Damage Characteristics of TiD$_2$ Films Irradiated by a Mixed Pulsed Beam of Titanium and Hydrogen Ions

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Abstract
Titanium deuteride is an important nuclear material used in the field of nuclear technology, and further research is needed into TiD$_2$ films irradiated by pulsed ion beams of the vacuum arc discharge with hydrogen. In the current study, these irradiated TiD$_2$ films have been investigated using scanning electronic microscopy and slow positron annihilation techniques. Both the thermal effect and irradiation defects of TiD$_2$ films were studied, following their irradiation with mixed pulsed ion beams of titanium and hydrogen ions. It is found that the thermal effect is trivial on the irradiated surfaces, and the dominant effect is irradiation defects which can be enhanced by repetitive shots and is characterized by the inner diffusion of irradiation defects.

Keywords: vacuum arc discharge at hydrogen, TiD$_2$ film, irradiation defect, slow positron annihilation

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(Some figures may appear in colour only in the online journal)

1 Introduction

Metal hydrides (MH$_x$) are typically subject to extreme non-equilibrium irradiations or a transient heating-up environment when used as a neutron moderator in nuclear reactors, ion sources in lasers, as a target material for neutron generators, and in other similar devices [1,2]. Pulsed ion beams generated from a vacuum arc discharge with hydrogen impregnated electrodes possess characteristics of lower power density and longer pulse duration [3] when compared with high intensity pulsed ion beams (HIPIB) [4,5]. In spite of the fact that vacuum arc discharge at hydrogen impregnated electrodes has already been studied [6], further research is needed into TiD$_2$ films irradiated by pulsed ion beams of the vacuum arc discharge at hydrogen.

2 Experimental setup

The schematic of the pulsed beam test bench is displayed in Fig. 1, and a detailed description of the experimental setups is stated in Ref. [3]. A pulsed discharge voltage of 10 kV is applied to the ion source, and a negative high voltage of 120 kV is applied to the film to achieve the pulsed irradiation. The parameters of the pulsed ion beams are listed in Table 1.

The ions were detected with a time-of-flight (TOF) mass spectrometer. As listed in Table 2, the components of the plasmas are actually a mixture of both H$^+$ and Ti$^+$ ions [6].

![Fig.1 Schematic of the pulsed beam test bench](image)

<table>
<thead>
<tr>
<th>Table 1. Parameters of the pulsed ion beams</th>
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<td>Maximum energy (keV)</td>
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<td>120</td>
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<th>Table 2. The component of the pulsed ion beams</th>
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<td>Categories</td>
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<td>Percentages</td>
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The Ti thin films used in the experiment were deposited onto a Mo substrate (80 mm in diameter, 2 mm in thickness) using an electron gun evaporator, and
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then were impregnated with deuterium under controlled temperature and pressure conditions [7].

A JSM-5600LV scanning electron microscope (SEM) was used to observe surface and cross section morphologies of the films. Evolution of irradiation defects was studied using a slow monoenergetic positron beam with energies from 0.18 keV to 20 keV. The PAS (positron annihilation spectroscopy) results were described with $S$ and $W$ parameters [8].

3 Results and discussions

3.1 Analysis of thermal and sputtering effects

The SEM images of surface morphology in Fig. 2 show the original film revealing a micro-irregularity with micro-protrusions on the surface which is attributed to the deposited titanium particles. However, no noticeable differences in morphologies could be found from the three SEMs, and therefore direct compelling evidence of melting cannot be presumed for the irradiated samples.

![Fig.2](image1)

**Fig.2**: Surface morphologies of (a) the original TiD$_2$ film and the films after (b) 100 shots and (c) 200 shots

It is difficult to record the temperature profiles experimentally during irradiation for the extremely short duration of HiPIB, so we conducted a 2-D axisymmetric finite element numerical way to estimate. $P(r,t)$ is the time and radial distribution function of power flux on the surface, and the two-dimensional heat transport controlling equations are:

$$P(r,t) = 2P_0 / (\pi r_0^2) \cdot \exp \left(-\frac{2r^2}{r_0^2}\right) \cdot \sin \left(\pi \times \frac{t}{30}\right)^2,$$

$$\rho C(T) \frac{dT}{dt} = \frac{\partial}{\partial r} \left(\kappa(T) \frac{dT}{\partial r}\right) + \frac{\partial}{\partial z} \left(\kappa(T) \frac{dT}{\partial z}\right),$$

where $r_0$ corresponds to $e^{-2}$ times of the maximum beam power flux at the central point, and $P_0$ is the full power density of the beam; $T$ is the temperature, and $\rho$, $c(T)$ and $\kappa(T)$ are the mass density, specific heat and thermal conductivity, respectively.

Evolution of temperature with time at the top surface and temperature profile along $r$ direction are shown in Fig. 3. According to calculations, the highest temperature is only 205°C, sufficiently lower than the decomposition temperature of TiD$_2$ (350°C) [9,10], and temperature exhibits a dropping trend along the $r$ direction. The above results agree well with the phenomenon observed with SEM. However the hydrogen isotopes within the film may still be activated, and redistribute as a result of a locally inhomogeneous temperature profile in millimeter scale.

![Fig.3](image2)

**Fig.3**: (a) Evolution of temperature with time at the top central point of TiD$_2$ film irradiated by pulsed ion beam with 1 shot, (b) Temperature profile on the TiD$_2$ film along $r$ direction

Therefore, the surface morphology is not likely to be transformed by heat deposition. Instead, other factors such as sputtering may contribute to the changes in surface microstructure and morphology. Since the components of TiD$_2$ are lattice atoms of Ti and interstitial H, the sputtering yields, evaluated by TRIM for single incident H$^+$, Ti$^+$, Ti$^{2+}$, Ti$^{3+}$ are $4.74 \times 10^{-3}$,
1.84, 1.34, 1.03 Ti atoms/ion, respectively, suggesting a non-negligible degree of sputtering effect, especially after hundreds of accumulated shots. For a rough estimation, about $7.5 \times 10^{13}$ incident ions impinge on the surface in one shot, and the total sputtered Ti atoms are $6.43 \times 10^{13}$ for each shot, corresponding to about $5.14 \times 10^{-5} \mu m$ in thickness for the experimental irradiated sample. For 200 of the same shots, a rough thickness of $10.28 \mu m$ is eliminated from the TiD$_2$ surface afterwards, which leads to sputtered surface morphologies as shown in Fig. 2(b) and (c).

3.2 Analysis of irradiation characteristics

The plots of $S$ and $W$ parameters vs positron energy are demonstrated in Fig. 4, and can be mutually verified. The $S$-curve of the original sample reflects a comparatively uniform distribution of defects along the depth direction, while that of the irradiated samples exhibits a fluctuation from 0 keV to 12 keV, indicating concentrations of defects have been largely elevated. Peaks of the $S$ parameters of the irradiated samples are located at around 3 keV, and then extend to the higher positron energy after a gradual decrease, in which the $S$-value for 200 shots is higher.

![Fig.4](a) $S$ and (b) $W$ parameters vs the incident positron energy

The number of displacements, shown in Fig. 5(a), predicts that vacancies induced by Ti$^+$, Ti$^{2+}$, Ti$^{3+}$ are mostly located within $0.4 \mu m$, while that of H$^+$ is mainly located at $0.7 \mu m$, because the projective ranges of Ti$^+$, Ti$^{2+}$, Ti$^{3+}$ are mainly within $0.4 \mu m$, beyond which H$^+$ induced defects are predominant. It seems logical to expect that the irradiation defects increase with the implantation dose, and therefore exhibit a higher value of $S$ parameter beyond $0.2 \mu m$, as shown in Fig. 5(b). However, the same $S$ parameter at $0.1 \mu m$ reflects that the concentrations of vacancy-type defects somehow reach a saturated value in the two cases, simply because the order of magnitude of collision cascades, displayed in Fig. 5(a), between Ti$^+$, Ti$^{2+}$, Ti$^{3+}$ and lattice Ti, is more effective than that between H$^+$ and lattice Ti. The $S$ parameter profile of the 100 shots shows defects almost terminating beyond $0.8 \mu m$, fitting well with the results of TRIM that show that the total vacancy type defects mainly distribute within $0.8 \mu m$. The TRIM simulation of vacancy type defects of the 200 shots, however, indicates that irradiation induced defects tend to diffuse to a deeper region, suggesting a radiation enhanced diffusion effect under repetitive shots.

![Fig.5](a) Profile of vacancies vs depth simulated with TRIM, (b) A comparative plot between the $S$ parameters deducted by the original blank value and the simulated profile of vacancies

On the premise that a strong interaction between hydrogen isotopes and defects has been recognized [11,12], it is of significance that the irradiation produced large amount of vacancies along the ion tracks that may sufficiently facilitate the diffusion of D atoms towards the surface, and may profoundly influence the redistribution of D within the film, in that D tends to accumulate towards defects and dislocations. It implies that, in the subsequent successive pulsed beam shots, it would become easier for the release of D from TiD$_2$ than before.
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4 Conclusions

a. The sputtering effect mainly contributes to the transformed morphology of TiD$_2$ films especially after hundreds of successive shots. Thermal effects during the pulsed irradiation are not significant, and would not be a predominate factor in facilitating the thermal decomposition of TiD$_2$.

b. Although irradiation defects are mainly distributed within 0.8 µm along the depth direction, repetitive shots enhance its diffusion to a deeper region. It would influence the redistribution of D within the Ti film, especially after hundreds of pulsed shots.

References


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