

Research Article

Upper Bound in the Fusion Products and Transmutation Enhancement in Alloys

Ankit Kumar, Pankaj Jain, K. P. Rajeev

Department of Physics, Indian Institute of Technology Kanpur, India

Raj Ganesh Pala

Department of Chemical Engineering and Materials Science Programme, Indian Institute of Technology Kanpur, India

Abstract

Significant elemental transmutations occur on cathodes comprised of either pure metals (Ni, Cu) or the alloys Kanthal ($\text{Fe}_{0.74}\text{Cr}_{0.21}\text{Al}_{0.05}$) and Ni-Fe ($\text{Ni}_{0.93}\text{Fe}_{0.07}$) in light water electrolysis highly biased (~ 30 V) against a pure graphite anode. The utilization of graphite as an anode is advantageous because it avoids anode-driven contamination and establishes a distinct upper bound in the fusion products (Cu), which is different from upper bounds in previous studies (Pb or Ag) with a Pt anode. Whenever isotopic shifts were measured following transmutation, they differed from the natural ratios. Pure Ni cathode predominantly transmutes to Fe and Cu, pure Cu cathode transmutes to Fe and Mg, Kanthal shows the appearance of Cu and Mg and the Ni-Fe alloy transmutes to Mg and Cu. While the extent of transmutation in pure cathodes is a few tens of percent after about twelve hours of Cathodic Hydrogen Loading (CHL), Kanthal and Ni-Fe alloy show comparable or larger amounts of transmutations in less than an hour of CHL.

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1. INTRODUCTION

In the past thirty years, multiple studies have shown nuclear signatures in various low-energy physical and chemical processes [1]–[3]. The history and motivation of most of these studies trace back to the work of Fleischmann and Pons, who reported that their electrochemical apparatus generated anomalous heat (“excess heat”) beyond what can be attributed to chemical reactions [4]. In addition, small amounts of nuclear reaction byproducts (including neutrons and tritium) were also detected in their electrochemical experiments wherein Pd was the cathode. As the focus of the initial studies was mainly on excess heat obtained from H or D fusion, not much attention was paid to the nuclear transmutations of the cathode itself. In addition, to ascertain the conditions for reproducibility of nuclear events, the emphasis was on working with the purest forms of Pd “bulk” cathodes [5].

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Low Energy Nuclear Reactions (LENR) experiments involving electrochemical conditions have been extensively explored, mostly with heavy water and less often with light water [6]. While the role of electrochemical potential in Cathodic Hydrogen Loading (CHL) is beyond debate, arguments have also been made that electronic density in the cathode plays a significant role in reducing the Coulomb barrier, thereby increasing the likelihood of a nuclear reaction at the cathode [7]. The importance of surface and micro/nanostructure has been documented by the codeposition experiments and those involving thin films and metal nanoparticles [8]–[11].

It has also been suggested that there is a critical lower bound to cathodic hydrogen loading (CHL) below which excess heat effects are absent [12]. The viability of light water as opposed to heavy water-based electrolytes for LENR is yet to be established unambiguously. Circumstantial evidence for nuclear phenomena under electrochemical conditions has been documented via the detection of charged particles and gamma radiation [13]–[15]. Still, the precise conditions necessary for reproducible detection of nuclear products are unclear. However, a review of LENR electrochemical experiments suggests that nuclear transmutations of the cathodes might offer reproducible experimental evidence for nuclear phenomena under electrochemical conditions.

The primary objective of the present work is to explore the contrasting nuclear transmutation behavior of alloys, as opposed to pure metals, when they are utilized as cathodes in highly biased light water electrolysis with a graphite anode. We used Ni, Cu, Kanthal ($\text{Fe}_{0.74}\text{Cr}_{0.21}\text{Al}_{0.05}$) and Ni-Fe ($\text{Ni}_{0.93}\text{Fe}_{0.07}$) alloy as cathodes and pure graphite was the anode in all the experiments. Transmutation of the cathode is explored as a function of bias voltage (10–30 Volts), current (a few milliamps to 3 A) and time duration (1–24 hours) of CHL. Our experiments with graphite anodes establish a distinct upper bound for fusion products, Cu, in contrast to other fusion products observed in earlier electrolysis driven transmutation with Pt anodes. The spot analysis with Energy Dispersive Spectroscopy (EDS/EDX/EDAX) shows that part of the Ni cathode gets transmuted post-electrolysis to Cu, Mg, and Fe. We also observed that Cu cathode was partially transmuted to Fe and Mg. Transmutation rates are significantly higher in alloys compared to rates in pure metal cathodes. We found that a substantial fraction of Kanthal transmutes to Cu and Mg within one hour of electrolysis for CHL. Similarly, a significant percentage of the Ni-Fe alloy transmuted to Cu and Mg in one hour of CHL electrolysis. The isotopic ratios in Ni and Cu after electrolysis are found to be different from natural abundance ratios. Our experiments also show that the macroscopic control of electrochemical conditions (such as potential bias, time and current) does not adequately control the transmutation outcomes. That is, with the same macroscopic parameters, there are variations in certain features of nuclear transmutation. We are currently exploring upper bounds in fusion products in other cathodes and the underlying reasons for the effectiveness of alloys in enhanced nuclear transmutation.

2. EXPERIMENTAL PROCEDURE

Light water electrolysis is performed in a covered borosilicate glass beaker[†] with generated H_2 vented out. Various mechanical and chemical cleaning processes are employed to prepare the samples for the experiment. Cleaning procedures are crucial as the experiment is sensitive to inorganic and organic contaminants. The cathode (wire of diameter 0.5 mm) is first mechanically cleaned using fine emery paper. Next, we chemically clean the cathode by dipping the cathode into a dilute acid (sulphuric or hydrochloric acid) to remove oxides present on the metal. Any organic contaminants are washed off using acetone. The cathode is rinsed with deionized water after every chemical treatment. The cathodes are stored in a desiccator. A pure graphite (99.99% by EDS) cylindrical rod (radius 0.3 cm and total length 10 cm of which 1 cm is dipped in electrolyte) is used as the anode. The use of a graphite anode, instead of metallic anodes such as Pt, is an innovation we introduced. We will elaborate on the implications of graphite anodes for our experimental results later in the discussion section. In addition, using a graphite anode serves to react away the generated O_2 , avoiding the dangerous and explosive combustion of H_2 and O_2 , thereby significantly improving the safety of the experiment. Pure Ni, pure Cu and the alloys Kanthal and Ni-Fe in wire form (1 cm of cathode wire dipped in electrolyte in all cases) are utilized as cathodes in 1 molar potassium carbonate solution (99% minimum assay with

0.03% chloride, 0.01% nitrate, 0.02% sulphate, 0.001% iron, and 0.1% sodium as maximum impurity), made using deionized light water (pH 7), as the electrolyte. The typical current density range is from 15–22 A/cm². The high voltage bias generates a significant amount of gases at electrodes, resulting in a significant increase of the electrolyte's resistance, leading to an appreciable increase in temperature of the electrolyte. Some amount of electrolyte is added periodically to compensate for evaporation. EDS/EDX/EDAX, Wavelength Dispersive Spectroscopy [16] (WDS) are used for elemental analysis and Time of Flight Secondary Ion Mass Spectroscopy (ToF SIMS) [17] is employed for isotopic analysis. The instrument used for EDS and WDS is Electron Probe Micro-Analyser (EPMA), JXA-8230 from JEOL and the ToF-SIMS used is PHI TRIFT V NANO ToF from Physical Electronics.

3. RESULTS

The results of the elemental analyses are presented in figures 1 to 4. We used WDS and EDS to characterize cathodes and we present the more precise and accurate WDS data in those cases where they were available. We point out that EDS and WDS are surface analysis techniques, where the electron beam penetrates to a depth of about a micrometer and hence the compositional information extracted using these techniques would be applicable to the near-surface regions. We choose four or five locations (or spots), each of size less than a micrometer in diameter on the surface of the sample for spot analysis.

Figure 1 shows a comparison of the WDS of the pure and transmuted Ni cathode. The purity of the Ni electrode, from Alfa Aesar, is nominally 99.5%. Figure 1(a) shows that the Ni purity determined from WDS varies in the range 98.70% to 99.42% from four data points with an average of 99.20%. We also found trace amounts of Cu (0.50%), Fe (0.12%), Mg (0.10%), Cr (0.04%) and K (0.01%). These trace elements are not visible in Fig. 1(a) because of their tiny size. Figure 1(b) presents the elemental analysis after CHL electrolysis. We find the production of 10.97% Cu and 5.39% Mg on average at four spots of the cathode with 11 hours of electrolysis for CHL.

Figure 2 shows a comparison of the EDS of a pure and transmuted Cu cathode (1 M K₂CO₃ at 30 Volts, 3.5 A for 12 hours). Figure 2(a) shows the pre-sample analysis of the cathode, showing the presence of 100% Cu at all the five locations chosen on the sample. Post-electrolysis analysis Fig. 2(b) shows the presence of 6% Fe and about 2.2% Mg (on average) on the transmuted sample.

Figure 3 shows a comparison of the EDS of a pure and transmuted Kanthal (Fe-Cr-Al alloy) cathode (1 M K₂CO₃ at 30 Volts, 3.5 A for 1 hour.). Figure 3(a) provides the elemental composition of pure Kanthal, which closely matches the expected standard composition of Kanthal. Figure 3(b) shows the composition of the electrolyzed sample. In just one hour, we find Cu (20–53%) and Mg (6–11%) generated in large mass ratios due to transmutations.

Figure 4 shows a comparison of the WDS of a pure and transmuted Ni-Fe alloy (1 M K₂CO₃ at 25 Volts, 3.5 A for 1 hour). Figure 4(a) shows 6% Fe and 94% Ni in the cathode before electrolysis. Figure 4(b) shows that, on average, about 1.8% Cu and 5.9% Mg appear in the cathode within an hour of CHL electrolysis.

In Table 1, we present the isotopic abundance of Cu and Ni at three different locations on the cathode after electrolysis of the Ni-Fe alloy with that of their natural abundance obtained using ToF SIMS measurement. We note that the experimental ratios are quite different from their natural ratios. At all three locations, the abundance of ⁶⁵Cu has reduced and that of ⁶³Cu has increased significantly. Similarly, ⁵⁸Ni, ⁶⁰Ni and ⁶²Ni abundance has reduced while that of ⁶¹Ni and ⁶⁴Ni has increased in the reacted cathode. We note that the increase in the case of ⁶⁴Ni is substantial.

The main observations of this work are:

1. Fusion products heavier than Cu are not generated in any of the highly biased light-water electrolysis conducted with four different cathodes, namely, either pure metals (Ni, Cu) or alloys Kanthal (Fe_{0.74}Cr_{0.21}Al_{0.05}) and Ni-Fe (Ni_{0.93}Fe_{0.07}) against graphite anode. This sharply contrasts the observations of other heavier fusion products when Pt is used as the anode [18]–[19].

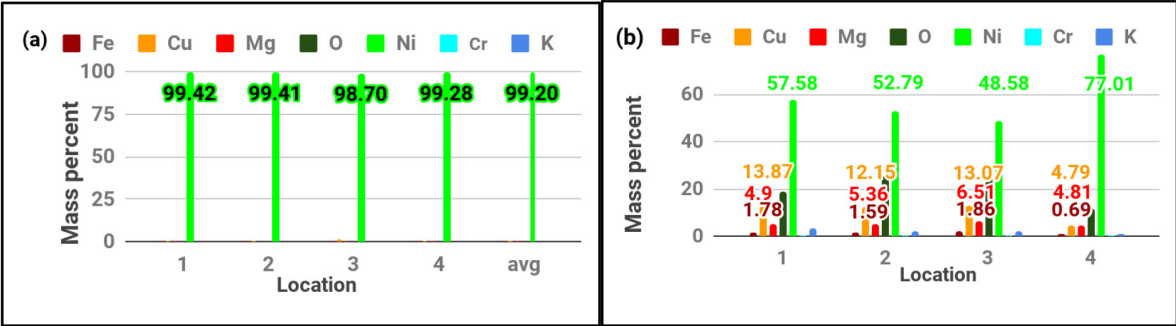


Figure 1. WDS of a Ni cathode before (a) and after (b) electrolysis in 1 M K₂CO₃ at 30 Volts, 2.5 A for 11 hours.

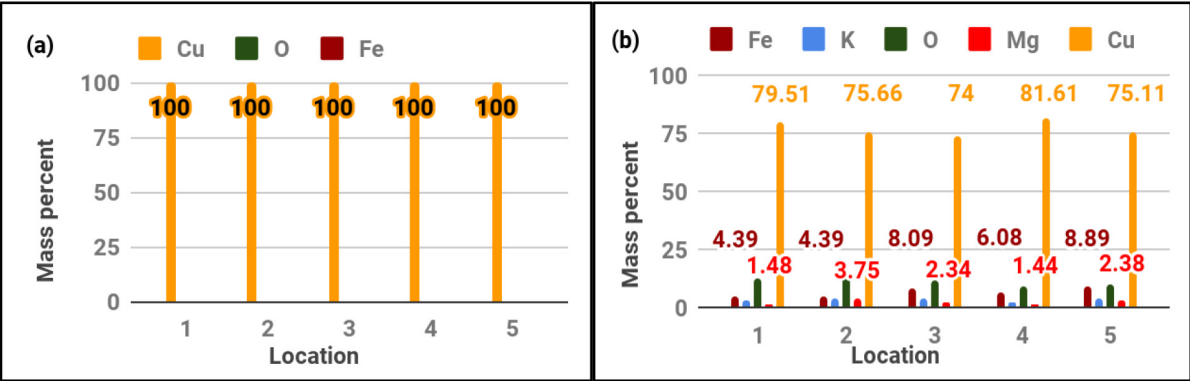


Figure 2. EDS of a Cu cathode before (a) and after (b) electrolysis in 1 M K₂CO₃ at 30 Volts, 3.5 A for 12 hours.

Table 1. Comparison of Cu and Ni isotopic ratios at three different cathode locations post electrolysis with that of natural ratios (ToF SIMS).

Isotope	Natural ratios	Location 1	Location 2	Location 3
⁶³ Cu	69.17	78.91	75.31	77.46
⁶⁵ Cu	30.83	21.08	24.68	22.53
⁵⁸ Ni	68.07	67.63	55.98	67.38
⁶⁰ Ni	26.22	24.37	19.57	24.26
⁶¹ Ni	1.13	1.63	1.91	1.72
⁶² Ni	3.63	3.4	2.15	3.54
⁶⁴ Ni	0.95	2.97	20.33	3.1

2. The utilization of graphite as an anode is beneficial not only in establishing the distinct fusion product (Cu) from the present highly biased electrolysis experiments, but also in avoiding other anode driven contamination.
3. With an Ni cathode, lower mass number Ni isotopes are depleted while there is a build-up of higher mass number Ni isotopes (Fig. 1, Table 1). In the case of Kanthal, composed of Fe, Cr and Al to begin with, the

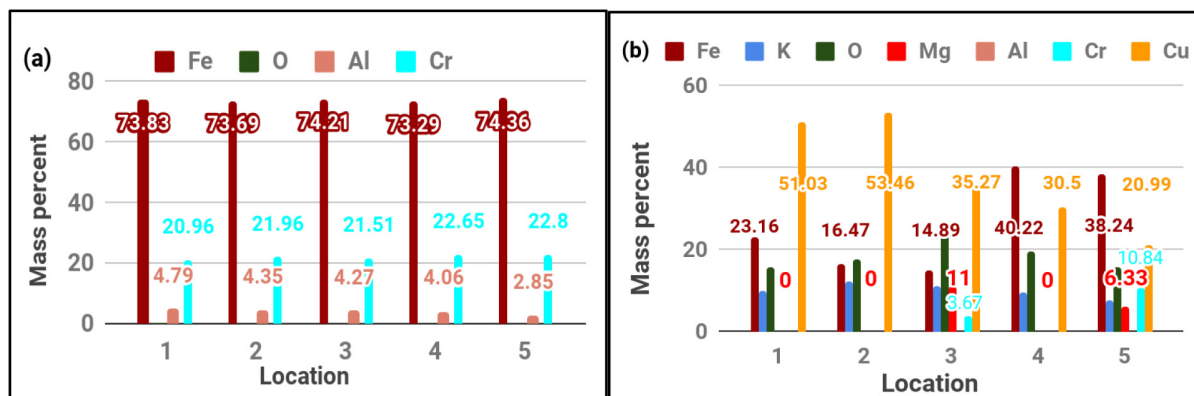


Figure 3. EDS of a pure and transmuted Kanthal cathode before (a) and after (b) electrolysis in 1 M K₂CO₃ at 30 Volts, 3.5 A for 1 hour.

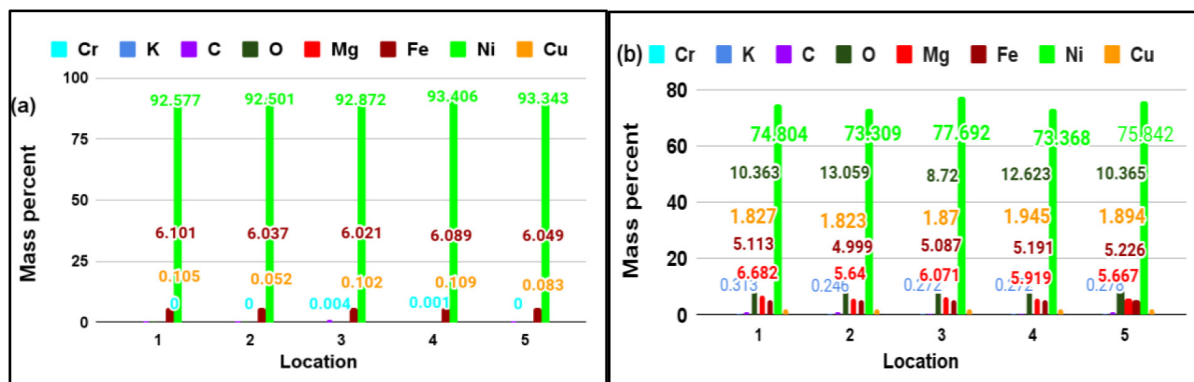


Figure 4. WDS of Ni-Fe cathode before (a) and after (b) electrolysis in 1 M K₂CO₃ at 25 Volts, 3.5 A for 1 hour.

heavier element Cu and also lighter elements such as Mg are generated. Nuclear transmutation into lower mass number elements such as Mg (all figures), Fe (Fig. 2) is also observed in the case of Cu cathode.

4. Observation of transmutation in pure Cu cathode (Fig. 2) goes against the general belief of the LENR community.
5. Alloys require less time for cathodic H loading than pure metals, as seen in the case of enhanced transmutations in Kanthal (Fig. 3) and Ni-Fe alloy (Fig. 4) in contrast to pure Ni (Fig. 1) and Cu (Fig. 2).

4. DISCUSSION

First, let us consider whether contamination can explain the presence of the new elements found in the cathodes. It is very difficult to conceive of a possibility that the new elements found on the cathodes are coming from the contaminants in electrolytes, electrodes or beakers. Even if one considers the possibility of contamination, the amount of contaminants should certainly be many times less than the observed amount of elements found in the cathode. Furthermore, the presence of contaminants would make it impossible to explain the isotopic shifts away from the

natural ratios because the contaminants will tend to move these abundance values towards their natural values. Hence, we conclude that some novel nuclear reactions are definitely occurring in the cathodes in our electrolysis experiments, resulting in the transmutation products observed in the cathodes.

Now we begin our discussion of the main observations of this work listed at the end of the previous section.

4.1. Elemental Transmutations

Elemental transmutations were observed in all four experiments, whether we used pure metal or alloy cathodes. However, we point out that not all of these processes are easy to explain in terms of standard nuclear reactions, even if we ignore the problem of the Coulomb barrier.

In recent papers, the authors have proposed that a deuteron can undergo a nuclear reaction with a heavy nucleus in which the deuteron gets broken up, leading to neutron capture by the heavy nucleus [20], [21]. Hence, we may propose the reaction $\text{Ni}^{60} + \text{d} \rightarrow \text{Ni}^{61} + \text{p}$ to rationalize the increase in ^{61}Ni abundance over its natural abundance. But we note that such a mechanism does not explain how ^{58}Ni can get transmuted to Ni^{60} . We may conceive of the reaction $\text{Ni}^{58} + \text{d} \rightarrow \text{Ni}^{59} + \text{p}$ with subsequent weak decay of Ni^{59} into Co^{59} , however, the ground-state of Ni^{59} has a half-life of 76000 years! It may be possible that some excited states might be involved and such possibilities are to be explored in the future. Also, we do not find any experimental evidence for any nuclide with a mass number 59 in our ToF SIMS data. If we admit the possibility of deuteron capture, then we have $^{58}\text{Ni} + \text{d} \rightarrow ^{60}\text{Cu} \rightarrow ^{59}\text{Ni} + \text{e}^+$ with ^{60}Cu having a lifetime of only 24 minutes. We note that the availability of deuterons is not an issue because the abundance of deuterons in hydrogen is about 150 ppm and we expect this fraction of deuterons to be available in light water. However, it is not clear how deuterons can get captured by a heavy nucleus because of the large Coulomb repulsion between them.

We find transmutation products with mass numbers ranging both below and above the atomic mass number of the host metal in the cathode. To explain how elements with lower mass numbers can appear in the cathode, we have to invoke the possibility of nuclear fission. The presence of Mg probably can only be accounted for in this fashion.

The presence of K in the cathode in Kanthal is probably due to the K^+ ions in the electrolyte (K_2CO_3 in water) being driven into the cathode during the electrolysis process. The carbon and oxygen found on the cathodes could probably be accounted for as contaminants picked up from the atmosphere or during handling. We note that they could also be transmutation or fission products.

4.2. Shorter (Longer) Hydrogen Loading Time in Alloys (Pure Metals)

From the data presented in Figs. 1–4, we find that the pure metal cathodes require about ten times more time of CHL to produce roughly the same amount of transmutation products compared to alloy cathodes Kanthal and Ni-Fe. We found a conversion of about 40% of the Nickel from the nickel cathode in about 11 hours of electrolysis for CHL (Fig. 1), whereas in the case of Kanthal and Ni-Fe about 60% and 20% respectively of the original material gets transmuted to other elements in one hour of CHL (Figs. 3 & 4). We repeated this experiment many times and found that the alloys always take much less time for CHL than pure metals in showing transmutations. The reason for this behaviour of the alloys remains a mystery. Typically, alloys, in comparison to pure metals are better hydrogen evolution electrocatalysts [22]. Hence, greater amount of hydrogen at higher chemical potential will be available when alloys are utilized as cathodes. In addition, under the high bias in the light-water electrolysis, K^+ from the electrolyte will be cathodically reduced and alloyed with the cathode. Such an addition of electropositive metals to transition metals has been shown to enhance the hydrogen loading capacity of the alloys [23]. Please note that the CHL time corresponds to the time of electrolysis. While it is most likely that transmutation occurs during CHL/electrolysis, the possibility that transmutation occurs post-electrolysis (say during EDS/WDS) cannot be neglected. Currently, our experiments are directed towards addressing this issue.

4.3. Copper as Cathode

We find transmutation products with pure Cu cathodes (Fig. 2). After 12 hours of CHL electrolysis, about 5% Fe and 2% Mg show up in the Cu electrode. The formation of these elements can probably be accounted for via a fission process. We point out that this may be the first time a LENR electrolysis experiment with a pure copper cathode has been conducted and the evidence of fission of copper nuclei presented. Most of the electrolysis experiments in LENR have been performed with Pd or Ni cathodes and the majority of the LENR community has been under the impression that nuclear reactions can occur only with these two elements as cathodes.

4.4. Graphite Anode Limits Contamination and Transmutation Artifacts at Cathode

The heaviest transmuted element found in all of our experiments with a graphite anode (irrespective of whether the cathode is Ni, Cu, Kanthal or Ni-Fe alloy) is Cu. In other electrolysis studies with Ni cathodes, and Pt anodes, the heaviest element found in the Ni cathode was Pb [11], [18]–[19]. Contrasting these studies suggests that the anode is not a passive participant in these electrochemical activated nuclear reactions. Based on the Pourbaix diagram of the anodes, we anticipate that under sufficient anodic potential, the anodes like Pt will get oxidized and dissolve in the electrolyte [24]. Subsequently, these ions can get reduced/deposited on the working cathode and may undergo transmutation and fission. Also, if the metal anode contains any impurities, they may get deposited on the cathode, which might lead to confounding experimental results due to the nuclear reactions of the impurities. Our data suggest that the graphite anode indirectly prevented the formation of heavier elements (beyond Cu) at the cathode. Overall, to avoid contamination from the anode and associated artifacts in transmutation products at the cathode, we suggest the utilization of a graphite anode.

4.5. Upper Bound on Fusion Products at the Cathode

In an effort to understand some general trends, we contrast the fusion products from our experiments in light of previous experiments. We observe two fusion reactions: 1. Ni to Cu and 2. Fe or Cr to Cu. The transmutation products from Pd/D co-electrodeposition on Au foil by Mosier-Boss et al. [25] yielded Cr, Fe and Ni only and hence, no fusion products were observed. A similar Pd/D co-electrodeposition experiment by Dash and Ambadkar [26] using Pd (anode) and Pd (cathode) in $\text{H}_2\text{SO}_4\text{-D}_2\text{O}$ electrolyte yielded Ag as a fusion product. The light water-electrolysis by Miley and Patterson with multilayer thin films of Pd and/or Ni cathode and Pt anode generated a large variety of fission/fusion transmutation products [19]. Four broad peaks occurring around $Z = 6\text{--}18$ (Mg-Si), $Z = 22\text{--}30$ (Fe-Zn), $Z = 44\text{--}50$ (Ag-Cd), and $Z = 75\text{--}85$ (at Au) with Pb as the heaviest element found on the beads. The D_2 gas permeation over Cs, Sr, Ba, or other elements deposited over Pd/CaO by Iwamura [27] displayed transmutation of Cs into Pr, Sr into Mo, Ba into Sm, and W into Pt and the heaviest fusion product was Pb. Heavy water electrolysis by Mizuno et al. [28] yielded Pb as the heaviest fusion product.

First, in all transmutations observed via electrolysis, there is an upper bound of observed fusion products. Neither the model of Widom and Larsen [29] nor other models for transmutation [30]–[31] provide guidelines for rationalizing the upper bound for fusion products.

Second, as none of the above experiments were performed with graphite as anode, it will be insightful to repeat these experiments without a Pt anode to evaluate if the fusion products differ in the two scenarios.

5. CONCLUSION

We demonstrate nuclear transmutation in cathodes made of pure Ni, pure Cu, Ni-Fe alloy and Kanthal via light water electrolysis undertaken at a voltage bias of 25–30 V against graphite anodes at about 100°C and atmospheric pressure,

for 1 to 12 hours of cathodic hydrogen loading. Cu is the heaviest fusion product on the cathode, and fission products such as Mg are observed. Cu used as a cathode with a graphite anode leads to induced fission processes in the Cu cathode. Graphite used as anode suppresses the formation of certain heavy elements on the cathode observed in earlier electrolysis transmutation experiments, implying that the anode actively participates in LENR processes occurring in the cathode. Results also demonstrate that alloys as cathodes are better than pure elements in inducing transmutations.

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ENDNOTES

† Borosilicate composition: we use beakers from Borosil™ brand the composition of which is, from their website, SiO₂ 81%, B₂O₃ 13%, Na₂O / K₂O 4%, Al₂O₃ 2% by weight).

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