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UNEXPECTED ELEMENTS FROM SATURATED PALLADIUM HYDRIDES

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Abstract

This work reports on the transmutation of elements obtained in saturated palladium hydrides. Deuterium layers increased the screening effect which was calculated to be 13.2. After the localization process of the deuterium, atoms or molecules of mass number 1, 2, 3 and 4 were found at the inner of the samples. However, the more surprising phenomenon was observed when the palladium was perturbed by air reaching an incandescent temperature (850 °C). The maximum specific energy emitted from palladium resulted of about 8.1 kJ/cm^3. During the experiments a low neutron emission was measured from localized deuterium samples and no incandescent phenomenon and no neutron emission were observed when deuterium gas was substituted with hydrogen gas. Further, the Pd processed samples were analyzed by a scanning electron microscope and an electron probe microanalyzer finding many deformations and a high concentration of Al, C, Ca, Fe, Mg, Na, O, and Si, while Ca, Sn and Am were found on the cell windows.

I. INTRODUCTION

Since the Fleischmann and Pons, and Jones et al. experiments were announced[1,2], many cold fusion experiments have been performed all over the world utilizing different systems. Several experiments reported the generation of variable amounts of heat using heavy water in electrochemical cells. Other experiments, using molten salt, reported an excess power at least 600% over the input power[3]. Cold fusion reactions have also been observed by electrolysis in light water[4-5] where an excess energy was also observed[6].

The well-known fusion reactions providing excess heat and involving the deuterium atoms are the following:

$$D + D = {}^{3}He(0.8\ MeV) + n(2.5\ MeV)$$
$$D + D = T(1.0\ MeV) + p(3\ MeV)$$
$$D + D = {}^{4}He + \gamma(23.8\ MeV).$$

Instead, the fusion reactions responsible of the sun heat and involving the hydrogen atoms are the following:

$$p + p = D + e^{+} + \nu(1.442\ MeV)$$
$$p + p + e^{-} = D + \nu(1.442\ MeV).$$

In cold fusion experiments the excess heat measured was referred to the nuclear reactions instead other authors referred the excess heat to different phenomena. A chemical interpretation of generated heat was proposed by Rittner et. al.[7], while a mechanical interpretation was proposed by AbuTaha[8].

White et al.[9] studied the Coulomb barrier between two deuterons within the octahedral cage of crystalline palladium and they found that the repulsive forces were always higher than the corresponding forces between deuterons in molecular D_2 concluding that these forces cannot be used to explain the results of the excess heat.

The situation isn't very clear especially when many cold fusion experiments recorded only fusion products without any excess heat[10-15]. A theoretical work studied the boson tendency to clump deuterons in palladium[16] and it found that the attractive

force supplied kinetic energy to deuterons moving toward the center of the cluster and for cluster that included 24 deuterons the results gave good fit to the experimental results published by Fleischmann et al[1].

Then, to understand the involved processes it was very important to control the D concentration within the Pd crystal and to find a technique for improving the rate of the phenomenon.

From the power law[17] and from Jenoes results[2] the minimum distance between the nuclei for nuclear reaction is about 3pm. Besides, the minimum distance *d* for hot fusion reactions for central collision is calculed by:

$$d = 1.43x10^{-11}/E \qquad (1)$$

with *d* is the distance measured in cm and *E* is the energy between the deuterons given in units of 10keV. Assuming that the deuterium atoms within the palladium host crystal essentially are freely moving as classical plasma particles[18], the localization process of the deuterium inside the palladium could decrease the time for fusion reactions due to the screening factor increase.

For localizing gas atoms within the palladium host crystall, a new technique, used for direct silicon nitridation, was proposed using cells containing gas and excimer laser irradiation[19]. The deuterium layers were formed by means of the plasma created near the palladium surface as a result of direct laser irradiation of the target in deuterium atmospheres. An XeCl laser with wavelength of 308 nm and a photon energy of 4.02 eV was siutable to dissiciate deuterium or hydrogen atoms.

During the experiment many phenomena were observed: deformation of palladium, transmutation of elements (Al, C, Ca, Fe, Mg, Na, O, Si, Ca, Sn and Am), incandescence of the satured palladium, presence of atoms or molecules of mass number 1, 2, 3 and 4, emission of neutrons and breaking of cell windows.

II. EXPERIMENTAL SETUP

In this experiment five stainless steel chambers were used long 20, 25, 37, 29 and 14.5 cm called respectively A, B, C, D and E. Each chamber was equipped with a quartz window at a its end. A locally built XeCl excimer laser (308 nm) provided the

UV beam for the irradiations. Its characteristics have been described previously[20]. It is formed by two discharging regions utilizing two capacitor charge transfer type circuits with nominal primary and secondary capacitance of 60 and 25 nF, respectively. The output laser beam was 2 x 1 cm centered between the electrodes of 100 mJ, 20 ns (FWHM) pulse duration and 5 mrad divergence. The laser beam was led into the cells by a 30 cm focal length lens, as can be seen from Fig.1. A palladium wire was coiled and placed at the inner of chambers and supported by an inox strip or by two small supports. The laser beam was focused into the chamber with the focus position very near to the palladium, with an energy density of 0.5 J/cm^2 and 2 Hz repetition rate.

The neutron emission was measured with a Victoreen Neutron Survey Meter 488A and CR-39 polycarbonate detectors. An Ametek quadrupole gas analyzer MA100 was used in order to measure the residual particles present in the gas chambers. During the gas analysis the chamber containing the palladium was connected to a turbo-molecular vacuum system. After the experiments, the Pd wires were analyzed by a scanning electron microscope (SEM) and an electron probe microanalyzer (EPMA) which was able to detector the atoms present on targets.

III EXPERIMENT RESULTS and DISCUSSION

The meaningful experiments were performed with the chamber D (vol. 312 cm^3) and with a palladium wire of 13 cm long and 1 mm in diameter. A thermocouple was placed very near to the target to measure the temperature. When the chamber was filled up to 2310 mbar deuterium, after a few days the pressure fell down to 1920 mbar, which inferred an atomic concentration D/Pd$\approx$ 0.77. After D_2-loading for 30 days, the chamber was opened and the palladium came into contact with air. Its temperature only increased by less than 5 °C. After a day the chamber was refilled again with deuterium at 2310 mbar and successively the palladium was irradiated with a laser beam of about 0.5 J/cm^2 per 60 min/day at a repetition rate of 2 Hz. In this experiment deuterium gas was localized inside the palladium crystal. After D_2-localizing for 30 days the chambers were opened and some palladium samples became incandescent for about 30 s reaching a maximum temperature of 850 °C. When a new palladium sample (no-soaked) was placed inside the chamber with deuterium at 2310 mbar and successively the laser beam was applied, the total

pressure did not change sensitively and after 20 days the total pressure only fell down of a few mbar. This behavior can be explained by the laser action that favoring the layer formation it did not allow to the deuterium to come in inside the palladium. The maximum temperature value reached in these experiments remains very interesting as well as the role of the air. However, similar behaviors were observed by Ray et al.[21] when their cathode came in contact with air and the temperature increased only tens degrees. Temperature of about 475 °C were found in experiments with D_2-loaded nickel samples but with a 50 W heater power input[22]

To get success the laser intensity and the spot position were also important as well as the loading and the localizing time duration. Then, many attempts were done on D_2-loading days and on D_2-localizing days. Experiment with less than 30-loading and 30-localizing days were performed and in this case the palladium did not become incandescent. When the loaded palladium with D_2 for 30 days was not in contact with air but the chamber only was refilled with D_2 up to 2310 mbar and successively the laser beam action was applied for 30 days, no Pd-incandescence was observed when the chamber was opened. Other experiments were performed with the chamber called C but with 35 loading days, 1 day in contact with air and 35 localizing days. In this case small explosions were observed when the chamber was opened. Several experiments were also performed with chamber B but only with much more than 35 loading days and much more than 35 localizing days. Only some palladium samples exploded when they came in contact with air.

The residual gas contained into the no-exploded palladium was analyzed by the quadrupole gas analyzer MA100. The chamber gas was pumped up to 10^{-7} bar by a turbo-molecular pump and its residual gas was successively analyzed. From Fig. 2 it is possible to observe the presence of particles of mass number 1, 2, 3, 4, 20 and 21. Instead the gas analysis of the exploded sample show a concentration of mass number 3 higher than the concentration of mass number 4, Fig.3. The analysis of the chamber without the palladium is shown in Fig.4, where the peaks relative to mass number 3, 4, 20 and 21 are absent although the chamber was contaminated by D_2 gas. The presence of peak 20 might be due to the formation of heavy water while the peak 21 might be due to the heavy water formed with T atoms.

In order to understand the influence of the air, the residual gas of the Pd samples was analyzed after contact with only nitrogen or only oxygen. Namely, 1 bar nitrogen gas was put into the chambers having the palladium with D_2 localized and

after one hour the residual gas was analyzed (see Fig. 5). The percentages of the particles with mass number 3 and 4 were higher than the percentage of mass number 1 and 2, while the peak of mass number 21 was absent. Instead, when 1 bar oxygen was put into the chamber containing the same Pd sample and after one hour the gas was analyzed, the percentages of the particles with mass number 3 and 4 were again higher than the percentage of the mass number 1 and 2, see Fig. 6. In this last two experiments the percentage of mass peak 20 remained the same.

As Ray et al.[22] have found the oxygen plays a vital role in the experiment. It reacted likely with the deuterium present on palladium surface causing the come out of particles present in the target. The gas analysis of the chamber containing palladium for only 30 loading days, so without laser action, were very interesting. These experimental results were very similar to those obtained from chamber without the palladium and shown in Fig.4.

Even if this last result was unexpected especially for the absence of the molecule D_2, this behavior could be ascribed to the absence of layers that did not allow the localization of the deuterium, and at 10^{-7} bar of total pressure the residual gas easily comes out from sample. Instead in palladium samples containing deuterium localized, nuclear reactions could be occurred due to the formation of layers and as a consequence the production of new particles. This hypothesis was justified by neutron measurements. In fact, different values of emitted neutron were found from chambers having palladium loaded with D_2 (#1), from chambers having palladium with D_2 localized by layers (#2) and from chambers having only 2310 mbar of D_2 without palladium (#3). These measurements were performed during the night in order to reduce the noise influence due to electromagnetic irradiation of laser discharges during the day. Each chamber was tested many times (>6). The average counts were:

#1 21.96 ± 0.48 neutrons/hour (Chambers having D_2-loaded palladium)

#2 22.56 ± 0.79 neutrons/hour (Chambers having D_2-localized palladium)

#3 20.76 ± 0.73 neutrons/hour (Chamber having only 2.3 bar D_2)

Although the low count values, there was a difference among the three cases. By considering the experimental set up and the Victoreen neutron meter position, the

total neutron emission from targets is 20 times higher than the measured values. However, even if the neutron flux was very low and statistically small meaningful the average values obtained were different and the value of the experiment #3 was supposed to be due to the ambient neutrons. In this experiment the total ambient neutrons was not investigated as well as their energy. The value measured in the experiment #2 seems to be higher of about 1.80 ± 1.52 n/h than the value measure in the experiment #3 and, and the total neutron flux was estemed:

$$A = 119x\ 10^{-3}\ \ n/s$$

This result can be ascribed to the screening effect which favored the tunnel process through the Coulomb barrier[17]. In fact, considering the total deuterium atoms absorbed, $D_i = 5.23\ x\ 10^{21}$, the classical collision frequency for deuterons was:

$$f = 1.4\ x\ 10^{-8}\ n_i / T^{3/2} = 1.4\ x\ 10^{17} s^{-1} \qquad (2)$$

where n_i (5.23 x 10^{22} cm^{-3}) is the deuteron density and T the temperature measured in eV.

The probability for a fusion reaction[23] at the above frequency is $A / fD_i = 1.6x10^{-40}$. This value is 34 times higher than the D-D fusion rate per second for D_2 molecule[2]. By the Boltzmann expression.

$$exp(-E/KT) \qquad (3)$$

the above probability is reached at a room temperature of 0.03 eV with deuteron energy of 2.75 eV. From Eq. (1) the minimum distance for central collision of 3 pm is reached for energy of 470 eV. This result can be explained considering the scrining factor, namely the distance d is modified to

$$d = (1/S)^2\ 1.43x10^{-11} / E \qquad (4)$$

Therefore, by Eq. (4) a screening factor $S = 13.2$ was reached due to the layers formed on the palladium surface.

The lower neutron emission measured from loaded palladium, #1, (1.16x20 n/h) could be explained by the absence of layers that did not localize the deuterium atoms inside the palladium.

The layer formation might contribute also to increase the plasmon energy in the Preparata's plasmon model increasing the fusion reaction rate[24]. Other experiments were performed utilizing CR-39 polycarbonate detectors to investigate the neutron generation. In this case three chamber were used: A, B and D containing only D_2, D_2-loaded palladium and D_2-localized palladium, respectively. In each chamber two CR-39 were placed inside. After 30 days the detectors were developed and analyzed and the results were:

Chamber A = 1 tracks/mm
Chamber B = 1.2 tracks/mm
Chamber D = 1.5 tracks/mm

However, the difference in the density of the measured tracks between the chambers A, B and D was meaningful. It indicates that 0.5 neutrons/mm^2/month were detected by the CR-39 surface.

No incandescent phenomena as well as no neutron emission were observed when the experiments were performed with hydrogen gas instead of deuterium

During the experiments X-ray films Phil-X 30 were placed near the chambers and no radiations were recorded as well as no γ ray emission was observed from processed Pd samples by a γ-spectrometer which analyzed a energy spectrum up to 300 MeV.

The maximum energy emitted from incandescent palladium was estimated using a high current power supply connected to a 13 cm long new palladium wire, see Fig. 7. Fixing the current at 30 A the temperature and voltage values were recorded. The initial voltage and temperature were 0.6 V and 25 °C, respectively. The sample became incandescent (850 °C) after 9 seconds and the voltage increased up to 1.4 V. To maintain the 850 °C temperature the current was lowered at 18 A and the voltage was established at about 1 V. From these data the total emitted energy from the sample was estimated to be about 810 J and then the specific energy emitted from palladium during the experiment with the cell D results of 8.1 kJ/cm^3. However, this excess temperature for 30 s was not observed before.

All processed palladium samples that did not explode were analyzed with the SEM and the electron probe. By this investigation either the samples processed with D_2 or the samples processed with H_2 showed modifications of their surfaces. Similar results were obtained in liquid electrochemistry[25]. The deuterated samples showed in particular many pits, see Fig 8. Aiming the electron probe on the pit many elements were recorded, Al, C, Ca, Fe, Mg, Na, O and Si, Fig 9. Fig 10 shows the spectrum of virgin palladium.

Transmutation of elements was also observed on the windows utilized to seal the cells. Because more windows during the experiments were broken, it will seem useful to analyzed their surface by the SEM and the electron probe. From these analysis Ca, Sn and Am were found. Considering that the windows were placed at about 10 cm from the palladium sample then, the new particles present on the window surface ougth to have a sufficient energy to reach the window.

IV. CONCLUSION REMARK

In this work new phenomena during cold fusion experiments have been observed using an excimer laser. The layers formed by laser action introduced a screening factor which reduced the Coulomb repulsion. The most singular phenomenon was the incandescence of the palladium and the transmutation of elements in gas loading instead of liquid electrochemistry. The new elements found were: Al, Am, C, Ca, Fe, Mg, Na, O, Si and Sn. Some new elements were also found on the cell windows.

V. ACKNOWLEDGMENTS

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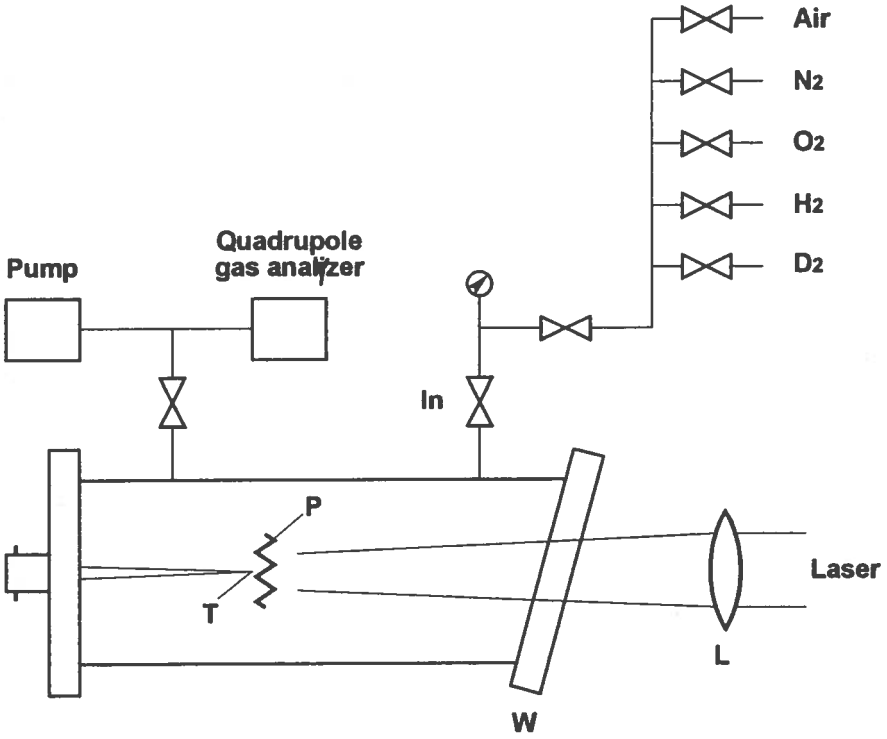

Fig. 1 Experimental setup. P: palladium sample, T: thermocouple, W: quartz window, L: 30 cm focal length lens.

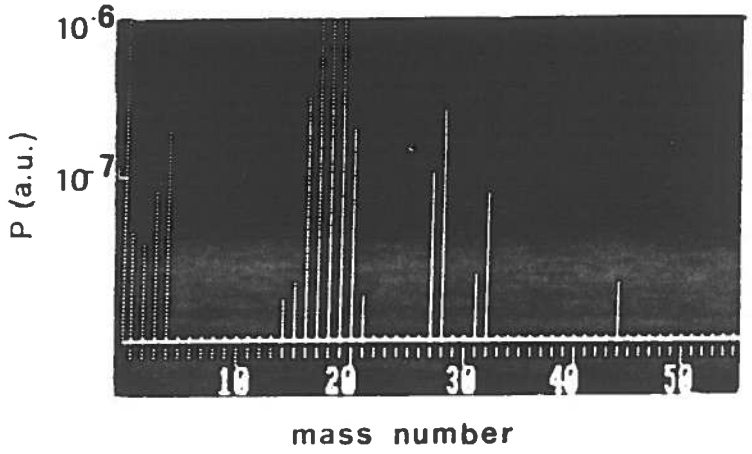

Fig. 2 Mass spectrometer results of the residual gas contained into no-exploded palladium sample with D_2 localized.

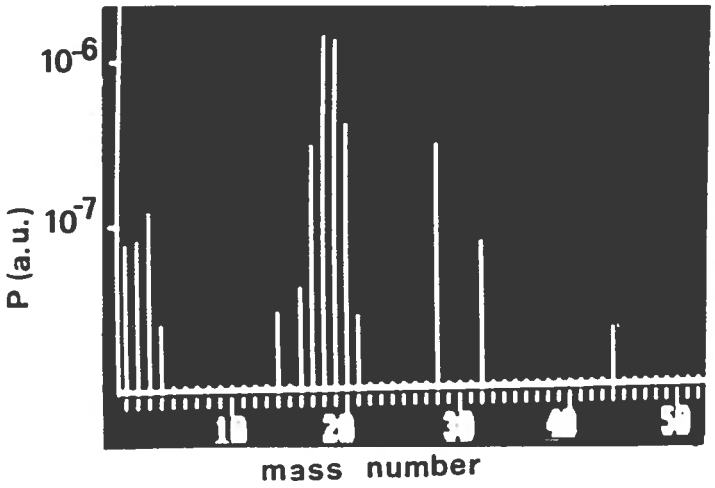

Fig. 3 Mass spectrometer results of the residual gas contained into exploded palladium sample with D_2 localized.

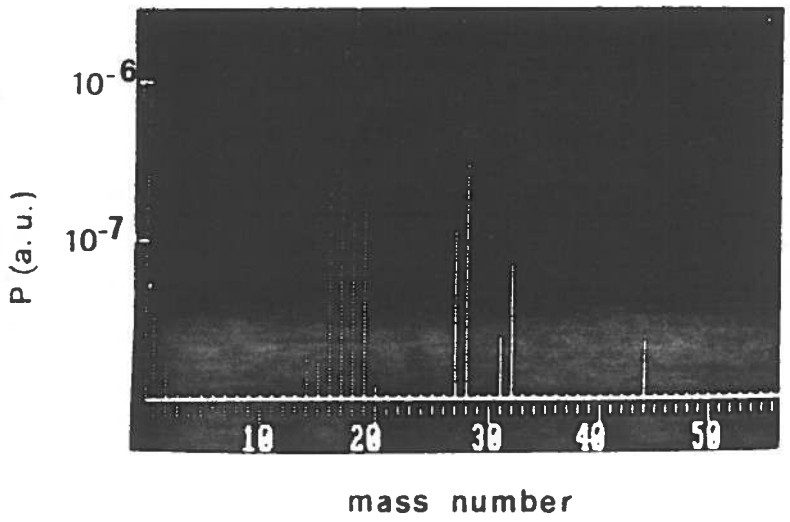

Fig. 4 Mass spectrometer results of the residual gas of the chamber contaminated by D_2 but without the palladium. The peaks relative to mass number 3, 4, 20 and 21 are absence.

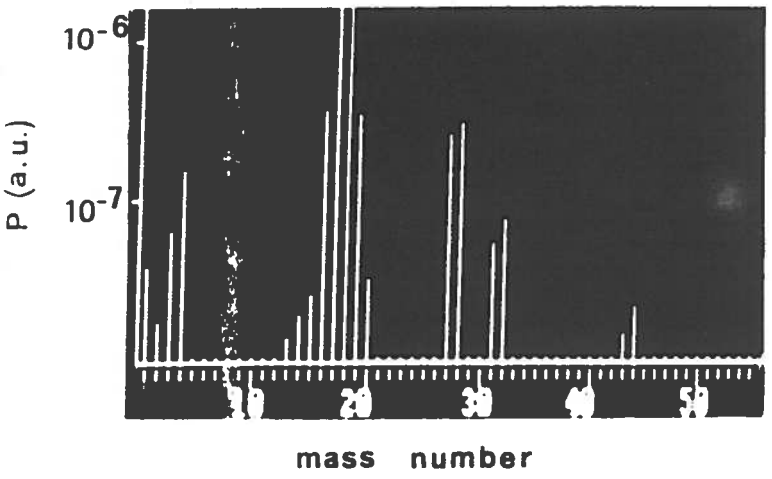

Fig. 5 Mass spectrometer results of the residual gas contained into the palladium with D_2 localized after one hour of contamination by N_2.

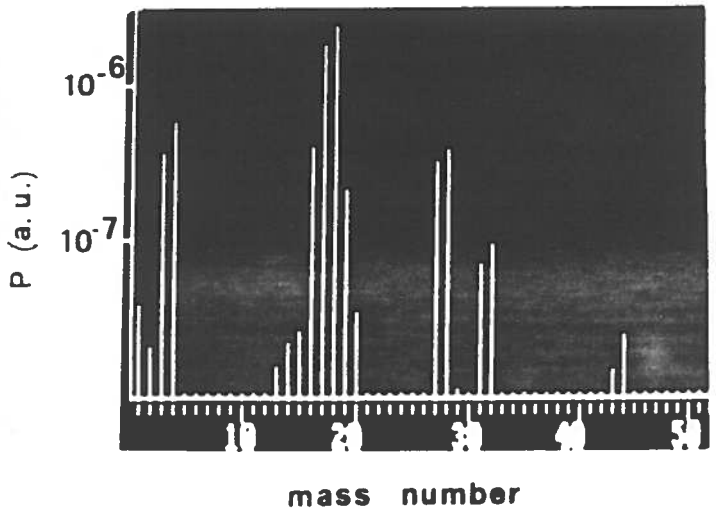

Fig. 6 Mass spectrometer results of the residual gas contained into the palladium with D_2 localized after one hour of contamination by O_2.

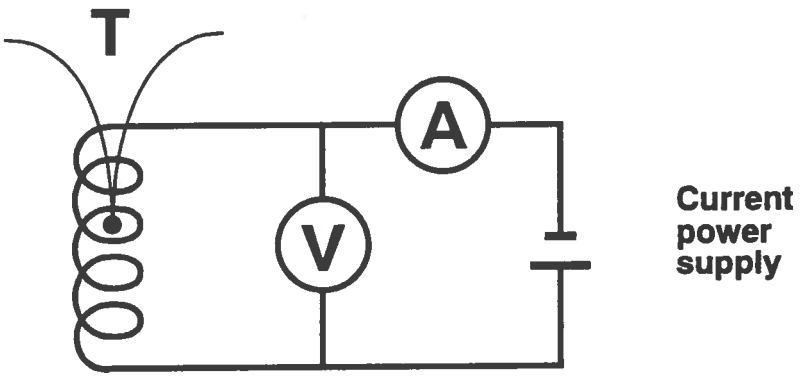

Fig. 7 Electric circuit for the measuring of the excess heat. T: thermocouple, V: V-meter and A: A-meter.

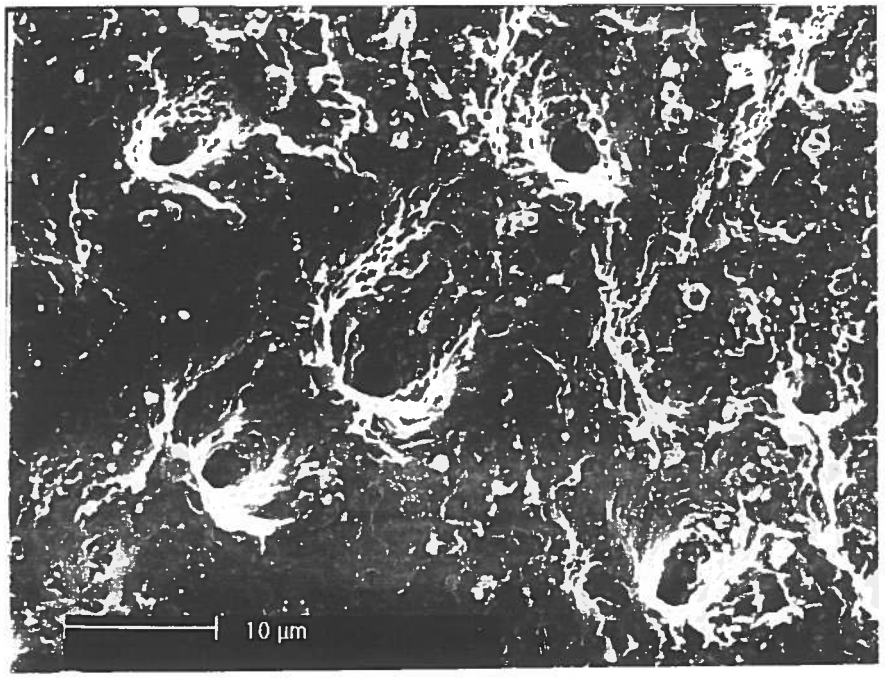

Fig. 8 SEM micrograph of a D_2 processed palladium sample. Many pits are present.

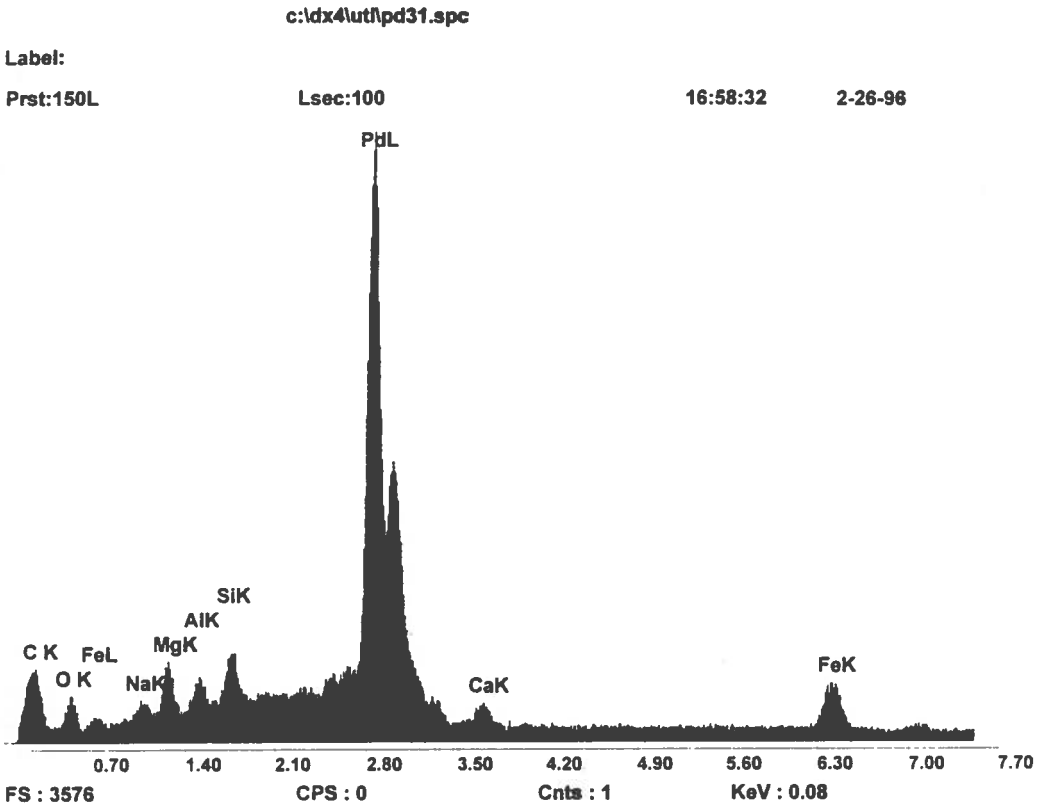

Fig. 9 Spectrum by electron probe of a processed palladium. The emission of the *K* and *L* line of many elements are present.

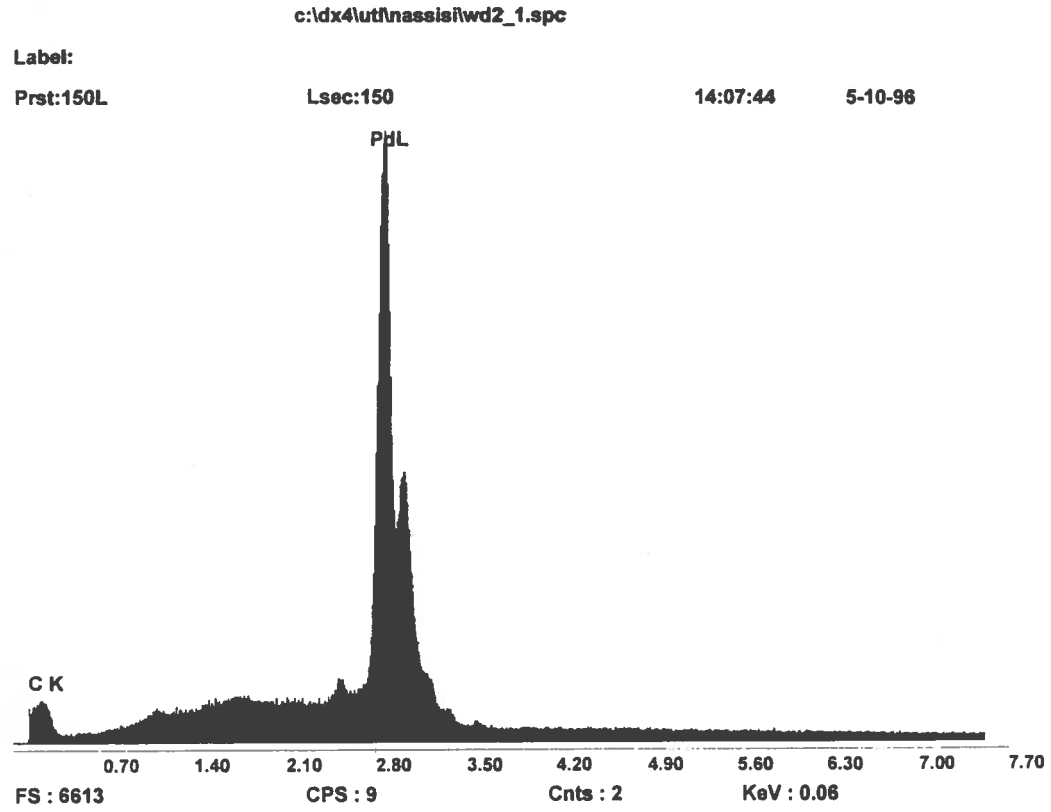

Fig. 10 Spectrum by electron probe of a virgin palladium.