

Cluster Reactions in Low Energy Nuclear Reactions (LENRs)

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ABSTRACT

Cold fusion reactions can be divided into two broad classes: deuterium reactions producing excess heat and helium; and transmutations of host metal atoms, termed Low Energy Nuclear Reactions (LENRs). Here we review some unique facts about a special class of LENRs leading to an array of transmutation products in thin-film electrodes of palladium (Pd) or nickel (Ni) highly loaded with hydrogen (H) or deuterium (D). This phenomena is explained in terms of the formation of an intermediate compound nucleus, one of which is identified as the double magic number compound nucleus $= {}^{306}\text{X}_{126}$. The formation of this nucleus requires a multi-body reaction between the D (or H) and the host metal. This is explained in terms of a proposed D (H) cluster formation. Such clusters can react collectively with palladium nuclei leading to a compound nucleus.

Introduction

Cold fusion was initially observed in electrochemical loading experiments when a very high concentration of deuterium was dissolved into the metal palladium lattice giving heat production that could be explained only by nuclear reactions [1]. Later a very convincing experiment was performed using deuterium gas loading with heat generation which continued for hours after the gas had been removed (“life after death”) where an energy release of $\sim \text{keV/Pd-atom}$, much above levels possible from chemical reactions, was confirmed [2]. It was assumed that cold D-D fusion reactions producing helium were responsible for the energy generation.

A later discovery was that the conditions could be created where highly loaded palladium and other hydrides produce nuclear transmutation reactions (termed “Low Energy Nuclear Reactions,” or “LENRs”) [3-5]. It was concluded that this type of reaction is quite different from the D-D type cold fusion since reactions with the metal lattice atoms are involved[6]. We will leave the question about the difference between these classes of cold fusion reactions open. Here we mainly concentrate on the issue of how LENR processes can produce heavy elements, suggesting multi-body reactions occur.

As pointed out earlier, the complete array of products observed in the University of Illinois thin film experiments appears to involve some direct nuclear reactions plus fission of three distinctive compound nuclei [3, 4]. Here we focus on some very distinctive properties of the portion of reaction products associated with the heaviest compound nucleus in order to give further definitive insight into this process. Important insight can be gained from the similarity between the measured distributions of nuclei [3, 7-10] generated by LENRs with the well known fission-fragment yield curve observed in the fission of uranium. We analyse these results to derive a new sequence for nuclei magic numbers which is consistent with fission of a double magic number nucleus ${}^{306}\text{X}_{126}$. Such reactions are explained by the reduction of Coulomb

screening in the swimming electron layer, leading to formation of reactive deuterium clusters. Other evidence that such cluster can form comes from separate studies of superconductivity states created by programmed loading of Pd-hydride.

It is important to distinguish the deuterium cluster model proposed here from neutron clusters proposed earlier by some prior cold fusion researchers [11]. Screened deuterons react like neutrons up to a radius in the range of picometers, generating clusters that behave like neutron clusters. However an essential difference is that the very high density of deuteron clusters makes them very reactive compared to the smaller size of neutron clusters.

Maruhn-Greiner Fission Model

The distribution of the nuclei after fission of uranium or plutonium shows a minimum at a nucleon mass A about one-half of the mass of the initial nuclei, e.g. at $A \sim 119$ in Fig. 1 [8]. This distribution with a broad absolute minimum, however, refers to fission of an unexcited (low temperature) initial nucleus. In the case that the originating nucleus is excited to higher temperature in the MeV range, as shown in Fig. 2, the distribution exhibits a sharp local maximum superimposed on the minimum of the curve for an unexcited nucleus. This behaviour has been explained by Maruhn and Greiner [9] using calculations based on collective mass parameters from the BSC formulation with the parameter λ of the length in the Schrödinger equation for splitting of heavy nuclei [12] taken from the theory of fragmentation dynamics in nucleus-nucleus collisions.

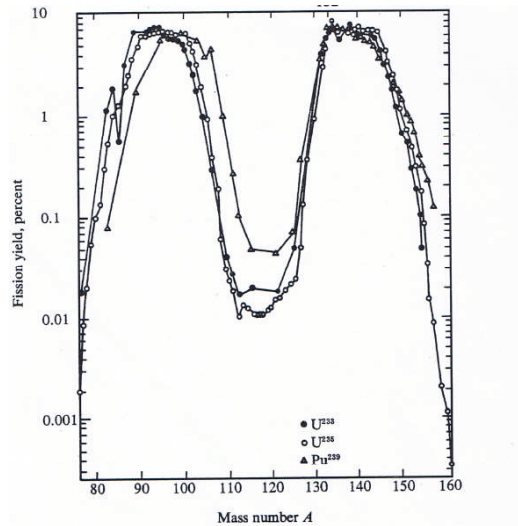


Figure 1. Fission mass distribution curves as measured for ^{233}U , ^{235}U and ^{239}Pu [8]

Figure 2 shows the resulting fission mass distribution for ^{236}U for the elongation $\lambda = 1.8$ at different excitation temperatures of the fissioning nucleus. It is significant that a local maximum occurs at $A \sim 118$ when the fissioning nucleus is excited to > 1 MeV temperature. This behaviour is a crucial point for the following discussion of product yield curves in LENR experiments.

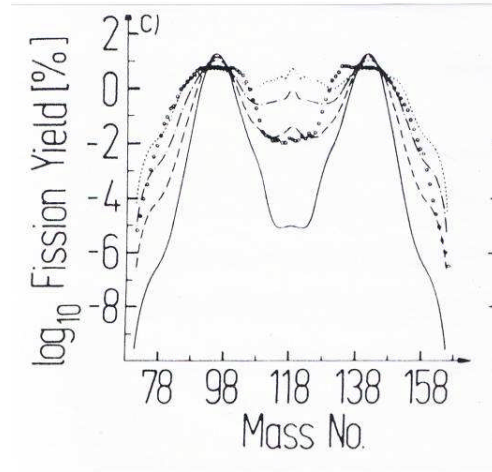


Figure 2. Fission mass distribution curves for ^{236}U calculated assuming the nucleus at the time of fission is excited to temperatures of 0, 0.5, 1, and 7 MeV respectively (upward sequence in plots) for the length parameter $\lambda = 1.8$ in the Schrödinger equation [9].

LENR Results

A key experimental result on LENRs with deuterium loaded thin-film palladium electrodes [3, 4, 10] was the product distribution dependence on proton number Z as shown in Fig. 3. The maxima of this distribution follows a Boltzmann probability distribution $N(Z)$ of the form [13]

$$N(Z) = N' \exp(-Z/Z') \quad (1)$$

where $Z' = 10$ provides a good fit as shown in Fig. 3. Other near-by numbers for Z' (9 or 11) do not fit well. This is especially important for the following consequence for the new kind of evaluation of magic numbers. A similar distribution occurs in the standard abundance distribution (SAD) of the elements in the Universe for elements above iron (see Fig. 10 of [14]). Below iron, the distribution is different due to the well-known exothermic fusion reactions. The endothermic synthesis of nuclei above iron is considered a crucial problem in astrophysics. It can be understood at the big bang theory with a Debye-layer model for the confinement of protons and neutrons in nuclei [15, 16].

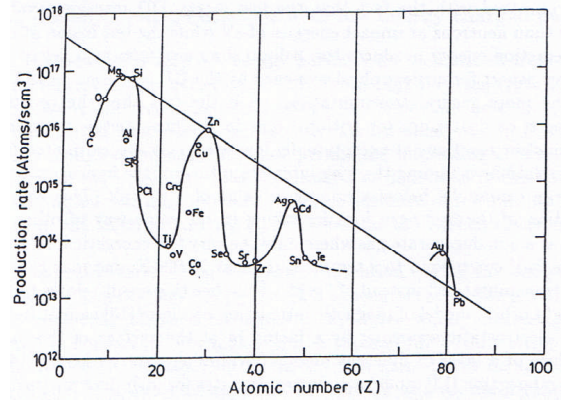


Figure 3. Dependence of the measured production rate on nuclei proton number [3.4]. The line represents a Boltzmann distribution, cf. Eq. (1).

The ratios for the maxima of $N(Z)$ was sorted out earlier to obtain the magic numbers of nuclei as [13]:

$$R(n) = 3^n \quad (n = 1, 2, 3, \dots) . \quad (2)$$

This suggests saturated shells follow a three-multiplicity of a quark property in the nuclei. This earlier result, however, had to take into account that there was a jump between the numbers 20 and 28 (see Table 1 of [13]), i.e., between the following two sequences of magic numbers:

$$M_{an} \in \mathbf{2, 8, 20}, 40, 70, 112 \quad (3)$$

$$M_{bn} \in 2, 6, 14, \mathbf{28, 50, 82, 126}. \quad (4)$$

Earlier Bagge [17] produced these series from speculative numerical combinations consistent with the observation that the numbers of the electron shells in atoms follow a $2n^2$ -law following the Schrödinger equation with the spin included (as derived by Dirac). The jump between the bold printed numbers in sequences (3) and (4) was explained by Jensen and Maria Goeppert-Mayer [18] by different properties of spin and spin-orbit combinations in the nuclei. In contrast to this explanation, the derivation of (2) explains the jump between the sequences (3) and (4) directly.

From this procedure [10,13], higher magic numbers can be derived with confidence. This results in:

$$\text{new magic numbers: } \mathbf{180, 246, 324} , \quad (5)$$

consistent with the rather linear relation shown in Fig. 4.

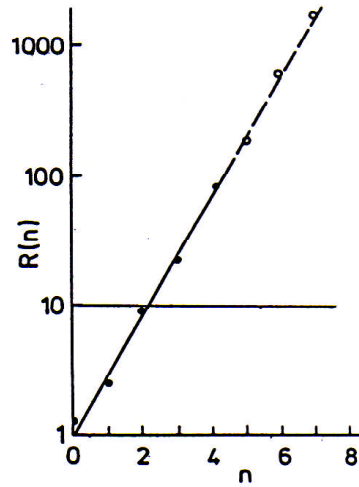


Figure 4. Ratio $R(n)$ from Eq. (2) for the sequence of the magic numbers $n = 1, 2, 3, \dots$ with the fitting value $Z' = 10$ for Eq. (2) given by the line, dots and circles. An exception is caused by the jump between the Bagge sequences (3) and (4)

Another important measurement of LENR is the detailed mass distribution of the resulting nuclei near the nucleon number $A = 153$ or 155 as seen from Fig. 5 [3, 4, 7]. This yield curve exhibits an absolute minimum similar to the fission of uranium, Fig. 1, along with a local maximum similar to the fission of excited uranium nuclei shown in Fig. 2. Later we use this remarkable observation along with the new higher magic numbers to identify the intermediate compound nucleus. (Other significant experimental phenomena observed in LENRs such as magnetic anomalies, charged particle and x-ray emission [19] may play a role but are not discussed here.)

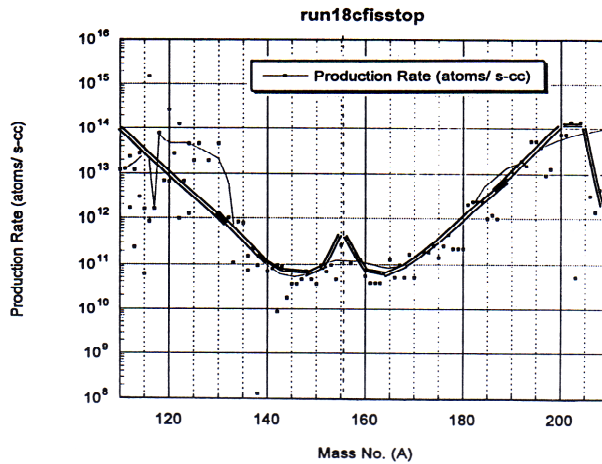


Figure 5. Detailed nuclear mass spectrum of the LENR generation probability at the highest minimum of Fig. 3.

We next consider the potential role of “cluster reactions” in this process. By this we mean the formation of very high density states of H or D in highly localized regions in the electrode.

LENRs are expected to occur due to these reactive states. However, as pointed out by Storms [20], the widely held belief is that “clusters of deuterium cannot form spontaneously” has inhibited the search for such clusters. Thus, before delving into a theoretical model, we will review experimental evidence that supports the possibility of cluster formation.

Indirect Evidence for Cluster Formation

A number of cold fusion researchers (Dash, Mizuno, Shoulders, Ohmori, etc.) have reported local “volcanic” like eruptions on electrodes that strongly suggest reactions occur in localized areas. (See, for example, the distinctive creator formation on an electrode in the cover photograph of the book by T. Mizuno, [21]). Also, early studies by M. Srinivasan [22] showed a pattern of small glowing spots on a fluorescent plate placed over an electrode after operation. SPAWAR researchers have presented a movie from an IR camera view of an operating electrode showing a dynamic “dance” of small glowing hot spots. All of these observations are strongly suggestive of localized cluster-like reaction sites. However, the most definitive measurement that shows the existence of a high-density cluster-like phase in a loaded hydride comes from work on superconductivity states at the University of Illinois [23]. We briefly review that work next, then discuss a proposed theory for cluster reactions in LENR.

Earlier Heuser et al., [24] discovered, by using small-angle neutron scattering (SANS), that dislocation cores caused by hydrogen cycling in palladium films are sites of high hydrogen absorption and concentration. The concentration of hydrogen (x = atomic ratio H/Pd) inside the dislocations depends on the hydrogen distribution with respect to distance from the core or the hydrogen binding energy. After cycling a palladium single crystal film with deuterium gas, a large number of dislocations ($N_d \sim 2.7 \times 10^{11} \text{ cm}^{-2}$) were generated. Without annealing, the residual hydrogen left in the palladium sample was α -phase with a concentration of 5 to 6 deuterium atoms per Å of dislocation line. This high level of residual hydrogen was associated with a low hydrogen binding energy of $\epsilon_H = 0.2 \text{ eV}$. But a much lower amount of residual hydrogen ($x \sim 10^4$) was found to have a much higher binding energy ϵ_H of $\sim 0.7 \text{ eV}$. In that case, all the absorbed hydrogen atoms are tightly bound inside the deep dislocation cores [25].

Obviously, the loading ratio in the deep dislocation cores is significantly larger than the stoichiometric H/Pd value that characterizes the palladium hydride β -phase in a regular lattice. Simple estimates show that a condensed deuterium phase in deep dislocation cores along a dislocation line in a PdH_x sample may have a concentration as high $C_H \geq 0.4 \text{ mol/cm}^3$. Such cores would be a sort of “nanotube” with an effective diameter of about two Pd Burgers vectors. Notice that the pressure inside such deep cores of the edge dislocation should be comparable to the local palladium bulk modulus, that is, up to 120 GPa. So, both of the conditions for hydrogen “precompression” and external pressure impelling would be fulfilled. A very conceptual picture of this compressed state is shown in Fig. 6 to illustrate the concept.



Annealed Pd-metal does not demonstrate superconductivity, at least above 3.0 mK. However, palladium films with defects produced by alpha-bombardment at low temperature did exhibit superconductivity at $T_c \sim 3.2$ K [26]. It was speculated that the quantum fluctuation of electron spins will not allow structurally perfect palladium metal to transit to the superconducting state, but specially introduced disorder serves to suppress those fluctuations. In contrast, palladium hydrides and palladium deuterides with a loading ratio $x > 0.8$ are superconductors with a relatively high $T_c \sim 10$ K. Though the superconducting effects in palladium hydrides, such as the reverse isotope effect, are still not fully explained, the arguments suggesting this superconductivity are based on a strong electron-phonon coupling to the optical modes and require a suppression of the spin fluctuation in the palladium lattice and sd-hybridization of hydrogen and palladium electrons. Anomalous transport properties over the temperature range below 300 K in highly loaded palladium hydrides [27-29] were detected earlier. In particular, in [29] it was shown that palladium foil electrochemically cycled with hydrogen possessed metallic conductivity down to a temperature of 4.2 K even though it was severely deformed.

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super-stoichiometric palladium hydride above the critical temperature of the normal hydride PdH_x (with $x \sim 0.8$). The observation of a diamagnetic signal from a hydrogen phase in the palladium matrix dislocations is possible when two necessary conditions are fulfilled:

- There is a sufficiently large number of dislocations in a palladium crystal that contain tightly bound compressed hydrogen, and
- The network of dislocations is organized in the form of closed loops that can carry a persistent current.

To verify this concept experimentally we have studied the structural and magnetic properties of PdH_x with a very low loading ratio $x \sim 4.0 \times 10^{-4}$ that was produced by cycling an annealing pure palladium single crystal in H_2 atmosphere [19, 23]. Thermal desorption analysis (TDA) shows that hydrogen exists only as a condensed phase captured in the deep dislocation cores. Magnetic measurements were carried out with a sensitive 1T SQUID and recorded the appearance of a strong diamagnetic contribution of the condensed hydrogen phase [$\text{PdH}_x\text{-Pd}$] in the palladium matrix at $T < 30$ K (in a weak magnetic field, $H \leq 5.0$ Oe), and of antiferromagnetic behavior in a higher magnetic field. The thermal desorption analysis and the SQUID measurements together suggest that the dislocation nanotubes in the hydrogen cycled palladium sample contains a diamagnetic phase of condensed hydrogen or superstoichiometric hydride at low temperature.

In summary, there is strong suggestive evidence that high density H(D) states and/or H(D)/Pd states can exist in properly loaded hydrides. If so, this would be consistent with observations from the University of Illinois LENR experiments and experiments by others suggesting localized reaction sites. Thus we explore a theoretical model for such LENRs in following sections.

Swimming Electron Layer Effects

The theory for nuclear reactions with high concentration deuterium in palladium or similar host metals needs to be viewed in light of a few key experimental facts [30] before more details may be explored, otherwise incorrect speculations may block the steps towards the truth. We first must realize that the usual hot fusion reactions (D-T, He-3 up to p-B-11) are fundamentally different from the usual nuclear reactions.

The original discovery that the reactions of the very light nuclei occur at beam energies around and above 10 keV only was a significant discovery. This was in contrast to the usual beam energy of a MeV required in order to move the nuclei against the electrical Coulomb repulsion to distance of their diameter around femtometers (fm; 10^{-13}cm). One of the tools for these experiments were the multi-million-volt accelerators, e.g. that of Cockroft and Walton. Cockroft was sufficiently adventurous to use – against all the knowledge for applying many million volts – at what happens when only 100 to 200 kilovolts were used: there the light nuclei did react [31], e.g. protons with boron. It was then Oliphant's gas discharge technique for 100 keV beams that provided the high currents necessary to obtain more precise results, e.g. the correct energy gain from the proton-boron reaction [32] as the prelude to the discovery of the fusion reactions[33, 34]. These reactions can be summarized as:

$$D + D = \begin{cases} T + {}^1\text{H} + 4.03 \text{ MeV (50\%)} \\ {}^3\text{He} + n + 3.27 \text{ MeV (50\%)} \end{cases} \quad (6a)$$

$$D + {}^3\text{He} = {}^4\text{He} + {}^1\text{H} + 18.3 \text{ MeV} \quad (7a)$$

$$T + D = {}^4\text{He} + n + 17.6 \text{ MeV} \quad (7b)$$

It has to be realized that these “hot fusion” reactions at 10 keV impact energy (corresponding to 100 million degrees temperatures) happen at *distances about hundred times larger (!!)* than the fm distances for all the usual nuclear reactions. This cannot be explained by a Gamow factor. The measurements of the involved fusion reaction cross sections are available now with very high accuracy. Nevertheless there has not been a theory for explaining them, apart from numerical fitting, e.g. with 5 parameters [35] or more. That does not represent physics but is only an empirical type solution. It was not before Li et al. [36] that a reasonable theory was developed using a Schrödinger potential with an imaginary part that the cross sections could be best reproduced using the input of two obvious parameters the resonance energy and the resonance width.

To understand cold fusion or LENR we assume that the deuterons (or hydrogen) in the palladium are behaving like a Maxwellian gas on the background of the degenerate electron gas between the ions. The Debye length for screening the deuterons is given

$$\lambda_D = 743(T/n_d)^{1/2} \text{ cm}, \quad (8)$$

with the deuteron density n_d in $\text{\#}/\text{cm}^3$ and temperature in eV results in ~ 3.8 pm for room temperature and solid state density for deuterons taking into account their non-degenerate state. For the special case of a two-dimensional geometry of the metal surface, the Debye length is reduced by a factor $2^{-1/2}$ as known for surface plasmons against the usual plasmons. This means that the Coulomb field of the deuterons is neutralized at a radius of ~ 2 pm and the deuterons behave like neutral particles, e.g. like neutrons at such a distance.

Alternatively and independently to this fact, it was clarified [37] from very early cold fusion experiments [38] that this results in a Coulomb screening by a factor 14 – compared to a factor 5 which is well known in high temperature plasmas (see Ichimaru in Ref. [10]). Such screening results in a screening again in the range of 3 pm. From the measured reaction times and deuteron distances for hot fusion, from muon-catalysed fusion and the calculated fusion probability in a D_2 molecule, a reaction time in the range of kilo- to megaseconds for the 3 pm distance could be concluded to fit with reaction rates [38]. These times agree with the K-shell electron capture by nuclei for Bohr radii in the pm range. The megasecond range then is a reasonable time scale for LENRs with palladium [3, 4].

With these facts in mind, we next consider a possible cluster generation mechanism in effort to explain the result of Fig. 6 in terms of a compound nucleus.

The screened deuterons are mutually repulsed by their Coulomb field at distances less than ~ 2 pm. However, due to their screening they move like neutral neutrons at larger distances. The attraction between the screened deuterons by the Casimir effect [39] is too small to significantly

decrease this distance. But our calculation of the gravitational attraction for the deuteron masses at ~ 2 pm distance suggests compression to about ten times higher energy density than for thermal motion at room temperature. This is the reason that once a high initial deuteron concentration is achieved in the palladium, cluster formation occurs. Clusters of 100 deuterons have a size of ~ 10 pm and move within the electron clouds of the palladium around the palladium nuclei. Consequently few pm nuclear reactions between a cluster and a palladium nucleus can take place on the megasecond time scale. Under the assumption that the screened deuterons in the cluster form a Bose-Einstein state, the deuterons (as bosons) are non-distinguishable. Thus a reactive interaction, e.g. by the deuterons with palladium nuclei, becomes a collective process of the whole cluster. This then allows the formation of compound nuclei via a multi-body type reaction.

Since the clusters are tighter in two-dimensional geometry, they are expected to appear preferably near the surface of the swimming electron layers; i.e. at a few 100 pm thickness from the surface of palladium or at interfaces in multilayer systems (e.g. in layered Pd-Ni thin film electrodes) due to the difference of the electron Fermi-Dirac energy between the metals. The swimming electron layer is an extension of the Debye layer for a plasma-material surface generalized to apply to the degenerate electron cloud at a metal surface [40]. This immediately explains the work function for electron emission from metals and explains the quantum theory of the surface tension of metals in agreement with measured values (versus “synthetic” theories of surface tension which can lead to unrealistic negative work function values [40]).

The tighter deuteron clusters within the swimming electron layers would explain why the low energy nuclear reactions LENR in multi-layers occur at higher rates than in bulk palladium electrodes [3, 4, 10]. This is consistent with the observation [10] that power levels exceeding a kW/cm^3 in thin film multi-layered electrodes can occur during LENR experiments.

Compound Nucleus Reactions

Compound nucleus formation was proposed earlier to explain how the reactions of deuterium in palladium produce an array of nuclei [41]. Section 7 of Ref. [10] presents examples for how a reaction of ^{101}Nb and ^{106}Pd can form ^{207}Fr as an intermediary excited state. ^{207}Fr then splits into ^{117}In and ^{90}Sr exothermally with an energy gain of 1.65 MeV. The same can be shown with the ^{238}Am compound nucleus where the energy gains can be calculated for the accurately known masses of the nuclei involved.

It is important to remember the fact that the fission of the compound nuclei will result in a local maxima within the absolute minimum of the mass distribution curves as shown earlier in Figs. 1 and 2 for uranium fission. The question then is what compound nucleus is predicted by the mass distribution measurement shown earlier in Fig. 5 for these LENRs. The atomic half mass for the compound nucleus in this case is seen to be $A = 153$. Using the new magic numbers of Eq. (5), we expect a relatively stable very heavy nucleus as one with double magic numbers (similar to ^{208}Pb with 82 and 126). The sequence and data suggest this to be:

$$\text{Compound nucleus} = {}^{306}\text{X}_{126}. \quad (9)$$

This identification is supported also by the fact that the search for stable (or relatively stable) trans-uranium elements predicts an element number 126 [42]. The main support for our LENR

case is indeed the data of Fig. 6 where the local maximum at $A = 153$ is indeed very significant for the identification. There is no question but that, if the compound nucleus (9) is produced for a very short time (less than 10^{-20} sec), it will be a very excited nucleus, similar to MeV excited consistent with a temperature for the compound nucleus in the MeV range.

There was earlier speculation [10] about which reactions could lead to the compound nucleus in (9). This may be possible based on the heuristic conclusion of the preceding section assuming large deuteron clusters form under preferred conditions in the swimming electron layer. The action of a cluster with 156 deuterons reacting collectively could lead to the generation of the compound nucleus in (9) by the following reaction:



Expressing the mass per nucleon m_X by proton masses in X, we arrive at:

$$m_X = 1.004946 \text{ minus the related part going to E.} \quad (11)$$

This mass is not unexpected to be large due to the very low value $m_{\text{Fe}} = 0.9988376$ compared to the value of $m_{\text{U}} = 1.0001868$ when splitting into ^{121}Sb with $m_{\text{Sb}} = 0.99824$. Comparable values would be for the splitting of the compound nucleus X into ^{153}Eu with $m_{\text{Eu}} = 0.9988375$. The energy per nucleon in $^{306}\text{X}_{126}$ is 5.73 MeV minus the contribution going into the reaction energy E.

The splitting of the compound nucleus leading to the yield curve around $A = 153$ in Fig. 5 can be represented by the following equation with varying integers x and y , and a number N of neutrons n :

$$^{306}\text{X}_{126} = ^{153+x+N}\text{A}_{63+y} + ^{153-x}\text{B}_{63-y} + Nn + E_{xy} . \quad (12)$$

This implies that few or no neutrons will be emitted, and indeed to date none have been observed in the LENR experiments using reasonably sensitive neutron diagnostics. However, further detailed searches for neutrons are warranted. Also more studies such as the preliminary ones in [43,44] to examine the correlation of the reaction products and excess heat, i.e. E_{xy} , would provide further confirmation of this concept.

Concluding comments

In summary we have shown that the detailed reaction product data obtained from a certain class of LENR experiments employing thin film electrodes suggest LENR reactions can proceed through a compound nucleus to produce a distinctive array of products. This striking observation of a sharp maximum in the product yield superimposed on a broad minimum indicates formation of a double magic nucleus $^{306}\text{X}_{126}$. Such a reaction would require the collective interaction of a large number of H(D) nuclei with the host palladium, suggesting involvement of H(D) clusters. Localized cluster reactions have been suspected previously based on the observations of local hot spots and crater formations on electrode surfaces. More direct measurements indicating the formation of a cluster state come from studies of superconducting behaviour in dislocation loops formed in loaded palladium.

While further studies are needed to definitively tie these observations together with LENRs, they do form a basis for hypothesizing the cluster reactions which have been described here. Further experiments are needed to fully verify this theory, but it provides a provocative “road map” for future exploration.

Assuming the cluster concept is correct, two immediate key questions arise: Are clusters of this type involved in other cold fusion reactions beyond the specific type of LENRs studied here (i.e. do they create the NAE sought by Storms [20])? Is it possible to design experiments to enhance this effect? We must leave the first question for future study after more relevant experimental data is accumulated. However, a preliminary response to the second question can be offered. The initiation of clusters was achieved in the superconductivity experiments cited here by repeatedly loading and de-loading to create a large density of dislocation loops. In the transmutation experiments cited, thin films, sometimes with multi-layers, were used to create the initiating swimming electron layer regions. And, it would seem that the same interfaces giving these layers must have had a high density of dislocation loops. Thus it is reasonable to assume that a combination of swimming electron layers and dislocation loops need to be created in order to trigger cluster formation. Along these lines, Miley suggested earlier the use of a special nanopore structure for the electrode [45]. Basically this electrode uses a micro-mesh nickel layer sandwiched between sputtered palladium films. The mesh-film interface is intended to create the simulated swimming electron layer – dislocation loop type conditions needed for cluster formation. This concept is currently under investigation.

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