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A SEARCH FOR THE EMISSION OF X-RAYS FROM ELECTROLYTICALLY CHARGED PALLADIUM-DEUTERIUM

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Abstract

This paper reports on attempts to measure 21.1keV X-rays from palladium and a palladium-lithium alloy, electrolytically charged with deuterium, with the aim of verifying the recent claimed observations of cold fusion in various metal-hydrogen alloys. No X-rays were seen within the limits of sensitivity of our experiment. We can thus put an upper limit on the number of nuclear events in our sample of 10^5 $cm^{-3}s^{-1}$.

Introduction

The recent claimed observations of nuclear fusion[1][2] in various metal-hydrogen systems when charged with deuterium has generated much excitement in recent months. We, like many other research groups around the world, have attempted to verify these claims. The experiment reported here is one of an intensive series that was started soon after the first claims were published. Unlike most of the experiments reported to date, this experiment is not looking for the primary radiation products of the nuclear reaction, neutrons, protons, γ-rays and so on, but for the X-rays produced when high energy charged particles slow down in condensed matter. The X-rays are generated by the refilling of the K shell electron orbits ionized by the passage of the charged particle. Since most materials are quite transparent to their own X-rays, the detection is not particularly difficult. The 21.1 keV X-rays produced by palladium can also easily penetrate the electrolyte and cell wall between the palladium cathode and the detector.

The problem with looking for the primary radiation products from cold fusion is that it presupposes knowledge of which reaction is actually occuring. Whilst this work is at the verification stage a technique such as the present, which is sensitive to all nuclear events, is more useful. Any nuclear event depositing its energy in the lattice will in theory produce X-rays. There has been much discussion of the discrepancy between the small numbers of neutrons apparently being produced (typically 0.6 $s^{-1}cm^{-3}$)[2] and the large heat output observed in some of the calormetric experiments. The discrepancy is of the order of 10^9 fewer neutrons than would be expected. Several exotic aneutronic reactions have been proposed to account for this, including: so-called Mössbauer fusion– $d + d \rightarrow \alpha + 24$ MeV[3][4], where the excess energy is absorbed by the lattice; the $^6Li + d \rightarrow 2\alpha + 24$ MeV reaction[6], due to Li being incorporated into the lattice electrolytically along with the deuterium. Others have proposed mechanisms whereby the $d + d \rightarrow t + p$ channel dominates over the $d + d \rightarrow {}^3He + n$ channel, both are equally likely in normal d + d fusion[5]. If the reaction is largely aneutronic then the only practical way to measure the energy deposited in the lattice is by measuring the X-ray flux. The large heat generation, if nuclear in origin rather than being chemical or an artefact of the calibration of the calormetric experiment, would imply that the X-ray flux should be very large.

This short paper discusses preliminary results from two sets of experiments. One set was similiar to the arrangement used by Fleischmann and Pons; a palladium cathode in a Li/D_2O electrolyte. The other set was designed to see if the reaction was in fact $^6Li + d$ rather than the d + d reaction. Here a Li/Pd alloy cathode was used in an electrolyte enriched with ^6Li.

Experimental Details

The electrolytic cell used in these experiments was designed to bring the active cathode as close to the detector as is reasonably possible. The 21.1 keV X-rays produced by the Kα electrons in palladium are quite penetrating and so this was easily achieved without having to sacrifice all the electrochemical niceties. The cell design is shown in figure 1. The cathode is mounted on a screw-threaded stud so that it can be carefully positioned less than a millimetre from a thin Mylar window. The detector can then be placed on the other side of this window so that only a small amount of electrolyte and Mylar separate it from the possible source. The electrolyte was pumped past the electrodes using a peristaltic pump to prevent hydrogen bubbles trapped between the cathode and the window inhibiting the normal charging process.

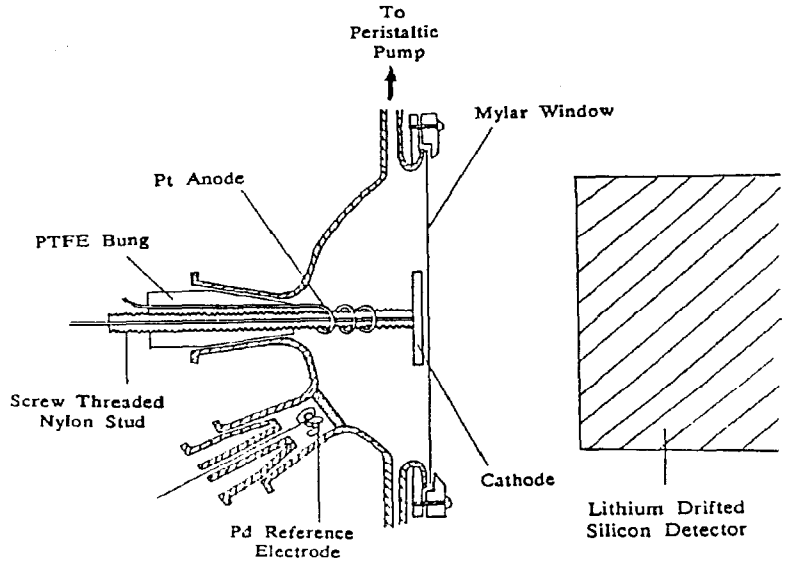

Figure 1: The thin walled electrolytic cell used to look for X-rays from palladium and palladium-lithium alloys when electrolytically charged with deuterium.

Since the effect may well be associated with the surface rather than the bulk, we forced the cathode to charge from its front face by painting the rear side with an inert resin (Lacomit). The anode used was a platinum wire mounted to the rear of the cathode. A palladium wire was used as a reference electrode with the aim of measuring the change in cell voltage as the palladium charged.

As has already been explained, two types of experiment were attempted. The Fleischmann/Pons style experiments used a pure palladium cathode in a 0.1M solution of Li dissolved in 99.8% purity D_2O. The second set used a Li/Pd alloy for the cathode in a 0.1M solution of ^6LiOH in D_2O. The alloy was made by rolling a sandwich of the two metals (with lithium as the meat), and heating to 800°C to allow the lithium to diffuse into the palladium. X-ray diffraction later showed the material to be single phase. Both of the cathodes were then rolled into discs about 1.5mm thick and 20mm in diameter. Before electrolysis began the discs where heated in a vacuum to remove any adsorbed gases and were then vacuum annealed at 600°C overnight. Lastly the surfaces were prepared by etching in nitric acid just prior to electrolysis. The detector used was a lithium-drifted silicon detector which had been calibrated using various known X-ray sources.

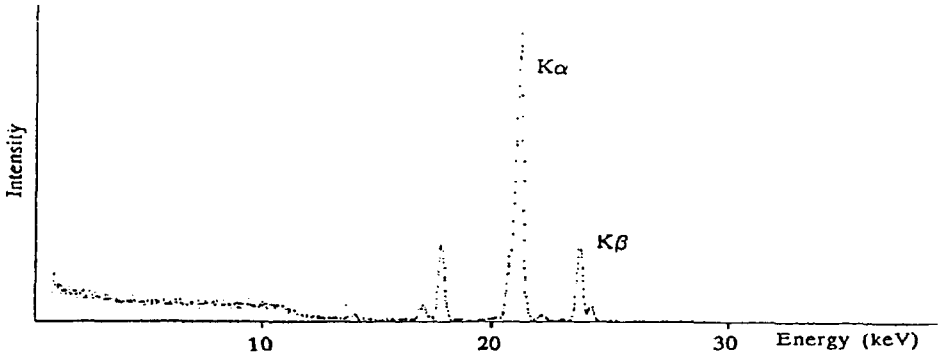

Figure 2: A calibration run using a 150nCi α source fixed to the back of a palladium foil. The Kα and Kβ lines of palladium can clearly be seen. The extra peaks at lower energies are the L shell x–rays from neptunium in the americium decay chain.

Results and Discussion

Several experiments were performed using current densities of about 150 $mAcm^{-2}$ and a driving voltage between anode and cathode of 15–20V. Each sample was run for 48hrs or more. Backgrounds were taken with an inactive cell and without the cell present. Background levels are extremely low. Most of it is caused by background γ–rays which have been Compton scattered to low energies. In all the background and experimental runs, a small peak can be seen at

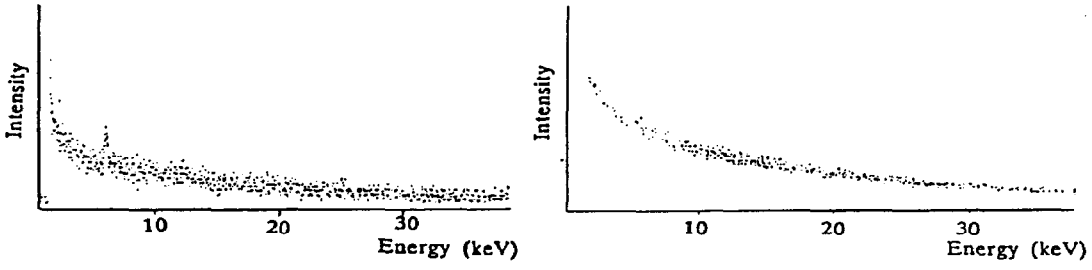

Figure 3 a) Background run with an inactive cell present. b) Spectrum from a palladium cathode being charged with a current density of 150 mA cm^{-2}. Both show a small peak at 5.8 keV which is probably due to previously induced activity in the aluminium collimator.

low energies. This is probably due to previously induced activity in the aluminium collimator used on the detector and is certainly nothing to do with the cell or the palladium. In the calibration run performed with a 150nCi (5.55 kBq) ^{241}Am α source placed against the rear of the 0.1mm palladium foil which was itself placed against the detector, palladium X–rays can be seen without difficulty *(Figure 2)*.

The self absorption of palladium X–rays in palladium can be calculated using tabulated data for the photo absorption cross section[8]. For example the intensity falls by a factor e in a distance $d \approx 54 \mu m$. This is enough to be able to see well into the bulk of the palladium. Theoretically we could have used much thinner cathodes without significant loss in the numbers of X–rays reaching the detector. However, it was still necessary to use a thick foil to minimize the distortion that occurs when Pd foils are charged with deuterium. The absorption of 21.1keV X–rays in the intervening Mylar, heavy water and air can be calculated in the same way to be about 6%. This is negligible compared with the absorption in the palladium.

The yield of X-rays at the detector per charged particle slowing down in a foil of thickness w can be calculated from

$$Y \approx N\sigma^{x-ray} t C_{abs} (d/w)(1-e^{-w/d})\varepsilon\Omega$$

where N is the number density of palladium atoms and σ^{x-ray} is the cross-section for X-ray emission (of the order 2-8 barns for the particles and energies proposed to result from cold fusion[9]). t is the distance over which slowing down occurs and C_{abs} is an absorption correction factor which takes into account absorption in the D_2O and Mylar. The $(d/w)(1-e^{-w/d})$ term is the integrated probability for X-ray escape from the foil assuming a uniform X-ray production rate. ε is the efficiency for the detection of the characteristic X-rays of palladium and is close to 100% and Ω is the solid angle subtended by the detector at the foil. For a 1.5mm foil and our cell geometry and the proposed reaction products, Y typically has a value of 10^{-5} X-rays detected per particle. This is in excellent agreement with the value obtained from the calibration measurements (*figure* 2).

In none of the runs was it possible to see a peak at 21.1 keV. Assuming Poisson statistics, an upper limit for the number of X-rays detected was 0.14 s^{-1} which is equivalent to 10^4 nuclear events in our sample or approximately 10^5 events $cm^{-3}s^{-1}$.

Our sensitivity for X-ray detection is comparable to that reported by other groups[2] for neutron detection. However, X-ray detection has the advantage of also recording the aneutronic reactions already mentioned. The nuclear reaction rate reported above is quite inconsistent with the large heat production reported by Fleischmann and Pons[1].

Other groups have attempted similiar measurements, but no one has yet reported seeing X-rays from electrolytically charged hydrogen-metal systems[7].

Conclusion

The lack of significant numbers of palladium X-rays indicates that no appreciable amount of nuclear fusion of any kind is occurring in our experiments, certainly not to the levels that would be necessary to account for the heating seen in some of the calorimetric experiments. Even the levels of neutrons reported[1] should produce measurable numbers of X-rays from the other products of the reaction. We should stress, however, that this does not rule out the possibility that cold fusion exists. It only shows that we did not observe it in our particular experimental arrangement.

Acknowledgements

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