

Research Article

# Statistical Analysis of Transmutation Data from Low-energy Nuclear Reaction Experiments and Comparison with a Model-based Prediction of Widom and Larsen

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## Abstract

Nuclear transmutations were reported in many low-energy nuclear reaction (LENR) experiments. In the present study, we analyzed (i) whether three available nuclear transmutation data sets show a consistent pattern and (ii) whether this pattern correlates with a model-based prediction of Widom and Larsen. Our analysis revealed that the data sets (i) exhibit a similar pattern and (ii) correlate with the predicted function. The last three peaks as a function of atomic mass  $A$  (intervals: 64–70, 116–129, 191–208  $A$ ) were significantly ( $p < 0.05$ ) correlated with the averaged data despite great differences in the experiments.

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## 1. Introduction

Experimental data are the basis for both scientific and commercial interest in low-energy nuclear reactions (LENR). There are four classes of evidences for the ability to induce nuclear reactions by the use of chemical energies: excess heat, transmutation products, energetic particles and radiation, and other low-energy phenomena. Excess heat data have received the most attention, but many studies of transmutation products are also available. A recent review summarizes some of the most relevant work reporting transmutations [1].

There are two major kinds of evidence for transmutations. The simplest, and possibly best-accepted, involves changes in the distribution of isotopes for specific elements, as measured with a mass spectrometer. Such data is useful even on a relative basis, that is, absolute calibrations and measurements are not needed. Measured relative distributions

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of isotopes are compared with natural distributions. Significant differences are taken as evidence of nuclear reactions, that is, transmutations. For example, Iwamura et al. apparently transmuted Sr into Mo [2]. The isotopic distribution of the Mo found after deuterium permeation of a complex Pd foil was similar to that of the initial Sr and very different from that of natural Mo.

The second type of transmutation data involves measurement of specific elements before and after LENR experiments. This type of LENR transmutation data requires sophisticated elemental analyses, which are open to more arguments than the relative data on isotope distributions. The analytical measurements can be done in either of two ways, with or without spatial resolution. Specific locations on samples can be examined analytically with a scanning electron microscopy (to get the location), which has the ability to do X-ray analyses (to obtain the elemental composition). Alternatively, analyses can be performed without spatial resolution by use of unfocussed instruments, or by dissolution of the surface or more of the materials from a LENR experiment followed by either wet or instrumental chemical assays. Sometimes, such spatially unresolved analyses are done for elements of particular interest [2]. Other times, they are performed for a wide range of elements spanning the periodic table. This paper is focused on the last type of transmutation data.

Miley [3–5], Mizuno [6,7], Little [8], Yamada [9] and their colleagues have published independent experimental transmutation data for elements across the periodic table. Their plots of nuclear transmutation production rates or final concentrations as a function of atomic mass number ( $A$ ) appear to be qualitatively similar despite great differences in the experiments. However, these important data sets have never been compared with each other quantitatively.

In 2006, Widom and Larsen presented a model to predict the nuclear transmutation production rate [10] (here called ‘WL-prediction’). Although they pointed out that their prediction seems to correlate quite well with the experimental data of Miley [3,4], a statistical analysis for the agreement between the theoretical prediction and experimental data has not been done so far.

Thus, in order to gain further insights into the process of transmutations, and LENR in general, we analyzed (i) whether three available nuclear transmutation data sets show a consistent pattern, and (ii) whether this pattern correlates with the WL-prediction. We consider this study to be an example of ‘data mining’, a process which could be more widely applied to diverse data available from LENR experiments.

## 2. Data and Methods

### 2.1. Laboratory Data

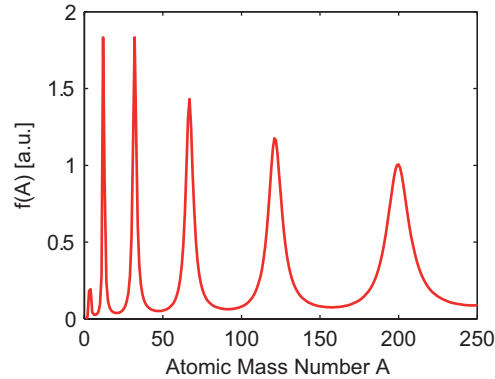
The following transmutation data sets from electrolysis experiments were selected from publications and used for the present analysis: data obtained by (i) Miley (*Miley data set*), (ii) Mizuno (*Mizuno data set*), and (iii) Little and Puthoff (*Little-Puthoff data set*).

Very brief synopses of the methods and results for each of the three experiments follow.

Miley et al. [3–5] electrolyzed plastic beads coated with Pd and Ni in a packed bed configuration through which a light water ( $H_2O$ ) electrolyte circulated. They used four analytical techniques, Secondary Ion Mass Spectrometry (SIMS), Auger Electron Spectroscopy (AES), and both Energy Dispersive X-Ray (EDX) and Neutron Activation Analyses (NAA), some both before and after 14-day runs. Production rates in atoms/s/cm<sup>3</sup> were reported as a function of mass  $A$ .

Mizuno et al. [6,7] electrolyzed a Pd rod in a closed cell containing a heavy water ( $D_2O$ ) electrolyte at high pressures, temperatures, and current densities for 32 days. They used the same analytical methods as Miley et al., except electron probe micro-analysis was employed in place of neutron activation analysis. Data were reported in SIMS count rates and gms of deposited material as a function of  $A$ . Both Miley et al. and Mizuno et al. observed excess heat and measured anomalous isotope ratios.

Little and Puthoff [8] used the same configuration as Miley et al. for runs of 2 weeks. They employed X-ray



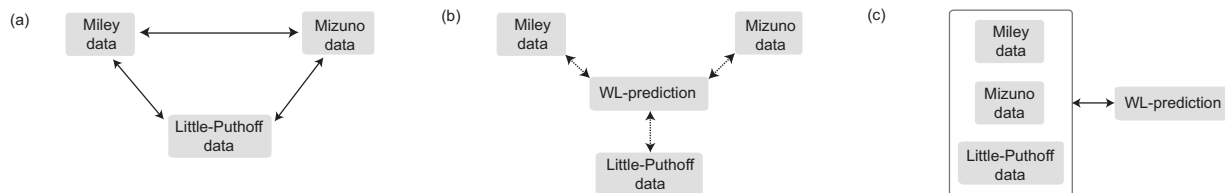
**Figure 1.** The  $f(A)$  function according to Eq. (3) for  $A \in \{1, 2, \dots, 250\}$ .

fluorescence analysis, which has a relatively high minimum detection limit. Miley analyzed their materials using SIMS and obtained absolute production rates generally similar to what his team observed.

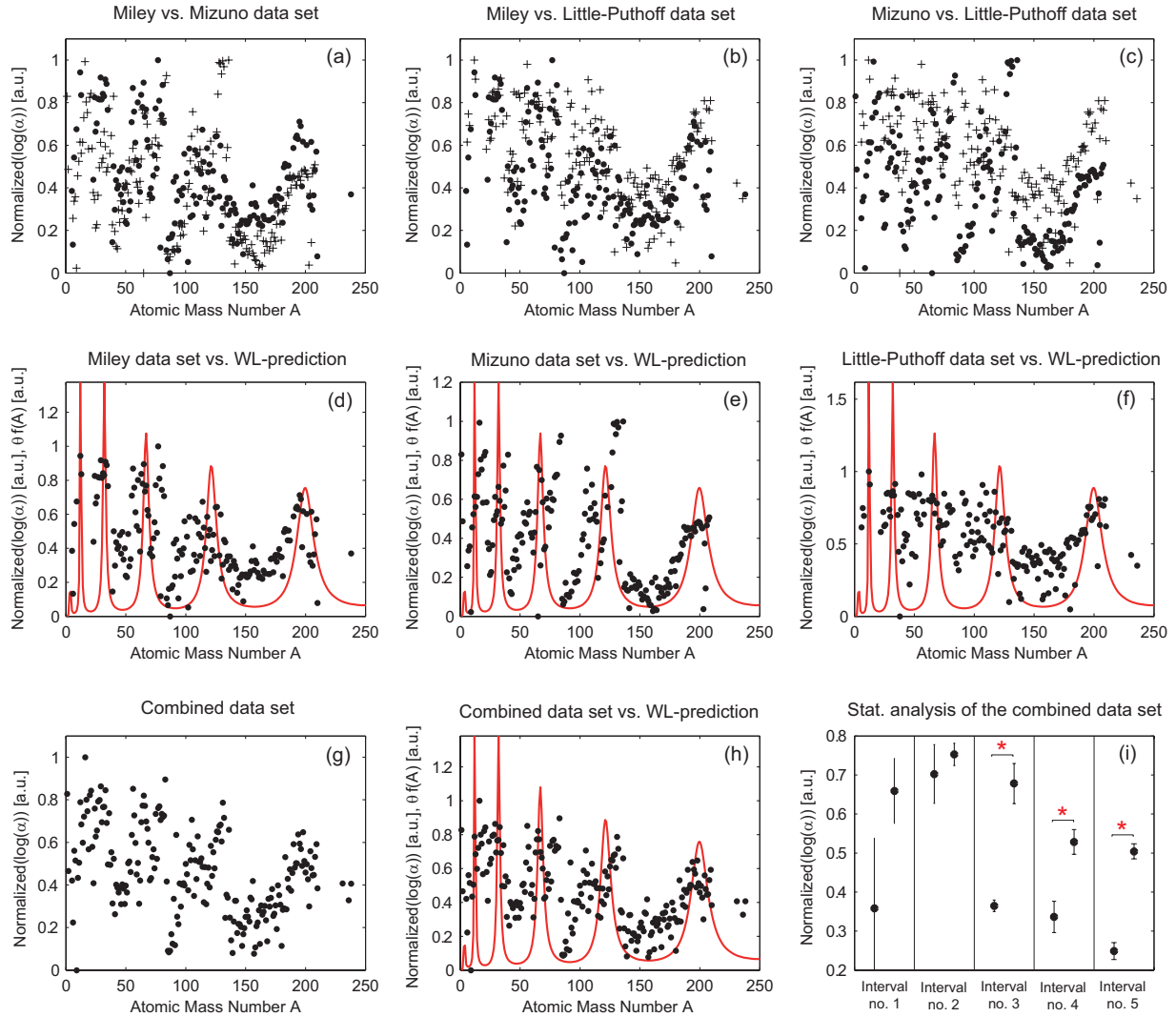
Since these data were not available as data files we used a Matlab (Mathworks, Natick, MA, USA) software (‘grabit.m’) written by J. Doke to extract the data from the published figures. For the *Miley data set* we used the figure given in [5], which is of higher quality than in [3], where the same data were reported. It consists of six runs (run numbers: #5, #7a, #8, #11, #13, and #18c) of the same experiment. We averaged over all six individual data sets to create the final data set. The *Mizuno data set* was created by using the figure in [7], consisting of two measurements (isotopic distribution of the top and the side of the electrode). We averaged over those two individual data sets to create the final data set (‘combined data set’). The *Little-Puthoff data set* was created by using the corresponding figure given in [8].

### 2.2. Widom–Larsen prediction

That collective electron and proton or deuteron plasma modes on surfaces of fully loaded metallic hydrides produce ultra-low momentum neutrons ( $< 10^{-10}$  eV) was put forward in a theoretical work in 2005 of Widom and Larsen [11]. According to this hypothesis, the reaction of radiation energy with electrons ( $e^-$ ) gives rise to heavy electrons localized near the metal hydride surface ( $\tilde{e}^-$ ). They can react with a proton ( $p^+$ ) resulting in the formation of an ultra-low



**Figure 2.** Visualization of the three different types (a–c) of data analyses performed in the present study.



**Figure 3.** (a)–(c) Comparison of the each data set with each other.  $\alpha$  is the scaling of the original data sets (isotopic abundance, ...). (a): Miley ( $\bullet$ ) vs. Mizuno data set (+). (b): Miley ( $\bullet$ ) vs. Little-Puthoff data set (+), (c): Mizuno ( $\bullet$ ) vs. Little-Puthoff data set (+). (d)–(f): Comparison of the each data set with the WL-prediction. Values obtained for the scaling parameter  $\theta$ : case (d),  $\theta = 0.751$ ; case (e),  $\theta = 0.655$ ; case (f),  $\theta = 0.3703$ . (g)–(h): Comparison of the sum of all data sets with the WL-prediction. (g) Combined data set. (h) Combined data set vs. WL-prediction. (i) Result of the interval analysis. \* Indicates statistically significantly ( $p < 0.05$ ) differences.

momentum neutron ( $n$ ) and an electron neutrino ( $\nu_e$ ):

$$\Delta E + e^- \rightarrow \tilde{e}^-, \quad (1)$$

$$\tilde{e}^- + p^+ \rightarrow n + \nu_e. \quad (2)$$

According to Widom and Larsen [11], the ultra-low momentum neutrons interact with the atoms on the metallic hydrides and cause nuclear transmutations.

Widom and Larsen proposed that the transmutation rates depend on the atomic mass number of the reacted element [10]. Based on an optical potential model for the effective neutron amplitude, they derived a function for the neutron scattering strength:

$$f(A) = \Im m \left[ \frac{\tan (zA^{1/3})}{z} \right] \quad (3)$$

with  $z = 3.5 + 0.05i$ . In their paper [10], they showed a remarkable similarity of the theoretically derived  $f(A)$  function and results from two LENR experiment where a change in the isotopic distribution was observed [3,4].

Figure 1 shows the  $f(A)$  function according to Eq. (3) for  $A \in \{1, 2, \dots, 250\}$  comprising a spectrum with five main peaks at  $A = 12, 32, 67, 121, \text{ and } 200$ .

### 2.3. Data Analysis

We performed three types of analyses: (i) comparison of each of the three data sets with each other, (ii) comparison of each data set with the WL-prediction (i.e. the function), and (iii) comparison of the combined data set with the WL-prediction. A visualization of these three analyses can be seen in Fig. 2.

All data sets used were first normalized to the range [0,1] in order to enable the next data processing steps, i.e. to make the data comparable to each other.

#### 2.3.1. Comparison of each data set with each other

Since the three different data sets contain missing values and show strong scattering, an interpolation function was calculated for each data set in order to be able to quantitatively compare the data sets. For the interpolation, a fully automated smoothing procedure based on a penalized least square method introduced by Garcia [12] was used. The correlations between the data sets were then quantified by calculating the Pearson correlation coefficient ( $r$ ) and the significance level ( $p$ ) of the correlation. A value of  $p < 0.05$  was considered as statistically significant.

#### 2.3.2. Comparison of each data set with the WL-prediction

First, the  $f(A)$  function for  $A \in \{1, 2, \dots, 250\}$  according to Eq. (3) was computed. Then, the  $f(A)$  function was fitted to each data set by using a classical least squares optimization method. The fitting was realized by multiplying the  $f(A)$  function with a scaling factor  $\alpha$ , which then was changed until the sum of squared errors between the  $f(A)$  function and the data was minimized. Finally, the correlation between each data set with the specific  $f(A)$  function was quantified as described in the previous section.

#### 2.3.3. Comparison of the combined data set with the WL-prediction

For this analysis, all three data sets were combined and an interpolation function was determined, according to the approach presented in [12], to obtain the missing values. Then, two types of data analyses were performed. *First data analysis:* The  $f(A)$  function was fitted to the new data set and the strength as well as statistical significance ( $r$ ,  $p$ ) were determined. *Second data analysis:* The new data set was segmented into intervals corresponding to the peaks and troughs of the  $f(A)$  function. Thus, ten intervals were determined. The borders of the intervals were chosen to cover the peak and troughs position of the  $f(A)$  function optimally. The borders for the peak intervals were: 11–13 A,

30–34 A, 64–70 A, 116–129 A, and 191–208 A. The borders for the trough intervals were: 7–9 A, 19–22 A, 44–50 A, 88–101 A, and 153–180 A. To test whether the mean values of the new data set in the peak and trough intervals are statistically significantly different, a *t*-test was used.

### 3. Results

#### 3.1. Comparison of each data set with each other

The three comparisons between the data sets revealed that all three combinations tested showed a positive correlation coefficient and a statistically significant correlation: (i)  $r = 0.3823$ ,  $p < 0.001$  (Miley vs. Mizuno data set), (ii)  $r = 0.5392$ ,  $p < 0.001$  (Miley vs. Little-Puthoff data set), and (iii)  $r = 0.3545$ ,  $p < 0.001$  (Mizuno vs. Little-Puthoff data set). The graphical visualizations of the data analysis results are given in Fig. 3(a)–(c).

#### 3.2. Comparison of each data set with the WL-prediction

The comparison of each data set with the WL-prediction showed that each data set was significantly correlated with the predicted function. The obtained values were: (i)  $r = 0.4360$ ,  $p < 0.001$  (Miley data set vs. WL-prediction), (ii)  $r = 0.2338$ ,  $p < 0.001$  (Mizuno data set vs. WL-prediction), and (iii)  $r = 0.3703$ ,  $p < 0.001$  (Little-Puthoff data set vs. WL-prediction). The graphical visualizations of the data analysis results are given in Fig. 3(d)–(f).

#### 3.3. Comparison of the combined data set with the WL-prediction

This analysis revealed that the sum of all data sets ('combined data set'), shown in Fig. 3(g), is statistically significantly correlated with the WL-prediction as depicted in Fig. 3(h) ( $r = 0.3409$ ,  $p > 0.001$ ). Fig. 3(i), which shows that when summing up the values for the five peaks and troughs according to the WL-prediction, the last three peaks are present in the data and correspond to the WL-prediction.

### 4. Discussion

This discussion addresses three topics. The first deals with the experimental challenges of transmutation studies. Next, we briefly summarize what was found in the present study, and an associated question raised at the conference (ICCF-17). Then, we consider how the peaked distributions seen in the three data sets and in the WL prediction can occur conceptually.

As a practical matter, getting transmutation rates experimentally is very challenging and expensive. Transmutations of one element into another are due to reactions. The amounts of materials that result from reactions, chemical as well as nuclear, depend both on the amounts of materials at the outset and the reaction rates. Hence, the reaction rates can only be obtained by measuring the amount of a given element or isotope present at the beginning and the end of a LENR experiment and forming the ratio of those numbers. That is possible in principle, but challenging in practice because of the sampling that is required. Rigorously, the same region (area and depth) of the sample must be analyzed before and after any nuclear reactions.

It is possible to do before and after analysis with non-destructive methods, such as X-ray analysis, but it is not possible for analytical methods that require consumption of the sample. Inductively coupled mass spectrometry is remarkably sensitive, but requires digestion of the sample. That is, pre-run analysis changes the sample, and it is that changed sample which goes into and out of the experiment. Of course, whatever analytical technique is used, it has to have an adequately low minimum detection limit, and offer good signal-to-noise performance for the analyte levels both before and after an experiment. It should be highlighted that the NAA method is able to examine the whole analyte

while also giving absolute values. Therefore, analysis with NAA is recommended for future LENR transmutation experiments. It remains remarkable that the three experimental data sets we studied have such great similarities in the face of these challenges.

In summary, we found that all three data sets, and the WL-prediction, correlated well with each other. Further, we found that peaks in three intervals of atomic mass are well correlated: 64–70 *A*, 116–129 *A*, and 191–208 *A*.

The fact that the data sets of the teams lead by Miley and by Mizuno, and also the Little-Puthoff data, all correlate well, each with the others, would seem to add strength to the value of these data. These LENR experiments, which differ widely in what was done and measured, apparently tell the same story, namely nuclear transmutations occur with results that resemble each other. However, there are two reasons to be cautious in adopting that viewpoint. At ICCF-17, right after presentation of this paper, David Kidwell of the Naval Research Laboratory raised a question about the data sets themselves [13]. He stated that, if all three of the transmutation experiments we studied were contaminated with dirt, which reflected the distributions of elements on the earth's surface, their similarity would be understandable and artificial. That is, the peaked data sets might be due to impurities in the experiments and not reflective of transmutation rates, as stated by the authors. The absolute amounts of material for any given element in the transmutation experiments are very small. Kidwell's idea might seem preposterous to some, since the data that were used in this study were obtained by careful experimenters. However, the fact that the data sets are so similar, even though the experiments and analytical methods are so different, does give some people pause for thought. The anomalous isotope ratios measured by the teams lead by Miley and Mizuno argue for the probability that transmutations were actually produced. We plan to perform a statistical comparison of the same three data sets, as used above, with the distribution of elements and isotopes in the earth's crust. Such an analysis could indicate whether (i) the LENR transmutation results are artifacts due to contamination with dirt, dust, etc., (ii) or whether both processes (LENR transmutation and natural elemental abundances) share a common physical mechanism.

The other reason for being cautious in drawing conclusions from the correlations we found is conceptual. The measured peak distributions as a function of atomic mass could be due to peaks in the initial distribution of elements, peaks in the reaction rates or both these factors. As already noted, analyses before and after an experiment can eliminate the effects of the initial variations in sample composition, and give the mass-dependent reaction rates. The theoretical prediction of WL about the rates of reactions of nuclei with what they call ultra-low momentum neutrons deals only with the reaction rates, and not the initial distributions of the elements in an experiment. The good correlations of their reaction rates with the experimental data can lead to the conclusion that the initial distributions of elements before the three experiments might be quite flat, that is, independent of atomic mass. But, that is highly unlikely.

The discussion to this point assumes that there is only one reaction per starting nucleus. However, there might be sequential (cascaded) reactions during an experiment to produce the final measured distribution. Assume for the sake of discussion that the WL-prediction is correct. If there are enough ultra-low momentum neutrons for long enough times, there might be sequential nuclear reactions involving isotopes lighter than the positions of each peak. Any isotope to the left of a peak would react to give a larger *A* value, and then that product would again react, etc. The final elemental distribution would be the result of a cascade of sequential reactions. The probability of reactions would increase as the peaks are approached from the left. When masses at the peak values are achieved, then the declining probabilities for reactions at larger *A* values would leave a high concentration in the region of the theoretical valleys, where the reaction rates are lowest. Then, most atoms would be in mass regions away from the peaks, contrary to experiment.

If this scenario happened, then the final distribution would be either less dependent on the initial distribution of elements, or even independent of it. Because of the possibility of cascades of reactions, we sought to find papers that dealt with this scenario. There seems to be only one such study in this field [14,15]. Mishinsky and Kuznetsov consider individual transmutation reactions, which can involve several input nuclei as reactants and up to three output nuclei products. Different types of LENR experiments with different reaction times and energy densities were considered. Setting aside concerns about the mechanistic details of such reactions, we note that the authors do take into consideration

the necessary conservation requirements for exothermic single and sequential transmutation reactions. For the sequential reaction case, distributions of elements are shown as a function of time for multi-hour runs, implying knowledge and use of reaction rates. It should be possible to test experimentally the results computed by these authors.

There is one well-studied topic in science in which sequential nuclear reactions occur, namely stellar nucleosynthesis [16–19]. The basic idea in the astrophysical field is that all heavy elements in the universe are the result of cascades of nuclear reactions, which occur within stars, both during their lifetimes and also during the explosions (supernova) at the end of their lives. A great deal of modeling has been performed on cosmic nucleosynthesis. It is based on conventional nuclear reaction physics. The results have no direct utility to the understanding of LENR. However, the computational methodologies employed to understand the natural distributions of elements and isotopes are worth studying to see if those techniques could be applied to understand the distributions of interest in this paper.

The results we obtained in this study do not validate the basic concept of the Widom–Larsen theory of LENR [10,11, 20–26]. Their theory for the occurrence of LENR depends on the production of extremely high fields (100 V/nm) on the surfaces of lattices. According to Widom and Larsen, such fields would “mass renormalise” electrons in the surface region, making it possible for the reverse of the familiar neutron decay reaction to occur. This would, in their view, produce ultra-low momentum neutrons, which could then react with nearby elements during LENR. The WL-prediction, which we employed in this paper, is ancillary to their basic theory. That is, the correlation of their prediction with the results of three transmutation experiments does not validate the occurrence of the postulated very high surface fields, the production of heavy electrons or the appearance of ultra-low momentum neutrons.

If it were shown computationally that there are (a) appropriate conditions in LENR experiments to produce high densities of ultra-low momentum neutrons and (b) enough time for many cascaded reactions, then the WL prediction might be the basis for understanding the peaked experimental distributions, as discussed above. Then, the observed peaked distributions could be taken as evidence for the validity of the WL-prediction we used. That, in turn, would indicate the occurrence of ultra-low momentum neutrons, which would buttress the case for their production by mass renormalisation of electrons, which would indicate that the super-high surface fields might exist, as theorized. Of course, it would also be good to design experiments that might be able to more directly produce evidence, which can only be explained by the presence of such immense fields.

It would be also interesting to compare the findings of the present study with prediction of other theories about LENR. However, we are not aware of any other theory that predicted the pattern of peaks in LENR transmutation rates.

Although our study proved that there is a similar pattern in the LENR transmutation data, this study has raised more questions than it has answered. Knowledge of the quantitative comparisons we obtained is a useful advance in our understanding of LENR experiments that reportedly produce transmutations. However, we are left with two large questions about the observed distributions of elements across the periodic table. The first is the important, albeit disturbing, issue that was raised by Kidwell. He asserted that it might be possible for the measured distributions now available to be the result of contamination and not transmutations. That concern can be addressed experimentally by use of very pure initial samples, or more ordinary samples with very well characterized compositions. Doing such experiments would require the employment of sophisticated sampling and analytical methods before and then after the LENR experiments. It is expected that such experiments will not be possible until adequate funding is available for the field.

The second large question can be addressed theoretically and computationally. It deals with the possibility that the measured distributions are indeed due to transmutations and that they are attained by cascaded nuclear reactions, such as occur in stars by understood nuclear reactions. The question is whether or not it LENR can also occur by cascaded reactions. This is basically a scientific question. However, it might have practical consequences. If many sequential reactions occur in commercial LENR energy generators, then the materials left after operation would be different than if only one or very few reactions can occur, one after the other. The point is that the residual materials from operation of LENR power sources would depend on the answer to this question about cascaded reactions. The reclamation of



elements from spent LENR energy generators would be different both chemically and economically without or with sequential nuclear reactions. If such new generators do become as widely used as many hope and some expect, the character of their residues would be significant. These considerations also apply to the hope on the part of many people that LENR can be used to process current radioactive materials from fission reactors into something more benign, while extracting additional energy in the process.

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