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ANELASTICITY IN 20 keV PROTON-IMPLANTED TANTALUM

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Abstract: Internal friction measurements have been carried out versus temperature in a tantalum specimen after hydrogen implantation obtained by a proton beam of 20 keV.

The result shows a thin layer on the exposed surface of the specimen with a large concentracion of hydrogen with respect to the bulk concentration. From the experimental data is it possible to obtain some information about the »range« and the diffusion parameters of the implanted ions. The effect of »supertail« is also observed.

1. Introduction

The internal friction measurements on interstitially doped b c c metals are widely employed to study the diffusion mechanism and to obtain the diffusion parameters[1-5]. In this paper we report the experimental results concerning a tantalum specimen doped by the ion-implantation technique.

These results confirm the data available in the literature on the diffusion phenomena, as for example the activation energy and the diffusion constant. Moreover they allow to estimate the range of the implanted particles. From this point

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of view the analysis of the internal friction suggests a new procedure, additional to the more appropriate methods known in the literature[6,7], to estimate the range distribution.

2. *Experimental apparatus and procedure*

The tantalum specimens employed* in the measurements have the shape of a circular plate of 30 mm diameter and 3.2 mm thickness. The first one (in the following referred as S1) after annealing and degassing by a 800 °C thermal treatment in vacuum has been doped by means of a 20 keV proton beam extracted by a Duoplasmatron ion-surce[5,8]. The source and its equipment has been located on an insulated platform raised to a positive potential by a H. V. d. c. power supply (30 kV, 10 mA).

The extractor, the accelerating system (an einzel-lens of three elements, the first being the extractor), the analysing 60° magnet and the implantation set-up are grounded and placed in a horizontal disposition. The magnetic field intensity has been adjusted to focus the doping ions on the specimen. The beam analysis revealed that the spurious ions were less than 10%. The tantalum plate has been mounted in a vacuum chamber and has been surrounded by a cylindrical electrode at — 200 V, in order to collect the back-scattered ions. The current intensities crossing the tantalum target and the back-scattering electrode, have been recorded and afterwards integrated. During implantation the specimen temperature, measured by a copper-constantan thermocouple, never exceeded 200 °C. The doping and the back-scattering beam intensities were 100 μA and 40 μA, respectively. The specimens have been implanted on each side with a dose of 10^{18} ions/cm^2, which would correspond to an average atomic concentration of about 10^{-4}. After implantation the elastic energy dissipation coefficient, Q^{-1}, and the vibration frequency have been measured as a function of the temperature by means of a resonance technique[9], in which the flexural vibrations of the specimen are excited by an electrostatic driving force. The values of Q^{-1} are obtained by measuring the logarithmic decrement of the free vibrations. In the experimental set-up the tantalum plate was supported in a horizontal plane by three pins located in small holes on a nodal circle. The driving force was supplied by the electrostatic pressure between the specimen and a small electrode placed just above its center. The sample was enclosed in an evacuated container surrounded by a Dewar vessel in which liquid nitrogen could be frozen. Two of the supporting pins constitute a copper-constantan thermocouple with the specimen used as point. This arrangement allows to measure the specimen temperature with an accuracy greater than 0.2 K, introducing only a negligible additional damping. The contribution to Q^{-1} due to system supporting the plate has been evaluated to be less than $3 \cdot 10^{-7}$.

To make the implanted dose homogeneous, the internal friction measurements have been alternated with annealing treatments in vacuum.

For comparison sake the measurements carried out on S1 have been repeated on another tantalum specimen (S2), annealed at the same temperature and doped by an electrolytic hydrogen-charge (200 mA for 15 minutes).

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* Haynes Stellite Co 99.99% polycrystalline tantalum.

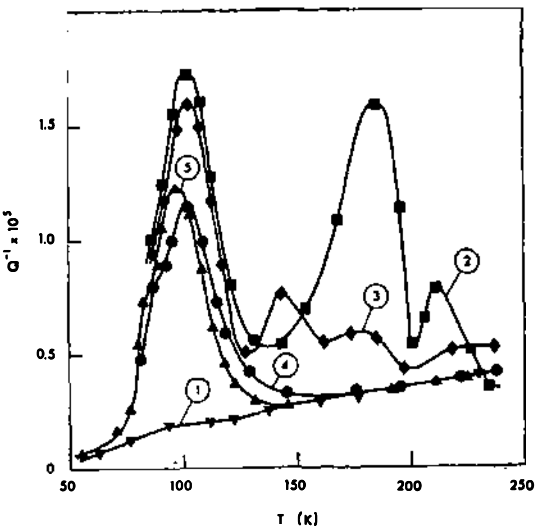

Fig. 1. Internal friction coefficient of S1 versus absolute temperature:

curve 1 (▼) after 10 hours at 800 °C,
curve 2 (■) after first implantation,
curve 3 (◆) after 6 hours annealing at 280 °,
curve 4 (●) after further 6-hours annealing at 280 °C,
curve 5 (▲) as curve 4 at a different resonance frequency.

3. *Experimental results and discussion*

After charging by ion-implantation, but without annealing, the internal friction coefficient Q^{-1} versus absolute temperature has been plotted in the range from temperature to liquid nitrogen temperature for a vibration frequency of about 71 kHz. The results are shown in Fig. 1 (curve 2). The curve shows a sharp peak around 100 K plus some maxima at higher temperatures caracterized by a marked instability in height and in the corresponding abscissa value, with respect to the annealing treatments. The effects of two successive annealings at 280 °C during 6 hours are shown by the curves 3 and 4 of the same figure. The low temperature peak may be attributed to the Snoek effect[10], namely to the stress induced by ordering of the interstitial hydrogen atoms occupying the tetrahedral sites, or alternatively, according to the more recent conclusions[11,12] (the problem is under discussion) the low temperature peak may be attributed to the hydrogen atoms which are trapped to oxygen interstitial and jump around their trapping centers. Let us assume the existence of the Snoek peak. According to the theory of C. Wert and C. Zener[13] in the hypotheses that jumps of the hydrogen atoms occur between tetrahedral sites, the following values for the diffusion constant D_0 and for the activation energy W have been obtained

$$W = 0.12 \pm 0.01 \text{ eV}, \qquad D_0 = 0.6 \cdot 10^{-5} \text{ cm}^2/\text{s}.$$

They are in agreement with the data available in this range of temperature[2].

The higher temperature maxima depend to some extent on the particular technique employed in doping S1, as confirmed by the behaviour of Q^{-1} versus temperature of S2 electrolytically charged, shown in Fig. 2. Considering both the low energy of the doping particles and the thermal recovery during the implantation, the influence of the radiation damage may be disregarded and then the maxima at higher temperatures can be attributed to a surface layer in the doped sample with a concentration greater than that in the bulk. This hypothesis is suggested by the behaviour after annealing (curves 3 and 4) and confirmed by the experimental data shown in Fig. 3. This diagram gives the Q^{-1} factor versus temperature after a second implantation followed by an annealing of 48 hours at a temperature of 220 °C (larger than the value reached during implantation). In Fig. 3 the curve 1 has been obtained measuring Q^{-1} while cooling the specimen to 80 K; the curve

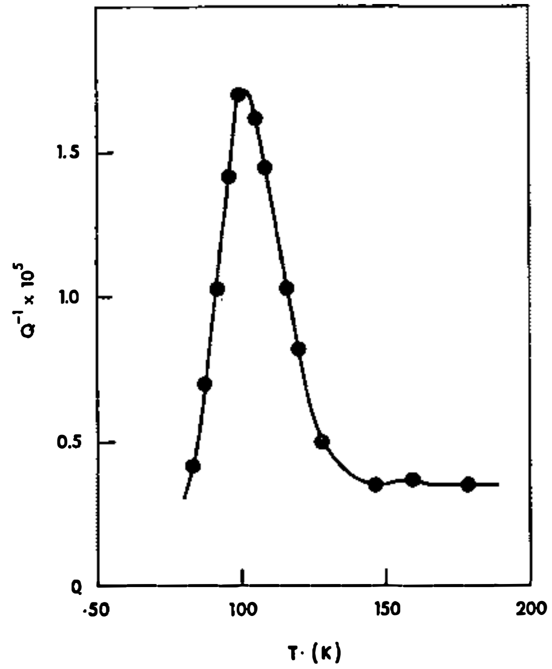

Fig. 2. Internal friction coefficient of S2 versus absolute temperature after electrolytic charging.

2 while subsequently warming it to room temperature. As shown by G. Cannelli and F. M. Mazzolai[14]) the maximum at higher temperature can be attributed to an $\alpha \to \alpha + \beta$ phase transition, that is, a hydride precipitation below a characteristic temperature T_c. The temperature T_c and the corresponding atomic concentration C are connected by the equation

$$\ln C = (2.0 \pm 0.2) - \frac{(0.102 \pm 0.005)}{k\,T_c}\ \mathrm{eV}.$$

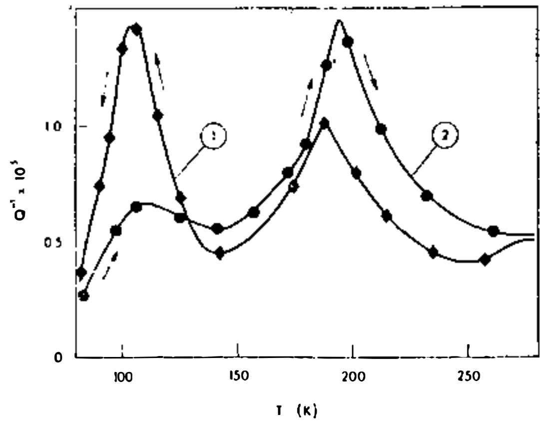

Fig. 3. Internal friction coefficient of S1 versus absolute temperature after the second implantation and a subsequent 48-hours annealing at 220 °C:
curve 1 (◆) cooling the specimen,
curve 2 (●) warming the specimen.

The evaluation of the average hydrogen concentration using the previous equation leads to an approsimate estimation of the projected range R_p of the implanted particles. By assuming, as usually done[7]), a layer thickness about twice the length of the projected range R_p, we may write

$$C = \frac{n_\mathrm{H}}{n_\mathrm{Ta}} \simeq \frac{10^{18}}{\varrho_\mathrm{Ta}\, 2\, R_p\, N/M_\mathrm{Ta}} \simeq 0.05,$$

where n_H and n_Ta are respectively the number of atoms of hydrogen and tantalum in the volume of the surface layer, ϱ_Ta and M_Ta the density and the atomic weight of tantalum and N is the Avogadro's number. We obtain

$$R_p \simeq 1.8\ \mu\mathrm{m}.$$

The theoretical value of the projected range from LSS theory[7,15,16] calculated taking into account only the electronic stopping power, is

$$R_{\mathrm{LSS}} \simeq 0.11\ \mu\mathrm{m}.$$

The discrepancy between R_p and R_{LSS} cannot be attributed to channeling effects (the sample is polycrystalline) or to the heating during implantation. In fact an annealing at a temperature lower than 250 °C gives no remarkable influence on the diffusion of the implanted ions as follows from the comparison between curve 2 of Fig. 1 and curve 1 of Fig. 3. The value of 1.8 μm obtained for R_p may be attributed to a supertail in the distribution of the implanted particles. Similar effects have been already observed and also theoretically expected in the theory of the »deep multistream diffusion« due to M. Sparks[17].

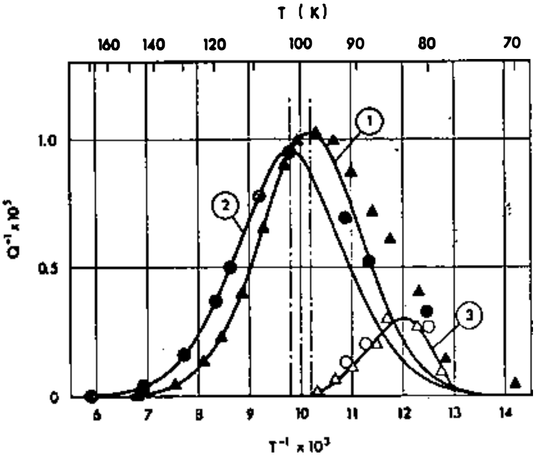

Fig. 4. Internal friction coefficient of S1 versus T^{-1} after subtraction of the background dissipation (points). The curves 1 and 2 are obtained by symmetry with respect to abscissa of Q^{-1}. The curve 3 represents the smoothed hunches shown in the left sides of the curves 4 and 5 of Fig. 1.

Let us now observe that the left sides of the curves 4 and 5 of Fig. 1 show a smoothed hunch in the range of temperature below 100 K. The phenomenon is better evidenced in Fig. 4 in which the experimental values of Q^{-1} (points of curves 4 and 5 of Fig. 1) are reported after subtraction of the background dissipation. In Fig. 4 the right side of the curves 1 and 2 are obtained by symmetry with respect to the abscissa of the maximum value of Q^{-1}. The curve 3 of this figure has been obtained by subtracting the theoretical values of Q^{-1} from the experimental points, to give evidence to the smoothed hunches in the left sides of curves 4 and 5 of Fig. 1.

This anomalous bump could be attributed chiefly to a precipitation effect due to residual concentration in the surface layer[14].

4. *Conclusions*

The experimental results show that the internal friction measurements on samples implanted by accelerated particles are a good tool for determining the diffusion parameters and for a rough estimation of the range of the particles.

The results concerning the diffusion parameters are in good agreement with the data available in the literature, whereas the range values differ from the theoretical previsions of LSS theory by more than one order of magnitude. These »supertails« seem to support the hypothesis of M. Sparks[17] which gives theoretical values greater than the LSS values.

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