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Diffusion Coefficient of Helium in Mo-Assessed by the Internal Friction Technique

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Abstract Diffusion behavior of helium in molybdenum was investigated by means of the internal friction method. An apparent relaxation internal friction peak associated with helium long-range diffusion was observed around 475 K at a resonant frequency of 56 Hz. In terms of the Gorsky relaxation model and the shift of the peak position with the measurement frequency, the activation energy and pre-exponential factor of the diffusion coefficient of the helium atoms in molybdenum were deduced as 0.63 eV and $6.5 \text{ cm}^2/\text{s}$, respectively.

Keywords: helium diffusion, molybdenum, internal friction

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1 Introduction

Plasma diagnostic systems are known to be very crucial for the international thermonuclear experimental reactor (ITER). Compared with the present conventional or superconductor tokamaks, both levels and fluxes of the H^+ , D^+ , T^+ , and α radiation in ITER will be much higher. Therefore it is impossible to diagnose the plasma directly through conventional optical windows or optical fibers. Metals, including beryllium^[1], copper^[2], molybdenum^[3], tungsten^[4] and stainless steel, are widely used as mirrors in experiments to diagnose the plasma. Because these metal mirrors stand nakedly in the high temperature plasma, it is impossible to avoid plasma irradiation.

In addition, high Z-materials tungsten^[5] and molybdenum^[6] are also leading candidates for the divertor materials in ITER due to their high melting point and high plasma erosion resistance. Under burning plasma conditions, plasma-facing materials (PFM) suffer irradiation of helium in addition to that of hydrogen isotopes. The behaviors of helium in metals are characterized by its fast migration through the lattice as well as very strong attractive interaction with defects such as vacancies, vacancy clusters, impurity atoms and even among themselves. So it is important to know the diffusion coefficient of helium in such metals acting as the plasma-facing mirror or divertor materials.

The internal friction measurement is very sensitive to the diffusion or movement of light atoms, such as H, He, C, and N, in metals mainly owing to two effects. One is the well known Snoek effect^[7], in which the short-distance diffusion of interstitial light atoms from one

site to a neighboring site in a body centered cubic lattice gives rise to the dissipation of mechanical energy. The other is the so-called Gorsky effect^[7], where the fast-moving light atoms migrate a long distance back and forth in the sample or in a grain of the sample. In general, C and N atoms exhibit the Snoek effect while H and He atoms exhibit the Gorsky effect, since the latter moves much faster than the former. There are many investigations into hydrogen diffusion in metals or alloys by measuring the Gorsky effect, such as hydrogen diffusion in V^[8], Ti^[8], Nb-Ta alloys^[9] and Zr^[10]. In this work, we investigated the helium diffusion behavior in molybdenum by the Gorsky effect.

2 Experimental details

The samples in our experiments were prepared as follows. Firstly, cold-rolled molybdenum sheets ($50 \times 5 \times 0.1 \text{ mm}$) were annealed at 1423 K for 6 h in a vacuumed quartz tube. Secondly, the Mo strips were heated at 1223 K for 10 h in a flowing high purity helium atmosphere in order to introduce helium atoms into Mo lattices.

The internal friction (IF) of the samples was measured on a vibration reed setup in the free decay mode with a strain amplitude of 4×10^{-6} and a heating rate of 2 K/min. The vibration direction was perpendicular to the largest surface of the Mo strip. The microstructure of the Mo sheet was characterized by a field emission scanning electron microscope (FESEM, Sirion 200 FEG). In order to obtain the fresh section of Mo, the pre-annealed pure Mo strip was heated at

573 K in a 300 MPa hydrogen atmosphere for 120 h and then quenched to room temperature, so that the Mo strips were brittle enough to be broken down. However, samples for IF measurements were never heated in hydrogen atmosphere.

3 Results and discussion

Fig. 1 gives the FESEM image of the fresh section of a Mo strip. It can be seen that the Mo strip has a layer-structure in which the layer plane is parallel to the largest surface of the strip. This kind of structure is frequently observed in the cold-rolled samples since the grains are elongated along the roll direction during the cold-roll process. In our case, the average thickness of the layer is about 2 μm .

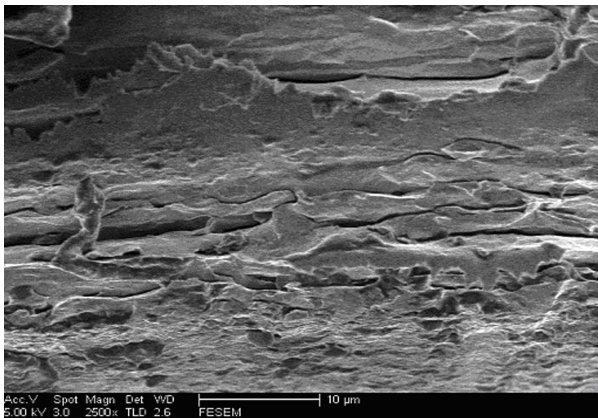


Fig.1 SEM image of the fresh section of pure Mo: the Mo strip exhibits a layer structure with an average thickness of about 2 μm

In Fig. 2, the temperature dependence of the IF and the square of the resonance frequency (proportional to the dynamic Young's modulus) for pure Mo and He-containing Mo strip are shown. In the pure Mo sample, the IF increases and the square of the resonance frequency decreases monotonously with the increase in temperature and no IF peak is detected in the temperature range from 300 K to 650 K. In the He-containing Mo specimen, however, a peak (designated as P_{He} peak) is observed around the temperature of 500 K at a measurement frequency of 113 Hz. Around the peak temperature, the square of the resonance frequency changes dramatically. Since no such peak is detected in pure Mo, this peak may be related to the impurities introduced in the annealing process, such as helium, oxygen, and nitrogen atoms. However, the association of this peak with the oxygen and nitrogen impurities can be excluded, because oxygen can not dissolve in Mo and the N Snoek peak in Mo appears around a temperature of 590 K for the measurement frequency of about 100 Hz, which is 90 K higher than the peak temperature of the present peak [12]. So it can be concluded that the P_{He} peak appearing in the Mo-He samples must be associated with helium in the Mo strip.

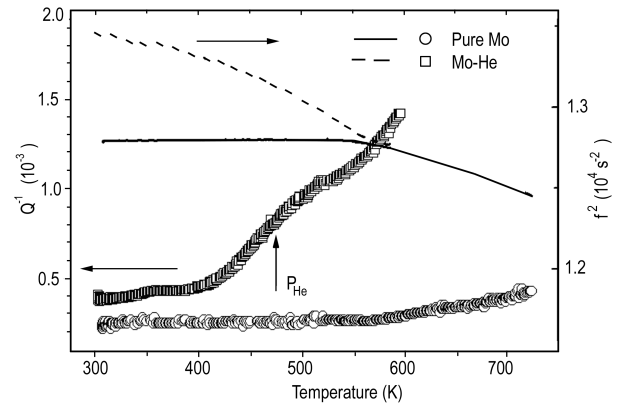


Fig.2 Temperature dependence of internal friction (Q^{-1}) and the square of resonance frequency (f^2) for pure Mo and the He-containing Mo sample measured at a frequency of about 113 Hz.

The result of different frequency measurements through changing the length of the sample indicates that the IF peak in the He-containing Mo sample is of the relaxation type. As an example, Fig. 3 shows the IF spectra of the Mo-He strips at two different frequencies of 56 Hz and 113 Hz, respectively, in which the peak position is seen to shift clearly towards a higher temperature with the increase in frequency. To obtain the relaxation parameters of the IF peak accurately, the curves of IF versus temperature as shown in Fig. 3 were fitted by the superposition of an exponential background and a Debye peak with distribution in relaxation time [11]. In Fig. 3, only the detailed fitting result at 113 Hz is shown for clarity. The obtained peak temperature is 476 K for 56 Hz and 502 K for 113 Hz, respectively. In the fitting process the distribution parameter β of the relaxation time, which is a measure of the peak width, is obtained to be 0.5 (the case of $\beta=0$ corresponds to the standard Debye relaxation peak with a single relaxation time).

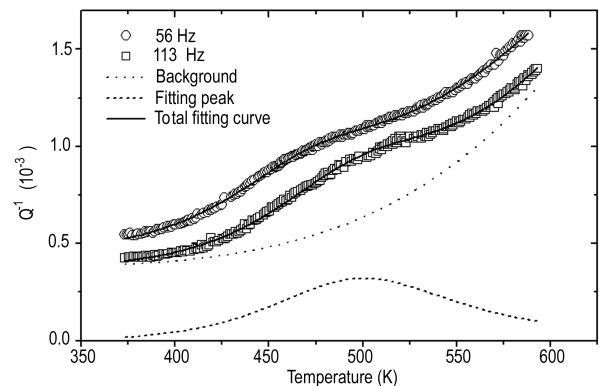


Fig.3 IF spectra of the He-contained Mo strip at two different frequencies of 56 Hz and 113 Hz, respectively. The fitted results are shown for the data at 113 Hz for clarity.

According to the Arrhenius relation, the relaxation time τ can be expressed as

$$\tau = \tau_0 \exp(E/k_B T), \quad (1)$$

where τ_0 is the pre-exponential factor (or the relaxation time at infinite temperature), E is the activation energy of the relaxation process, T is the absolute temperature, and k_B is the Boltzmann constant. For the case of a Debye relaxation peak, $\omega_p \tau_p = 1$ is fulfilled at the peak temperature T_p , where $\omega = 2\pi f$ is the angular frequency of measurement and the subscript p denotes values at the peak position. So if we plot $\ln(\tau_p)$ as a function of $1/T_p$ (Arrhenius plot), a linear relation will be obtained. In this case, the relaxation parameters E and τ_0 can be determined from the slope and the intercept of the line. Fig. 4 shows the Arrhenius plot of the IF peak for the He-Mo sample, from which E and τ_0 are evaluated as 0.63 eV and 6.2×10^{-10} s, respectively.

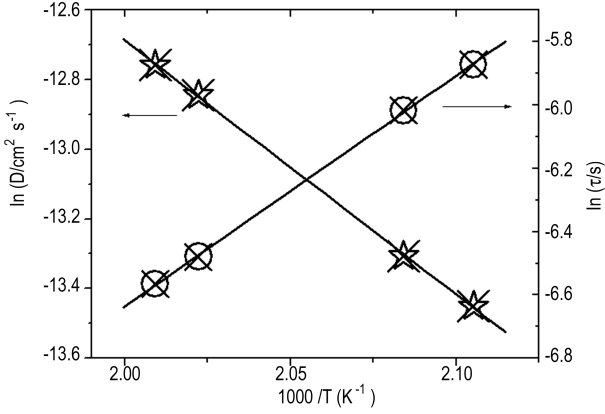


Fig. 4 Temperature dependence of the relaxation time (τ) of the IF peak and the diffusion coefficient (D) of He in the He-contained Mo sample, where the symbols are experimental data and the lines are linear fitting

As is well known, the value of τ_0 for the Snoek effect (short distance diffusion) is in the range from 10^{-15} s to 10^{-14} s, such as the nitrogen diffusion in Mo^[12] and the oxygen diffusion in Niobium^[13]. However, in our case, the value of τ_0 for the P_{He} peak ($\sim 10^{-10}$ s) is 4~5 orders of magnitude larger than that for the Snoek effect, which excludes the possibility that the P_{He} peak is due to the short-range diffusion of helium atoms. So it is straightforward that the P_{He} peak in the Mo-He materials is originated from the Gorsky effect, i.e., the long range diffusion process of helium atoms in Mo metal.

In the process of IF measurements, the He-Mo sample bends and the strain varies linearly across the depth of the specimen (perpendicular to the largest surface of the sample), inducing a strain gradient in the sample. Since helium atoms prefer the location with larger dilatation owing to the minimization of free energy, the He atoms will diffuse along the direction of the strain gradient to reach a new equilibrium distribution of helium in the Mo strip. This long range diffusion of helium gives rise to an additional time-dependent strain and a relaxation IF peak (the P_{He} peak in the case) if a sinusoidal stress is applied. Since our samples have a layer structure, the He atoms diffuse only within one layer, resulting in the so-called intercrystalline Gorsky effect^[14].

According to the theory about the Gorsky effect, the relaxation time τ of long range diffusion for a specimen with a rectangular section can be expressed as^[7,9]:

$$\tau = d^2/\pi^2 D, \quad (2)$$

where d is the thickness of the specimen for the normal Gorsky effect or the grain size for the intercrystalline Gorsky effect, and D is the diffusion coefficient of the diffusion atoms.

Combining Eqs. (1) and (2), D of helium in Mo can be given by

$$D = (d^2/\pi^2 \tau_0) \exp(-E/kT) = D_0 \exp(-E/kT), \quad (3)$$

where $D_0 = d^2/\pi^2 \tau_0$ is the pre-exponential factor of the diffusion coefficient.

If one considers the peak as a normal Gorsky effect, that is, the He atoms diffuse from one side to the other side of the Mo sample, the value of d should be adopted as the thickness of the sample (100 μ m), and the value of D_0 calculated from Eq. (3) is about 1.6×10^4 cm²/s which is about 4 orders of magnitude larger than that reported in stainless steel^[15]. This argument excludes the possibility to explain the peak as a normal Gorsky effect. Therefore, in our case, the relaxation peak shown in Figs. 2 and 3 is the intercrystalline Gorsky effect, and the value of d can be roughly estimated as the average thickness of the layer in the Mo strip, namely $d \approx 2$ μ m. This point can be further confirmed from the fact that the peak can be well fitted by a Debye peak with a distribution in relaxation time ($\beta = 0.5$), since the thickness of layers may have a distribution while the thickness of the sample has a single value.

The temperature dependence of D for helium diffusion in Mo is shown in Fig. 4, from which the value of D_0 can be evaluated as about 6.5 cm²/s, which is of the same order of magnitude with the pre-exponential factor of diffusivity of interstitial helium atoms in stainless steel^[15]. It is worthwhile pointing out that the activation energy of helium atom diffusion in Mo (0.63 eV) deduced from the internal friction measurement in this paper is close to the calculated activation energy of helium atom diffusion in Ag (0.65 eV), Ni (0.66 eV), and Cu (0.63 eV)^[16].

4 Conclusions

A relaxation internal friction peak was observed in the helium-containing molybdenum sample. The peak is originated from the long range diffusion of the helium atoms in Mo (the Gorsky effect). The activation energy and the pre-exponential factor of the relaxation time were deduced as about 0.63 eV and 6.2×10^{-10} s, respectively. According to the theory of Gorsky relaxation, the diffusion coefficient D of helium in Mo was obtained. The activation energy and the pre-exponential factor of D were deduced as about 0.63 eV and 6.5 cm²/s, respectively. The measurement of the

Gorsky effect offers a suitable method to investigate the diffusion behaviors of fast-moving atoms (such as hydrogen and helium atoms) in metal and alloys (such as tungsten and stainless steel) that will be considered as candidates of structural materials as well as plasma facing materials in future fusion devices (ITER).

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