June 1995, Mizuno of Hokkaido University, carried out a systematic analysis of his post-run Pd cathodes that had earlier been electrolyzed in heavy water solutions and to his pleasant surprise also found a four humped yield spectrum similar to that of Miley (see Fig. 4). Mizuno has elaborated on the details of his transmutation quest both in his book [10] as well as a recent review paper [11].

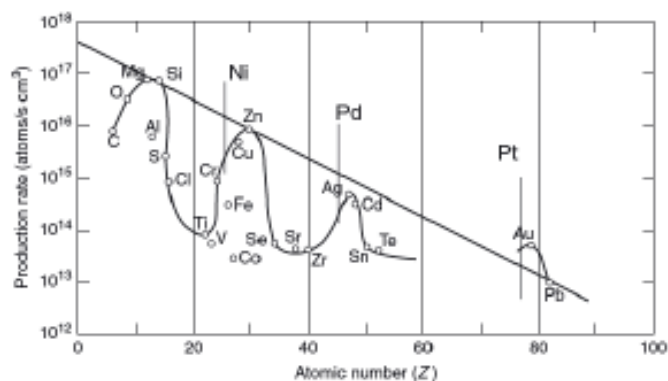


Figure 3. Reaction product yield vs. atomic number (Miley).

## 5. Deuterium Gas Permeation (Iwamura)

Possibly the most spectacular of transmutation findings in the CF/LENR field are those of Yasuhiro Iwamura and his colleagues at the MHI Laboratories in Japan who have been systematically studying the nuclear products formed during the loading and diffusion of deuterium in single and multilayer nano-structured Pd foil complexes, for close to two decades. Suspecting that impurities could play an important role in the nuclear processes, Iwamura incorporated CaO in the form of alternate layers of CaO and Pd in a multilayer foil complex. CaO was selected because of its low work function [12]. In an early experiment they mounted such a multilayer foil complex at the bottom of an electrolytic cell and evacuated the chamber underneath the cell to promote diffusion of deuterons towards the outer evacuated face. Subsequent post run examination of the inner (electrolyzed) face showed the presence of large amounts of Ti

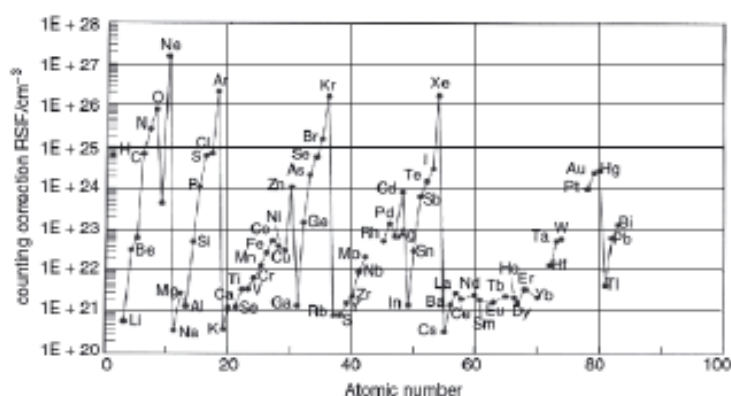


Figure 4. Yield of elements formed on Pd surface (Mizuno).

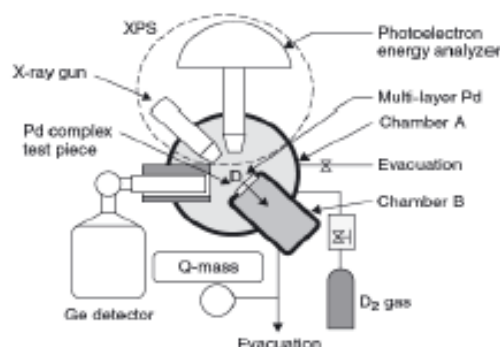
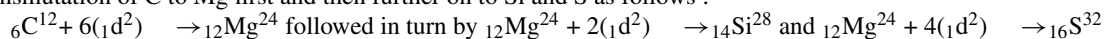


Figure 5. Iwamura's gas diffusion apparatus.

and ( $\text{Fe}^{57}/\text{Fe}^{56}$ ) ratios as high as 1.8 in selected spots, compared to the ratio's natural abundance value of 0.023. This surprising finding set the stage for Iwamura's long "Transmutation journey".

Having experienced the advantage of vacuum assisted permeation, they reverted back to pure gas loading, incorporating instrumentation which enabled *in situ* elemental analysis (using XPS) and measurement of isotopic ratios (using SIMS) without having to take the sample out for analysis, thereby eliminating the possibility of contamination.

Figure 5 is a schematic diagram of the apparatus used by them in most of their subsequent investigations. In one of their first experiments with this set up they observed carbon as an initial impurity on the surface prior to filling  $\text{D}_2$  gas in the chamber, a phenomenon well known to those handling vacuum systems. But it was what they observed after  $\text{D}_2$  gas permeation over a period of a few days that surprised them. They had obtained evidence [13] for the sequential transmutation of C to Mg first and then further on to Si and S as follows :



In these experiments some nuclides present on the surface appear to be absorbing deuterons in pairs of 2 or multiples thereof such as 4 or 6. Such multiheavy ion nuclear reactions have never been observed even in the most advanced

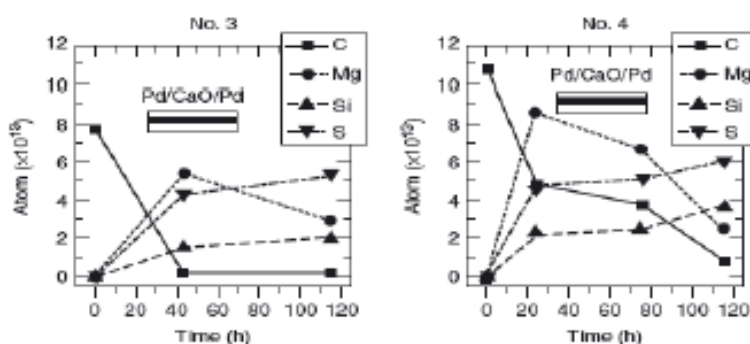


Figure 6. XPS measurement of time dependence of C, Mg, Si, and S on surface of multilayer Pd samples.

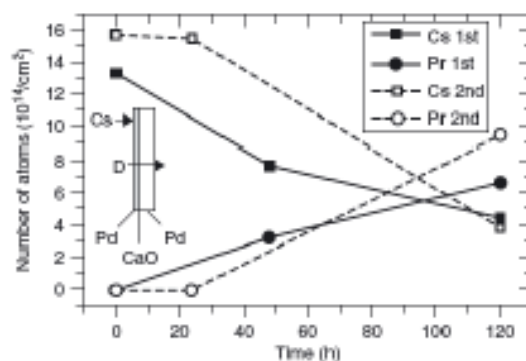


Figure 7. Variation of Cs and Pr concentration.

nuclear physics laboratories elsewhere, let alone the fact that this unbelievable reaction seems to be taking place during the simple act of deuterons diffusing through a foil complex, that too at room temperature (actually they found keeping the foil complex at a slightly higher temperature helped deuteron diffusion rate).

With a view to further confirm occurrence of such multi deuteron capture transmutations, in the next step they deposited a thin layer of a test element on top of the multilayer complex and followed the progressive formation of new elements and isotope species during the  $\text{D}_2$  permeation, over a period of 1 or 2 weeks, through periodic *in situ* measurements, using XPS and SIMS.

In the first of such “designed transmutation” studies they coated  ${}^7\text{Li}$  as a dopant on the surface and observed the production of  ${}^{19}\text{F}$  following capture of six deuterons which then went on to become  ${}^{27}\text{Al}$  following a further capture of four more deuterons. In a subsequent series of experiments [13] they studied the conversion of  ${}^{133}\text{Cs}$  to  ${}^{141}\text{Pr}$  following four deuteron captures. This is brought out in Fig. 7.

In a recent overview [14] of their two decade long pursuit of LENR transmutations, Iwamura has placed his findings in perspective. He confesses that he still does not understand why exactly addition of  $\text{CaO}$  (or  $\text{Y}_2\text{O}_3$ ) works but not  $\text{MgO}$  and why such multideuteron captures work mostly only with intermediate layer coatings of alkali and alkaline earth elements and not others.

## 6. Biological Transmutations

In the 1960s a book was published [15] by a French medical doctor titled “Biological Transmutations” which summarized the results of a number of prior experiments conducted over the previous couple of centuries that seemed to suggest that elemental (nuclear) transmutations do occur in plants and animals and even human beings. Of course the techniques for analysis of elemental composition of substances in those days were rather crude and as such the scientific community did not take the claims of Biological Transmutation seriously. With availability of improved analytic methods, evidence for this Biological phenomenon has continued to be reported. Since the cold fusion era began many such papers have been presented at various ICCF series conferences, especially on the topic of Microbial Transmutation. In recent years with mass spectroscopic instrumentation becoming more commonly available, isotopic anomalies in Biological Transmutation experiments have also been published. During the last decade Vladimir Vysotskii of Keiv, Ukraine, has conducted a series of systematic measurements carrying forward the field from where Kervran left it. The most impressive of Vysotskii’s recent work reports on the transmutation of radioactive  ${}^{137}\text{Cs}$  to stable  ${}^{138}\text{Ba}$  using microbial colonies in light water based cultures. Due to space restriction we are however unable to discuss more about Vysotskii’s



work here and refer interested readers to his review paper published in the ACS LENR Source book Vol. 1 [16] and his book published in 2009 [17].

Biberian has recently published a review article [18] and a book in French [19] on the historical evolution of “Biological Transmutations”.

## 7. Conclusions

In this brief overview we could discuss the work of only about half a dozen researchers, but there are many more. We selected examples each from glow discharge, electrolysis, gas loading and biological transmutation. Almost all the results reveal a pattern of essentially similar behavior :

In all the experiments the newly found elements or isotopes appear to be explainable through occurrence of multiple deuteron captures in one or more of the isotopes of the high-Z elements in/on the cathode, followed by fission of some of the complex intermediate compound nuclei.

In Iwamura's  $D_2$  permeation runs, 2, 4 or 6 deuterons are absorbed by certain test elements deposited on the surface, during the simple act of diffusion. (However Iwamura has not reported observing any evidence for occurrence of fission following multiple deuteron captures.)

All the results, clearly point to the phenomenon occurring mostly in highly localized spots. Russian investigators have suggested that these spots appear to be associated with grain boundaries, in the case of bulk metal electrodes. Several researchers have observed microcraters on the surface and along whose rims transmutation products are found.

Although the best results are obtained with Pd and D, glow discharge experiments show that other combinations of cathode material and fill gas environments also seem to support such transmutation reactions.

The question arises as to whether these transmutations are merely an “academically interesting side show” in the overall field of CF/LENR or do they play a central role in macroscopic (industrial) scale energy production? While in PdD systems Helium appears to be the main nuclear product, in the case of Ni–H systems the question is still open. Miley had earlier suggested that transmutation products are most likely the source of excess heat in their thin film cathode light water runs. Similarly Karabut and Savvatimova started looking for transmutation products primarily because neutrons, tritium and helium could not be detected in quantities commensurate with the observed excess heat in their glow discharge runs. Andrea Rossi has recently gone on record to state that transmutation of Ni to Cu is the source of nuclear heat in their Ni–H devices. Reliable experimental data from analysis of spent Ni fuel powders is keenly awaited to settle this question.

Vysotskii's microbial transmutation experiments are the only reliable measurements to date wherein radioactive nuclei have been transmuted to stable nuclei, to the best of this authors knowledge, although there were many experimental attempts and claims of electrolytic remediation of Thorium radioactivity in the mid-1990s.

The occurrence of transmutation reactions in simple LENR configurations has however a more profound implication, namely it questions a 300 year old “dogmatic belief” of Science, prevalent from the days of Lavoisier, that one cannot transmute one element into another in any simple laboratory experiment, no matter what you do, such as heating, cooling, applying pressure, passing a current, etc., other than by bombarding the target nuclei with nuclear particles such as neutrons or high energy alpha particles or using particle accelerators.

These results are clearly suggesting that the age old claims of Alchemy may perhaps be true after all, however unpalatable it may be to the Scientific community [20]. In this context it may be worth mentioning that at least two groups [21,22] of LENR researchers have during the last one year come up with exploratory proposals to manufacture rare and valuable stable elements including gold, deploying LENR based processes.

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