NEUTRON ISOTOPE REACTIONS

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For discussion and review.

Abstract. We explore the possibility that transfer of neutrons from neutron isotopes to ordinary nuclei, followed by beta decay of the neutron-enriched nuclei, facilitates a class of low-temperature transmutations. We have tested this possibility by comparing the implications of neutron isotope theory with the transmutations reported by Iwamura and associates. We find that experiment quantifies and supports the theory, and that theory clarifies and supports the experimental observations.

1. Introduction

The field of low temperature nuclear reactions has made slow but steady progress. Evidence has accumulated for production of energy at the level of a few watts, for production of helium in proportion to energy, for energetic particles, and for transmutations of elements. But there is no generally accepted theory for these phenomena. Progress requires a body of experimental evidence and a candidate theory through which theory and experiment can gain mutual support and acceptance.

Energy, helium, and energetic particles have not provided discrimination among theories, nor do they offer data of such nature as to constrain and test theory. Transmutations however offer a possibility. Iwamura and his associates have investigated the transmutation of a number of target elements. In these experiments the quantity of a target element declines while simultaneously the quantity of the transmuted element rises. Only a fraction of potential transmutations are observed. Theory must be able to predict which are observed and which are not.

Our theory is based on the idea that transmutation occurs by transfer of neutrons from a neutron source to a target nucleus, followed by beta decay of the target to a stable transmutation product. We hypothesize that the neutron sources are neutron isotopes and apply this theory to the transmutation data, in order first to determine the extent to which parameters of the theory can be deduced from the data, then to determine the extent to which theory so refined can improve our understanding of the transmutation process.

We have found that neutron isotope theory and transmutation experiment do in fact support each other, improving the theoretical basis for understanding the transmutation process and for predicting which transmutation products are observed. This finding supports the belief that the observed transmutations are indeed nuclear reactions of a new kind, and strengthens the idea that neutron isotopes are active participants in these reactions.

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We hope this partnership between experiment and theory will help to justify, to motivate, and to guide new experimentation. The field of neutron isotope physics holds promise of being as rich as that of neutron physics in expanding our knowledge of nuclear phenomena, and it opens the potentiality of applications to energy production, nuclear medicine, nuclear waste remediation, and other fields that parallel and extend the fields currently available to nuclear physics.

2. Experimental

Iwamura and his associates (subsequently referred to as Iwamura) have made extensive investigations of low-temperature transmutations [1]–[12]. The active element in their reactor is a palladium sheet 0.1mm thick and about 25mm square, the top surface of which is coated with a thin CaO/Pd complex protected by a 400Å overlay of palladium. In some experiments the complex was formed by simultaneous sputtering of CaO and Pd to a thickness of 1000–3000Å. In other experiments the complex consisted of alternating 20Å layers of CaO and 180Å layers of Pd to a thickness of about 1000Å. The Pd sheet is mounted in equipment that exposes the top surface to deuterium from electrolysis using the plate as cathode, or to deuterium gas at about one atmosphere pressure. The bottom surface is exposed to vacuum. During operation deuterium enters the film at the top surface, flows through the CaO/Pd complex and supporting Pd sheet, and emerges from the vacuum surface.

Under these conditions a low-temperature nuclear reaction was initiated and sustained. Measurement of energy output in excess of input established that the reactor was generating energy. Observation of X-radiation suggested that the energy was nuclear. Observation of transmutations confirmed that the reaction was nuclear. In some experiments atoms of a target element were deposited in a layer of atomic thickness on the top surface of the CaO/Pd complex. In others the components of the complex served as target elements. After mounting the complex in the reactor the vessel was sealed and reaction was initiated. The duration of reaction was approximately one or two weeks. Transmutation reactions were observed and quantified by various experimental methods following and sometimes during reaction, providing overall a challenge to theory.

3. Theory

Neutron isotope theory [13] suggests that interactions between neutron isotopes and deuterium can support a chain reaction that maintains a concentration of ambient neutron isotopes within an active reaction volume. (This is analogous to the interaction between neutrons and uranium that maintains a concentration of ambient neutrons in an ordinary nuclear reactor.) We accept this as our starting point, and explore the reactions that are expected from interactions of ambient neutron isotopes with target elements.

Chain reaction. Let ^{A}n represent a neutron isotope containing A neutrons. In our view a neutron isotope reactor is activated by a chain reaction that includes an exothermic isotope growth reaction such as

$$^{A}\mathrm{n} + {}^{2}\mathrm{H} \longrightarrow {}^{A+1}\mathrm{n} + {}^{1}\mathrm{H} \tag{1}$$

that transfers a neutron from ^{2}H fuel to a neutron isotope. It also includes exothermic isotope fission reactions such as the sequential beta/beta/alpha combination

$$^{\mathrm{A}}\mathrm{n} \longrightarrow {}^{\mathrm{B}}\mathrm{n} + {}^{\mathrm{A-B-4}}\mathrm{n} + {}^{4}\mathrm{He} \tag{2}$$

Together these reactions can support a chain reaction that builds and maintains the environment of ambient neutron isotopes.

Transmutation. In the general transmutation reaction a neutron isotope interacts with an ordinary nucleus $^{\mathrm{B}}$Z by transferring one or more neutrons to or from it in a reaction of the form

$$^{\mathrm{A}}\mathrm{n} + {}^{\mathrm{B}}\mathrm{Z} \longrightarrow {}^{\mathrm{A-C}}\mathrm{n} + {}^{\mathrm{B+C}}\mathrm{Z}. \tag{3}$$

In this reaction the number of transferred neutrons C is usually positive and $^{\mathrm{B+C}}$Z is a neutron-rich isotope of $^{\mathrm{B}}$Z.

Physically we expect that a neutron isotope diffuses at random throughout the active reaction volume, occasionally encountering an ordinary nucleus that serves as a target nucleus for transmutation. It then recoils elastically or it reacts by neutron transfer. We assume that the concentration of ambient neutron isotopes is stable, and that neutron transfer reactions occur at a steady rate that can be quantified by a fixed mean lifetime for survival of target nuclei, which we denote by t_m. Following transfer of neutrons, a target nucleus becomes a heavier isotope of the target element. That isotope may be stable, or it may undergo decay to an isotope of a new element, and so on through a chain of decays until it reaches a stable isotope.

Quantitative analysis requires knowledge of the mass excesses of neutron isotopes. We write $\Delta(^{\mathrm{A}}\mathrm{n})$ for the mass excess of isotope $^{\mathrm{A}}$n. We expect that a neutron isotope can be described by a liquid drop model in which

$$\Delta(^{\mathrm{A}}\mathrm{n}) = \mathrm{A}\,\Delta(\mathrm{n}) - a_v\,\mathrm{A} + a_s\,\mathrm{A}^{2/3} \tag{4}$$

over the range of A for neutron isotopes participating in a steady state reaction. In this formula $\Delta(\mathrm{n})$ is the mass excess of a neutron, $a_v > 0$ is the binding energy per neutron, and $a_s > 0$ is a surface energy parameter. The change in mass excess on adding one neutron is

$$\theta = \Delta(^{\mathrm{A+1}}\mathrm{n}) - \Delta(^{\mathrm{A}}\mathrm{n}) = \Delta(\mathrm{n}) - a_v + (2/3)\,a_s\,\mathrm{A}^{-1/3}. \tag{5}$$

Because the parameter θ is a slowly-varying function of A that grows smaller as A grows larger, we consider a range of values of θ that corresponds to the range of values of A for the neutron isotopes that participate in the chain reaction.

The transmutation reaction (3) releases energy E that depends on the value of C,

$$\begin{aligned} E &= \Delta(^{\mathrm{A}}\mathrm{n}) - \Delta(^{\mathrm{A-C}}\mathrm{n}) + \Delta(^{\mathrm{B}}\mathrm{Z}) - \Delta(^{\mathrm{B+C}}\mathrm{Z}) \\ &= \mathrm{C}\,\theta + \Delta(^{\mathrm{B}}\mathrm{Z}) - \Delta(^{\mathrm{B+C}}\mathrm{Z}). \end{aligned} \tag{6}$$

For any target isotope $^{\mathrm{B}}$Z having charge Z, and for any value of θ in an allowed range there is a value of C that produces maximum released energy. If the maximum is positive we assume that reaction (3) occurs and target isotope $^{\mathrm{B}}$Z is transmuted to product isotope $^{\mathrm{B+C}}$Z. There may be a single value of C for θ in the allowed range, leading to a single transmutation product, or there may be different values of C within contiguous subranges of θ, leading to a mix of transmutation products. These neutron-rich isotopes generally

are unstable and each undergo a series of beta decays to stable or long-lived transmutation products ${}^{\mathrm{B+C}}(\mathrm{Z}+\Delta\mathrm{Z})$. When this process is repeated many times, each time with a new target nucleus and a new random value of θ, the supply of target isotope ${}^{\mathrm{B}}\mathrm{Z}$ is gradually depleted and the supplies of transmutation products ${}^{\mathrm{B+C}}(\mathrm{Z}+\Delta\mathrm{Z})$ are correspondingly increased.

Products of neutron transfers. The neutron transfer reaction (3) can be rewritten

$$ {}^{\mathrm{A}}\mathrm{n} + {}^{\mathrm{B}}\mathrm{Z} \;\longrightarrow\; {}^{\mathrm{A+B-D}}\mathrm{n} + {}^{\mathrm{D}}\mathrm{Z} \tag{7} $$

where $\mathrm{D} = \mathrm{B} + \mathrm{C}$ is the mass number of the product isotope. Similarly, we can express the energy released by the transfer of neutrons in terms of D,

$$ E = \mathrm{D}\,\theta - \Delta({}^{\mathrm{D}}\mathrm{Z}) + \big\{\Delta({}^{\mathrm{B}}\mathrm{Z}) - \mathrm{B}\,\theta\big\}. \tag{8} $$

We see that for any value of θ the value of D that maximizes the energy released is independent of B (although the amount of energy depends on B). In general (for any value of θ), all isotopes of a target element lead to a common product isotope of that element.

Calibration. To match the lifetimes of these processes to the timescale of natural decay values we note that the target survival mean lifetime t_m can be determined approximately from the Iwamura data. Overall t_m increases as the target mass number B increases, roughly according to the relationship $t_m = 0.05\,\mathrm{B}$ for time measured in days. The expression for t_m indicates that initiation of neutron transfer is less probable for larger targets, suggesting that the alterations in nuclear structure required for neutron addition become more complex and therefore less likely.

Our analysis of the data suggests that θ lies in the allowed range $2.95 < \theta < 3.82$. (The rationales for the lower and upper bounds are given in Sections 4.2 and 4.10, respectively.) In the absence of guiding evidence we assume that the values of θ are uniformly distributed throughout this range.

Summary. In summary our theory is based on equations (1–6) constrained by the parameters $t_m = 0.05\,\mathrm{A}$ and $2.95 < \theta < 3.82$. With these equations and parameters, utilizing the system of differential equations described in Appendix A, we are able to calculate the theoretical evolution of neutron transfers and isotope decays through successive generations of transmutation for any target element or isotope.

4. Analysis

We now proceed to discuss the transmutations of target elements into product elements. The elements Pd, Ca, O of the CaO/Pd complex were present as potential target elements in every Iwamura experiment. In most experiments an additional target element (C, Li, Na, Sr, Cs, Ba, or W) was provided in the nuclear active region. For each target element we determine quantitatively the progress of transmutation as predicted by theory; we review the data obtained by Iwamura; and we discuss the extent to which the theory is able to account for the data.

In Table 1 we list for reference the various detection methods used by Iwamura. Note that some methods determine mass numbers and others identify elements. In the text, we present figures for transmutations showing theoretical concentration curves (as functions of

TABLE 1. Methods of detection and properties detected.

Method	Acronym	Property
Secondary Ion Mass Spectrometry	SIMS	
Time of Flight SIMS	TOF-SIMS	mass number
Inductively Coupled Plasma Mass Spectroscopy	ICP-MS	
X-ray Photoelectron Spectroscopy	XPS	
X-ray Absorption Near Edge Structure	XANES	
Energy Dispersive X-ray Spectrometry	EDX	element
Wavelength Dispersive X-ray Spectroscopy	WDX	
Auger Electron Spectrometry	AES	
X-ray Fluorescence	XRF	

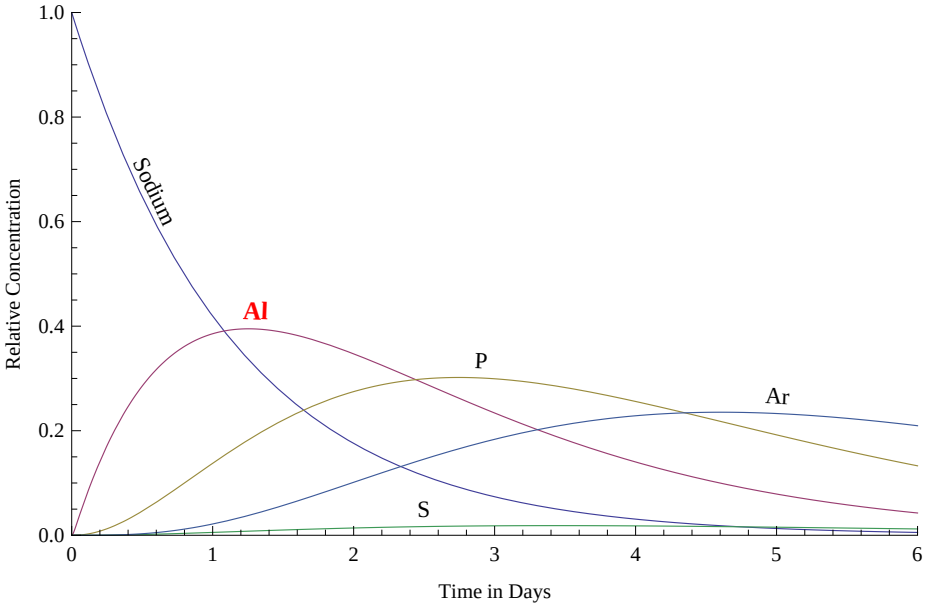

FIGURE 1. **Sodium**. Theoretical concentration curves are shown for the target element sodium and for the first three generations of elemental transmutation products (Al, P+S, Ar). Iwamura [9] detected a chemical signal for aluminum as indicated with bold red print.

time). In each figure we highlight (in bold and red) the degree to which the experimental observations of mass numbers and elements confirm the theoretical expectations.

4.1. **Sodium transmutation.** We begin the comparison of theory and experiment by consideration of the transmutations of sodium. In their sodium experiment [9] Iwamura dissolved sodium in the electrolyte of an electrolytic reactor. Following reaction they ascertained by chemical analysis of the electrolyte that the sodium concentration had decreased while at the same time the aluminum concentration had risen significantly from a negligible initial value, indicating transmutation of sodium to aluminum.

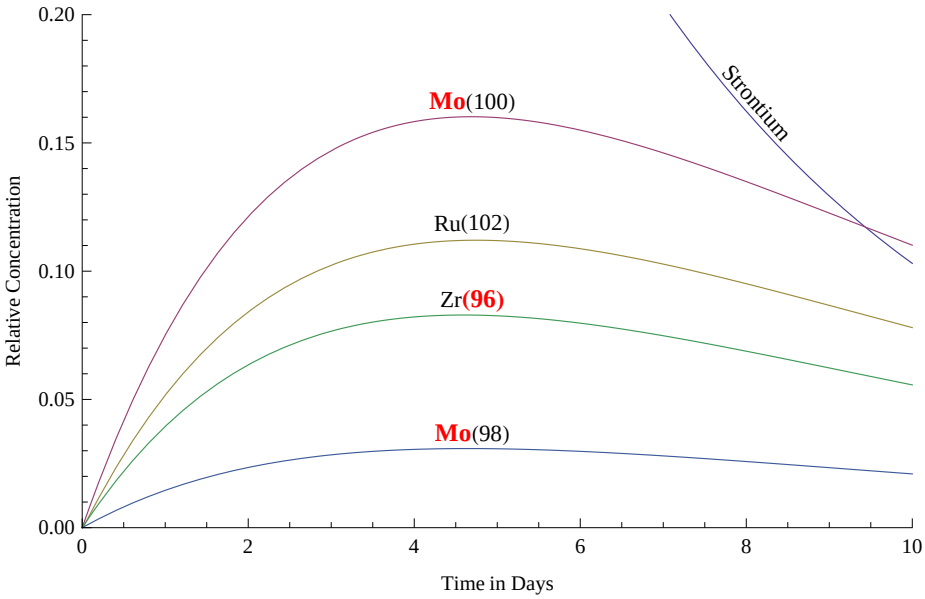

FIGURE 2. **Strontium**. Theoretical concentration curves are shown for first generation transmutation product isotopes. Iwamura [7] detected XPS data for molybdenum and SIMS data for mass-96, both indicated with bold red print.

The theoretical transmutation reactions begin with the stable sodium isotope ^{23}Na. When this target isotope is exposed to a neutron isotope environment in an Iwamura reactor, neutrons are transferred from neutron isotopes to sodium targets via reaction (3) with $^{B}Z = {}^{23}$Na. For each transfer a value of θ is chosen at random from the range $2.95 < \theta < 3.82$. The number of neutrons in a transfer is the number required to maximize the energy released in that transfer (6). For sodium all values of θ lead to the transfer of four neutrons and to the product ^{27}Na:

$$^{A}\mathrm{n} + {}^{23}\mathrm{Na} \longrightarrow {}^{A-4}\mathrm{n} + {}^{27}\mathrm{Na}. \tag{9}$$

The ^{27}Na undergoes beta decay to ^{27}Mg with a half life of 300ms and the ^{27}Mg then undergoes beta decay to stable ^{27}Al with a half life of 9.5m. The ^{27}Al is available for observation and quantitative measurement, and also to serve as target for a second generation of neutron transfers. The half lives of ^{27}Na and ^{27}Mg are so short that these isotopes do not significantly accumulate before they decay, and their influence on the overall transmutation reaction is negligible. In consequence the neutron transfer and beta decay reactions can be represented together in the overall reaction

$$^{23}\mathrm{Na} + 4\,\mathrm{n} \longrightarrow {}^{27}\mathrm{Na} \Longrightarrow {}^{27}\mathrm{Al}. \tag{10}$$

In this representation the symbol "$\Longrightarrow$" indicates a series of short-lifetime beta decays for which there is negligible accumulation of participating isotopes in comparison with the accumulation of a long-lived product isotope (in this instance ^{27}Al).

As the concentration of ^{27}Al builds up, this isotope becomes a significant target for transmutation in a second generation of neutron transfer reactions. The ^{27}Al can undergo either of two transmutation reactions, depending on the value of θ (the per-neutron change in neutron isotope mass excess). For θ in the range 3.21–3.82 (which constitutes 70% of the total allowed range 2.95–3.82) transfer of 6 neutrons is the most energetic, while for θ in the range 2.95–3.21 (30% of the total allowed range) transfer of 4 neutrons is the most energetic. Thus the branching ratio for 6 neutrons is 0.7 and the branching ratio for 4 neutrons is 0.3.

The second generation of neutron transfer reactions is characterized by

$$^{27}\mathrm{Al} + 6\,\mathrm{n} \longrightarrow {}^{33}\mathrm{Al} \Longrightarrow {}^{33}\mathrm{P} \Longrightarrow {}^{33}\mathrm{S} \tag{11}$$

$$^{27}\mathrm{Al} + 4\,\mathrm{n} \longrightarrow {}^{31}\mathrm{Al} \Longrightarrow {}^{31}\mathrm{P}. \tag{12}$$

In (11) the long-lived isotope ^{33}P has a half life of 25 days, during which a portion undergoes beta decay to stable ^{33}S. In consequence both ^{33}P and ^{33}S are significant targets for transmutation in a third generation of neutron transfer reactions,

$$^{33}\mathrm{S} + 9\,\mathrm{n} \longrightarrow {}^{42}\mathrm{S} \Longrightarrow {}^{42}\mathrm{Ar} \tag{13}$$

$$^{31}\mathrm{P} + 8\,\mathrm{n} \longrightarrow {}^{39}\mathrm{P} \Longrightarrow {}^{39}\mathrm{Ar} \tag{14}$$

$$^{33}\mathrm{P} + 6\,\mathrm{n} \longrightarrow {}^{39}\mathrm{P} \Longrightarrow {}^{39}\mathrm{Ar}. \tag{15}$$

Figure 1 shows the theoretical time dependence of the first three generations of sodium transmutation reactions. Product S does not significantly accumulate because most of its precursor ^{33}P(25d half life) undergoes neutron transfer to ^{39}P before it has time to beta decay to ^{33}S. Iwamura [9] detected a chemical signal for aluminum as indicated with bold red print. Theory and experiment agree for transmutation of Na to Al.

4.2. **Strontium transmutation.** Figure 2 shows the theoretical transmutation of strontium deposited on the surface of the CaO/Pd complex in a deuterium gas reactor. First generation products ^{100}Mo, ^{102}Ru, ^{96}Zr, and ^{98}Mo rise to their maxima at about 5 days. Experimentally Iwamura [7] reported XPS evidence for molybdenum, and SIMS evidence for mass 96 where no such signals existed prior to transmutation. The experimental evidence for molybdenum and for mass 96 is flagged by bold red type. Iwamura interpreted these observations as evidence for ^{96}Mo. We interpret the observations as evidence for ^{96}Zr, ^{98}Mo, and ^{100}Mo. The data support both interpretations. Overall the theory is in good agreement with the observations of Mo and of mass 96.

The experimental data also show that no mass-94 nuclei are produced during transmutation. Theory predicts that ^{94}Zr is produced if $\theta < 2.95$. Hence to obtain agreement between experiment and theory it is necessary that the range of θ be restricted to $\theta > 2.95$ as we have done. With this restriction theory and experiment are in good agreement.

4.3. **Cesium transmutation.** In order to demonstrate the transmutation of cesium deposited on the surface of the CaO/Pd complex, Iwamura recorded *in situ* XPS signals during the transmutation process [7]. They identified a cesium signal that decreased during transmutation, and they observed a praseodymium signal that appeared and grew. From measurements of the magnitudes of the XPS signals they deduced that the decline of the

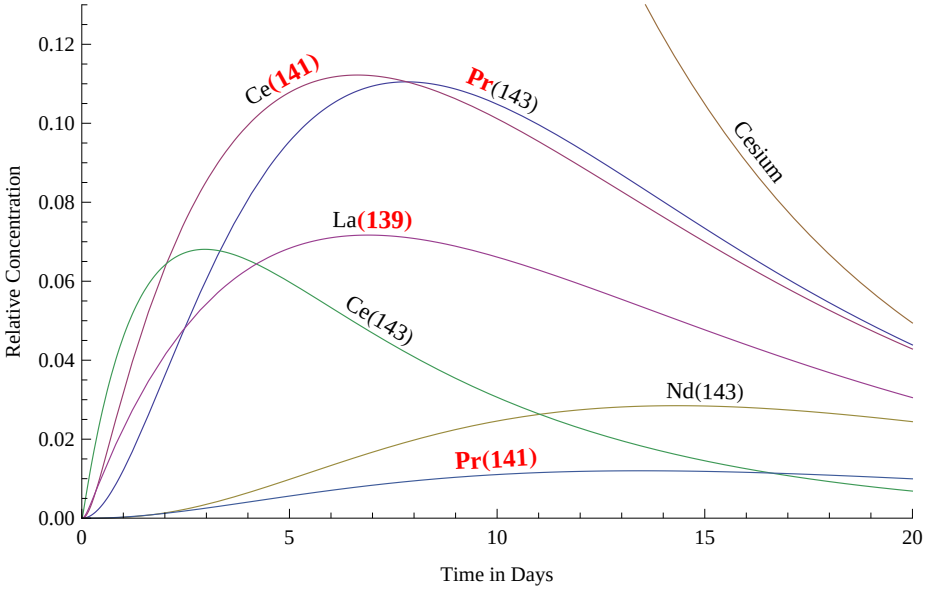

FIGURE 3. **Cesium**. Theoretical concentration curves are shown for first generation transmutation product isotopes (mass numbers 139, 141, 143). In separate experiments Iwamura has detected XPS evidence for praseodymium [7], and SIMS evidence for mass numbers 139 and 141 [9], as indicated with bold red print.

cesium signal matched the growth of the praseodymium signal within the accuracy of their measurements. In an experiment with a cesium target in an electrolytic reactor [9] they found SIMS evidence for mass numbers 139 and 141. Figure 3 shows that the theoretical transmutation of cesium agrees with these experimental observations.

Based on the evidence for a concentration of Pr that increased as the concentration of Cs fell, and for a mass-141 product, Iwamura interpreted the product to be stable ^{141}Pr. Our theory provides an alternate interpretation, suggesting that the rising XPS signal measured during transmutation results from ^{143}Pr and the mass-141 signal results from ^{141}Ce.

This question of interpretation is examined in Figure 4 which examines theory for an experiment in which nuclear reaction is terminated at $t = 7$ days when the concentrations of ^{143}Pr and ^{141}Ce are near their maxima. Both ^{143}Pr and ^{141}Ce are radioactive and undergo beta decay. The half life for $^{143}\text{Pr} \rightarrow {}^{143}\text{Nd}$ is 13.6 days and the half life for $^{141}\text{Ce} \rightarrow {}^{141}\text{Pr}$ is 32.5 days. As ^{141}Ce decays, ^{141}Pr rises and maintains a constant total for mass-141 products. And as ^{143}Pr decays, the rise in ^{141}Pr maintains an approximately constant concentration of Pr. Owing to the somewhat different half-lives of ^{143}Pr and ^{141}Ce, about 40 days after nuclear reaction is terminated the total Pr concentration sags to about 80% of its starting and finishing concentrations. Iwamura also reports [10] the results of XPS, TOF-SIMS, XANES, XRF, and ICP-MS tests at various times following transmutation, confirming the presence of praseodymium and mass 141 for periods of time up to months after nuclear reaction was terminated.

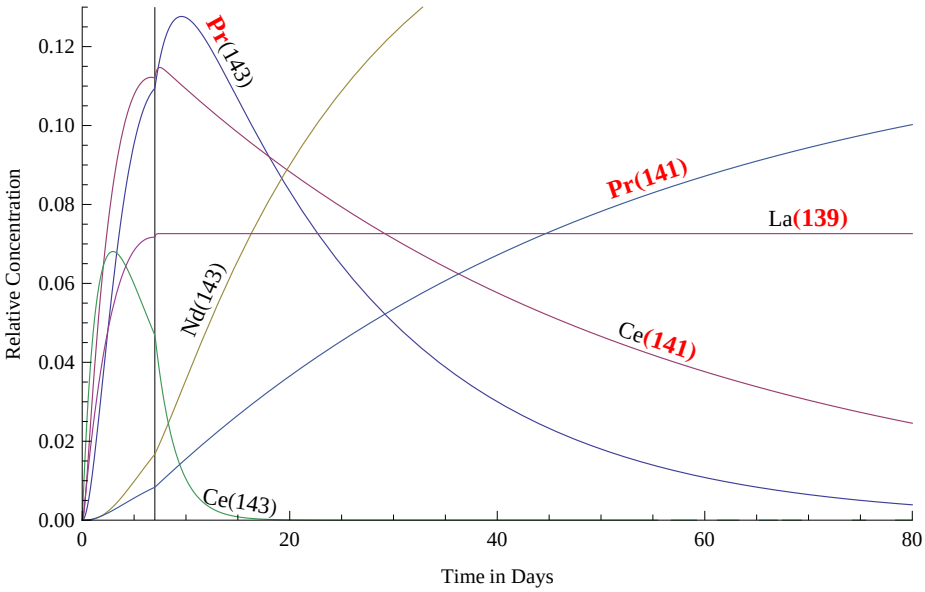

FIGURE 4. **Cesium** (interrupted nuclear reaction). Comparison of theoretical and experimental transmutations of cesium in an experiment with nuclear reaction terminated after 7 days (near the peak concentrations of ^{143}Pr and ^{141}Ce). Iwamura evidence [see the caption to Figure 3] is indicated by bold red print.

We conclude that there is good evidence for the slow beta decay of first generation cesium transmutation products ^{143}Pr + ^{141}Ce into stable products ^{143}Nd + ^{141}Pr after nuclear reaction has been terminated, conserving mass number 141 and approximately conserving the concentration of praseodymium in satisfactory agreement with experiment.

4.4. **Barium transmutation.** We now consider the transmutation of barium deposited on the surface of the CaO/Pd complex. Iwamura investigated this process with both XPS and SIMS [11]. In an XPS experiment the concentration of samarium Sm3d was observed to rise while the concentration of barium Ba3d declined, providing strong evidence of transmutation to samarium. In a wide-range XPS spectrum (an enlargement of Figure 4a in [11]) additional XPS evidence was presented for samarium Sm4d and Sm4p. The wide-range spectrum also contained evidence for the Auger signal GdA(362eV) for gadolinium, and for the Nd3d$_{5/2}$(983eV) signal for neodymium, although these signals were not noted in the text. The SIMS experiment showed evidence for mass numbers 146, 148, and 150. In Figures 5 and 6 we plot the theoretical evolution of transmutation isotopes and compare with the XPS and SIMS observations. To minimize clutter we plot the first generation transmutation products in Figure 5 and the second generation products in Figure 6.

Using a target of 91% enriched ^{137}Ba, a signal was observed at mass 149 [11]. This can be interpreted as the molecule ^{137}Ba^{12}C resulting from carbon contamination, or as evidence for a clean isotope of a transmutation product. However it is shown (see the upper portion of Figure 7 in [11]) that in the absence of carbon the ^{137}Ba component of natural

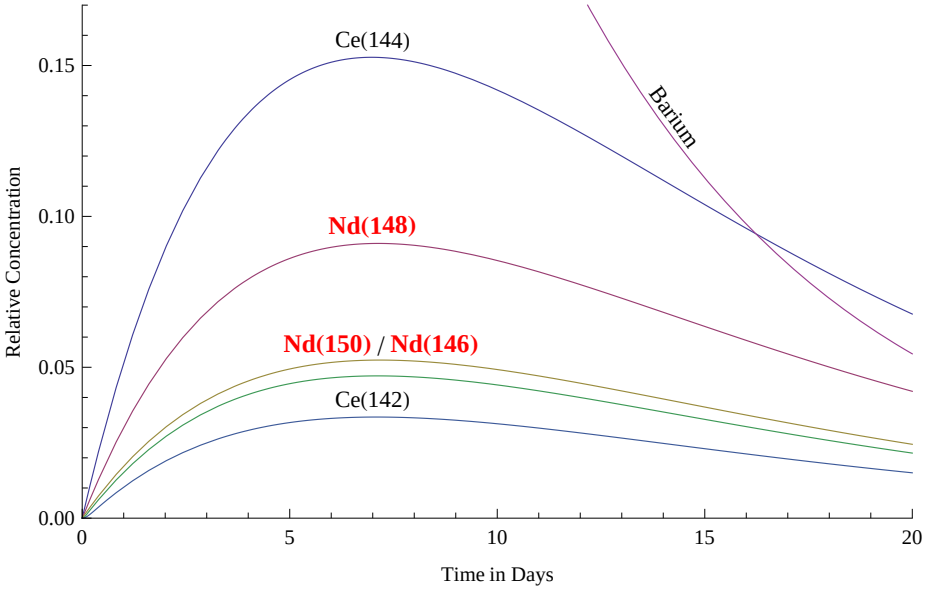

FIGURE 5. **Barium** (first generation). Theoretical concentration curves are shown for first generation transmutation product isotopes. Iwamura [11] detected XPS data for neodymium, and SIMS evidence for masses 146, 148 and 150, as indicated with bold red print. Second generation products are shown in Figure 6.

barium does not transmute to a mass 149 product. Hence the interpretation of the mass 149 product as a $^{137}Ba^{12}C$ molecule is confirmed to result from carbon contamination.

Although theory and experiment are in good agreement for transmutation of barium to the elements neodymium, gadolinium and samarium, we note that the theoretical concentrations of neodymium isotopes do not agree with the experimental SIMS concentrations. Experimentally the relative concentration of ^{150}Nd is 0.78 of total Nd, whereas the theoretical relative concentration is only 0.30 of total Nd. It may be that the theory is in error, or it may be that there are errors in the tabulated mass excess values on which the theory relies. Application of the theory to barium requires knowledge of the mass excesses of extremely neutron-rich barium isotopes. Examination of the most recent data [15] shows that the experimental uncertainties of the mass excesses of these isotopes are on order of 0.5 MeV, more than enough to account for the differences between theory and experiment regarding relative quantities of transmutation product isotopes. We anticipate that as more accurate mass excess values become available, more meaningful isotope comparisons will be possible.

4.5. **Tungsten transmutation.** Most recently Iwamura has reported transmutation of tungsten deposited on the surface of the reactor into osmium and platinum [12]. They looked for transmutation products having mass numbers in the range 182–196. Within this range mass numbers 182, 183, 184, 186 correspond to stable W isotopes and mass numbers 192, 194, 195, 196 correspond to Pt isotopes known to be significant impurities in the target W. In consequence only mass numbers 185, 187, 188, 189, 190, 191, 193 are empty

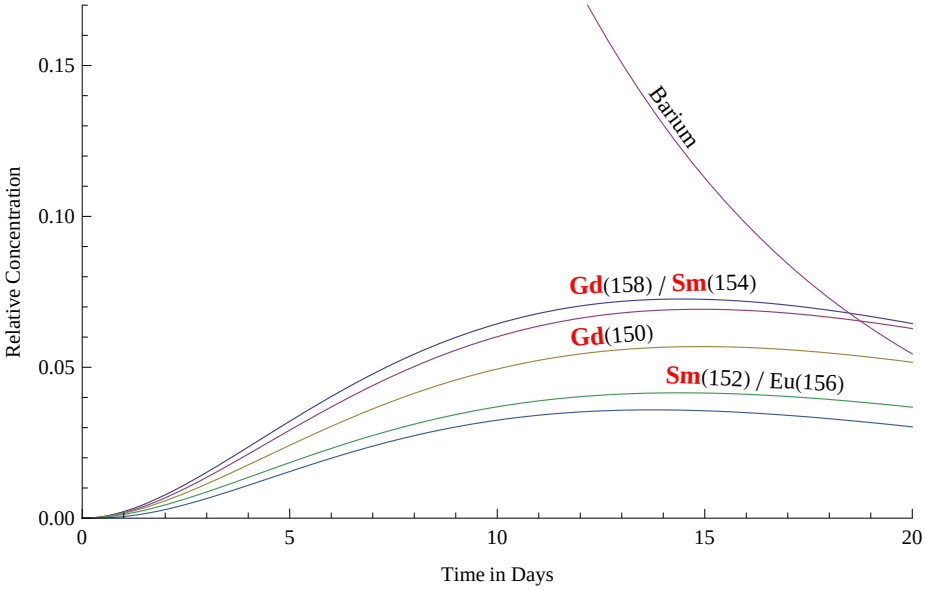

FIGURE 6. **Barium** (second generation). Theoretical concentration curves are shown for second generation transmutation product isotopes. Iwamura [11] detected XPS data for gadolinium and samarium products as indicated with bold red print. First generation products are shown in Figure 5.

and available prior to initiation of nuclear reaction. After nuclear reaction Iwamura found that masses 185, 187, 188, 190 had been produced. The mass-190 signal was tentatively identified as ^{190}Pt or ^{190}Os. No comment was made concerning the mass 185, 187, 188 signals.

We cannot immediately apply our theory to tungsten transmutations because the required mass excess data for neutron-rich tungsten isotopes have not been measured beyond mass number 190 [16]. However it may be that reliable extrapolation from measured data to more massive isotopes is possible. To check this possibility we drew on the model and extrapolation of the table of isotopes created by W. J. M. F. Collis [17]. Adopting the Collis mass excesses for tungsten isotope mass numbers 191–200 we calculated the first-generation transmutation products of tungsten to be ^{192}Os, ^{194}Os, ^{196}Pt, ^{198}Pt. These all were masked by pre-existing Pt impurities in the W target and were not identifiable. We interpret the signals at mass numbers 185, 187, and 188 to be signals for molecular SIMS products ^{183}WD, ^{183}WDD, ^{186}WD, all expected in the presence of deuterium D in an active reactor.

Hence overall we find that the tungsten experiment does not provide an opportunity for comparison of theory and observation. Yet we see that in similar experiments with other targets it may be possible in this way to extend our knowledge of physical properties to previously inaccessible neutron-rich isotopes.

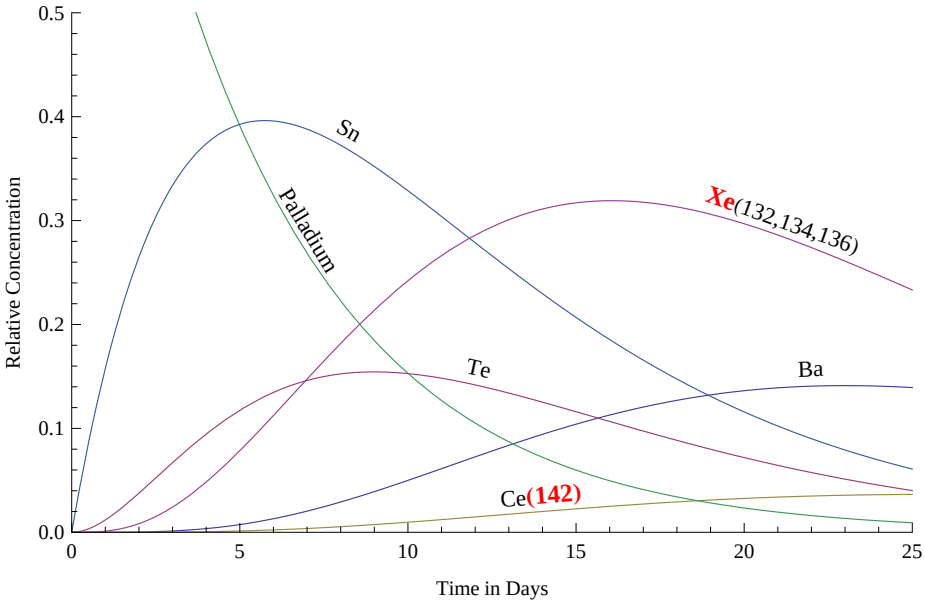

FIGURE 7. **Palladium**. Theoretical concentration curves are shown for palladium and for selected transmutation product isotopes. The ^{132}Te decays to ^{132}Xe with a half life of 3.2 days. Without comment Iwamura published XPS data for xenon [7, 11] and SIMS data for mass 142 [9] as indicated with bold red print.

4.6. **Carbon and lithium transmutations.** Theory predicts transmutation of carbon to ^{14}C and no further, in the reactions

$$^{A}\mathrm{n} + {}^{12}\mathrm{C} \longrightarrow {}^{A-2}\mathrm{n} + {}^{14}\mathrm{C} + (2\,\theta - 3.02)\,\mathrm{MeV}$$
$$^{A}\mathrm{n} + {}^{13}\mathrm{C} \longrightarrow {}^{A-1}\mathrm{n} + {}^{14}\mathrm{C} + (\theta - 0.105)\,\mathrm{MeV}.$$

The product isotope ^{14}C is inert with respect to further transmutation. With the constraint $2.95 < \theta < 3.82$ the energy release lies in the range 2.88–4.62 MeV. Iwamura investigated the transmutation of carbon in two experiments [6]. The carbon was deposited on the surface of the CaO/Pd complex by adsorption of hydrocarbon contaminants of the deuterium gas. In one experiment the concentration of C had fallen to zero after 1.8 days of reaction, whereas in the other the concentration of C had fallen incrementally after 1.0 days, 3.2 days, and 4.8 days, with about 10% still remaining. The quantities of transmutation products (Mg, Si, S) were not proportional to the quantities of C that were lost. Being weakly bound to the surface, it is probable that carbon nuclei were dislodged by the energy released during their transmutation or by that of nearby constituents of the CaO/Pd complex. We believe that the products attributed to carbon can be understood as transmutation products of the constituents of the CaO/Pd complex, as we discuss below.

Theory also predicts transmutation of lithium to ^{7}Li and no further, in the reaction

$$^{A}\mathrm{n} + {}^{6}\mathrm{Li} \longrightarrow {}^{A-1}\mathrm{n} + {}^{7}\mathrm{Li} + (\theta - 0.82)\,\mathrm{MeV}.$$

The product isotope ^{7}Li is inert with respect to further transmutation. The energy release lies in the range 2.13–3.00 MeV. Iwamura investigated the transmutation of lithium in two experiments [6]. The CaO/Pd complex was electrolytically doped with lithium from a LiOD solution. Product elements were observed after several reaction times. Although these products were attributed to transmutations of lithium, no measurements of initial lithium concentration were provided, nor were any provided for lithium concentrations after the various reaction times. We believe that the products attributed to lithium also can be understood as transmutation products of the constituents of the CaO/Pd complex.

4.7. **Palladium transmutation.** We now consider transmutations of the components of the CaO/Pd complex, beginning with palladium. Figure 7 shows the theoretical transmutations of palladium in a deuterium gas reactor. For these transmutations we used the Collis tabulation to extend the measured mass excess values for palladium isotopes to include mass numbers 126, 128, 130 with mass excesses 52.78, 46.19, 31.97 MeV. Although palladium is the element having highest concentration in the Iwamura transmutation research, no transmutation product was attributed to it. However, Iwamura has published a wide-range XPS spectrum for transmutation of barium deposited on a CaO/Pd complex [11], and we have examined it for possible evidence of palladium transmutation products. (For this purpose we had the advantage of a full-page print of Fig. 4a of [11] made available to conference attendees.) Two of the three largest signals (aside from those for palladium) are a pair at 1088eV and 1113eV identified by Iwamura as $Sm3d_{5/2}$ and $Sm3d_{3/2}$, demonstrating the presence of samarium as a transmutation product of barium. The third large signal lies at 1157eV. Although it is the largest of the three, Iwamura does not mention it in their analysis. We suggest that it may be the Xe3s signal for xenon. We note also that the wide-range spectrum has a significant signal at 674eV corresponding to the xenon signal $Xe3d_{3/2}$. In another XPS experiment [7] Figure 2c shows a signal at 940eV corresponding to $Xe3p_{3/2}$. In a SIMS experiment [9] Figure 21 shows an isotope mass number 142 not present before transmutation. We attribute these signals to transmutation of palladium to xenon and to ^{142}Ce, and conclude that theory and experiment are in agreement for transmutation of palladium.

4.8. **Oxygen transmutation.** The theoretical time dependence of the chain of transmutation reactions of oxygen is shown in Figure 8. It illustrates that the transmutation process can continue for many generations before the transmutations of reactor components and the accumulation of transmutation products poison and terminate the chain reaction.

The expected elemental products include major products Ne, Mg, Si, S, Ar, Ca and minor products Na, Al, P. Because Ne and Ar are light rare gases, it is likely that they would diffuse out of the CaO/Pd complex and would not accumulate to detectable levels. Calcium is a component of the complex and cannot be considered as a transmutation product. Concentrations of the minor products Na, Al, P are too small for reliable XPS detection. (Although phosphorus could not be claimed by XPS evidence, more precise SIMS measurements [6] showed a high concentration of mass-33, an order of magnitude greater than would be expected for the natural ratio ^{33}S/^{32}S, providing evidence for ^{33}P being present and decaying to ^{33}S as predicted by theory.) Hence in oxygen XPS experiments Mg, Si, S and P comprise the list of transmutation products expected.

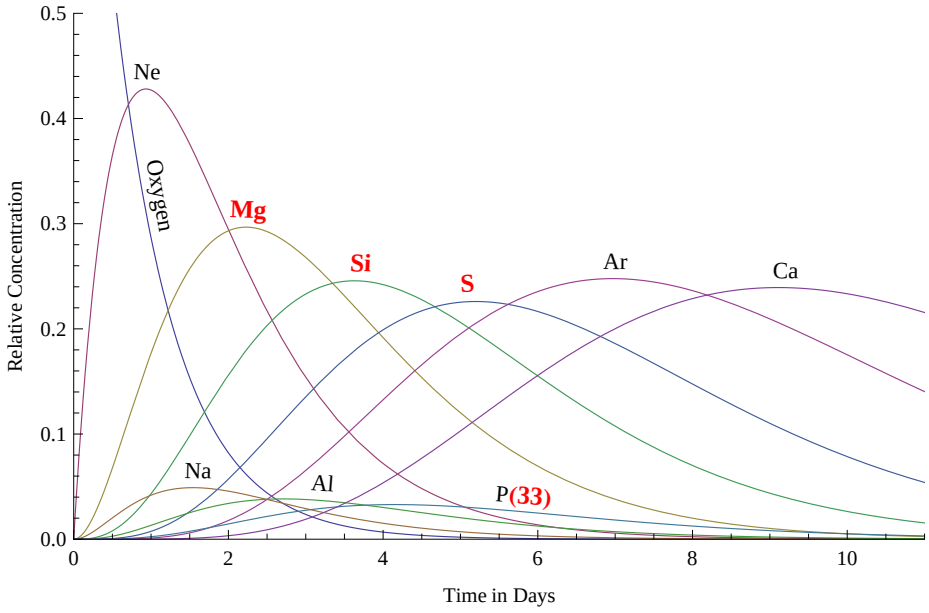

FIGURE 8. **Oxygen**. Theoretical concentration curves are shown for oxygen and for six generations of elemental transmutations products. Iwamura [6] observed XPS signals for elements magnesium, silicon, and sulfur (indicated with bold red print) and a small mass-33 SIMS signal was observed that we attribute to phosphorus.

In an experiment using *in-situ* XPS Iwamura observed the time-dependent concentrations attributed to Mg, Si and S [6]. In each of four runs the concentration of Mg rose more rapidly than the concentration of Si and reached a maximum in about 2 days. The concentration then declined to below the concentration of Si which continued to rise to near 5 days, in agreement with the theoretical concentrations in Figure 8. Data for S was provided in two of the four runs, in which the concentration of Mg declined to below the concentration of S which continued to rise to near 5 days, also in agreement with the theoretical concentrations in Figure 8. However the experimental S concentration was larger and rose more rapidly than the experimental Si concentration, in disagreement with theory. We considered the possibility that the XPS signal for S could have been enhanced by coincidence with another element of the CaO/Pd complex that shared the same XPS signal frequency. In Figure 7 we note that Te is a first generation transmutation product of Pd. The XPS signal Te4s(174eV) for Te nearly coincides with the S2p(168-170eV) signal for S. Because of their finite widths the S and Te signals overlap and merge. We suggest this happened in the experimental runs under consideration. The combined S + Te signal that was attributed to S became larger than the Si signal, thereby accounting for its more rapid rise.

Overall we conclude that there is good agreement between theory and experiment for the transmutation of oxygen to Mg, Si, S, and ^{33}P.

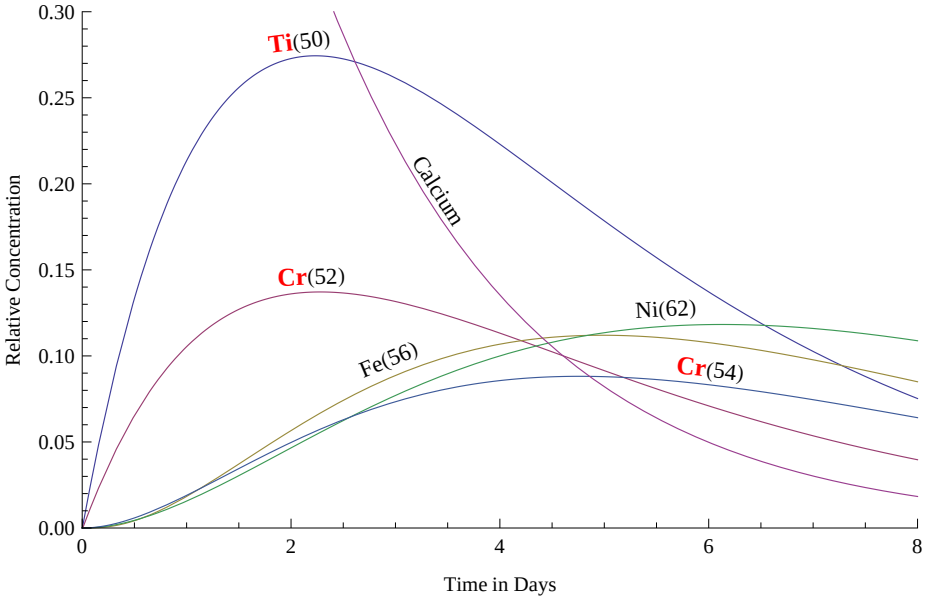

FIGURE 9. **Calcium**. Theoretical concentration curves are shown for calcium and for isotopes of transmutation products. Iwamura reported spectroscopic evidence for titanium [5] and for chromium [6] as indicated in bold red print.

4.9. **Calcium transmutation.** Calcium also is a component of the CaO/Pd complex. Figure 9 shows the theoretical transmutation of calcium, for which the first generation products are ^{50}Ti and ^{52}Cr and the second generation products are ^{54}Cr, ^{56}Fe and ^{62}Ni.

First we address the titanium product. In an electrolytic experiment [5] Iwamura reported a strong EDX signal for Ti in agreement with theory. After removing the CaO/Pd complex from the reactor, signals for Ti were confirmed by EDX, WDX, AES, and ICP-MS.

In their gas reactor *in situ* experiments [6] Iwamura reported XPS signals for F and Al in addition to the signals for Mg, Si, and S discussed above. Because neutron isotope theory does not predict either F or Al as a transmutation product of any component of the CaO/Pd complex, we explore the possibility of misidentifications for both signals. We suggest that Iwamura observed XPS signals near 700eV and near 78eV, and that they attributed one of them to the F signal F1s(694) and the other to the Al signal Al2p(77). We attribute them to the Cr signals Cr2s(702) and Cr3s(80). Recognizing that the experimental Cr concentration was counted twice, once by Cr2s and again by Cr3s, we take their average from the data in reference [6] as the best experimental measure for the time dependence of the concentration of Cr. The concentration of Cr then lies below that for Mg at 1–2 days and rises above it and above the Si signal to a maximum near 8 days, consistent with the theoretical curves in Figures 9 and 10 of Reference [6].

We conclude that overall there is good agreement between theory and experiment for the transmutation of Ca to both Ti and Cr.

4.10. **Deuterium transmutation.** Iwamura has shown that deuterium (^{2}H) is an essential reactant for sustaining the nuclear reaction in their reactor [7]. Two deuterium reactions are of particular interest. The first is transmutation of ^{2}H to ^{1}H by transfer of a neutron from ^{2}H to a neutron isotope as in equation (1),

$$^{A}\mathrm{n} + {}^{2}\mathrm{H} \longrightarrow {}^{A+1}\mathrm{n} + {}^{1}\mathrm{H} + (5.847 - \theta)\,\mathrm{MeV}. \tag{1a}$$

This is the essential reaction by which deuterium fuel supports neutron isotope growth and helps to sustain the chain reaction responsible for maintaining an ambient concentration of neutron isotopes. The second reaction is transmutation of ^{2}H to tritium (^{3}H) by transfer of a neutron from a neutron isotope to ^{2}H,

$$^{A}\mathrm{n} + {}^{2}\mathrm{H} \longrightarrow {}^{A-1}\mathrm{n} + {}^{3}\mathrm{H} + (\theta - 1.814)\,\mathrm{MeV}. \tag{16}$$

Although Iwamura did not report tritium as a reaction product many others have reported trace amounts [14]. In comparing the rates of competing reactions we have assumed that the most exothermic reaction occurs at the highest rate. Because the $^2\mathrm{H} \longrightarrow {}^1\mathrm{H}$ reaction is more rapid than the $^2\mathrm{H} \longrightarrow {}^3\mathrm{H}$ reaction we deduce that it releases the greater energy: $(5.847 - \theta) > (\theta - 1.814)$, or equivalently $\theta < 3.8305$. Because a trace of ^{3}H is released we deduce that the value of θ must approach 3.8305, and take $\theta < 3.82$ as our best estimate. Hence the full experimental constraints on θ are $2.95 < \theta < 3.82$ as we have assumed for our analysis.

4.11. **Fission.** It may be possible that neutron isotopes can facilitate fission of target elements. In an electrolytic reactor Iwamura found evidence for ^{57}Fe and copper as transmutation products for one of the elements oxygen, calcium, and palladium present in the CaO/Pd complex [5]. In a later gas reactor experiment Iwamura confirmed the ^{57}Fe observation [6]. However no neutron transfer reaction involving elements in the CaO/Pd complex leads to these products. Theory must address the situation, and to this purpose we consider extension of the basic neutron transfer reaction (3) to include fission of the target element accompanied and assisted by neutron transfer to the fission products.

In the absence of neutron transfer the fission of ^{102}Pd into equal fragments is

$$^{102}\mathrm{Pd} \longrightarrow {}^{51}\mathrm{V} + {}^{51}\mathrm{V} + 16.5\,\mathrm{MeV}. \tag{17}$$

Although this reaction is exothermic its rate of occurrence is negligible. When neutron transfer is allowed the most exothermic fission of ^{102}Pd into equal fragments is

$$^{A}\mathrm{n} + {}^{102}\mathrm{Pd} \longrightarrow {}^{A-12}\mathrm{n} + {}^{57}\mathrm{V} + {}^{57}\mathrm{V} + (12\,\theta + 0.47)\,\mathrm{MeV}, \tag{18}$$

which has energy release (35.9–46.3) MeV, two to three times greater. The ^{57}V transmutation products undergo beta decays to ^{57}Fe. Other ^{102}Pd assisted fission reactions include

$$^{A}\mathrm{n} + {}^{102}\mathrm{Pd} \longrightarrow {}^{A-16}\mathrm{n} + {}^{65}\mathrm{Mn} + {}^{53}\mathrm{Sc} + (16\,\theta - 9.46)\,\mathrm{MeV}, \tag{19}$$

which has energy release (37.7 to 51.7) MeV depending on the Pd isotope mass number. The product ^{65}Mn beta decays to ^{65}Cu and the product ^{53}Sc beta decays to ^{53}Cr. Assisted fission reactions for other Pd isotopes generate the same products ^{57}Fe, ^{65}Cu, ^{53}Cr but release less energy.

It may be that assisted fission reactions can account for the ^{57}Fe and Cu products observed by Iwamura. Unfortunately a proper analysis requires more than our present knowledge

of the associated nuclear reactions. We conclude that assisted fission is a promising but speculative possibility.

5. Research opportunities

We proceed now to discuss additional experiments that may have potential interest or value. For this purpose we explored a few additional transmutations that have potential to further test the applicability of the theory.

The transmutations so far reviewed have all resulted from the transfer of neutrons from neutron isotopes to target elements. In the general reaction (3) no restriction was placed on the parity of the product ^{B+C}Z relative to that of the target ^{B}Z. Yet every transmutation so far considered conserves parity. We seek an experiment in which the question of parity conservation may be tested. Consider transmutation of beryllium, which has the single stable isotope ^{9}Be,

$$^{A}n + {}^{9}Be \longrightarrow {}^{A+1}n + {}^{4}He + {}^{4}He + 6.5\,MeV. \tag{20}$$

This reaction does not conserve parity, which is negative for ^{9}Be and positive for ^{4}He. (Because the neutron parity is positive, we assume the parity of ^{A}n is positive for all A.) Hence if the beryllium reaction were to occur we would know that parity need not be conserved, and beyond that we would know that beryllium is a second fuel in addition to deuterium for supporting a chain reaction. This would be a discovery with important scientific and practical consequences.

On the other hand it could turn out that parity must be conserved and the beryllium reaction cannot proceed. A negative result of the beryllium experiment would suggest that beryllium is inert with respect to neutron isotopes. This too would be an important discovery, providing an inert structural material for neutron isotope reactor construction.

Many radioactive isotopes are predicted to be found in transmutation reactions. Some of these may be of value as tracers in nuclear medicine. Among those transmutations studied by Iwamura we found several with half-lives of hours or days including ^{143}Ce(33h) $\longrightarrow$ ^{143}Pr(14d) $\longrightarrow$ ^{143}Nd(stable) and ^{132}Te(3.2d) $\longrightarrow$ ^{132}Xe(stable). Others have shorter or longer half lives. Many useful isotopes may be identifiable by a more thorough examination of transmutations throughout the periodic table.

Beta decay plays an essential role in the neutron isotope theory of transmutation. In some instances, including transmutation of cesium, theoretical half lives are of sufficient duration that significant decay can continue after an exposed target has been removed from the reactor (see Figure 4). This possibility can be checked by examining an exposed target for radioactivity. If found, radioactivity would provide strong evidence in support of the neutron isotope theory. If not found its absence would suggest a serious flaw in the theory. The radiation following beta decay includes gamma rays whose energies uniquely identify the isotopes (both charge and mass) from which they originated. If this could be demonstrated for cesium, it would suggest that in situ gamma ray detection could reveal the full transmutation process for elements throughout the periodic table. For these reasons we believe that a search for radioactivity of cesium transmutation products would provide critical tests of the theory's validity and power.

It may be possible that some dangerous radioactive wastes can be economically transmuted to stable or other less dangerous products. Neutron isotope theory has capability to guide and quantify the search for economic means of waste remediation.

For heavy target nuclei the initial transfer reaction can result in a neutron-rich isotope of the target element that lies beyond the limits of what currently is known. This is the case for palladium and tungsten as discussed above, and may be the case for many other nuclei. It may be possible to quantify the properties of these currently unknown neutron-rich isotopes through measurements of their lifetimes and decay products, in a generalization and broadening of our analysis of palladium and tungsten transmutation products. In this way neutron isotope theory may open a new window for extending our knowledge of the properties of neutron-rich isotopes.

6. Summary

We have explored the possibility that nuclear transmutations observed by the Iwamura team can be understood as transfers of neutrons from neutron isotopes to target elements, followed by beta decay of the neutron-enriched target elements. By matching theory with observation for strontium and deuterium targets, it was possible to deduce key neutron isotope parameters that fit the transmutations of these targets, and that fit also the transmutations of cesium, barium, oxygen, calcium, tungsten, and palladium targets. Opportunities for future research have been suggested.

Acknowledgement

All of the experimental transmutation data that underlie and support our analysis were obtained by the efforts of the Iwamura team, whose members include Yasuhiro Iwamura, Takehiko Itoh, Mitsuru Sakano, Ichiro Toyoda, Nobuaki Gotoh, Satoshi Sakai, Shizuma Kuribayashi, Noriko Yamazaki, Yasuko Terada, Tetsuya Ishikawa, Jirohta Kasagi, H. Yonemura, K. Fukutani, D. Sekiba, N. Watari, K. Muta and S. Tsuruga. Members of this team have contributed to the various transmutation experiments as indicated in the list of references. We view the results they have obtained as the most accurate and comprehensive body of data extant against which to test and refine theory. We thank the team for their work, which has been of great value to us. And we hope that neutron isotope theory will prove useful to the team in their future work.

We also thank Mitsubishi Heavy Industries for sponsoring the Iwamura team, and for encouraging them to focus on fundamental research to elucidate the scope and promise of this new branch of nuclear science.

Appendix A. The system of ODEs

Let $f_i(t)$ denote the relative concentration of isotope i at time t. Let the initial amounts of the isotopes be given by $f_i(0) = \alpha_i$, where $\alpha_i \geq 0$ and $\sum_i \alpha_i = 1$. Let λ_i denote the inverse of the mean lifetime of isotope i associated with ordinary decay modes. (Let τ, λ, and $t_{1/2}$ denote the mean lifetime, decay rate, and half life respectively. Then $\tau = 1/\lambda$ and $t_{1/2} = \log(2)/\lambda$. Note $\tau > t_{1/2}$.) For ordinary decay modes, let the branching ratios from isotope i to isotope j be given by $\beta_{ij} \geq 0$. For unstable isotopes $\sum_{j \neq i} \beta_{ij} = 1$. For

stable isotopes $\beta_{ij} = 0$ for all $j \neq i$. For convenience let $\beta_{ii} = -\sum_{j \neq i} \beta_{ij}$. Thus for stable isotopes, $\beta_{ii} = 0$ and for unstable isotopes $\beta_{ii} = -1$.

Similarly, for polyneutron interactions, let γ_{ij} denote the branching ratios for isotope i into other isotopes (via the gain or loss of neutrons) and again let $\gamma_{ii} = -\sum_{j \neq i} \gamma_{ij}$. In addition, let $\mu_i = 1/t_m^i$, where t_m^i is the survival mean lifetime for isotope i (associated with the transfer of neutrons between polyneutrons and isotope i).[1]

Given this setup, the isotope concentrations satisfy a system of first-order homogeneous linear ordinary differential equations (ODEs) with time-dependent coefficients composed of equations of the following form:

$$f_j'(t) = \sum_i \beta_{ij}\, \lambda_i\, f_i(t) + 1_{\text{on}}(t) \sum_i \gamma_{ij}\, \mu_i\, f_i(t), \tag{21}$$

where

$$1_{\text{on}}(t) = \begin{cases} 1 & \text{if the reactor is } \textit{on} \text{ at time } t \\ 0 & \text{if the reactor is } \textit{off} \text{ at time } t \end{cases}. \tag{22}$$

The system can be written compactly as[2]

$$f'(t) = A(t)^\top f(t) \qquad \text{subject to } f(0) = \alpha \tag{23}$$

where $A(t)$ is a square matrix with

$$A_{ij}(t) = \beta_{ij}\, \lambda_i + 1_{\text{on}}(t)\, \gamma_{ij}\, \mu_i. \tag{24}$$

Appendix B. *Mathematica*

We use *Mathematica*'s `IsotopeData` function to obtain information about isotopes and decay modes.[3] In particular, `IsotopeData[`*`element`*`]` returns a list of known isotopes associated with the given element. `IsotopeData[`*`isotope, property`*`]` returns the property for the given isotope and `IsotopeData[`*`element, property`*`]` returns the list of the properties for the included isotopes. The element can be given as a string such as `"Cesium"` or by the element's atomic number such as `55`. An isotope can be given as a string such as `"Cesium133"` or by the pair of atomic number and mass number such as {`55, 133`}.

We use the properties

- `"MassExcess"`
- `"MassNumber"`
- `"AtomicNumber"`
- `"Parity"`
- `"Spin"`
- `"IsotopeAbundance"`
- `"HalfLife"`
- `"DaughterNuclides"`

[1] The branching ratios are determined according to the theory in Section 3. In addition, $\mu_i = (0.05\, A_i)^{-1}$, where A_i is the mass number of isotope i. This isotope data are acquired via *Mathematica* as described in Appendix B.

[2] "$\top$" denotes transpose.

[3] As of December 2012, the data returned by `IsotopeData` was taken from the 2005 Nuclear Wallet Cards [16].

- `"DecayModes"`, where decay modes are (currently) restricted to
  `"BetaDecay"`, `"ElectronCapture"`, `"AlphaEmission"`, `"BetaPlusDecay"`.

We also use *Mathematica* to solve the system of ordinary differential equations (ODEs) numerically. The main function is `NDSolve`.

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