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Microstructure and Deuterium Retention of Tungsten Deposited by Hollow Cathode Discharge in Deuterium Plasma *

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Tungsten has been chosen as one of the most promising candidates as the plasma-facing material in future fusion reactors. Although tungsten has numerous advantages compared with other materials, issues including dust are rather difficult to deal with. Dust is produced in fusion devices by energetic plasma-surface interaction. The re-deposition of dust particles could cause the retention of fuel atoms. In this work, tungsten is deposited with deuterium plasma by hollow cathode discharge to simulate the dust production in a tokamak. The morphology of the deposited tungsten can be described as a film with spherical particles on it. Thermal desorption spectra of the deposited tungsten show extremely high desorption of the peak positions. It is also found that there is a maximum retention of deuterium in the deposited tungsten samples due to the dynamic equilibrium of the deposition and sputtering process on the substrates.

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Thermonuclear fusion is regarded as mankind's ultimate energy source because of its unique advantages such as infinite fuel resources, no pollution and no high-level radioactive nuclear waste. The tokamak is one of the most promising devices to obtain nuclear fusion energy. However, there will be all kinds of complicated problems during the operation of a tokamak including the generation and re-deposition of dust particles in the vacuum chamber. Dust particles are produced in a tokamak by interaction between plasma and plasma facing material such as first wall, divertor and limiters.[1] Dust may be toxic, radioactive, and chemically reactive. An increase in the amount of dust will cause potential operation and safety issues.[2,3] On the one hand, the presence of dust particles in the vacuum chamber could influence the stability of fusion plasma. On the other hand, the dust particles could deposit on the surface of the first wall material to form a re-deposition layer, which may affect the performance of in-vessel diagnostic components.[4] Also, the re-deposition layer could contain deuterium or tritium and thus can affect fuel recovery systems.[5,6] Possible mechanisms of the formation of dust particles in magnetic fusion systems include nucleation of vaporized materials, blistering and fracturing of deposited layers, explosive ejection and brittle destruction of surface imperfections, arcing and generation of reactive species in edge plasma.[7]

As a high-Z material, tungsten (W) is an excellent candidate plasma facing material (PFM) for a tokamak because of its high threshold for physical sputtering energy, high thermal conductivity, high melting point, and good thermal shock resistance.[8–10] In the design of future fusion reactors, the use of tungsten as the first wall material has almost reached a consensus. As a result, the tungsten dust produced in the fusion device has attracted much more attention and investigations. However, the present investigations are mainly based on the dust particles collected from tokamaks during their scheduled maintenance time or milled powder.[11–13] It is important and valuable to prepare simulated dust of fusion devices to investigate its basic characteristics and the retention of the hydrogen isotope.

Since hollow cathode discharge (HCD) was first achieved by Paschen in 1916, it has attracted considerable attention in the field of low-temperature plasmas.[14] HCD is gas discharge between a cathode that contains a hollow structure and an arbitrarily shaped anode.[15] The mechanism of the hollow cathode effect is called 'pendulum electronic effect'. High-energy electrons pass through the negative glow region and then come back under the electric field of the opposite plate. The reciprocating motion of electron and the energy depletion in the excitation or ionization process is called the pendulum effect of the electron. The pendulum effect of the electron greatly enhances the ionization efficiency.[16,17] Compared with the normal glow discharge, HCD possesses high-energy electron beams and a high plasma density at the same current density.[15]

In this work, the hollow cathode discharge was used in deuterium atmosphere to simulate the sputtering and deposition process of tungsten dust in a tokamak. The hollow cathode discharge (HCD) device consists of the following several parts: vacuum system, electrode structure, sample stage, power supply system and cooling water system. Figure 1 shows

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the schematic diagram of the hollow cathode discharge device. The sample stage is placed at the center of the vacuum chamber, whereon is tungsten tube used as the hollow cathode and the source of sputtered tungsten atoms. The tungsten tube, whose inner diameter is 20 mm and the outer diameter is 30 mm, is connected to an external power supply (dc overlapped pulse power supply, JCL-SZDPY-10) at the cathode output while the entire vacuum chamber wall is connected at the anode output of the power supply. A ceramic tube with a slightly larger diameter is put surrounding the tungsten tube to guarantee that the discharge only takes place between the inner wall of tungsten tube and the anode. By applying electric potential difference between cathode and anode, high-density plasma can be obtained inside the tungsten tube. High-energy particles strike the surface of the tungsten tube. Tungsten atoms or clusters were generated and then deposited on the substrate. In this way, the deposited tungsten sample was obtained. The deposited tungsten sample is the simulation of the redeposited layers on the tokamak first wall surface.

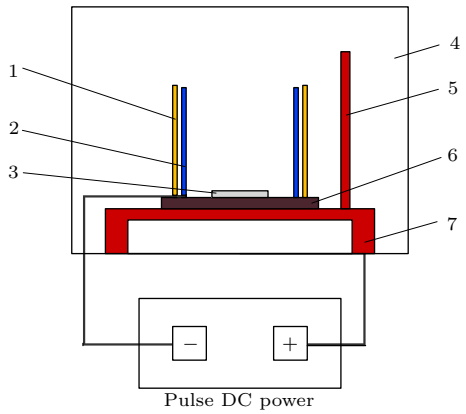

Fig. 1. Schematic diagram of the hollow cathode: (1) ceramic tube; (2) tungsten tube; (3) substrate; (4) vacuum chamber; (5) stainless steel rod; (6) ceramic spacer; and (7) sample stage.

A quartz plate (10 mm × 10 mm × 1 mm) was used as the substrate to collect the tungsten dust particles at the bottom of the tungsten tube. Before use, it was ultrasonically cleaned in acetone and absolute ethyl alcohol in sequence and finally dried by nitrogen puffing. The preceding step of preparing the deposited tungsten sample is to find out the parameters of a stable HCD state of the corresponding gas, such as discharge voltage, atmosphere pressure and gas flow rate. Then the deposited tungsten sample can be prepared with all these parameters known. Put the substrate which was already ultrasonically cleaned into the bottom of the tungsten tube. Evacuate the chamber to base pressure (4×10^{-4} Pa) and deuterium gas is then driven into the chamber at the flow rate of 14.4 sccm. In this study, we have operated the experiment at the deuterium pressure of 500 Pa. The electric potential difference is 400 V. The deposited tungsten samples are prepared under this condition for 1 h, 2 h, 3 h and 4 h, respectively.

The morphology and qualitative elemental composition of deposited tungsten samples were characterized by a scanning electron microscopy (SEM, S4800) and an energy dispersive spectrometer (EDS, IE300X). In addition, deuterium content of deposited tungsten sample prepared by HCD device was measured to estimate the deuterium inventory held in a tokamak. Thermal desorption spectroscopy (TDS) was used to analyze the desorption behavior of D. The sample was heated from 300 K to 1423 K at a heating rate of 1 K/s. A quadrupole mass spectrometer (QMS) monitor was used to record the mass 3 (HD) and mass 4 (D_2) signal in the residual gas of the chamber to determine the deuterium desorption during the heating process. The absolute HD and D_2 signals of the QMS were calibrated.

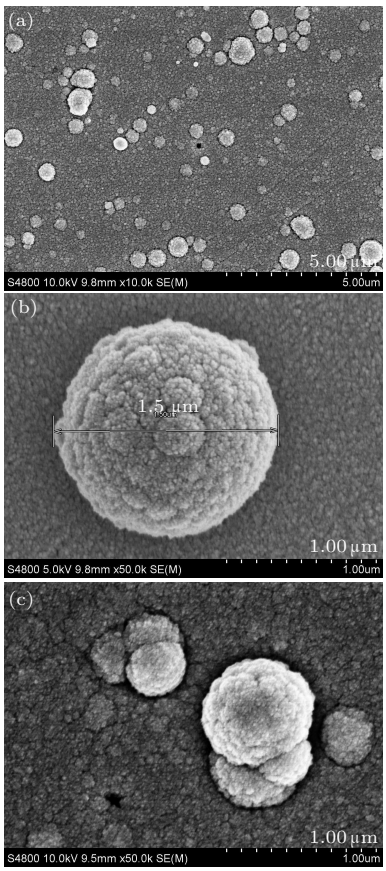

Fig. 2. SEM photographs of deposited tungsten sample prepared by hollow cathode discharge device at the deposition time of 2 h: (a) overall morphology of the tungsten sample, (b) higher magnification of the spherical particle, and (c) fusion of spherical particles.

The overall morphology of the deposited tungsten sample produced in the hollow cathode device is shown in Fig. 2(a). The relatively flat film is dotted with spherical particles of slightly different sizes. Figure 2(b) shows the details of spherical particles. These spherical particles are flocculent structure and their sizes are from about 0.1 µm to 2 µm. These spherical particles and the plain film are both composed of smaller spherical particles. Two or more particles

that are close to each other will gradually merge into a larger particle, as shown in Fig. 2(c).

Figure 3 shows the cross section of the deposited tungsten sample. The thickness of the sample prepared by hollow cathode discharge is around 400 nm. No clear dependence of thickness on the deposition time is observed. This may be due to the dynamic equilibrium of deposition and sputtering process on the substrate. The substrate is placed in the bottom of the gas discharge area. When the tungsten atoms were deposited on the substrate, some deuterium ions with high energy could bombard the surface of the deposited film. Part of the deposited tungsten atoms and trapped deuterium atoms could be detached from the deposited film to the gas phase. The deposition and detachment process may be repeated over and over again. Therefore, the deposition and growth of the film cannot be continuous with the extent of time, and the thickness of deposited film may reach a maximum value.

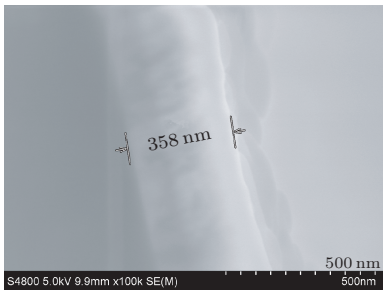

Fig. 3. SEM photographs corresponding to the cross section of deposited tungsten sample prepared by hollow cathode discharge at the deposition time of 2 h.

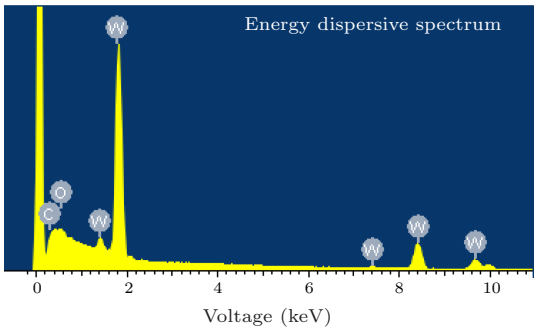

Fig. 4. EDS analysis of the deposited tungsten sample.

To further verify the elemental composition of the deposited tungsten sample, the energy dispersive spectrometer (EDS) analysis was obtained. As can be seen from Fig. 4, the elemental composition of the deposited tungsten samples prepared by the HCD device is mainly composed of tungsten element and a small amount of oxygen and carbon. The oxygen element may come from the mild oxidation of the inner surface of the tungsten tube, which plays the role of both cathode and source of sputtered tungsten atoms. The existence of impurities in tungsten could promote the deuterium retention to a certain extent.[18,19] It is reported that the precipitation of C on W surface could work as a barrier for re-emission of implanted H and could significantly increase H retention.[20,21]

The desorption of deuterium from the deposited tungsten sample was measured by TDS. The positions of desorption peaks correspond to the deuterium trapping energy. Generally, a low-temperature desorption peak is responsible for the low-energy trapping site, such as grain boundaries and dislocations, while high desorption peak is responsible for the high-energy trapping site, like vacancies and voids.[22–25]

Figure 5 shows the deuterium release peaks of the deposited tungsten sample prepared by the HCD device at the deposition time of 3 h. There are two kinds of gas signals. For D_2 signal, there is a distinctive release peak at around 440 K and a broad release peak spanning at 1000 K–1200 K. For HD signal, the corresponding release peak at low temperature is located at around 450 K while the broad release peak locates in the temperature range between 1100 K and 1300 K. However, the intensity of HD signal is much higher than that of D_2. Samples prepared at different deposition times have almost the same shape of TDS spectrum.

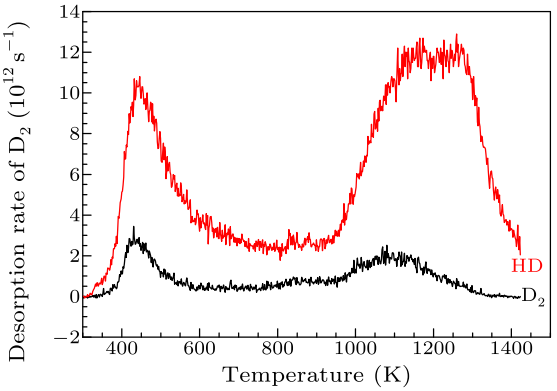

Fig. 5. The D_2 and HD release spectra of the deposited tungsten sample.

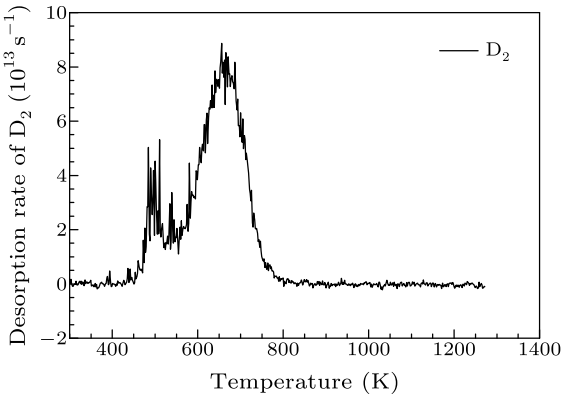

Fig. 6. The D_2 release spectrum of bulk tungsten sample.

As can be seen from Figs. 5 and 6, compared to the thermal desorption spectrum of bulk tungsten sample with the same size irradiated by deuterium,[26] whose

desorption peaks are located at 500 K and 660 K, the desorption peak of the deposited tungsten film prepared by the HCD device in deuterium gas is much higher. This may be due to their different mechanisms of deuterium retention. As for the former, deuterium atoms are captured in the intrinsic defects of the material or the newly introduced defects in the near surface area.[19] However, for the deposited tungsten sample in this work, the process of deposition of tungsten atoms and retention of deuterium atoms are synchronized. During the condensation and growth process of tungsten atoms, the deuterium irradiation will introduce a large number of defects to the tungsten lattice. These deep defects can also work as trapping sites, which make the desorption of the trapped deuterium much more difficult. Deuterium gas accumulated in the deep space needs to overcome the surface barrier to release to vacuum. Therefore, the desorption of the captured deuterium gas needs more energy, which is consistent with higher desorption peaks in the thermal desorption spectrum of the deposited tungsten sample in Fig. 5.

The signal corresponding to HD is much higher than the one obtained for D_2. This may be because of the porous structure of the deposited tungsten sample. Different from the magnetron sputtering process, the deposited tungsten film is prepared in the HCD device at a much higher pressure (500 Pa). Therefore, the tungsten atoms have much greater possibilities to collide with deuterium atoms and have great energy loss. Tungsten film deposited under such a condition tends to have porous structure, which could lead to water absorption after exposure to atmosphere. This may explain the very high HD signal level.

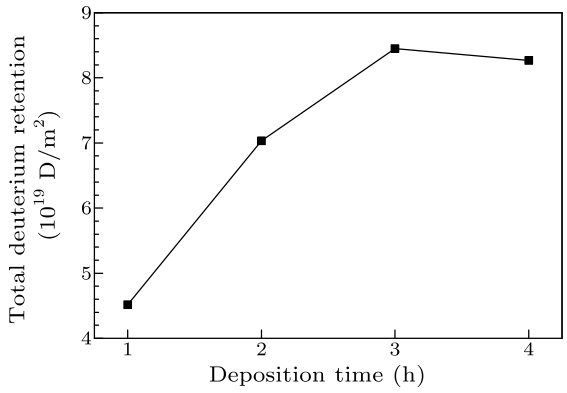

Fig. 7. Effect of deposition time on the total deuterium retention.

The total deuterium retention of deposited tungsten samples for different deposition times is shown in Fig. 7. Total deuterium retention is calculated as an integral of D_2 and HD desorption signals over the time. As we can see, with the extension of time, there will be a maximum deuterium retention within the deposition time of 4 h. The deuterium retention in the bulk tungsten sample calculated from Fig. 6 is about 2.36×10^{20} D/m^2. Therefore, the maximum deuterium retention amount in the deposited tungsten sample is still far less than that in the bulk tungsten sample due to the limited thickness of the deposited tungsten.

As discussed above, the deposition of tungsten, the retention of deuterium and the sputtering of deposited tungsten sample are synchronized, which make the process rather complicated. There is a dynamic equilibrium of the deposition and detachment of the tungsten and deuterium on the substrate. On the one hand, there may be a maximum value of the thickness of the deposited film. On the other hand, there is a maximum value of the deuterium retention in the deposited tungsten film. This is the inherent nature of the tungsten sample deposited under such a condition. In the operation of a tokamak, the re-deposition layer on the surface of the first wall material would take the place of the first wall material to undergo part of the particle bombardment.

In summary, deposited tungsten samples have been successfully prepared on the quartz substrates by the HCD device to simulate the production of dust in a tokamak. The morphology of deposited tungsten can be described as a relatively flat film with numerous spherical particles in different sizes. The existence of saturation retention is proved and comparisons are made of deuterium retention mechanism and retention amount between deposited tungsten and the bulk tungsten material. The higher desorption peak indicates that the desorption of trapped deuterium in the deposited tungsten needs much more energy. Meanwhile, because tungsten deposition and sputtering process are simultaneous, the thickness of the deposited tungsten and deuterium retention amount will not increase continuously. There will be a dynamic equilibrium of deposition and sputtering process, and a maximum deuterium retention can be reached within the deposition time of 4 h. However, the maximum retention amount of deuterium in deposited tungsten is still far less than that in the bulk tungsten material.

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