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Analysis of nuclear transmutations observed in D- and H-loaded Pd films

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Abstract

New experiments of transmutation of elements in Pd films loaded with deuterium and hydrogen and processed by an excimer laser have been performed. The films were deposited on pure Si wafers having rough and smooth surfaces. The samples were put into inox chambers filled with D_2 or H_2 at a total pressure up to 6 bar. Some samples were irradiated by an excimer laser after the loading process. At the end, the samples were analyzed by a scanning electron microscope and an electron probe microanalyzer. The film morphology was strongly modified for the formation of many cracks. Many of them intersected mutually, often with the presence of grains in the middle. The grains contained the products of element transmutation independently of the loading gas. The grain dimensions and their density were higher in the films processed with the laser than in those processed without laser action. Films on rough substrates also presented a rough surface and a very low formation of cracks, but it was not possible to detect other elements apart from Pd or Si. The samples loaded with hydrogen showed a higher concentration of cracks and of grains as well as of transmutation products.

Keywords: Hydrogen in solid; Excimer lasers; New elements

1. Introduction

In this work, we concentrated our attention on nuclei transmutation in palladium films. The nuclear transmutations were observed in bulk samples and also in loaded films [1–3]. Elements such as Mg, Cl, Fe, Al, Ca, K, and others in the PdH_2O electrolysis system [4] were found and radioactivity in PdD and Ni–H systems [5,6] was measured.

The element formation channels are still unknown and, because the thermal energy in play is very low to provoke nuclear reactions, only collective phenomena could justify the experimental results, which we suppose induce a high particle interaction probability.

The interaction of particles in condensed matter could be ascribed to coherent oscillation of the metal electrons (plasmon) [7]. Considering hydrogen or deuterium within a metal lattice in an ionic state, the oscillation of electrons generate an intense oscillating electric field which, in turn, affects the ions accelerating them. In this case, the distance between two ions reduces below 0.1 Å contributing to the increase in probability for nuclear reaction.

The increase of the probability of fusion in a Pd–D system could also be due to the deformation of crystalline lattice which increases the probability at least 2–3 orders of magnitude with respect to the probability of fusion on the surface of the lattice [8].

Phonon distribution can trigger plasma oscillations in H atoms cluster that separates positive and negative charges, producing free electrons in a condition to be accelerated and the nucleus can gain kinetic energy that can be sufficient to overcome the Coulomb barriers and as a consequence obtain a nuclear fusions [9].

In this work, in order to study the effect of the transmutation, we utilized palladium films deposited on rough and

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smooth Si wafers loaded with deuterium or hydrogen. Palladium was chosen because of its remarkable features in the absorbing gas. The use of Pd films, instead of bulk material, allowed us to know exactly the film's thickness which may be important for the easier understanding of the experiment results.

Lapped Si substrates with a rough surface were used in order to limit even the other two dimensions of the film. In this case, the evaporated Pd film was distributed in zones about 3 μm wide with an irregular surface following the roughness of the substrate profile. On the contrary, on smooth polished wafers, the whole Pd film surface was perfectly flat.

Following these considerations, we seek to understand the influence of the geometrical characteristics of the film, of the kind of loading gas and of the optical treatment on the experimental results. The measurements performed by means of the available instruments in our laboratories allowed to analyze in detail the film morphological modifications and to detect the formation of other elements apart from Pd, Si.

2. Experimental apparatus

The films were obtained by thermal evaporation of palladium wire (99.95% purity) from a tungsten boat on lapped (rough) Si(1 0 0) wafers and on polished (smooth) Si(1 1 1) wafers as substrates. The typical impurity concentration in the Pd evaporation charge was well below 50 ppm each for noble metals and other elements. The thickness of the evaporating films was continuously controlled by a Maxtek quartz microbalance in order to get the desired final value. The substrates were cut into pieces having an area approximately of 1 cm^2.

The reaction chambers were stainless steel cylinders having an internal diameter of 4 cm and a length of 20 cm. Before performing the experiments, the chambers were accurately washed with acetone and fluxed with pure nitrogen. A chamber end was closed by a quartz window to allow the ultraviolet (UV) laser beam accessing. In this work, we used four chambers and eight samples. In the same loading phase, each chamber contained two similar samples but only one was subject to laser process. The chambers were initially evacuated by means of a turbomolecular vacuum pump; then two of them (chambers A and T) were filled with deuterium and the other two (chambers B and F) with hydrogen. In this way, we covered all the possible combinations in dealing with rough or smooth films, D_2 or H_2 loading and laser or no-laser process. The typical impurity concentration in hydrogen is reported in the data sheet as 1 ppm for N_2 and water vapour and lower levels for other gaseous species. As far as the typical impurity concentration in deuterium is concerned they were <30 ppm for N_2, <15 for water vapour and lower level for other gaseous species.

Laser processing was performed after injecting the gas inside the chamber and awaiting a few days.

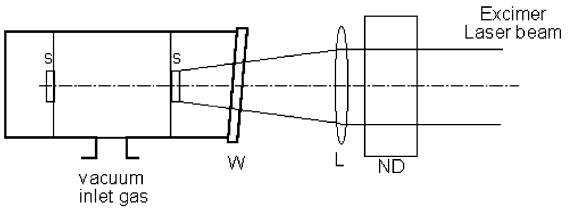

Fig. 1. Experimental set-up for laser irradiation. ND: Neutral density filters; S: sample, W: window; and L: convergent lens.

The laser was a UV excimer laser working at a wavelength of 308 nm. The UV light illuminating the samples in atmosphere of H_2 or D_2 favours multiphoton absorption processes, which localize the gas atoms within the Pd host crystal [10]. The laser beam was led into the chamber by a focalizing lens (L) and a set of neutral density filters. The experimental apparatus is shown in Fig. 1.

The scanning electron microscope (SEM) was a JEOL JSM 5410 LV equipped with an Oxford microanalysis system combined with a Link ISIS analytical software.

3. Experimental results

By evaporator device, we obtained eight samples formed by 110 nm thick Pd films deposited on two types of silicon pieces.

In the gas loading phase, the maximum gas pressure was fixed at 6 bar. After about 1 week of gas loading, each sample placed in the chamber near the quartz window was irradiated in order to create layers and to localize the gas atoms inside the palladium. The laser fluence was always lower than 25 mJ/cm^2 to avoid palladium ablation. The irradiation consisted of 400 laser shots at a laser repetition rate of 1 Hz per week and the laser process was repeated 5 times. On all processed samples, the total laser shots applied were 2000.

After opening the chambers, the samples were analyzed by SEM and by Energy Dispersive X-ray Spectroscopy (EDX) for studying the surface morphological aspect and the chemical composition. In all the measurements, among the revealed elements, Pd, Si and O were always found. In particular, the latter is due to natural oxidation of the silicon surface which occurs during the closing and the opening of the chambers.

None of the films deposited onto rough silicon surface showed significant morphological modifications, regardless of the loading gas and laser treatment. Fig. 2 shows their typical aspect: the film surface is highly irregular following the roughness of the substrate (with a typical size of about 3 μm) and it is evident the absence of cracks and grains. Only in very few points it was possible to find on the film surface some short cracks with a small grain (<1 μm in diameter). The microanalysis performed on these grains reported only Pd, Si and O.

On the contrary, all films deposited on smooth surface showed a morphological modification with the appearance of cracks, islands and grains. The loaded films were broken in islands separated by cracks evenly distributed across the surface (Fig. 3). In the intersections of cracks it was possible to observe localized grains, whose shapes corresponded to the contours of the cracks. This fact suggests a strong correlation between the break of the Pd film and the formation of grains (Fig. 4).

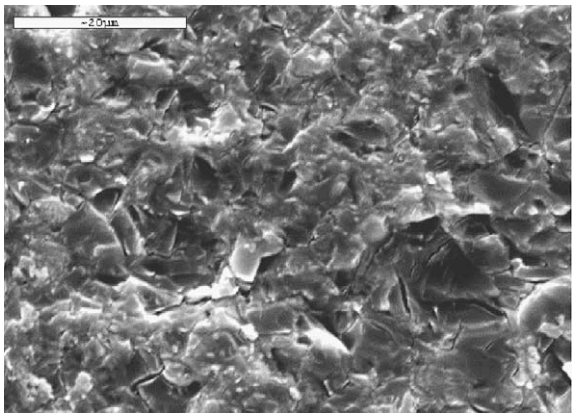

Fig. 2. SEM micrography of a Pd film deposited on the rough surface of a lapped silicon wafer, after the H_2 gas loading: the aspect was the same even for D_2-loaded film, regardless of the laser treatment.

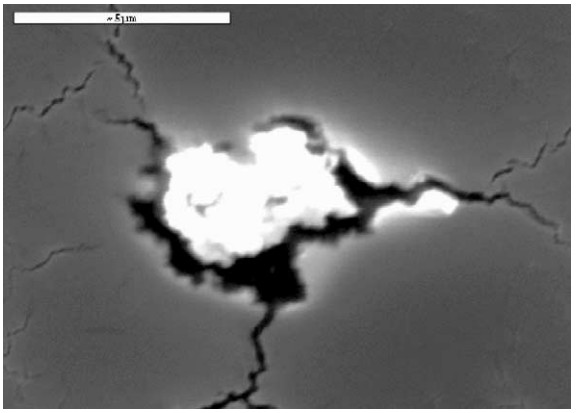

Fig. 4. SEM micrograph of a typical grain found at the intersection of cracks on the Pd film.

The microanalytical examination of the grains revealed in most cases the presence of other elements besides Pd, Si and O. The grain characteristics differ significantly depending on the type of loading gas and on the laser treatment to which the sample was subjected.

The main results obtained from the characterization of the films deposited on smooth surface are summarized in Table 1.

The deuterium loading resulted in the breaking of Pd films in islands whose size was about 7 μm. The laser treatment had the only effect to make the cracks more marked, and the

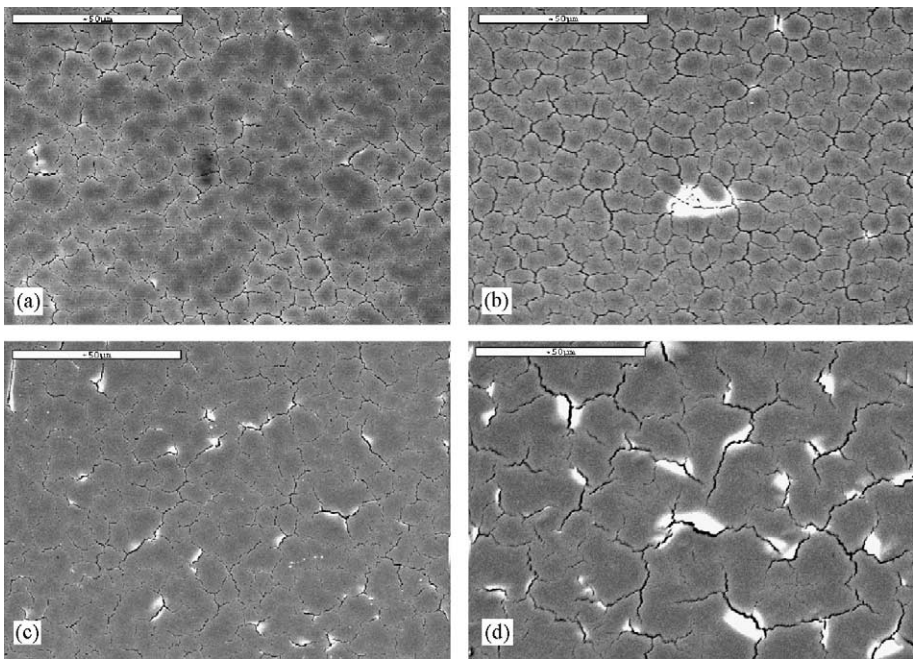

Fig. 3. Typical aspects of the surfaces of Pd film deposited on polished silicon wafers, after the completion of the gas-loading phase: (a) D_2 without laser treatment; (b) D_2 and laser treatment; (c) H_2 without laser treatment, and (d) H_2 and laser treatment.

Table 1
Sample characteristics, processing parameters and main morphological and analytical results

Sample characteristics, processing parameters					Morphological and analysis results										
Chamber	Sample	Silicon surface	Loading gas	Laser treatment	Island dimension (μm)	Grain density (n/mm^2)	Grain dimension (μm)	New elements found							
T	26	Rough	D_2	Yes	—	—	—								
T	27	Rough	D_2	No	—	—	—								
F	28	Rough	H_2	Yes	—	—	—								
F	22	Rough	H_2	No	—	—	—								
A	24	Smooth	D_2	Yes	10	10	1.7 ± 0.3	Ca	Fe						
A	25	Smooth	D_2	No	7	10	1.7 ± 0.3	Ca	Fe						
B	23	Smooth	H_2	Yes	15	10^4	3.6 ± 1.5	Ca	Fe	S	Zn	Ti	Cu	Cr	
B	29	smooth	H_2	No	10	10^3	1.6 ± 0.5	Ca	Fe	S	Ti				

Pd islands a little wider (about 10 μm) with the contours corresponding again to the cracks.

In deuterium-loaded film, the laser treatment had no effect on the grain characteristics. In fact, the density, the size and the chemical content of the analyzed grains were the same for both laser-treated and non-laser-treated film. The grain density was of about 10 grains/mm^2 of the film surface and the mean grain size was about (1.7 ± 0.3) μm. The X-ray analysis performed on the grains revealed the presence of only two new elements, Ca and Fe.

Even the hydrogen loading of palladium films deposited on smooth Si surface resulted in the formation of many islands. Their size was about 10 μm in the absence of laser treatment and about 15 μm for laser-treated samples.

The laser treatment had important effects on grain characteristics. The Pd film without laser treatment had a grain density of about 10^3 grains/mm^2 of the surface and the mean grain size was about (1.6 ± 0.5) μm value quite similar to the previous one. X-ray microanalysis in the grains revealed up to three new elements among Ca, Fe, S and Ti.

The laser treatment caused an increase in the density of the grains, in their dimension and in the number of chemical elements they contained. The grain density reached 10^4 grains/mm^2 and the size was about (3.6 ± 1.5) μm.

The new elements simultaneously found out by microanalysis in each grain are summarized in Fig. 5 which shows the density of grains as a function of detected elements for both laser and non-laser films. It is important to stress that scanning electron microscopy together with microanalysis system allows us to determine the presence of chemical elements in the small region of the sample but it cannot determine the concentration of new elements with respect to the Pd because of the very low density of grains.

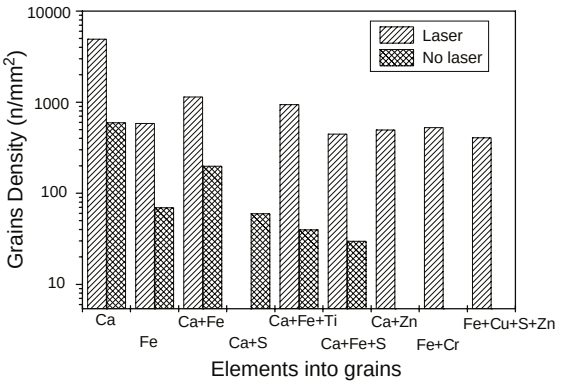

Fig. 5. Histogram showing the density of grains as a function of detected elements both for laser and non-laser films.

4. Discussion and conclusions

In this work, Pd films of the same thickness were deposited both on lapped (rough) and polished (smooth) silicon surfaces. After loading with H_2 or D_2 and processing with a UV laser beam, the films were subjected to a morphological and elemental analysis, that indicated the formation of new elements.

This result was strictly coupled to the transformation of a part of the film in grains, that occurred only at the intersections of cracks. In fact, these effects were clearly evident only in films, deposited on smooth silicon surfaces, that presented many cracks, whilst films deposited on rough surface showed neither cracks nor grains. In the latter case, the realization of a film with a discontinuous structure can limit the oscillation space of the plasmons and phonons, causing their damping. In this way, the enhancement of collective phenomena which we suppose to be the cause of nuclear reactions cannot be favoured.

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