

## **Copious low energy emissions from palladium loaded with hydrogen or deuterium**

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Palladium samples were loaded with deuterium and hydrogen using plasma focus and other loading techniques. Each sample, loaded so far, was observed to be emitting low energy, low intensity radiations. These radiations have been detected and analyzed by autoradiography and other supporting techniques.

The occurrence of anomalous emissions from certain metals (such as palladium and titanium), when loaded with deuterium, either electrolytically<sup>1,2</sup> or in gas phase<sup>3,4</sup> or in plasma phase<sup>5,6</sup>, has been reported by various laboratories. Most of these emissions have been found to be sporadic and are not easily reproducible. We report here emission of some low energy radiations emanating from palladium when loaded either with deuterium or with hydrogen, by any loading technique. *The phenomenon is reproducible and the low energy radiations have been observed from all the samples loaded so far.*

### **Loading of Deuterium/Hydrogen in Palladium**

#### ***Preparation and testing of input material***

The deuterium gas (D<sub>2</sub>) for loading was prepared by electrolysis of heavy water. The tritium content of this gas was estimated<sup>7</sup> to be  $\sim 10^{-4}$  Bq/mL. For hydrogen (H<sub>2</sub>) loading commercially available gas was used. The radioactive content of this gas was below the threshold of measurement ( $< 10^{-5}$  Bq/mL).

Commercially available palladium (Pd) was used without any pretreatment. Most of the palladium was procured from Hindustan Platinum Ltd, Bombay, India. The palladium foils were obtained from Johnson Matthey, USA. Each Pd sample was checked before loading (i) for low energy radiations by autoradiography, (ii) for charged particles by surface barrier detector, and (iii) for X-rays by a thin sodium iodide (NaI) detector (1 mm thick, 200  $\mu$ m Be window). No sample, before loading, showed any fogging during autoradiography. Charged particle and X-ray emissions were also below the detectable threshold.

#### ***Loading of gas***

The majority of the samples (used for subsequent measurements) were in the shape of disks, 16 mm diameter and 2 mm thick. These were loaded by affixing them on a plasma focus<sup>5,8</sup> (PF) central electrode. The plasma focus was evacuated and then filled with the desired gas (D<sub>2</sub> or H<sub>2</sub> at few mb) and discharged. This process was repeated typically 15 to 30 times for loading a sample.

To cross check if the phenomena take place with other loading techniques as well, several Pd needles were loaded using the Wada gas discharge technique, by D<sub>2</sub> or H<sub>2</sub> [a 10 kV (AC) discharge was struck between two pins in 600 mb of gas]. Several Pd foils (5 mm x 5 mm, 25  $\mu$ m thick), planchets (16 mm diameter and 2 mm thick) and several hundred Pd chips were loaded by first degassing in vacuum (600°C, 10<sup>-5</sup> mb), and then allowing them to cool in the D<sub>2</sub> or H<sub>2</sub> atmosphere (at 1 bar pressure, for 2 h). Table 1 summarises the information about loading.

The bulk loading obtained during gas loading could be estimated from the amount of gas absorbed by the sample. The estimated atomic fraction (the quantity  $x$  in PdD <sub>$x$</sub>  or PdH <sub>$x$</sub> ) varied between 0.1 and 0.6. It is possible that the loading in the surface layer may be much higher.

## Measurement of Radiative Emissions

The radiations emitted from the samples were predominantly of very soft nature and of comparatively low intensity (as will be evident from subsequent sections). They were short lived (maximum life of a few days). It was not possible to perform any measurements in vacuum, as this might have resulted in unloading of the sample and cessation of the phenomena. Under these circumstances, it was not feasible to utilize several of the conventional techniques used for measuring these kinds of radiations. Because of the low intensity, each measurement took several tens of hours, and therefore it was not possible to use all the diagnostic techniques (described in the following sections) on each loaded Pd sample.

Table 1 — Loading data

Method of loading	Sample loaded	
	<u>D<sub>2</sub> gas</u>	<u>H<sub>2</sub> gas</u>
a) Plasma focus by fixing 16 mm dia 2 mm thick disk on anode	16 disks	25 disks
b) Gas loading Sample degassed at 600°C and cooled in 1 atm gas for 2 h	4 foils ~10 <sup>2</sup> chips 3 disks	6 foils ~10 <sup>2</sup> chips 3 disks
c) Wada discharge struck a 10 kV AC discharge between electrodes in 600 mb gas	2 needles	2 needles

### Autoradiography

Autoradiography<sup>9</sup> was the most extensively used diagnostics. *All the D<sub>2</sub> or H<sub>2</sub> loaded palladium samples fogged the X-ray films.* For autoradiography the (bare) X-ray films were kept in contact or a few mm away from the sample, in normal atmosphere. For all autoradiographs INDU 'Polyester' or QX-16 X-ray films (from Hindustan Photofilms, India) were used. Both the films were screen type Superol (from May and Baker, India) brand chemicals were used for developing and fixing. The exposure time varied from 24 to 120 h. Normally, uniform fogging of the film was observed. Though all samples fogged the film, the fogging density varied from sample to sample. D<sub>2</sub> or H<sub>2</sub> loaded Pd samples yielded radiographs of approximately similar average density, under similar loading conditions. Fig. 1 shows a contact autoradiograph of a planchet loaded with D<sub>2</sub> using a PF device (30 discharge shot, 24 h exposure). Fig. 2 is an autoradiograph of a similar H<sub>2</sub> loaded sample (30 discharge shots, 90 h exposure) kept 0.2 mm away from the film.

To rule out a chemical reaction (resulting in fogging) between film and H<sub>2</sub> (or D<sub>2</sub>) gas, the film was kept in an atmosphere of H<sub>2</sub> and H<sub>2</sub>-air mixture for 92 h. No observable fogging of the film was detected.

The samples continued to fog the film for ~100 h. If the sample was heated to temperature  $\geq 60^\circ\text{C}$  for more than 1 h, then it no longer fogged the film. Once a sample stopped blackening the radiograph and it was reloaded (after sand paper polishing) with D<sub>2</sub> or H<sub>2</sub>, it again fogged the film approximately to the same extent.

*With filters* — Some of the samples were kept 2.2 mm (0.26 mg/cm of air) away from the film. Fig. 3 is the image of one such H<sub>2</sub> loaded (using PF) sample (15 discharge shots, 70 h exposure) kept 2.2 mm away from the film. In these, the average density (intensity) of the radiograph was approximately reduced to 50%. No fogging of the autoradiograph was observed when the following filters were kept between sample and the film:

- 2  $\mu\text{m}$  thick aluminised polycarbonate foil [0.25 mg/cm<sup>2</sup> (14%Al)]
- 50  $\mu\text{m}$  transparent polyester film (light transmission 98% in visible region  $< 4500 \text{ \AA}$ , 66% at 3400  $\text{ \AA}$  and no transmission at 3000  $\text{ \AA}$ )
- 7.5  $\mu\text{m}$  thick beryllium foil (1.35 mg/cm<sup>2</sup>)
- 3 mm thick quartz plate (light transmission 92% in visible and ultraviolet region  $< 2500 \text{ \AA}$ , 67% at 2000  $\text{ \AA}$  and 45% at 1800  $\text{ \AA}$ )

Fig 4 is the autoradiograph of a D<sub>2</sub> loaded (using PF) planchet (30 discharge shots, 115 h exposure) kept 0.2 mm (air gap of ~20 μm/cm<sup>2</sup>) away from the film. A small portion of one segment of this planchet was covered with a 50 μm thick transparent polyester foil, a larger segment was blocked by a 2 μm thick aluminised polycarbonate foil and the middle portion left open.

*With Polaroid film* — Loaded Pd samples were also photographed using Polaroid films (Viva, 3000 ASA) and films sensitive up to ultraviolet wavelength. In both the cases no fogging of the films was observed after an exposure time of 60 h. This test implies that the emission from the planchet may not be visible or ultraviolet radiation.

*With applied electric field* — The electric field between the loaded sample (disk type) and the film was applied by keeping them between two parallel copper plates connected to a power supply. The sample was in contact with the top copper plate, while the film was in contact with the bottom copper plate. The separation between the film and the sample was maintained by a perspex spacer, 1.2 mm thick, having an opening of 12 mm at its centre. Typically an experiment consisted of using three such assemblies. To the first sample positive voltage was applied, to the second no voltage was applied, while to the third negative voltage was applied. In different sets of experiments the voltage was varied from ± 1.5 V to ± 400 V (field varying from +3.3 kV/cm to -3.3 kV/cm). Application of the field very much increased the intensity of fogging of the autoradiographs. The fogging increased as the voltage was increased, saturating approximately at 100 V. Surprisingly, when positive voltage was applied to the sample (electrons being repelled from the film) the fogging was higher than when negative voltage was applied. Figs 5(a) and 5(b) are the autoradiographs of two identical plasma focus loaded (15 PF shots) planchets. The first one was with 100 V positive and the second one with 100 V negative supply. The exposure time for both the cases was 41 h.

*With applied magnetic field* — The radiations from two similar samples were collimated (126.1 mm diameter). One sample was kept in a magnetic field of 6.5 gauss (applied perpendicular to sample axis), and the other sample was kept without field. Both were autoradiographed with the films kept 2.2 mm away from samples, for an exposure time of 48 h. No observable fogging was observed on the autoradiograph in the former case. In the latter case a weak spot (corresponding to collimator opening) was observed on the autoradiograph.

### ***Thermoluminescent dosimeter (TLD) studies***

*Bare sample* — CaSO<sub>4</sub> based TLDs, in powder form, were sprinkled on the surface of the samples to detect relative emission from the sample. Typical observed activity was seven times (or 60 σ) above the background, for an exposure time of 72 h. These TLDs are not sensitive to chemical reduction or visible light, but are sensitive to electrons, heavy charged particles, ultraviolet (UV) and shorter wavelength electromagnetic radiations. Calibration of these TLDs, for the low energy radiations, is however not available.

*With fused silica filter* — One of the samples was partly covered with a fused silica plate (3mm thick, cut off wavelength 1800 Å), and TLD powder was sprinkled on both bare and covered parts. After 72 h, the TLD powder, sprinkled on the part covered with fused silica, did not show any luminescence above background, while the uncovered part showed significant luminescence (as described in the previous section). This indicates that the emitted radiations are neither visible nor ultraviolet radiation.

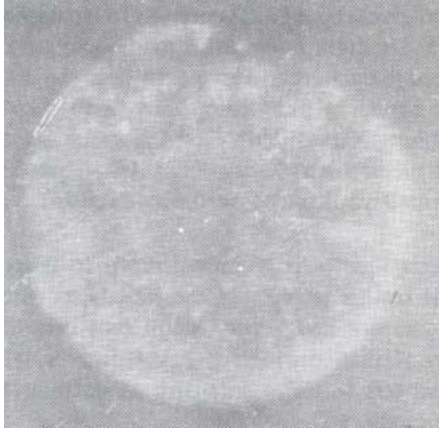


Fig. 1 - Autoradiograph (D<sub>2</sub> loaded, Sample in contact)

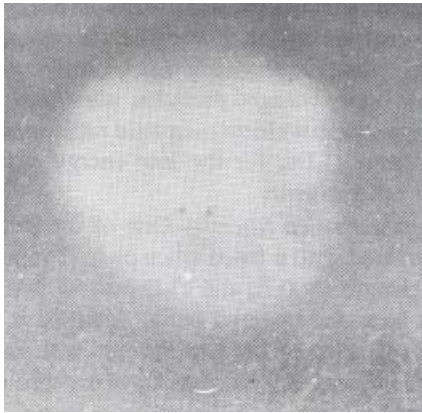


Fig. 2 - Autoradiograph (H<sub>2</sub> loaded, Sample 0.2 mm away)



Fig. 3 - Autoradiograph (H<sub>2</sub> loaded, Sample 2.2 mm away)

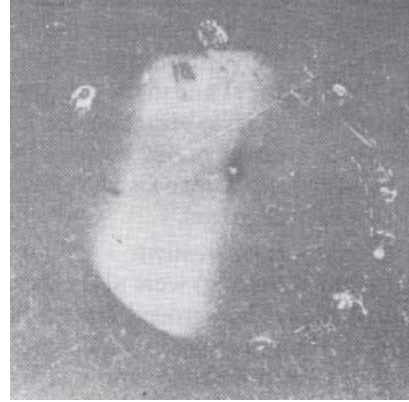


Fig. 4 - Autoradiograph (D<sub>2</sub> loaded, Sample 0.2 mm away)

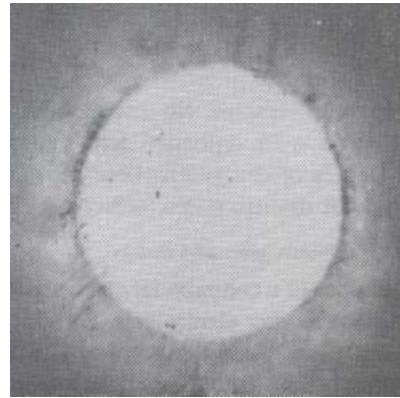


Fig. 5(a) - Autoradiograph (H<sub>2</sub> loaded, + 100 V, 1.2 mm away)

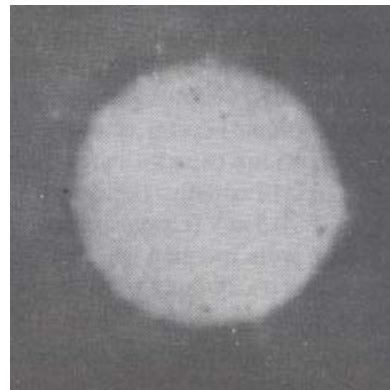


Fig. 5(b) - Autoradiograph (H<sub>2</sub> loaded, - 100 V, 1.2 mm away)

## ***X-ray emissions***

*Sodium iodide detector measurements* — All the D<sub>2</sub> and H<sub>2</sub> aided samples were checked for X-ray emission before and immediately after loading (for 1 h) by an X-ray sensitive NaI detector (1 mm thick, 200 μm Be window). This detector was found to be suitable for X-rays above 3 keV and had a threshold of 400 counts/s (1 h counting). None of the samples loaded so far has exhibited any X-ray emission above background.

*SiLi detector measurements* — Four samples (3 loaded, 1 without loading) were also checked for X-ray emission on a low background silicon lithium (SiLi) detector with a thin beryllium window (1 keV energy threshold). The measurement time was typically 18 h. PdL and K X-rays were observed with loaded as well as not loaded samples. These, however, are attributable to the excitation of the samples by background gamma radiations. Typical background around the Pd K peak (~21 keV) was  $0.02 \pm 0.003$  count/s. No sample showed emission of X-rays above background.

## ***Gas flow detector studies***

*2π geometry detector* — Three loaded samples (two PdH<sub>x</sub> and one PdD<sub>x</sub>) were also counted in a 2π geometry gas flow proportional counter detector. The detector had a background of 5 to 6 counts/s, the threshold of measurement was 126.015 count/s. The detector had an energy threshold of 126.05 keV (counts from particles below this energy, if any, were merging with the electronic noise). No counts above background were observed from any of the samples tested.

*Low background end window type detector* — Higher energy radiations emanating from the samples was also measured by an end window type gas flow detector (operating in Geiger Muller mode, 0.8 mg/cm<sup>2</sup> window — this window thickness would stop electrons below ~20 keV). The typical background of the detector was  $0.01 \pm 0.002$  count/s. Samples with D<sub>2</sub> or H<sub>2</sub> loading were observed to give  $0.02 \pm 0.002$  count/s (18 h counting after loading). Forty-eight hours after loading, this emission was reduced to background levels. If these radiations are electrons then, in principle, they would generate bremsstrahlung X-rays. However, the efficiency of conversion of electrons to X-rays is < 0.1%. Therefore, the X-rays emitted would be below the threshold of even SiLi detector.

This technique was complementary to autoradiography. The results imply that the radiations emanating from the samples had a very small (but measurable) component of relatively high energy. However, this component was too small in intensity to account for the autoradiographs.

## ***Heavy charged particles activity***

*Surface barrier detector studies* — Each sample was counted on a surface barrier detector (window of 500 Å aluminum) for 1 h before and after loading. This detector was suitable for detecting only energetic heavy charged particles (p, He<sup>3</sup>, t, α) and had a threshold of 0.003 particle/s (for 1 h counting). All the measurements were made in air and the samples were kept in contact with the detector. The detector was calibrated by an Am<sup>241</sup> α source.

Charged particle emission was observed rarely and that too only with D<sub>2</sub> loaded samples. It was sporadic and non-reproducible. Only two D<sub>2</sub> gas loaded and two plasma focus loaded (D<sub>2</sub>) samples (total of four samples) exhibited charged particle activity (Table 2). Because of fluctuations in intensity and spectrum of the emitted charged particles, it was not feasible to ascertain the species of the emitted particle. None of the H<sub>2</sub> loaded samples was observed to emit charged particles above the threshold.

*Solid state nuclear track recorder studies* — The deuterated samples which showed positive results with surface barrier detector were also kept in contact with solid state nuclear track detector (SSNTED) film (type LR 115 Kodak) for 18 h. Large number of tracks were observed confirming the emission of energetic charged particles from these samples. No tracks above background were observed with unloaded and hydrogen loaded samples. Details of charged particle detection measurements would be discussed in a forthcoming paper.

## ***Other loading methods, gases and metals***

Pd electrodes of a commercial electrolytic cell were autoradiographed after electrolysis. Part of the electrode of this cell was also deuterium gas loaded and autoradiographed. In both the cases we failed to observe any fogging on the radiograph. The electrodes of this cell were processed (activated) for fast migration of hydrogen. This could have inhibited the phenomena responsible for autoradiography. However, preliminary evidence of fogging of autoradiographs by Pd electrodes after electrolysis had been observed by Prof. Jones at Brigham Young University.

Table 2 — Surface barrier detector counting  
 [Deuterated palladium sample PDGL 31 Side A]

Experiment Description		Counts/hour for the energy ranges (MeV) (The numbers below the counts are the one $\sigma$ errors)									<u>Total</u>	
Set	Date	0-0.39	0.39-0.78	0.78-1.17	1.17-1.55	1.55-1.94	1.94-2.72	2.72-3.49	3.49-4.27	4.27-5.05	per h	per s
BKG1	20/3/91	27 3	1 1	0 0	0 0	0 0	0 0	0 0	0 0	0 0	29 3	008
BKG2	21/3/91	25 5	3 2	0 0	0 0	0 0	0 0	0 0	0 0	0 0	28 5	0.008
					<u>Net Counts</u>							
EXP1	20/3/91	26 6	11 3	3 1	1 0.5	0.6 0.3	0.6 0.3	0 0	0 0	0 0	42 6	0.011 0.002
EXP2	21/3/91	102 8	31 3	6 2	7 1	5 1	3 1	2 1	1 1	1 1	158 15	0.043 0.004
EXP3	21/3/91	38 9	20 4	8 2	2 1	0 0	0 0	0 0	0 0	0 0	68 8	0.019 0.002

BKG Background with palladium sample on the surface barrier detector; background was later reduced (by using new set-up) to  $7 \pm 3$  counts/h.

EXP Counts with deuterated palladium sample on the detector; higher energy ( $>3.3$  MeV) particles are perhaps due to their emission in (unresolved) sharp bursts. Lower energy particles are perhaps produced at some depth and thus lose a part of their energy in the sample itself.

One Pd sample was also loaded with deuteron implantation technique. The autoradiograph, from this sample, was fogged to the same extent as the other loading methods.

*Pd loaded with helium gas* — One Pd sample was also loaded with helium ( $He^4$ ) using plasma focus. The sample was subjected to 25 discharges (at 1 mb) in the device<sup>5</sup>. The plasma focus is capable of ion implanting helium into palladium, thus surface loading the Pd with helium. Subsequently the sample was autoradiographed. The film was kept 0.2 mm away from the sample and exposed for 72 h. No fogging on the film was observed. This implies that phenomena observed are specific to  $D_2/H_2$  loading only.

*Titanium loaded with hydrogen/deuterium* — As reported previously, out of several thousands of  $D_2$  gas loaded titanium samples, a few had tritium present in them. This caused subsequently strong fogging of the autoradiograph<sup>5,9,12</sup>.

In the current series of experiments, several dozen planchets of Ti were loaded in plasma focus by  $D_2$  or  $H_2$ . Several thousand small chips of titanium were also gas loaded by  $D_2$  or  $H_2$  and were subsequently subjected to liquid nitrogen treatment. None of the loaded (and treated) samples was observed to contain tritium<sup>13</sup>.

All the samples were autoradiographed for an exposure time of  $> 48$  h. Only from one sample ( $D_2$  loaded in plasma focus), which was cut from a crystal bar (the Ti was of very high purity), a very weak fogging of radiograph was observed (film exposed to sample for 48 h; subsequently no tritium could be detected in this sample).

There is also some preliminary evidence to suggest that loaded Ti samples (like Pd samples) may also fog a film, if the exposure time is very large (several weeks). Two plasma focus  $D_2$  loaded Ti samples were kept close to an X-ray film for approximately one month. A spotty radiograph was obtained from one of the samples.<sup>14</sup>

Measurements with NaI detector indicated that X-rays ( $> 3$  keV) might have been emitted from three Ti samples (out of several thousands) for a short duration of few tens of minutes (the emission was not sufficient to produce an autoradiograph). Two of these samples were D<sub>2</sub> loaded (one by plasma focus, other gas loaded) and one was H<sub>2</sub> loaded (by gas loading). A similar X-ray emission, for a short duration, from loaded Pd samples as well cannot be ruled out.

None of the titanium samples checked (loaded in this series of experiments) was observed to emit charged particles.

*Other metals loaded with deuterium* — A few samples of zirconium, hafnium and niobium-(Nb- Ti superconductor) were also loaded with deuterium using the plasma focus. All the samples were autoradiographed (film exposed for 48 h). No fogging of the autoradiographs was observed. Charged particle, electron or X-ray emissions were not observed to be emitted from any of the loaded samples.

## Nature of Radiation

As pointed out in earlier sections, only a small percentage of loaded Pd samples were emitting either high energy charged particles or other radiations. The intensity of these emitted radiations was, however, not sufficient to fog the film to the extent observed. Therefore, some other low energy radiations were also emitted by the samples. First, the nature of these predominant radiation(s) responsible for the autoradiographs is discussed.

### *Radiations responsible for fogging of the autoradiograph*

*H<sub>2</sub>/D<sub>2</sub> chemical reaction with the X-ray film* — Direct reduction of silver in the film by nascent hydrogen emanating from Pd sample (and Pd acting as catalyst) is ruled out as (i) the radiographs have also been obtained with samples kept 2.2 mm away from the film (this distance is too large for any catalytic action or for nascent hydrogen to survive), (ii) TLDs (which are unaffected by chemical reaction) have also given positive results, and (iii) the fogging was influenced by application of relatively weak magnetic and electric fields.

As indicated earlier, our film was tested and found to be insensitive to gaseous hydrogen and hydrogen-air mixture.

*Intensity of radiations* — Without knowing the exact nature and spectrum of the radiations, it is not possible to determine the intensity of radiations. However, a rough estimate may be made by comparing the autoradiographs with the ones obtained from ‘tritium loaded in titanium sources’ (TiT)<sup>15</sup>. TiT sources which fogged the autoradiograph to the same extent, in same exposure time, were observed to emit  $5 \times 10^4$  to  $5 \times 10^5$  particles (as counted in a  $2\pi$  proportional counter). Since the average energy of particles emitted from TiT ( $\beta$ /X-ray) is approximately 5 keV, about  $10^8$  to  $10^9$  events/s were required to fog the autoradiograph to the extent observed. Assuming that this calibration is valid for the radiations emanating from loaded Pd samples, a total energy of  $\sim 10^{13}$  to  $10^{15}$  eV was emitted by the samples in the duration of emission.

*Visible and ultraviolet radiations* — The radiations emanating from the sample get completely stopped by a polyester film as well as by a quartz plate, both transparent to visible light. Therefore, the fogging is not due to electromagnetic radiations in the visible range.

Radiations were completely stopped by quartz and fused silica plates (which had sufficient transmission at wavelengths  $> 1800$  Å). This was observed by autoradiography and confirmed by TLD measurements. Therefore, the samples were not emitting ultraviolet (UV) radiations with wavelength  $> 1800$  Å. If UV radiations  $< 1800$  Å were being emitted by the samples, then they would be completely absorbed in the air gap between the sample and film for autoradiography. Since emissions were getting influenced by electric and magnetic fields, we concluded that non-ionising electromagnetic radiations were not responsible for the observed fogging.

*Ionising radiations* — Low energy ionising radiations emanating from the sample (such as electrons, X, UV and heavy charged particles) could be responsible for the fogging of the film. These radiations, after emission, get absorbed in the air adjacent to the sample and ionise it. The secondary ions and electrons thus produced fog the film. Under an electric field (of any polarity) either secondary ions or electrons were collected more efficiently by the film (just as in an ionisation chamber), thus increasing the fogging of the autoradiograph. These very low energy, secondary ions/electrons also get deflected by a weak magnetic field. Any physical barrier (such as quartz plate, polyester, aluminised polycarbonate or beryllium film, etc.) would also easily stop these secondary ions/electrons resulting in cessation of fogging of autoradiographs.

With the present studies we are unable to determine whether the ionising particles emitted by the sample are photons, electrons or heavy charged particles (ions).

*Energy of the ionising radiations* — From the investigation carried out so far, it is not clear whether the emissions were continuous or in very sharp bursts or in both modes. This may have little effect on autoradiography and SSNTD studies. But the SiLi, gas flow detectors and surface barrier detector are not capable of handling large sharp bursts of radiations.

If the radiations were not in sharp bursts, then SiLi and  $2\pi$  proportional counter measurements suggest that there was no significant emission above 1 keV energy. Therefore, the ionising radiation energy was in the range of few tens of eV (minimum required for ionisation) to few hundreds of eV (as indicated by  $2\pi$  proportional counter measurements). The intensity of these emissions was  $10^5$  to  $10^7$  particles/s (assuming that the calibration using the TiT source was also valid for these radiations).

However, if the radiations were in bursts, then the energy could be deduced only from the autoradiographic and SSNTD studies. Since radiations emitted from the loaded Pd samples were stopped by a  $2\mu\text{m}$  aluminised polycarbonate filter, if the radiations consisted of electrons or electromagnetic radiations, then the energy was again  $< 1$  keV. On the other hand, if the radiations consisted of ions, then even higher energy ions may also be present. Since no ions from these samples (normally) were recorded on SSNTD, the energy of these ions should be  $< 10$  keV. It is intuitively felt that low energy ion emission from the loaded samples is unlikely.

*Non-ionising radiations* — Samples might also be emitting non-ionising radiations ( $< 10$  eV). However, as stated earlier no non-ionising electromagnetic radiations (visible, UV light) were present. Since these radiations were deflected by weak magnetic field non-ionising ions are unlikely to be present. Non-ionising electrons might also not be present as application of positive voltage to the sample increases the fogging of the film (autoradiograph fogging density should have decreased if radiations consisted of non-ionising electrons).

### ***Higher energy radiations***

Not considering the sporadic emission of charged particles and observations of tritium, the only positive evidence of higher energy radiations was from gas flow detector which had a  $0.8 \text{ mg/cm}^2$  beryllium window. Hence, apart from X-rays, only electrons, protons and as having energies  $\geq 20$  keV, 500 keV and 2 MeV respectively can cross this window. Since no X-rays and heavy charged particles were observed by SiLi and surface barrier detectors respectively, the activity observed could not be due to X-rays or heavy charged particles. Therefore, the gas flow detector counts were due to electrons with energy  $> 20$  keV. Since the end point energy of tritium  $\beta$  is  $\sim 18$  keV, these electrons could not be due to tritium present (if any) in the loaded Pd sample. In these experiments there was also evidence to suggest that counts were observed even after the window was covered by another filter of  $1.6 \text{ mg/cm}^2$  (electron energy  $> 50$  keV)

## **Possible Mechanisms of Emission**

### ***Chemical phenomena***

The energy released per recombination of hydrogen with oxygen is  $\sim 1.54 \text{ eV}$  ( $8052 \text{ \AA}$  — our film is not sensitive to this infrared radiation). Since the radiations emitted may have energy  $> 10 \text{ eV}$ , the chemical recombination could not give rise *directly* to the observed radiations.

Though direct fogging of film due to hydrogen oxygen recombination is ruled out, one cannot, a priori, rule out the role of this reaction in triggering or providing the energy for the radiations *indirectly*. However, there is no simple mechanism by which more than seven times the energy per reaction can get transferred to a single event.

### ***Lattice phenomena***

It is obvious that some of the electrons (and perhaps ions) got, by some kind of collective phenomenon, sizably more energy than the average thermal energy. This collective phenomenon could be short lived large energy fluctuations,<sup>16</sup> correlated collisions<sup>17</sup>, some coherent phenomenon<sup>18</sup> or some instability in metal plasma, etc. The consequent radiation emission from the sample fogs the film. Rarely (assisted by some external trigger such as temperature cycling or current density variation, etc.), some of the particles (ions and electrons) get relatively higher (in the range of tens of keV) energy. These particles may be responsible for keV level X-rays (by electron



bremsstrahlung, possible in both hydrogen and deuterium loaded samples) and fusion (possible only in deuterium loaded samples) which may result in emission of neutrons, charged particles, tritium, etc.

As mentioned in the earlier sections, the phenomenon occurs only if H<sub>2</sub>/D<sub>2</sub> were loaded in the metal (and not with bare metal). However, the condensed matter studies have indicated that in Pd there are no major lattice changes, if it is loaded moderately with H<sub>2</sub>/D<sub>2</sub>. Hence any lattice phenomenon has to be invoked with great care.

### ***Cold nuclear fusion***

The radiations could also be emitted as a result of cold fusion

D + D He + fusion energy to lattice radiations

He + n charged particles and neutrons

T + p charged particles

4 H catalyst He + fusion energy to lattice radiations

It is agreed that as per the present knowledge of physics, this explanation is far fetched. But viewed with the other so-called evidences of cold fusion (such as observation of excess heat and X-rays from light water electrolysis cells with K<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub> and NaCl as electrolytes <sup>19,20</sup>) this could also be a legitimate explanation. However, at this stage there is no direct evidence to substantiate this theory, and it is given here just for completeness.

### **Conclusion**

Palladium when loaded with hydrogen or deuterium emits low energy radiations, as long as the gas remains loaded in the sample (few tens of hours).

The phenomenon is 100% reproducible and is observed with any loading technique. It is detectable even with low loading of the gas.

The samples are reusable. That is, once the Pd stops radiating, because of the gas diffusing out of it, and it is reloaded, then it again starts emitting the radiation to the same extent.

The main part of the radiations perhaps consists of electrons in the energy range of few tens to few hundreds of eV with intensity of 10<sup>5</sup> to 10<sup>7</sup> particles/s. Though a small number of particle (10<sup>-2</sup>/s) till few tens of KeV have also been observed.

The radiation can be easily detected by the simple inexpensive technique of autoradiography.

These radiations could be due to some lattice phenomenon or cold nuclear fusion.

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## References

1. Jones S E, Palmer E P, Czirr J P, Decker D L, Jenson D L, Thome J M, Taylor S F & Rafelski J, *Nature (London)*, 338 (1989) 737.
2. Fleischmann M, Pons S, Hawkins M, *J Electroanal Chem*, 261 (1989)301.
3. De Ninno A, Frattolillo A, Lollobattista G, Martinis L, Martone M, Mori L, Podda S & Scramuzzi F, *Europhys Lett*, 9 (1989) 221.
4. Menlove H, Fowler M M, Garcia E, Miller M C, Pacicotti M A, Ryan R R, Jones S E, *J Fusion Energy*, 9 (1990) 495.
5. Rout R K Shyam A, Srinivasan M & Chitra V, *Fusion Technol*, 19(1991)391.
6. Wada N & Nishizawa K, *JpnJAppl Phys*, 28 (1989) 2017.
7. Krishnan M S, Malhotra S K, Gaonkar D G, Nagvenkar V G & Sadhukhan H K, *Fusion Technol*, 18 (1990) 86.
8. Mather J W, *Methods of experimental physics* (Academic Press, New York), Vol. 9B (1971) 187.
9. Rout R K, Srinivasan M & Shyam A, *Fusion Technol*, 18 (1990) 83.
10. Krishnan M S, Malhotra S K, Gaonkar D G, Srinivasan M, Sikka S K, Shyam A, Chitra V, Iyengar T S & Iyengar P K, *Fusion Technol*, 18(1990)35.
11. Private communication from Prof. S.Jones, Physics Department, Brigham Young University, Provo, Utah, USA (1991).
12. Kaushik T C, Shyam A, Srinivasan M, Rout R K, Kulkarni L V, Krishnan M S, Malhotra S K & Nagvenkar V B, *Indian J Technol*, 28 (1990) 667.
13. Kaushik T C, A report on second phase experiments with deuterated titanium and its alloys cycled in liquid nitrogen, Internal note (unpublished), Neutron Physics Division, BARC, Bombay, 1990
14. Private communication from Auluck S, The experiments were conducted at the plasma focus facility at IPF, University of Stuttgart, Federal Republic of Germany, (1990)
15. Srinivasan M, Rout R K, Misra S K, Lai M, Shyam A, Rao P S & Iyengar P K, *Fusion Technol*, 18 (1990) 88.
16. Dasannachary B A & Rao KR, *Remarks on cold fusion*, Report BARC-1500, Bhabha Atomic Research Centre, Bombay, India, (1989).
17. Beuhler R J, Friedlander G & Friedman L, *Phys Rev Lett*, 63 (1989)1292.
18. Hagelstein P L, "Progress report on coherent fusion studies," Preprint, MIT, Cambridge, Mass., USA (1990).
19. Mills R L & Steven K P, *Fusion Technol*, 20 (1991) 65.
20. Matsumoto T, *Fusion Technol.*, 17 (1990) 490.