

TRACE ELEMENTS ADDED TO PALLADIUM BY EXPOSURE TO GASEOUS DEUTERIUM

Thomas O. Passell* and Russell George**

* TOP Consulting **Saturna Corporation

Abstract

This is an experimental program to investigate possible trace element changes brought about in palladium (Pd) after extensive electrolysis in heavy water electrolytes as well as long time contact of particulate Pd with gaseous deuterium. Of particular interest are cathodes and particulate Pd which had experienced episodes of excess heat production beyond all electrical and other inputs. This paper details the careful analysis by neutron activation analysis (NAA) of a set of three samples of finely powdered Pd exposed to high deuterium pressures (hundreds of atmospheres) near room temperature at the core of hollow cylindrical Pd cathodes. A fourth sample of unused Pd powder from the same batch used in the cathodes was analyzed as a control. The most prominent change observed in the three active samples versus the virgin Pd was the Zn-64 content. The active samples showed an increase in the Zn-64 isotope of 6 to 14 times that in the virgin Pd. Speculation regarding the source of this increased zinc varies from contamination during electron beam welding (used to seal off the hollow core) to nuclear reactions generated by high pressure deuterium gas on the large surface area Pd particles in the core.

Introduction

Palladium (Pd) cathodes electrochemically charged with deuterium (D) have exhibited episodes of excess heat beyond all inputs. (1-5) To confirm or refute the suspicion of a possible nuclear reaction producing the excess heat, trace element changes were measured in particulate Pd exposed to very high deuterium pressures generated in the hollow core of cylindrical Pd cathodes in a series of experiments by Arata and Zhang.(1) Independently in a completely different type of experiment, Mo, Cai, Wang, Wang, and Li reported observations of increased zinc for Pd exposed to hydrogen and deuterium gas.(6) In addition they reported observing an altered ratio of Zn-64 to Zn-68 relative to that of naturally occurring zinc. They speculated that the extra zinc observed may have been produced by some nuclear process induced by the hydrogen or deuterium on the Pd metal surface, since it was unlikely that contamination by handling or experimental processes would have given zinc with an altered relative abundance of Zn-64 and Zn-68.

The present study attempted to check the hypothesis put forward by Mo et al (6) on other Pd exposed only to gaseous deuterium (with some mixture of light hydrogen inevitably present in deuterium).

Experimental

The samples of particulate Pd weighing from 5 to 16 milligrams each were sent to the University of Texas for NAA.(7). All elements susceptible to NAA at a flux of $1E12$ n/cm²-sec under an irradiation of 3 hours were detected by means of their emitted gamma rays as detected in an intrinsic germanium crystal operated at 77°K. Since NAA is isotope specific, certain changes in the isotopic ratios within palladium were also accessible by this method, for example the Pd-102/Pd-110 ratio. Analyses were mostly dependent upon the ratios of gamma ray integrated photopeaks within a given sample and hence were less subject to errors in sample weighing or comparisons between separate samples. Since not all the samples could be simultaneously gamma counted, small corrections had to be made for different times of decay after irradiation, as these different decay times affected the ratios of gammas from isotopes of different half lives.

Results

Table I shows the increases in the elements iridium (Ir) and gold (Au) in addition to the increase in zinc (Zn) relative to the virgin material (Sample D). In one instance, Sample B showed a DECREASE in Ir from the virgin material. The conventional explanation for such increases is the cathodic deposition of electrolyte impurities on the cathode surface. However, all these samples were protected from the electrolyte inside the gas-pressure-tight hollow core of the cylindrical cathode. The only remaining possibility of contamination would be in the process of cathode preparation. Once the Pd powder is placed inside the core of the cathode, it had to be electron-beam welded because the deuterium pressure could reach extremely high values during electrolysis. In fact, some cathodes failed in a burst mode during electrolysis due to this pressure if not properly welded. It is possible that the high temperatures experienced by the welded region could have vaporized various impurities in the Pd and the vapors carried down for deposition on the Pd powder. While this scenario is plausible for the relatively volatile zinc, it is less likely for gold and iridium. For example, zinc has a boiling point of 907°C, whereas iridium and gold have boiling points of >4800°C and 2600°C respectively.

The zinc concentrations observed in the powder are by far the highest of any observed impurity, ranging between 102 PPM to 248 PPM by weight assuming the zinc is natural zinc. Since we only measure the Zn-64 isotope via its neutron capture product isotope, Zn-65, the Zn-64 concentrations are about half the above numbers (Zn-64 is 48.6% of natural zinc).

The conditions of irradiation apparently obscured the appearance of the prominent gamma ray of the Zn-69m isotope from Zn-68 neutron capture, so the attempt to check whether the zinc observed had an unnatural ratio of Zn-64 to Zn-68 failed. The Zn-69m half-life is nearly the same as that of Pd-109 (both about 13.8 hours) and the gamma rays of the latter were so dominant that we failed to resolve the 438.5 keV gamma of Zn-69m. The above assumes that the Zn-68 isotope is actually present, even at some different ratio than that of natural zinc.

Discussion

The samples analyzed along with its virgin material were volunteered by Dr. Arata and Dr. Zhang of Osaka University. However, the precise amounts of excess heat produced by the cathodes in which the powdered Pd was contained has not yet been made available. Arata and Zhang's published work shows data from similar cathodes which produced about 30 to 40 megajoules of excess heat over the most active 2 month period of their electrolysis.(1) If one assumes that some nuclear process produced each excess Zn-64 atom at about 10 Mev per atom, and further that some 12 grams of powdered Pd was contained in each cathode hollow core, and further that our sample of 5 to 15 milligrams was a representative sample of the full 12 grams present, then one obtains an expected amount of excess heat of 20 Megajoules.

The only heat producing nuclear reaction capable of producing Zn-64 is the fission of palladium isotopes or proton capture in impurity copper. While some copper impurity is undoubtedly present, it is not readily measured by NAA under these circumstances and even if present, it is not likely to be so large as several hundred PPM. However, new short irradiations (to observe Cu-66 with its 5.1 minute half life) are planned to check for this possible copper impurity.

We are thus left with the possibility of Pd fission, a process giving about 20 to 30 Mev per fission (10-15 Mev per fission product atom). To check this possibility we attempted to measure any change in the ratio of Pd-102 and Pd-110 from that of the virgin material. Table I shows a roughly 20% increase in the Pd-110/Pd-102 ratio in the A and C samples but only about 5% in sample B, the latter not outside the limits of error. If the apparent depletion in Pd-102 relative to Pd-110 is confirmed, this may indicate that the lighter isotopes of Pd are more susceptible to fission than the heavier ones. Clearly these three experiments are only suggestive at present.

The proof of a nuclear source for the increased zinc (and possibly other multi-isotope elements), will be resolved by NAA or other methods that show the isotopic abundance ratios to be significantly different from those found naturally. Otherwise, a contamination source is indicated.

The small increases in iridium and gold in the active samples relative to the virgin sample find no ready explanation. Both elements are considerably less volatile than zinc and hence are less likely to be carried from the site of electron

beam welding to the powder. It may be we are seeing the natural variation of these impurity elements - that is - they are not uniformly distributed in the powdered Pd. In one case, (sample B in Table I) iridium shows a factor of 2 decrease rather than the factors of three and four increases observed for samples C and A respectively. Gold shows the smallest increase (10%) in sample B with increase factors of 1.7 and 5.3 for samples C and A respectively. No nuclear processes that have a positive Q value can be readily identified to suggest a nuclear reaction origin for these elemental impurity increases. However, a commonly observed impurity in palladium is platinum, from which both iridium and gold could be produced by positive Q nuclear reactions with protons or deuterons. However we observe at most only a few PPM of platinum in these powdered Pd samples.

Table I. Neutron Activation Analysis Results for Impurities in Palladium Exposed to Gaseous Deuterium

SAMPLE	Zn-64 Content PPM by Weight	Zn-64 Ratio Relative To Unused (Virgin) Palladium Sample D (One Sigma)	Iridium Content PPM by Weight	Gold Content PPM by Weight	Pd-110/Pd-102 Relative to the Ratio Observed in Unused Pd i.e., Sample D (One Sigma)
ARATA A	50	6.6 (1.6)	4.1	55	1.24 (0.11)
ARATA B	121	14.4 (3.2)	0.2	11	1.06 (0.06)
ARATA C	58	8.3 (2.1)	3.1	17	1.21 (0.11)
ARATA D (VIRGIN)	8	1.0 (DNA)	0.5	10	1.00 (DNA)

Conclusions

These measurements are so far only suggestive of possible nuclear reactions occurring in deuterated palladium. Some suggestive early results from gas loaded palladium was given by Mo, Cai, Wang, Wang, and Li (6). They found alterations in the Zn-64/Zn-68 ratios from gas-exposed samples relative to the same ratios in natural zinc. The most severe alteration showed an enhancement of the Zn-68 to Zn-64 ratio by 20% relative to the ratio for natural zinc. Their unexposed palladium showed no zinc present. Gas-exposed palladium is likely to be reacting at its surface and hence the products of such reactions should be concentrated at the surface. Experiments should be performed which separately measured surface material and material well below the surface. The study by Mo et al used thin wire samples rather than powder, in which such differentiation between surface and bulk would be more practical.

It is possible that conventional explanations will be found for these results, namely that of contamination during electron beam welding for gas tight closure of the hollow core. Experiments are currently under way for NAA of Pd powder that was exposed in the hollow core of a cathode producing NO excess heat under electrolysis with normal light water electrolysis may be persuasive in answering this question.

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