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Pd-110/Pd108 Ratios and Trace Element Changes in Particulate Palladium Exposed to Deuterium Gas

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ABSTRACT

Changes in Pd-110/Pd-108 ratios as well as the concentration of silver, gold, zinc, cobalt, iridium and lithium-7/6 ratios have been measured using neutron activation analysis (NAA) and Time of Flight Secondary Ion Mass Spectrometry (TOF-SIMS) on a set of four samples of particulate palladium exposed to high-pressure deuterium gas in the hollow core of Arata-Zhang cathodes. Three samples were from cathodes producing excess heat (10's of megajoules) over a period of several-months electrolysis, while the fourth was virgin powder from the same batch as that of the active samples. If a nuclear process is the source of these changes, then multi-isotope elements such as silver, zinc, and iridium should show significant deviations in their isotopic ratios from the natural terrestrial values. Surface trace lithium did indeed show such differences from that of the virgin material. The Ag-109/107 ratio is currently under study by accelerator mass spectroscopy (AMS) for the one sample showing the greatest difference in Ag-109 content from that of the virgin material. Since these variations may have explanations unrelated to nuclear reactions, these results are not yet definitive. The 8% increase in the Pd-110/108 ratio for one of the four samples relative to the virgin material is one of the most difficult for which to find a conventional explanation.

INTRODUCTION

This is an attempt to collect the results of some four years effort to find evidence of nuclear reaction products (ashes) in samples from experiments in which excess heat production has been reported (1-4). Focus on material exposed only to gaseous deuterium has the advantage that conventional electrochemical cathodic deposition from electrolyte impurities or extracts from anodes can be eliminated as a possible cause of observed changes. However, the closure process of the hollow center of the cylindrical cathodes, requires electron beam welding in which a part of the palladium annulus is melted at above 1550 degrees C. Thus lower melting impurities in the palladium shell could be volatilized and thereby reach the enclosed palladium powder. This path of possible contamination seems remote and in any case would preserve isotopic ratios in multi-isotope elements such as silver, zinc, and lithium. It would have no effect on the Pd-110/Pd-108 ratio of the three plus grams of contained powder.

RESULTS

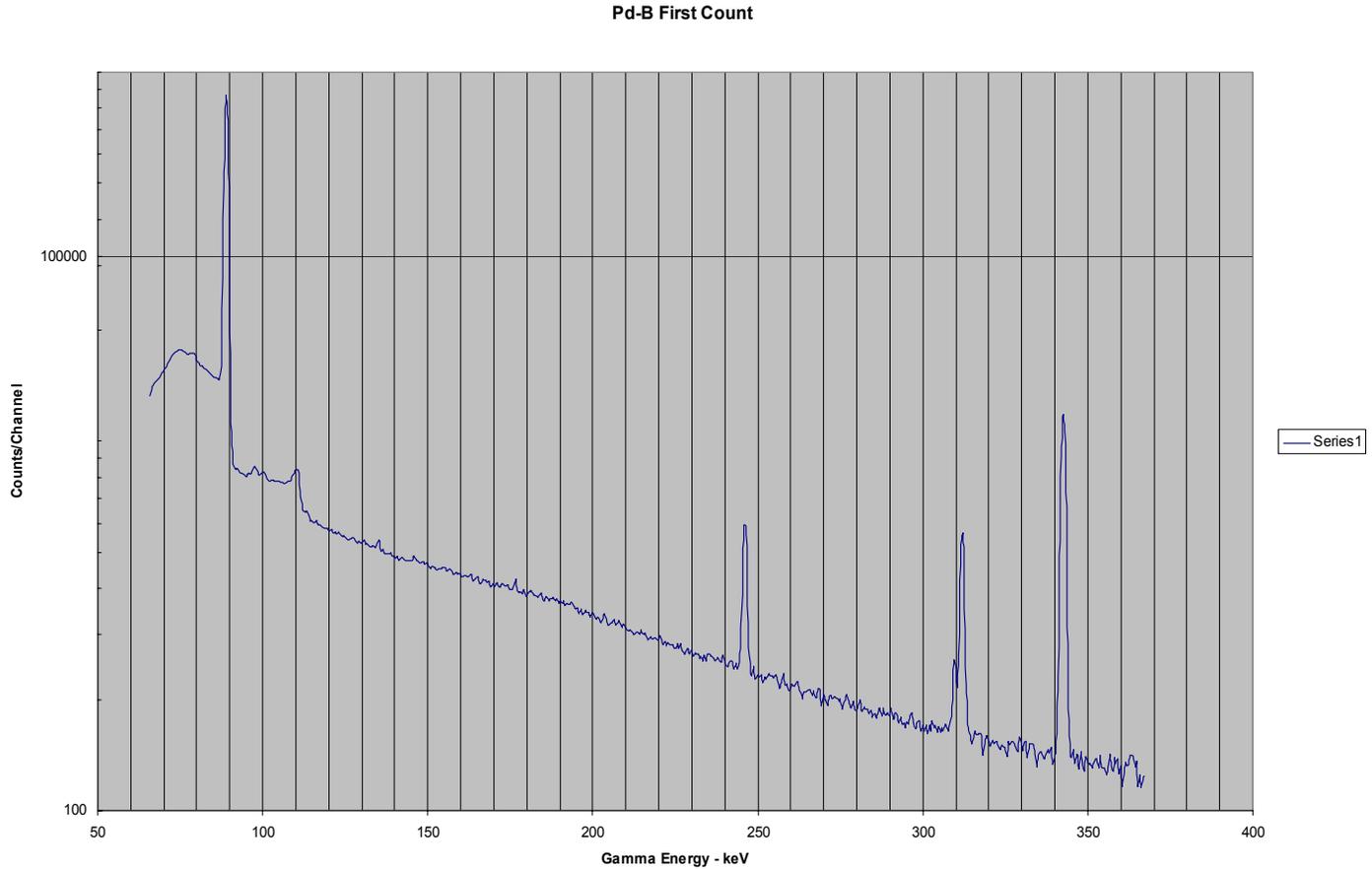
Table 1 summarizes the results obtained so far. In addition to the measured ratios of atoms the changes in the ratios from those of the virgin sample (Pd-D) are given as A/D, B/D, and C/D respectively. As an example of the original

NAA data upon which the Pd-110/Pd-108 ratio is based, Figure 1 gives the portion of the gamma ray spectrum of Pd-B most important to that determination. The two gamma rays giving the statistically most accurate ratio of 13.47 hour Pd-109 and 7.45 day Ag-111, products of neutron capture in Pd-108 and Pd-110, respectively, are seen as the prominent peaks at 88 and 342 keV. Other gamma ray peaks from these two radioactive isotopes were also employed to confirm the results of the 88/342 intensity ratio, which they did but with slightly less statistical accuracy.

TABLE 1. Summary of Results

| Sample Name | Pd110/Pd108 Atomic Ratio (One Sigma) | Ag109/Pd110 Atomic Ratio (X 10 ⁶) | Co59/Pd110 Atomic Ratio (X 10 ⁶) | Zn64/Pd110 Atomic Ratio (X10 ⁶) | Au197/Pd110 Atomic Ratio (X10 ⁶) | Ir191/Pd110 Atomic Ratio (X10 ⁶) | Li7/Li6 Atomic Ratio (One Sigma) |
|---------------|--------------------------------------|---|--|---|--|--|----------------------------------|
| Pd-D (Virgin) | Exactly 1 By Definition | 53 | 227.5 | 83 | 19.4 | 2.26 | 13.6(1.0) Nat. Li=12.5 |
| Pd-A (Active) | 1.037(.008) | 663 A/D=12.5 | 286 A/D=1.26 | 516 A/D=6.22 | 89.4 A/D=4.61 | 17.8 A/D=7.88 | 14.5(0.3) A/D=1.07(.08) |
| Pd-B (Active) | 1.089(.008) | 152 B/D=2.87 | 377.5 B/D=1.66 | 1259 B/D=15.2 | 20.6 B/D=1.06 | 0.84 B/D=0.37 | 22(1.4) B/D=1.62(.16) |
| Pd-C (Active) | 1.014(.009) | 488 C/D=9.21 | 730 C/D=3.21 | 608 C/D=7.33 | 24.1 C/D=1.24 | 1.5 C/D=0.66 | 16.2(0.1) C/D=1.19(.09) |

FIGURE 1. Partial Gamma Spectrum of Pd-B from File psn 2021



DISCUSSION

The most definitive of these data are those involving changes in isotopic ratios since these are not subject to suspicion of contamination. A previous paper discussed the uncertainties in the TOF-SIMS measurement of lithium 7/6 ratios (5). Uncertainties in NAA measurement of the Pd-110/Pd-108 ratio have not been previously addressed and hence will be dealt with here.

The key measurement is determining accurately the integral number of counts under the photoelectric peak of two gamma rays, one for each of the two radioactive isotopes produced by neutron capture in the target isotope whose abundance is in question. If the irradiation was under identical conditions of neutron flux for the same irradiation time, then the only variable requiring correction to the ratio of integral photopeaks is the time of decay since the end of irradiation. Fortunately, accurate half-lives have been previously determined for the two isotopes of interest here, namely Pd-109 (13.427 hours) and Ag-111 (7.45 days). The time of irradiation was only 80 minutes, a time short with respect to both the product isotope half lives, so that no significant correction need be made for differences from decay during irradiation. All samples were simultaneously irradiated, and since we are comparing RATIOS relative to the virgin sample, Pd-D, those small differences are of no consequence.

However, these small powder samples of some 5 to 15 milligrams each were packaged for irradiation in small polystyrene plastic containers and gamma counting was performed with the sample within these containers. Hence

small differences in geometry between samples could occur relative to the germanium detection crystal. However, unless these small geometric differences could influence the RATIO of the two gamma photopeaks, they would have no significant adverse consequences for this study. All the irradiation and counting was performed at the University of Texas, Austin, research reactor. The test performed to check such effects was the measurement of the ratio of two gamma rays from the SAME isotope which should remain constant through all possible permutations of geometric differences. Checks of such ratios (for example the 88 and 647 keV gammas of Pd-109 and the 245 and 342 keV gamma rays of Ag-111) were constant within a fractional standard deviation of 1.8%.

Probably the most significant uncertainty was the proper integration of each photopeak and the proper subtraction of the continuum background under those peaks. Such integration was performed manually by summing the channel counts to include all channels showing counts above the obvious continuum on both sides of the peak plus a few of the constant slope continuum on each side and subtracting the average count of some 4 or 5 channels of the continuum on each side throughout the integrated set of channels that included the peak and the two side continua. This method was clearly most satisfactory for the tallest peaks relative to the continuum and is the reason the best data on Pd-110/Pd-108 was obtained for the 88 keV gamma of Pd-109 and the 342 keV gamma of Ag-111.

Having focused on the isotopic ratio of Pd-110/Pd-108, consider how the sample Pd-B, showing the largest difference in that ratio from that of the virgin material Pd-D, (around 8%), correlates with other observations on the same sample (always relative to the Pd-D results). From Table 1 we observe that Pd-B exhibits the following pattern relative to virgin Pd-D: 1) 15.2 times the Zn-64 content; 2) 1.62 times the Li-7/Li-6 ratio; and 3) 0.37 times the Ir-191 content. These three factors are the extremes among the three samples measured. At the lower extreme among the three active samples, Pd-B shows the least change in the Au-197 content, (1.06) and the Ag-109 content, (2.87). In the intermediate range of alteration from the virgin material, the Pd-B content of Co-59 is 1.66 times that of Pd-D.

If one assumes all these changes are due to nuclear reactions, then fission of Pd-108 to Zn-64 and Co-59 would top the list. The Li-7/Li-6 ratio change is assumed unrelated to Pd-110/Pd-108 ratio changes but might be related to change by mechanisms suggested by Iwamura et.al.(6). A moderate amount of deuteron –/ proton-induced transformation of Pd-108 to Ag-109 may be present. It is interesting that the next largest deviation of Pd-110/Pd-108 from that of Pd-D, that of Pd-A (3.7%), is associated with the largest increase in Ag-109 (12.5), Au-197 (4.61) and Ir-191 (7.88), but a minimal change in the Li-7/Li-6 ratio (1.07). If nuclear processes must be called upon to explain such data, then a bewildering array of reactions with the palladium host material as well as with the significant elemental impurities must be taking place. For example, two samples show a 3-fold and 1.5-fold reduction in the Ir-191 content, respectively, versus the virgin material and one shows over a 7-fold increase! If these several elemental impurities are taking an active role in nuclear processes, then the lack of expected reaction products from D+D fusion is made more plausible.

Lipson, et. al. (7), describe an experiment in which the neutron capture cross sections of palladium isotopes in metallic foils are enhanced by a factor of 8 when the metal is in a plastically deformed state. They also show evidence for surface layers of the highly strained lattice exhibiting separation of palladium isotopes. It is possible that the palladium powders studied here could be in a plastically deformed state. If so then the present results could be explained by variations in the degree of plastic deformation of each sample, an uncontrolled variable. If the degree of strain of a palladium sample affects the neutron capture cross section, then NAA is no longer a reliable tool to detect possible nuclear reactions in the palladium-deuterium system. Also, near-surface strain-induced palladium isotope separations would vitiate any nuclear reaction interpretation of the present results, since the particle sizes (~40 nm diameter) are within the surface layer depth within which ratio changes among Pd isotopes were observed (6). However, no other strain-induced changes in neutron capture cross sections are known to this author for other metals.

CONCLUSIONS

It is hard to find conventional explanations for alterations in isotopic ratios among the parent palladium isotopes if the Lipson (7) observations are not operative here. An impressive test will be to find alterations in isotopic ratios of those possible nuclear reaction products, such as silver, which have more than one natural isotope. Some suggestive results among the past four years of this work, (8,9) have shown possible changes in the zinc isotopes as well as

more definitive results for surfaces of the powders in the Li-7/Li-6 ratios (5). Currently Hubler and associates at the Naval Research Laboratory are using accelerator mass spectrometry to determine the Ag-109/Ag-107 ratios in two of the samples described in Table 1.(10)

NAA has limited sensitivity to many elements but extreme sensitivity to others. Hence NAA gives a distorted view of the impurity content of these materials. For example the more important impurities may be such elements as beryllium and boron, which with their lower atomic numbers, have less formidable coulomb barriers than heavier elements like palladium, silver, zinc, gold, iridium, and cobalt.

Probably the greatest revelation in this work is the possibility that trace elements may be significant participants in nuclear reactions in solids such as Pd so that focusing entirely on D+D fusion is not necessarily the only path forward in understanding these phenomena.

Of course it is possible that some of the variations in impurity levels is simply random differences within a non-homogeneous batch of powdered Pd. Iridium and gold may be in this category, since no easy explanation for their changes have be found.

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