Initial Stage of Fiber-Form Nanostructure Growth on Refractory Metal Surfaces with Helium Plasma Irradiation

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The initial stage of fiber-form nanostructure growth on tungsten and molybdenum surfaces with helium plasma irradiation is investigated by a technique that allows time evolution of nanostructure growth to its spatial variation. The pitting of the original base surface resulting from successive hole formation is clearly demonstrated. The surface fine-grained miniaturization proceeds with increasing hole area. Then, loop-like nano-scale structures appear with some coherency, which are thought to be precursors of fiber-form tendrils. Loop raptures and branching out are considered as possible candidates of the initial stage of nano-fiber growth.

Keywords: fiber-form nanostructure, refractory metal, helium plasma, bubble, hole

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Since the discovery of fiber-form nanostructures on tungsten surfaces irradiated by helium plasmas [1, 2], a lot of attentions have been paid from many kinds of aspects, including the fundamental formation mechanism [3–7], the effect on the power transmission factor through the plasma sheath, the triggering and control of unipolar arcing, the effect of plasma heat load on surface morphology, and several industrial applications. Moreover, the tip of a tungsten Langmuir probe placed at a divertor target was found to be fully covered with a layer of nano-tendrils in helium plasma discharges in Alcator C-Mod [8].

In this letter, we focus on the initial stage of fiber-form nanostructure growth on tungsten and molybdenum surfaces with helium plasma irradiation, based on careful investigations of FE-SEM (Field Emission –Scanning Electron Microscopy) images.

Similar investigations of the initial stage have been reported [3, 4]. Figure 9 of reference [3] shows a schematic view of the initial stage through the observation with TEM (Transmission Electron Microscopy) associated with the FIB (Focused Ion Beam) technique. Figure 2 of reference [6] shows a viscoelastic model for the mechanism of fiber-form nanostructure growth, which is supported by a viscosity change due to the development of helium clusters in bulk tungsten [7].

We will show additional and new aspects of the initial stage precursor for the fiber-form nanostructure growth, through careful observation of FE-SEM images from the initial to the well-developed stage of formation. We use a technique employing a spatial gradient of helium ion flux on the tungsten surface. This technique converts the time evolution of nanostructure growth to its spatial variation that can be observed with FE-SEM.

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Figure 1 shows the experimental procedure (a) and FE-SEM images of a fractured cross section of a tungsten sample (b, c). Half of the surface of a rectangular tungsten sheet with a thickness of 35 μm was covered with a thin (15 μm) tungsten foil. After helium plasma exposure, the thin cover was removed from the main surface, as shown in Fig. 1 (a). Crossing the boundary zone between the original surface and the helium affected area, a fractured cross section is obtained along the broken line. Macroscopically, the boundary seems to be sharp, but that is not the case mi-

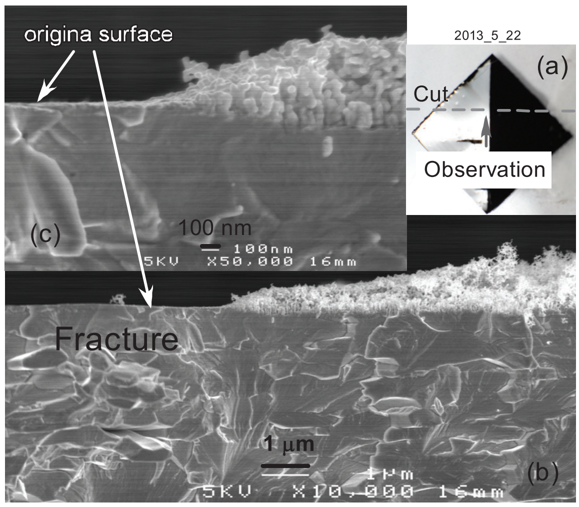

Fig. 1 Fractured cross section of the boundary zone from the original through the damaged surface. (a) Photo showing the macroscopic boundary zone and fractured line. (b) and (c) are FE-SEM images. Helium ion flux on the uncovered area is 3.8×10^{21} m^{-2}·s^{-1}, and its fluence is 2.7×10^{25} m^{-2} with an ion energy of 105 eV, and an initial tungsten surface temperature of 1300 K.

croscopically, as shown in Fig. 1 (b). The incident ion flux has a gradient, which in turn causes the ion fluence on the tungsten surface to have a gradient. This allows a time evolution of the nanostructure growth to be converted to a spatial variation. A detailed view is shown in Fig. 1 (c), showing clearly that the nanostructure formation is started by pitting the original surface, arising from the arrival of bubbles to the surface [9]. The pitting is about 200 nm deep in the present case, while the top of the defect has a diameter of ∼ 100 nm initially, which grows as nano-fibers with the diameter of a few tens nanometers.

Figure 2 shows a top view of the above mentioned boundary zone. Dark areas suggest holes generated by bubble migration to the surface from the interior of the bulk material. In the right-most region, many protrusions appear, growing as fibers. In order to clearly demonstrate an increase in the area occupied by the holes, binarization from gray to black or white was performed for three different zones, as shown in Fig. 3 from the zone (a) close to the originally unexposed surface to the zone (c) containing tungsten nano-fibers. Black areas may represent not only the real holes on the surface, but also the holes of loops or arches, which are not connected to the tungsten bulk. The area occupied by "holes" is 16%, 42% and 60% for (a), (b) and (c), respectively. The mean hole diameter is 40 nm, 56 nm and 200 nm, respectively. From this estimation, it is assumed that the remaining surface area becomes thin and fine-grained, as shown in Fig. 1 (c). These structural changes may serve as a background for the formation of loops or arches as precursors for the fiber-form nanostructure growth mentioned below.

The FE-SEM images obtained with similar experimental parameters to those in Fig. 1 correspond to a grazing view of the cross section made with the CP (Cross section Polishing) method using an argon ion beam, as well as the surface, as shown in Fig. 4, over the boundary zone between the low ion fluence (right side) and the high ion fluence (left side). Many loop- or torch-like structures were observed in the low fluence region whereas for well-developed nano-fibers were observed in the high fluence region. It seems that some loops stand almost perpendicu-

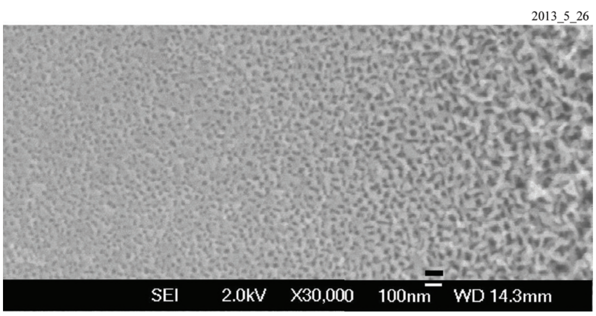

Fig. 2 Top view of the boundary zone.

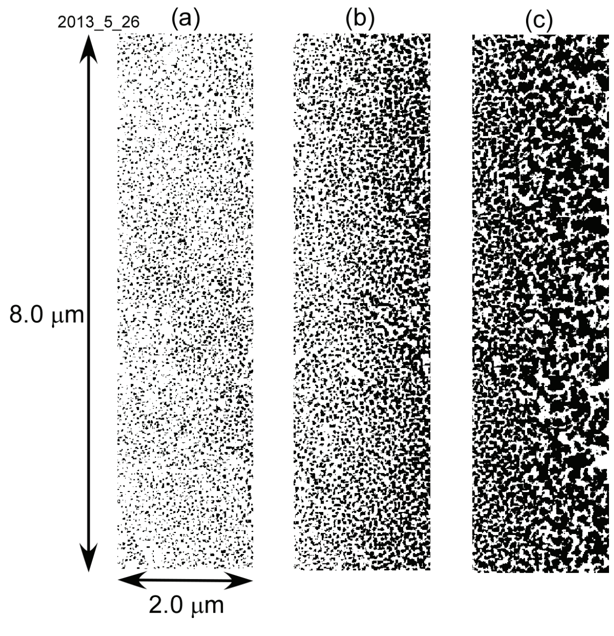

Fig. 3 Clear demarcation of hole area by binarization process of Fig. 2.

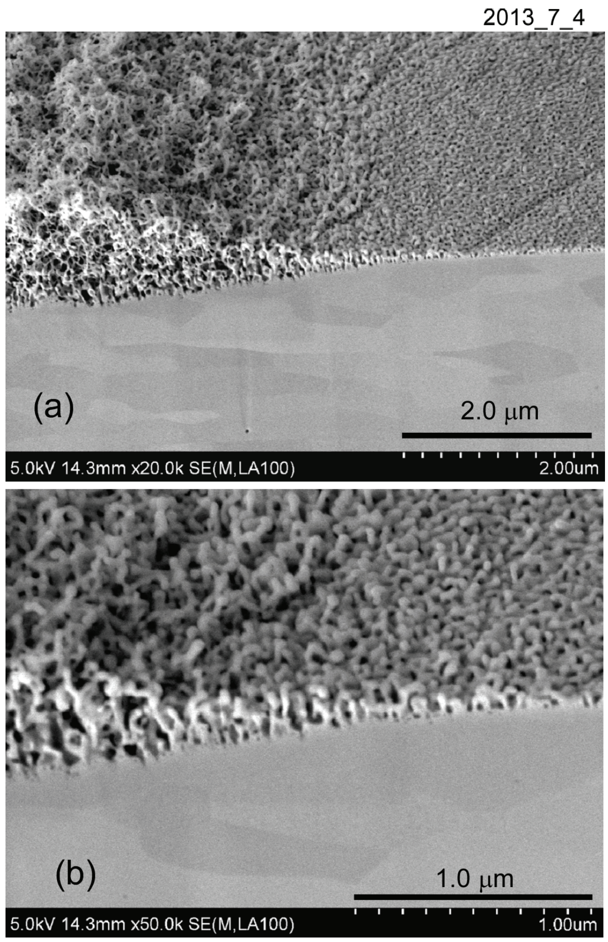

Fig. 4 Oblique view of boundary zone sliced with CP method: (a) a whole view over the boundary zone and (b) detailed image showing many loops.

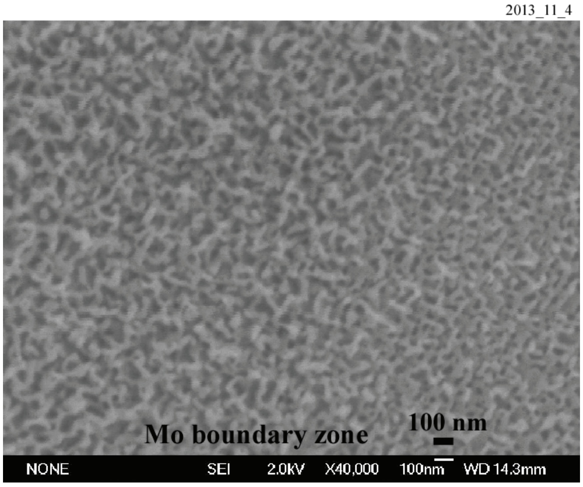

Fig. 5 Loops observed on molybdenum boundary zone. Helium ion flux is 2.4×10^{21} $m^{-2} \cdot s^{-1}$, its fluence is 8.6×10^{24} m^{-2} at a temperature of around 1000 K.

lar to the original surface, but others are at a different angle, and the orientation of the loop surface is random. However, the size of loops is somewhat coherent. In the low-fluence region, the outer diameter of the loops is ~ 70 nm and the inner diameter is ~ 30 nm, while in the high fluence region close to the nano-fiber growth area, the diameters become 150 nm and 70 nm, respectively.

Similar observations were obtained for molybdenum surfaces irradiated by helium plasmas, as shown in Fig. 5, which is an FE-SEM image of the boundary zone looked obliquely. We can reconfirm the presence of the loop- or torch-like nano-structures, the size of which are roughly twice as large as those on tungsten. The nano-fibers themselves have diameter about twice as large as those on tungsten.

From the above observation, it is thought that loop-like structure is a kind of precursor to the growth of nano-fibers. Currently, it is not clear how nano-fibers are developed from the loop-like structure. Possible mechanisms include loop rapture or branching growth from the loop, a kind of bifurcation.

In conclusion, we summarize the experimentally observed growth process as follows:

(1) A technique that allows one to observe time evolutions of fiber-form nanostructure growth in a single exposure of a refractory metal to helium plasma has been developed with a partial covering of the metal surface.

(2) The pitting of the original base surface, resulting from hole formation, is demonstrated clearly by the experiments. Bubbles inside the bulk migrate to the surface.

(3) The hole area becomes large as the helium fluence increases so that a fine-grained miniaturization of the surface proceeds, making a kind of nano-walls, which may become a background of the precursor formation.

(4) Before growth of nano-fibers, randomly oriented, loop-like structures appear as a precursor to the nano-fiber growth. The size increases as the fluence increases because of a possible surface tension exerted by the nano-bubbles inside.

(5) Currently It is not clear how the nano-fibers start to grow from the loop-like structures. Loop rapture, branching out from loops or both occurring together are possible candidates.

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