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REPRODUCIBLE, ANOMALOUS EMISSIONS FROM PALLADIUM DEUTERIDE/HYDRIDE

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Each and every palladium sample loaded/reloaded either with hydrogen or deuterium was observed to fog radiographic films kept in its close proximity in air. Strangely, even with ten layers of black paper (thickness ≈63 mg/cm^2) as a filter between film and sample, fogging was observed. On the other hand, no fogging could be observed even when thin beryllium foil (≈1.4 mg/cm^2), three layers of transparent polyester foils (≈10 mg/cm^2), or thin aluminized polycarbonate (0.3 mg/cm^2) were employed as filters. Several experiments have been performed to identify the phenomenon responsible for fogging. These experiments appear to rule out any of the known mechanisms, suggesting a new, strange, and unknown phenomena.

INTRODUCTION

In the last 6 yr, the deuterium/hydrogen metal system has been extensively studied. Several anomalous effects have been reported; however, most of them are not readily reproduced. Here, we report totally reproducible emissions from palladium deuteride/hydride (PdD_x/PdH_x), which are capable of fogging a radiographic film kept in proximity and through some filters.

EXPERIMENTAL

Autoradiography

For autoradiography, the X-ray film (bare) was kept either in contact with or a few millimetres away from the sample with the help of a noninteracting spacer (ring shaped). Figure 1 shows the schematic of autoradiography techniques used here. For all the autoradiographs. polyester-based "INDU" medical X-ray films (screen-type X-ray film of a thin protecting gelatin layer) were used. The exposure time for autoradiography was generally varied from 16 to 120 h.

Fogging of Films

Palladium samples with or without any pretreatment were loaded with H_2 or D_2 by any one of the following techniques:

1. electrolysis
2. heating by an external source, evacuating, and then cooling in gas
3. plasma discharge
4. ion implantation
5. self-heating/cooling in gas by passing current.

All the samples loaded to any extent (any deuterium/ hydrogen-to-palladium ratio) were always observed to fog radiographic films kept in contact or a few millimetres away in air for a few hours. The intensity of fogging increased with exposure time and deuterium/hydrogen-to-palladium ratio. The phenomenon is completely reproducible, and the samples were observed to fog the films every time they were reloaded. No difference in fogging could be observed between hydrogen and deuterium loading. Figure 2 shows the contact image of a hydrogen-loaded palladium foil of 100-μm thickness and 10- × 34-mm dimension.

Effect of Enveloping Gas

Strong fogging was observed only in the atmosphere of air or oxygen. Weak fogging was observed in the atmosphere of hydrogen. Fogging was not observed in the atmosphere of other gases and vacuum. Table I gives the results obtained under various enveloping gases. The density specified in the tables is averaged and normalized to 24-h exposure duration.

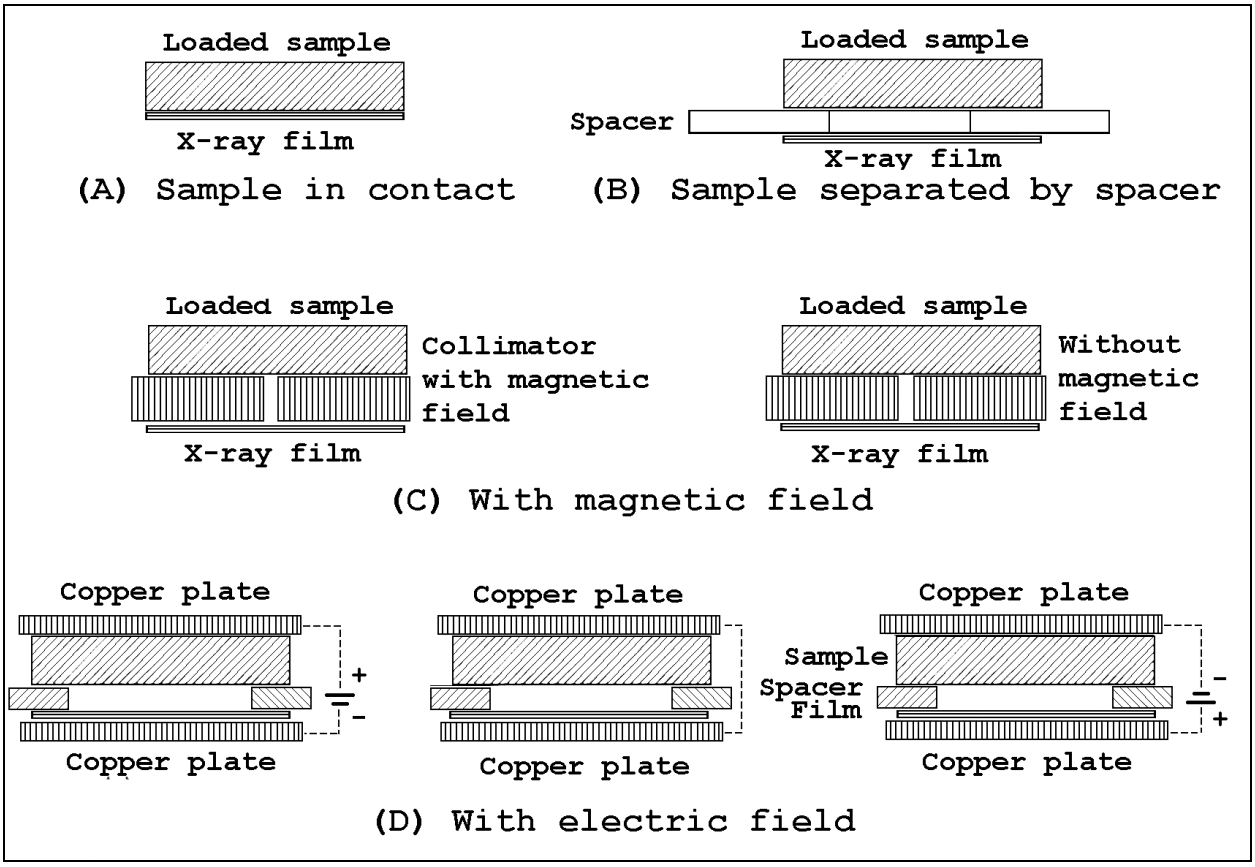

Fig. 1. Schematic of autoradiography.

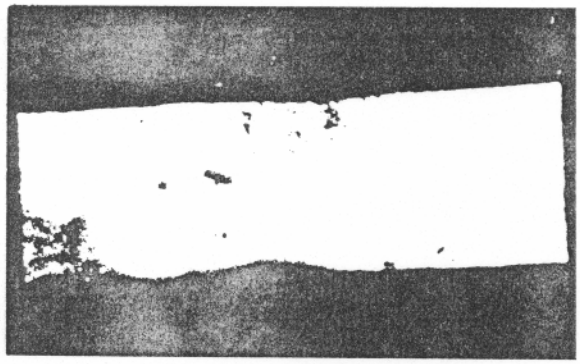

Fig. 2. Contact autoradiograph of a palladium foil (100 μm thick, 10 × 34 mm).

Other Metals and Loading Gases

No fogging was observed when palladium was loaded with helium (ion implanted). Among the other metals, weak fogging was observed with nickel and to a lesser extent with titanium. Table II tabulates the results.

Fogging in Presence of Fields

Application of a cross magnetic field (as shown in Fig. 1c) prevented the film from getting fogged. On the other hand, fogging of the film was enhanced when the electric field of either polarity was applied between-the sample and the radiographic film (Fig. 1d). Figure 3 shows the images of the loaded samples with different electric fields. Table III shows the variation in fogging densities.

TABLE I
Effect of Enveloping Gas*

Enveloping Gas	Fogging Density
Air	0.23
Oxygen	0.13
Hydrogen	0.03
Vacuum ($\leq 10^{-2}$ Torr)	0.00
Nitrogen	0.00
Helium	0.00
Argon	0.00

*Distance between film and sample, 200 μm; exposure time, 96 h; deuterium-to-palladium ratio, 0.5.

TABLE II
Other Metals and Loading Gases*

Metal	Loading Gas	Deuterium/Metal Ratio	Exposure Time (h)	Fogging Density	Reproduction (%)
Palladium	Hydrogen	0.5	48	0.23	100
Palladium	Deuterium	0.5	96	0.23	100
Palladium	Helium	Not known	96	0.00	---
Nickel	Hydrogen	0.2	96	0.10	50
Titanium	Hydrogen	1.8	96	0.00	---
Titanium	Deuterium	1	48	0.01	≈50
Zirconium	Hydrogen	Not known	48	0.00	---
Hafnium	Hydrogen	Not known	48	0.00	---
Copper	Hydrogen	Not known	48	0.00	---
Steel	Hydrogen	Not known	48	0.00	---

*Distance between the samples and film, 200 μm; enveloping gas, air.

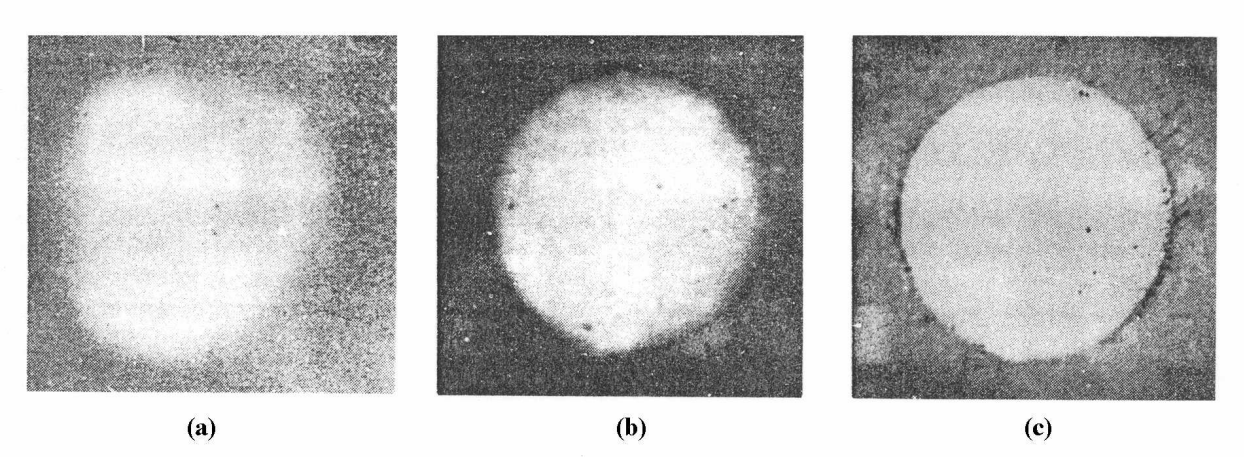

Fig. 3. Autoradiographs of palladium planchets with electric fields: (a) no field, 1.2-mm gap. (b) with -100 V, 1.2-mm gap, and (c) with +100 V. 1.2-mm gap.

Effect of Filters on Fogging

Several filters were employed between the film and the sample. The filter was used in place of the spacer as shown in Fig. 1b. The reduction in fogging density is shown in Table IV. The most important of these results are plotted in Fig. 4. The most startling observation is that while the emission is able to transmit through several layers of papers, it is unable to cross even a very thin metal foil. Also, the transmission through polyester foils (having approximately same average *z* as paper) was much less than that of paper. Figure 5 is the picture of both sides of a loaded palladium wire, one side of which was in contact with the film and the other side of which was seen through a step filter (10 steps), i.e., up to 10 layers of black paper.

Other Detection Techniques

Several other optical and nuclear techniques were utilized to determine the nature of the radiations. The results obtained are tabulated in Table V. In none of the techniques, except the thermoluminescent dosimeter, was the positive signal (strong enough to fog the film) obtained. It is to be noted that the thermoluminescent dosimeter, unlike other nuclear techniques, can detect emissions from energy as low as ~3-eV level.

TABLE III

Effect of Fields on Fogging*

Field Type	Fogging Density
No field	0.11
Electric field (+) 100 V	0.85
Electric field (-) 100 V	0.76
Cross magnetic field (10^{-3} T)	0.00

*Distance between film and sample, 1200 μm; exposure time, 41 h; deuterium-to-palladium ratio, 0.5; enveloping gas, air. The film was always at ground potential. The field was much lower than any breakdown/corona threshold.

DISCUSSION

A radiographic film can be fogged by various means. All these means (known to us) are given in the following discussion in reference to the fogging we observed.

Effect of Electromagnetic and Ionizing Radiation

Optical, Ultraviolet, and Near Infrared Radiation

The fogging here is not due to optical, ultraviolet (UV), or near infrared radiation for the following reasons:

1. The radiation/emissions could not cross thin glass and fused silica filters, which were transparent to optical, UV, and near infrared radiations.
2. The emissions could cross even 10 layers of black paper, which were completely opaque to these radiations.
3. None of the sensitive optical radiation measuring devices (at least two orders of magnitude more sensitive than X-ray film) showed any signal.

X-Rays and Gamma-Rays

The emissions/radiations are not X rays or gamma rays as they (emissions) were unable to cross 0.3 mg/cm^2 aluminized polycarbonate, but they were able to cross 80 mg/cm^2 paper. Also, none of the high- (as compared with X-ray film) sensitivity X-ray/gamma-ray measuring techniques (e.g., high-purity germanium detector) showed any presence of X rays/gamma rays above threshold.

TABLE IV
The Effect of Filters on Fogging Density*

Description of Filter	Thickness (μm)	Thickness (mg/cm^2)	Fogging Density
No filter	---	---	0.23
Air	1200	1.6	0.11
Aluminized polycarbonate	2	0.3	0.00
Beryllium	7.5	1.4	0.00
Pyrex glass[b]	100	20	0.00
Fused silica[c]	1000	200	0.00
Palladium foil	30	36	0.00
Polyester film, 1 layer	25	3.3	0.04
Polyester film, 2 layers	50	6.6	0.03
Polyester film, 3 layers	75	10	0.00
Undeveloped X-ray film	200	25	0.02
White paper,[d] 1 layer	85	8	0.22
White paper,[d] 4 layers	340	32	0.18
White paper,[d] 7 layers	595	56	0.08
White paper,[d] 10 layers	850	80	0.05
Yellow paper,[c] 1 layer	85	7.7	0.19
Yellow paper,[c] 4 layers	340	30.8	0.15
Yellow paper,[c] 7 layers	595	53.9	0.10
Yellow paper,[c] 10 layers	850	77	0.02
Black paper,[c] 1 layer	75	6.3	0.18
Black paper,[c] 4 layers	300	25.2	0.08
Black paper,[c] 7 layers	525	44.1	0.07
Black paper,[c] 10 layers	750	63	0.03

*Exposure time, 91 h; deuterium-to-palladium ratio. 0.5; enveloping gas, air.
[a] Dark spots with average fogging density of 0.013 were observed, but these were due to pinholes in the filter.
[b] The filter was transparent to optical radiations.
[c] The filter was transparent from near infrared to UV until 1700 Å.
[d] These were ordinary glazed papers used for photocopying.
[e] These papers are used to keep photographic films in light-tight condition and therefore are totally opaque to optical or UV radiations.

Radio-Frequency Radiations

The films used for radiography were subjected to radio-frequency radiations of several kilowatts emitted from a Marx bank and a capacitor bank. No fogging was ever observed, indicating the films were not sensitive to these radiations.

Electrons or Beta Particles

Though the emissions/radiations were found to be responding to electric and magnetic fields, these are not electrons or beta particles because of the following reasoning. The emissions were found to cross ten layers of paper; therefore, if they were electrons, their energy should be > 100 keV. These electrons (energy > 100 keV) should be able to cross beryllium (1.4 mg/cm^2) or aluminized polycarbonate (0.3 mg/cm^2) filters, but they were observed to be completely stopped. The gas (argon) flow proportional counter also failed to record any signal—even though it was at least two orders of magnitude more sensitive than X-ray films (for ionizing electrons or beta particles).

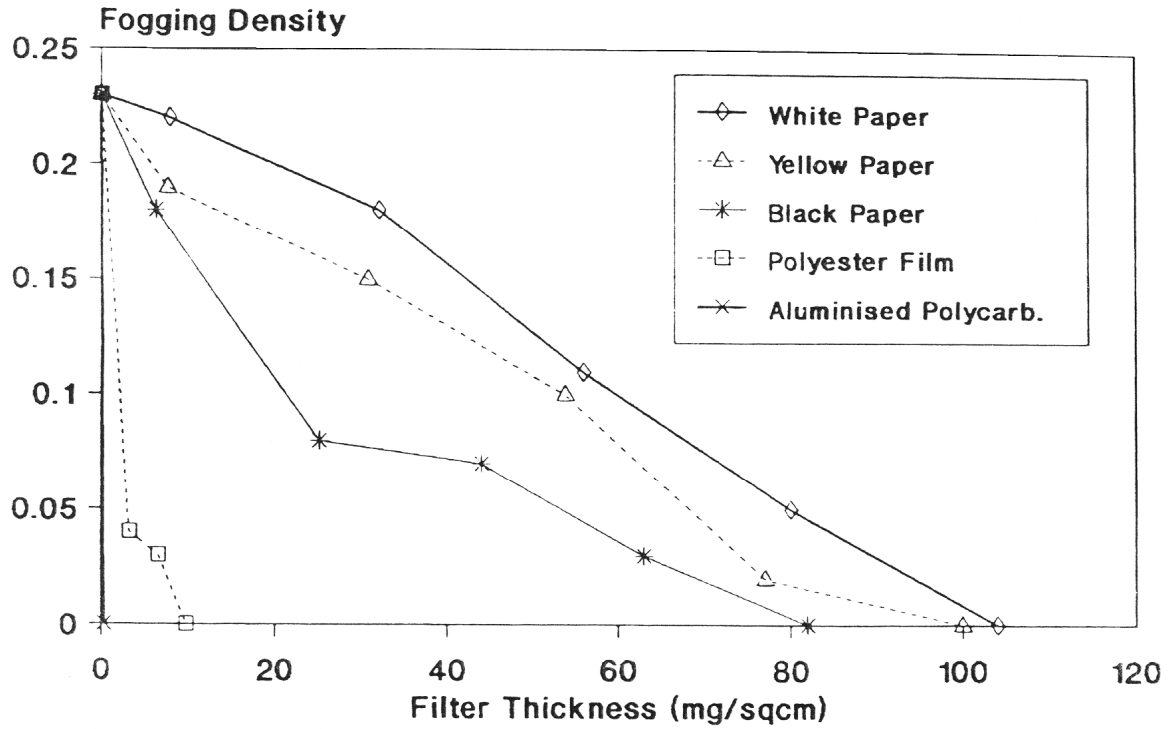

Fig. 4. Fogging of the films by emission(s) from palladium hydrides/deuterides through different filters.

Charged Particles

Charged particles (e.g., H^+ or proton, T^+ He^+ or alpha, etc.) have very low ranges. Their energies should be several mega-electron-volts to cross several layers of papers. If these particles were emitted, they should have been easily detected by several other techniques, which were much more sensitive as compared with the film (such as solid-state nuclear track recorders, film covered with ZnS scintillator, and gas flow proportional counters). Therefore, the fogging was not due to charged particles.

In general, the emissions were able to cross 80 mg/cm^2 of paper but were stopped by 10 mg/cm^2 of polyester film (having approximately the same effective *z*) and 0.3 mg/cm^2 of aluminized polycarbonate film; therefore, these emissions could not be any (known) ionizing radiation.

Thermal or Fast Neutrons

The films were found to be insensitive to fast or thermal neutrons (tested until 10^3 n/s·cm^{-2}). The samples were found not to be emitting neutrons when tested using a BF_3, detector setup (threshold ≈0.5 n/s) and fast neutron detector (threshold ≈0.01 n/s). Hence, the fogging could not be due to neutrons.

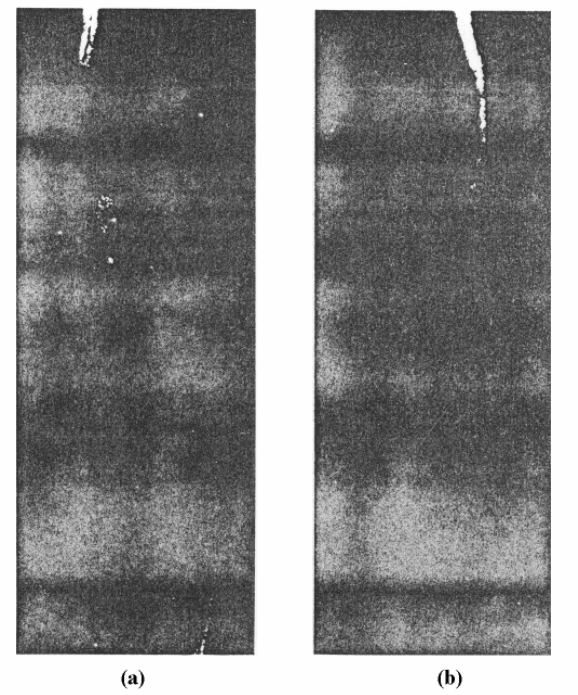

Fig. 5. Autoradiograph of a 125-μm-diam palladium wire: (a) image in contact and (b) image through a step filter (l to 10 layers).

TABLE V
Optical and Nuclear Radiations*

Description of Technique	Detecting Capability	Duration of Measurement (h)	Detection Threshold	Detected[a] Signal
Thermoluminescent dosimeter	UV, X ray, alpha, beta, gamma	72	0.1 mR	7 times
Photomultiplier tube, fused silica	Optical, UV	12	1000 ph	No signal
High-sensitivity photon detector	Optical	2	50 ph	No signal
Scintillator Detector NaI 75 mm	Gamma	10	700 ph	No signal
Detector NaI 3 mm	X ray	10	500 ph	No signal
High-purity germanium	X ray, gamma	10	60 ph	No signal
Scintillator detector, organic	X ray, alpha, beta, gamma, fast *n*	10	Variable	No signal
BF_3 detectors	Thermal *n*	10	500 n	No signal
Screen and X-ray film	X rays	96	1 mR	No fogging
Imaging plate	X ray, beta, gamma	96	Few pt	No signal
Air ionization chamber	Ionizing radiations	10	0.03 pA	No signal
Gas flow Proportional Argon	Ionizing radiation	10	120 ct	No signal[b]
Gas flow Proportional Air	Ionizing radiation	Detector unstable		
Differential Geiger Müller 12-μm polycarbonate window	Ionizing radiation	10	150 ct	360 ct[c]
Nuclear track Recorders CR-39	Charged pt	96	50 pt	No signal
Nuclear track Recorders LR115		96	100 pt	No signal
Surface barrier	Charged pt	10	10 pt	No signal
Film covered with ZnS scintillator	Charged pt, X rays, etc.	10	100 pt	No signal

* Hydrogen-to-palladium ratio, 0.5.

[a] All the detectors were calibrated by appropriate nuclear/optical sources, which fogged the films. The signal obtained from the detectors, using these sources were at least two orders of magnitude higher than the threshold (ph = photons, n = neutrons, and pt = particles).

[b] The phenomena does not occur in an argon atmosphere.

[c] Signals (counts) obtained are too small and are inadequate to fog the film (ct = counts).

Physical Effects

Pressure

All photographic films are pressure sensitive. However, pressure of several tens of bars are required to fog the films. These may be locally generated by scratching, etc. In our case, the film was not fogged due to these kinds of external pressure (e.g., sample pressed close to the film) for the following reasons: (a) none of the controls (unloaded palladium samples kept exactly in the same manner as the loaded samples) exhibited any fogging, and (b) fogging was observed even if the film was kept several millimetres away from loaded samples.

The film was also found to be fogged by strong sound/ultrasound (pressure) waves. The hydrogen-loaded material was also observed to produce these waves (perhaps due to microcracking). However the level of ultrasound/ sound produced by the samples was at least two orders of magnitude lower than the level required to fog the film.

Temperature

The films were also found to be temperature sensitive. The films used in the experiments were observed to be fogged when they were kept in contact with a vessel containing water at a temperature ≥60°C for 18 h.

However, the fogging of the film, kept in close proximity to loaded palladium, was not due to thermal effects. This possibility was ruled out by the following experiments:

1. The temperature of the loaded palladium sample was monitored for 90 h, and the detector did not record any change of temperature (<35°C).
2. A palladium sample (similar to ones that were loaded, but without loading) was electrically heated to 60°C and was kept in close proximity to the film (in the same way as the loaded palladium samples were kept) for 90 h. No fogging could be observed. Similar futile tests were also done with copper and nickel wires. It may be noted that the energy used to electrically heat the sample (for 90 h at 60°C) exceeded the total energy that could be produced by recombination of hydrogen in a palladium sample.
3. No fogging was observed through beryllium foil, yet fogging was observed through ten layers of paper, even though the thermal conduction through beryllium foil was >10 times higher than that of paper.

Chemical Effects

Hydrogen/Deuterium

The film was kept in the atmosphere of hydrogen/ deuterium gas for 96 h, but no fogging could be observed. Therefore, the film used here was not sensitive to these gases. The film may be sensitive to nascent hydrogen; however, hydrogen cannot remain in nascent form after passing through filters. Therefore, the fogging was not due to hydrogen/deuterium.

Hydrogen Peroxide

It has been claimed[1] that oxidizing freshly fractured/ abraded metal surfaces (in the presence of water vapors) could produce hydrogen peroxide (H_2O_2), though it is not clear to the authors how

H_2O_2 can be produced in a reducing atmosphere. However, if H_2O_2 is produced, it can fog the film.[1]

This possibility was ruled out by replacing a PdH_x sample by a H_2O_2 solution and observing the effect through layers of paper and polyester foils. No effect could be observed after two layers of paper or after one polyester foil. The emission from PdH_x was observed to cross even ten layers of paper and two polyester foils, and hence, the fogging is not due to H_2O_2.

The foils (of paper, polyester, etc.) used as filters were found to be unaffected (not bleached) after radiography. This rules out the production of any bleaching agent either of the reducing (nascent hydrogen) type or of the oxidizing type (hydrogen peroxide, ozone, etc.) by the loaded sample.

It may be noted that in all the radiographs the sample was observed to be imaged on the film. It is unlikely that any chemical could cross several layers of filter and form the image.

CONCLUSIONS

As the presence of air/oxygen in “envelope gas” gives strong fogging, probably the energy released during oxygen-hydrogen recombination is responsible for these emissions in some way. It is likely that this energy provides the necessary nonequilibrium conditions that are known to enhance anomalous effects. The energy of the emissions from palladium hydride appears to be small, as it is able to affect radiographic films (>2 eV) and thermoluminescent dosimeters (>3 eV) but did not ionize (>10 eV, average 30 eV/ion pair) gases.

The fogging (emission?) appeared to be enhanced by application of an electric field of either polarity, but it was suppressed by a crossed magnetic field. These emissions were previously assumed[2,3] to be of low energy (a few tens of electron volt electrons). This possibility is now ruled out as the emission was able to cross ten layers of paper.

The major difference between paper (highest transmission), polyester foil (low transmission), and beryllium foil (no transmission) is the degree of porosity of the material. However, as per our observation, the paper is not porous enough to let H_2O_2 or solutions of other chemicals pass after two layers. A small amount of hydrogen may pass through the filters, but hydrogen was found not to affect the film.

The phenomena, though most easily reproducible in palladium, are perhaps more universal and may also be occurring when H_2/D_2 is loaded into other metals.

All the mechanisms (known to us) which might have fogged the films, were considered and ruled out. Therefore, it is proposed that some new, unknown agency emitted from loaded palladium is responsible for fogging. It is felt that further study will not only give some additional insights for understanding this phenomenon, but it may also provide explanations for other anomalous effects observed in metal-hydrogen systems.

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