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EVIDENCE FOR PRODUCTION OF TRITIUM VIA COLD FUSION REACTIONS IN DEUTERIUM GAS LOADED PALLADIUM

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Introduction

After the first announcement reporting the observation of cold fusion¹⁻² further evidence supporting the same has appeared in scientific literature although many other groups have failed to obtain positive results. Palladium and titanium loaded electrolytically¹⁻² and titanium loaded directly with deuterium gas³ have been reported to emit neutrons. Interestingly gas loading experiments involving Pd-D have not been reported so far. Such experiments were therefore conducted recently in our group. Tritium measurements in gas loaded Pd-D targets have been carried out. The present paper summarises the results obtained so far to ascertain whether cold fusion reactions occur in gas loaded Pd targets also.

Preparation of Pd Samples

For loading deuterium gas in Pd, two types of samples were used. One was Pd-Ag alloy supplied by M/s. Johnson-Matthey and was directly used without further surface treatment. The other type of sample used was Pd-black powder which was prepared from PdCl₂. Absorption of deuterium by Pd-black was very fast and readily gave a stoichiometry of D:Pd = 0.6 as deduced from the drop in gas pressure during loading. But in case of Pd-Ag alloy the absorption was rather slow and also the amount of deuterium absorbed was much less than that corresponding to a D:Pd ratio of 0.6.

D₂ gas Loading Procedure

The D₂ gas used for gas loading was prepared from D₂O procured from Heavy Water Plant at Baroda (Gujrat state). This D₂O had a tritium activity of 0.075 nCi/ml. The gas generated from it by reducing with Na in a vacuum system under stringent conditions, was stored in a s.s. cylinder under pressure and liquid N₂ cooling in the presence of activated charcoal. The D₂ gas thus produced was not further analysed for tritium as it was expected to contain not more than 0.038 nCi/l activity. This corresponds to a (T/D) isotopic ratio of 3×10^{-14} .

The schematic drawing of the experimental set up used for gas loading is given in Fig. 1. It essentially consists of a vacuum system equipped with a rotary pump and an oil diffusion pump giving a vacuum of 10^{-6} torr. The s.s. reaction vessel C containing the Pd sample is connected to the vacuum system through an s.s. buffer tank B. Deuterium cylinder D is connected to the vacuum system through needle valve V1. The system is also equipped with a pressure gauge G and a manometer/ pressure gauge VG. The entire system was tested for a vacuum of 10^{-6} torr and pressure of 100 Kg/cm^2 . A weighed amount of Pd-black or Pd-Ag alloy was taken in the vessel and heated to 600°C for 2 hours under a vacuum of better than 10^{-5} mm. After cooling to room temperature, deuterium gas was filled at 1 atm. pressure and the system sealed off to attain equilibrium.

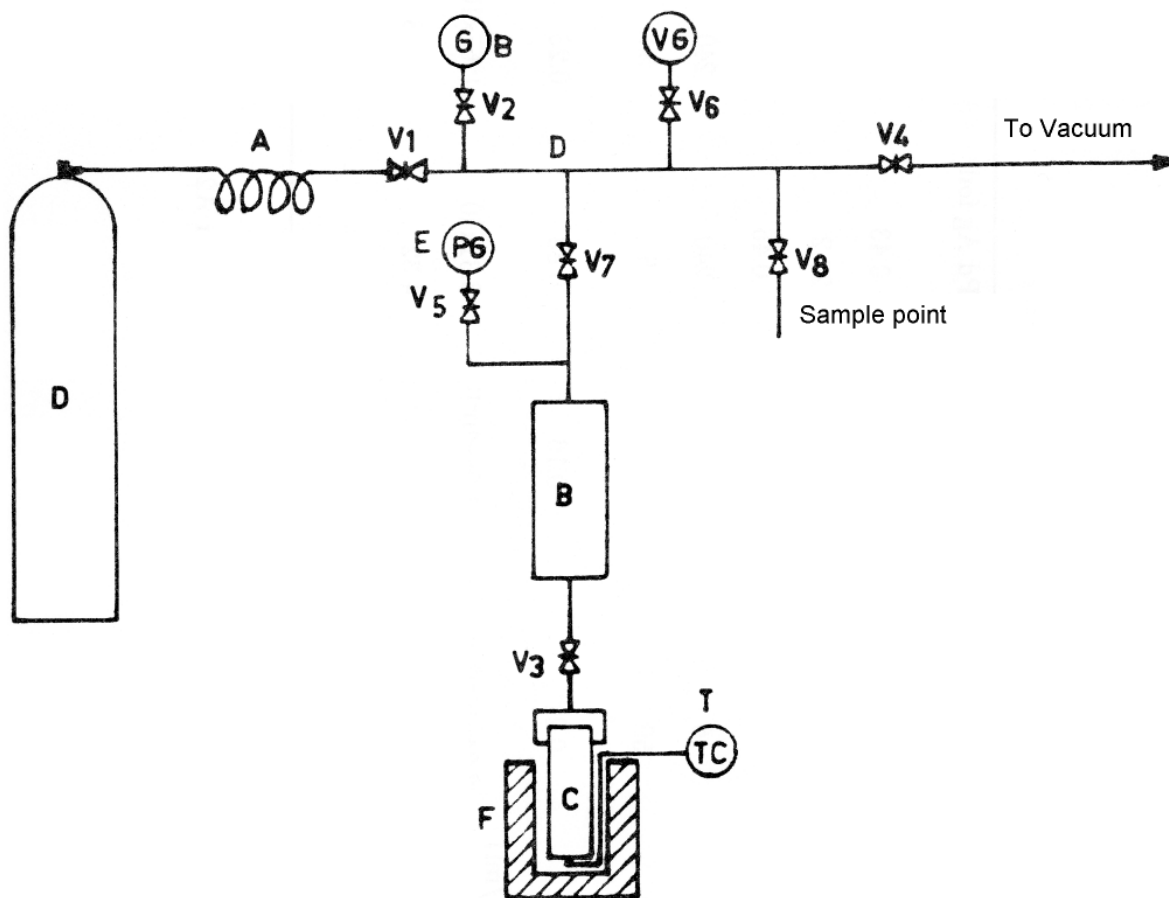


Fig. 1. Schematic diagram of equilibration system.

After completion of gas loading the vessel containing the Pd sample was isolated from the filling system and transferred into a closed glass container in a dry enclosure, free of moisture or oxygen and kept for equilibration for several hours. Adequate precautions were taken to avoid inadvertent exposure to moisture since this would lead to catalytic recombination of absorbed D_2 gas with O_2 , accompanied by considerable increase in temperature. The entire deuterium absorbed in Pd would be lost if dire precaution is not taken. This was confirmed in one set of experiments where the loaded sample was accidentally exposed to air and the resultant water sample equilibrated did not show any activity.

Tritium Analysis

In order to measure the tritium if any produced due to cold fusion reactions the deuterated Pd samples were kept in contact with distilled water for a few hours to extract the tritium by isotopic exchange into the water. The water samples containing tritium were later sent to the Tritium group of the Health Physics Division for liquid scintillation counting using procedures described in this report elsewhere.

In converting the measured tritium activity in the distilled water to a calculated tritium activity originally present in the Pd samples, there is a conversion factor to be used which is computed as follows:

Taking into consideration the exchange reaction



and applying laws of Chemical Equilibrium, one obtains the following relation for low tritium concentration:

$$Y_i \cdot n_g = n_l [X_e(1/K+1) - X_i] \quad (2)$$

where X & Y refer to tritium atom fraction in the absorbed and the liquid phases respectively and subscripts i and e refer to initial and equilibration conditions respectively. n_g & n_l are the gm moles of gas absorbed in metal and gm moles of water taken for equilibration. K is the equilibrium constant for reaction (1) and was taken to be the same as for exchange of tritium between hydrogen and water, since at equilibrium the system consists mainly of H both in the absorbed and the liquid phases. The value of K is 6.128 at 30°C . Y_i calculated from equation (2) gives the number of tritium atoms produced for every deuterium atom absorbed in the metal. The tritium atom fraction X can be calculated from the tritium activity A applying equation 3.

$$x_e = \frac{A}{d \times 3200} \quad (3)$$

where A is activity in Ci/ml and d is density of water in gm/ml ($d = 0.996542$ at 27°C).

Results and Discussion

Table I summarises the results of the tritium measurements on the four samples studied by us. The first three rows give the experimental parameters viz. mass of sample, volume (STP) of deuterium absorbed and the volume of water employed for isotopic exchange. The fourth row gives the D/Pd ratio. The fifth row shows the time for which the gas loaded Pd samples were kept undisturbed in the same pressure at which D₂ gas has been loaded. The seventh row gives the tritium activity of the water after equilibration. The last two rows give the idea of absolute amounts of tritium present in the metal before equilibration. The T/D ratio is about two to three times more in the case of Pd-Ag foils as compared to Pd black (6th row). It should be noted that the T/D ratio in the targets is in the range of 10⁻¹² to 10⁻¹¹ which is more than two orders of magnitude higher than that in the initial deuterium gas used for loading.

TABLE I Summary of Tritium Measurements

1 Nature of Sample	Pd black powder	Pd-Ag foil*	Pd Ag foil*	Pd Ag foil [‡] (single foil)
2 Mass (gms)	20.0	0.96	0.43	10.9
3 Volume of D ₂ gas absorbed (ml)	1325	34.5	20.2	518.4
4 D/Pd ratio	0.63	0.46	0.45	0.45
5 Time of equilibrations (hrs)	16	16	240	240
6 Vol. of water used for isotopic exchange (ml)	50	6	5	50
7 Tritium activity of water after equilibration (nCi/ml)	0.22	0.16	0.88	0.23
8 (T/D) ratio	3.24×10 ⁻¹²	1.08×10 ⁻¹¹	8.31×10 ⁻¹¹	8.67×10 ⁻¹²
9 Absolute tritium activity (nCi)	11.05	0.97	4.32	11.58
10 Number of tritium atoms in Pd	2.31×10 ¹¹	2.02×10 ¹⁰	8.96×10 ¹⁰	2.4×10 ¹¹

*These were the triangular foils also studied by Autoradiography.

[‡]This was a single 11.5 cm dia foil; same as cathode used in cell of paper A2.

A fresh lot of Pd black (10 gms) and Pd-Ag foils (0.43 gms) were equilibrated, with D₂ gas and after this accidentally they were exposed to air. These became intensely hot and on equilibration with distilled water and analysis of T in the equilibrated water, the T activity observed in both was below detectable limit. The same sample of Pd-Ag foil (0.43) was reactivated (650°C, 10⁻⁵ mm) and again loaded with D₂ gas which shows the results as in the 3rd

vertical column, proving that the residual activity in Pd is nil. It is also seen that when the equilibration time is more, more tritium production is observed.

The method of isotopic exchange for extraction of tritium from metal to water phase is less cumbersome and the T/D ratio obtained from equation (1) gives at least a conservative estimate of tritium produced.

Autoradiography of Samples

The Pd-Ag foils were also subjected to autoradiography to obtain images of tritium distribution as described in detail elsewhere in this report⁵. It essentially consists of keeping the gas loaded sample on x-ray film and developing it after allowing adequate exposure time. The Pd-black samples could not be subjected to Autoradiography as the powder got stuck to the film. But in case of Pd-Ag foils it gave unmistakable fogging of x-ray film corresponding to the geometrical shape of the foils, thus indicating the emission of some radiation from these foils and the only radiation being emitted from these foils which can be thought of is the β -rays from tritium and the characteristic X-rays of the metal excited by tritium β s.

Summary and Conclusions

Gas loaded Pd samples have provided evidence for the first time of the presence of tritium, strongly suggesting the occurrence of cold fusion reactions. The Pd-D system does not require high pressure of D₂ gas and also no external perturbation is required to create non equilibrium conditions as suggested by De Ninno et al³. Although no quantitative comparisons can be made between electrolytically loaded and gas loaded Pd experiments. The present results confirm that electrolysis is not the only approach to inducing cold fusion in Pd lattice. Unfortunately no neutron measurements were carried out in the present work. A correlation between neutron production if any and tritium production would contribute significantly towards understanding the mechanism of cold fusion.

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