Introduction to isotopic shifts and transmutations observed in LENR experiments

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This article presents a brief introduction to the topic of transmutation reactions which occur in a variety of LENR configurations wherein the 'host metal' nuclei (Pd, Ni, Ti, etc.) interact with the loaded deuterium or hydrogen nuclei, resulting in the formation of new stable elements or isotopes not present in the system prior to the experimental run.

Keywords: Cold fusion, isotopic anomalies, multi-deuteron capture reactions, transmutation reactions.

Introduction

THE term 'transmutations' as used in the low energy nuclear reactions (LENR) field has come to refer to the occurrence of nuclear reactions between the loaded deuterium (deuterons) or hydrogen (protons) on the one hand, and nuclei of the host metal such as Pd, Ni, Ti, etc. or of other high Z-components 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. Nuclear reactions that take place among the hydrogenous isotopes themselves, such as p or d present in the reactive zone, are referred to as 'fusion' reactions.

Possibly the most spectacular of transmutation findings in the cold fusion (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 on the surface during the simple act of diffusion of deuterium in multilayer nano-structured Pd foil complexes, for close to two decades. These are discussed in the immediately following paper in this issue. Also, there are some examples of living organisms/species 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, although protons (and deuterons from natural abundance) are present in biological substances. This phenomenon, referred to as 'biological transmutations', is discussed in two papers in this special section. In the present article we discuss the transmutation reactions observed by two representative groups, one in glow discharge experiments and other in electrolysis experiments with coated thin-film cathodes. These examples are extracted from a comprehensive review paper[1] on LENR transmutations jointly authored with George Miley and Edmund Storms and published in the Wiley Encyclopedia of Nuclear Energy in 2011 and a shorter version published in the *Journal of Condensed Matter Nuclear Science*[2].

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Typical experimental procedure

Transmutation experiments generally involve two steps: The experimental run during which a target or test sample is first loaded with deuterons or protons by a suitable technique such as electrolysis, gas/plasma loading or any other procedure and then triggering it appropriately to cause nuclear reactions. In the second step, the post-run test sample is analysed to determine if there is any evidence for transmutation reactions having occurred. This involves measuring the elemental composition and/or isotopic distribution of various elemental components which are present in the reaction zone before and after the loading. In the case of electrolysis and glow discharge experiments, the cathode would be the one mainly studied. Obviously, mere detection of traces of a 'new' element which was not present prior to the run does not confirm 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 various impurities could have easily 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 that such type of contamination is not the cause of the observations through appropriate control runs.

However, if the isotopic distribution of the 'newly found elements' or for that matter of any of the materials previously present in the system, is 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 referred to as 'isotopic anomalies'. Advanced mass spectrometric analytic tools 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 genuine.

Lastly, the well-known phenomenon of non-uniformity of the LENR reactions has to be recognized while interpreting the transmutation results. Storms has propounded the interesting concept of nuclear active environment or NAE for short, to explain this characteristic of LENR (papers by Storms in this Special Section). Consequently, the LENR reactions are invariably found to occur in one spot but not in a neighbouring one. Also, a systematic variation of the magnitude of the phenomenon is often discerned as one goes from outer layers to deeper layers of the test sample. Thus, depth profiling of new element concentrations and isotopic ratios has contributed to establishing the genuineness of the transmutation results.

Russian glow discharge experiments

Karabut and Savvatimova were among the earliest researchers to study LENR transmutation phenomenon using glow discharge[3,4]. Figure 1 is a schematic of their glow discharge apparatus, which is basically a double-walled quartz vacuum chamber with a Mo anode and a cathode. The design of the set-up permitted use of different cathode materials for study. 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 ~1 cm^2 in area. Applied voltage varied from 50 V to 1.2 kV; discharge current was ~100 mA.

The chamber and electrodes were separately water-cooled to perform calorimetric measurements. The authors have reported observing excess heat consistently with near 100% reproducibility[4]. The quantum of excess heat increased with applied input power, reaching a maximum of ~15 kW when the input was ~120 kW. But the experimenters did not detect the normal (d–d) fusion reaction products such as neutrons, tritium, or even helium, commensurate with the magnitude of the heat generated. Hence their continued quest for finding transmutation products.

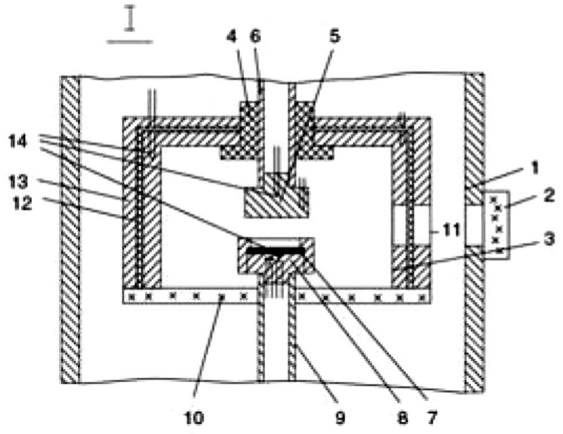

Figure 1. Glow discharge apparatus used by Karabut *et al*.

Prior to commencement, the impurity content of the virgin Pd cathode material was confirmed to be under 0.01%. Post-discharge Pd cathode buttons were analysed using surface topography by scanning electron microscopy; elemental and isotopic composition using spark mass spectrometry, SIMS, thermal ionization mass spectrometry (TIMS) and XRF; and autoradiography for evidence of any remnant radioactivity. Results have consistently indicated significant deviations from natural abundance values for most elements, while the virgin cathode materials showed only natural isotopic abundance.

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 Pd. The top 1 μm layer of the Pd sample was examined at several spots in the front portion, back portion and shielded area with a spatial resolution of 1 μm using an X-ray microprobe analyser. It was found that the content of some elements increased by tens to hundreds of times relative to initial content in virgin Pd in some spots.

At ICCF 5 held in Monaco in 1995, they reported finding significant spot-to-spot variations using an X-ray microprobe analyser. 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 are not present in any of the construction materials used in the experimental apparatus, was in the range 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 and Karabut[4] reported new results obtained by subjecting the discharge device to an 'impulsive periodical power source' (pulsed voltage), which led to the generation of intense 'X-ray laser beams'. This puzzling observation is discussed in detail in a companion paper by Hagelstein in this Special Section. The 'impurity nuclides' (with more than 1% content) 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 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_2. In general, however, with cathodes other than Pd, 'impurity' element yield was significantly lower. Their paper also presents details of the 'impurity

content' yield variation following peeling-off of top layers of the cathode material using plasma etching.

Electrolysis experiments

It was the Miley–Patterson paper first published in 1996 that perhaps really opened the door to acceptance of transmutation as being 'possibly real' even within the CMNS community[5]. Industrial chemist James Patterson had invented a pebble-bed cathode, circulating electrolytic solution 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 4 or 5-layered bed constituting the cathode. Li_2SO_4 solution served as the electrolyte as well as coolant. Figure 2 is a schematic of this cell. As this cell showed excess heat with both D_2O-based as well as H_2O-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 laboratory after fabricating his own version of multilayer, thin-film-coated, flat-plate cathodes as well as a second-generation electrolytic cell using no metallic components, to preclude the possibility of trace elements from entering the solution inadvertently and causing contamination.

An advantage of thin-film cathodes (coating thickness varied in the range 500–3000 Å) is that high deuterium or hydrogen loadings could be attained 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 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, whereas NAA gave a quantitative measure of eight key elements, namely Al, Ag, Cr, Fe, Cu, V, Co and Zn, present in a gross sample 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 smoothens 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 of elemental analysis confirmed the presence of a wide gamut 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. In some of the runs, as much as 40% of the initial metal atoms of the thin-film coating was transmuted. The increase in mass of the eight key elements appearing in the thin films was noted. Figure 3 shows 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). Miley speculates that each of these group 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 (anode material). It can be seen that Ni ($Z = 28$) goes up the Z ladder to Cu ($Z = 29$) and Zn ($Z = 30$),

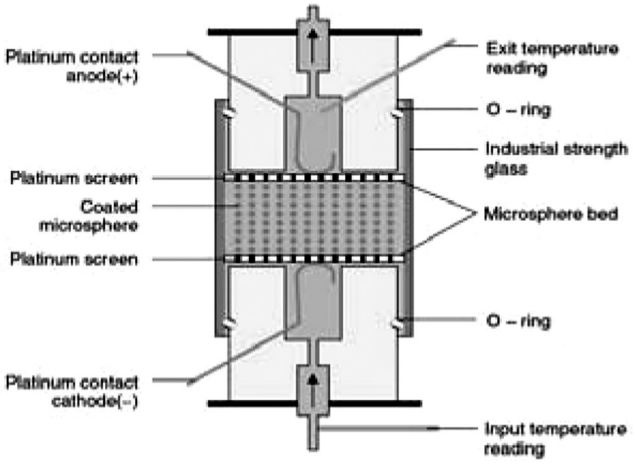

Figure 2. Schematic of the Patterson power cell.

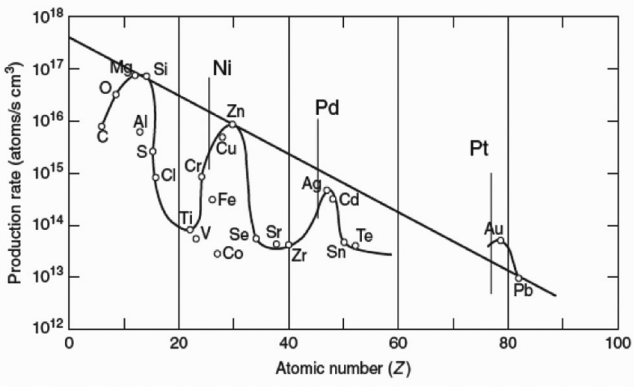

Figure 3. Reaction product yield versus atomic number (Miley).

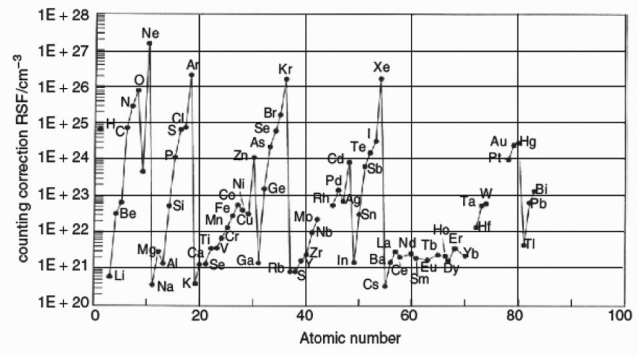

Figure 4. Yield of elements formed on the cathode surface (Mizuno).

while Pd ($Z = 46$) goes up to Ag ($Z = 47$) and Cd ($Z = 48$). Likewise, the anode material Pt ($Z = 78$) presumably plated out on the cathode surface during electrolysis, gets transformed to Au ($Z = 79$). The major low-Z peak comprising Si ($Z = 14$) and Mg ($Z = 12$), although close to sulphur ($Z = 16$), may actually be the result of fission of Ni and may not have arisen from the transmutation of sulphur.

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 systematics in the isotopic shift results, since there was considerable scatter in the isotopic ratios depending on the location of the bead in the cathode bed.

Miley's papers[5,6] also discuss 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 and D_2O runs. The similarity of the four-humped yield curve with the well-known double-humped yield curve observed in the neutron-induced fission process has led to the speculation that there could be a similar 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, Hokkaido University, Sapporo, carried out a systematic analysis of his post-run Pd cathodes that had earlier been electrolysed in heavy-water solutions; he also found a four-humped yield spectrum similar to that of Miley (Figure 4). Mizuno has elaborated on the details of his transmutation quest both in his book[8] as well as a recent review paper[9].

Concluding remarks

In this brief article we could discuss the work of only two research groups. It would seem that the newly found elements or isotopes can be explained through the 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. Although the best results are obtained with Pd and D, the Russian glow discharge experiments show that other cathode materials and certain rare gas environments also seem to support such transmutation reactions.

The confirmation of occurrence of transmutation reactions in simple LENR configurations has a profound implications, since it questions a 300-yr-old 'dogmatic belief' of science, prevalent from the days of Lavosier, that one cannot transmute one element into another in any simple laboratory experiment, no matter what one does, 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

These results clearly suggest that the age-old claims of alchemy may perhaps be true after all, however, unpalatable it may be to the scientific community.

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