愛知工業大学研究報告
第 51 号　平成 28 年

Observation of Microwrinkle Structure on Tungsten Surfaces irradiated by Neon Plasmas

ネオンプラズマ照射を受けたタングステン表面に形成された微細皺構造の観察

Shuichi TAKAMURA†, Yoshihiko UESUGI††, Miyuki YAJIMA†††,
Hirotaka KOBAYASHI††††, Shiro MAENAKA††††, Kazunori FUJITA††††
髙村　秀一†，上杉　喜彦††，矢嶋　美幸†††，小林　裕孝††††，前中　志郎††††，藤田　和宣††††

Abstract Microwrinkle structure on tungsten material surfaces has been observed in plasma-wall interaction of fusion related configuration. Incident neon ions have kinetic energies of a few tens eV, just above the threshold of physical sputtering. The pitch of wrinkle, 100 ~ 600 nm, has a strong temperature dependence between 1100 and 1550 K. The formation mechanism has been discussed on the basis of hard surface layer buckling model with a required lateral force due to thermal stress.

1. Introduction

Since the discovery of fiber-form nanostructure formation on tungsten (W) material surfaces irradiated by helium (He) plasms[1), a lot of interests on characteristics of such surfaces and physical formation mechanism including other refractory metals have been studied[2) because He is a fusion product of Deuterium (D) – Tritium (T) nuclear fusion reaction.

Tungsten is the most important material for divertor target of magnetic confinement fusion reactors. In the fusion DEMO reactor, plasma heat flux on divertor plate is expected to be too large to sustain divertor material for plasma heat removal so that a plasma heat reduction at upstream would be essential by using a radiation cooling with noble gases like neon (Ne) or argon (Ar)[3, 4).

Therefore, the interactions between other noble gas ions and W surfaces are also important in addition to helium effect on divertor material[5). In this research work we investigate the effect of Ne plasma irradiation on W surface at elevated temperatures.

2. Surface Morphologies of Tungsten irradiated by Ne Plasmas

2.1 Typical Surface Modification

Typical surface morphology of thin PM-W (Powder Metallurgy Tungsten) irradiated by Ne plasma at the surface temperature of 1500 K and the ion incident energy of 45 eV is shown in Fig.1. Microwrinkle structure with fairly uniform orientation across grains can be recognized. In more detail, Figure 2 shows that it has a pitch of about 0.5 μm, and that the wrinkle is not quite straight but it has rugged lines somewhere locally.

Similar but different microwrinkle structures on relatively thick PM-W substrate at a slightly lower temperature of 1380 K are shown in Fig.3, where the structure including wrinkle orientation depends on grain as is shown in (a) and the wrinkle pitch is slightly smaller than that of Fig.2 as shown in Fig.3(b).

2.2 Details of Wrinkle on PM-W

---

†　Faculty of Engineering, AIT, Toyota 470-0392
††　Kanazawa University, Kanazawa 920-1192
†††　National Institute for Fusion Science, Toki 509-5292
††††　YUMEX Inc., Himeji 671-2114

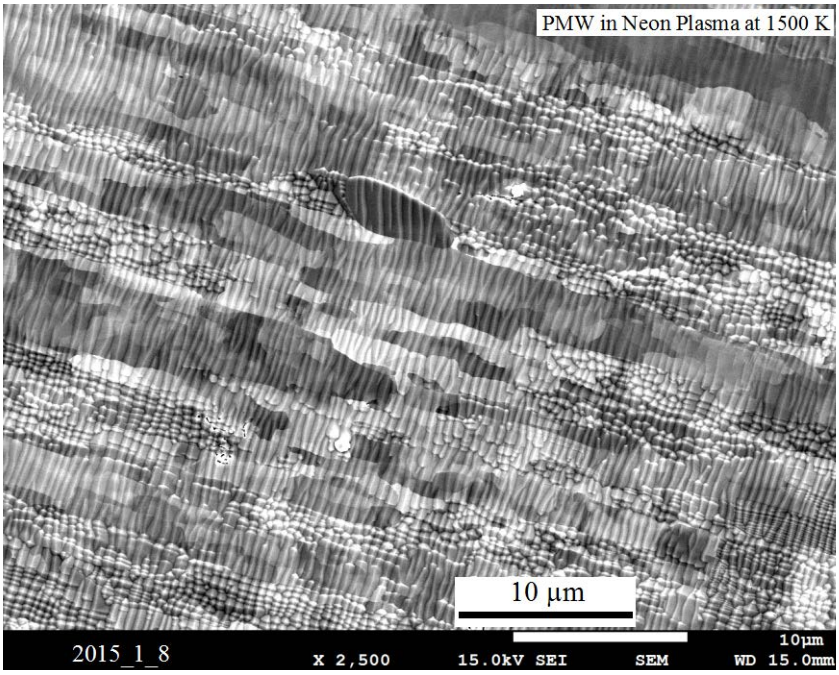

Fig.1 Typical surface morphology of thin PM-W (10×15×0.015 mm) irradiated by Ne plasma at the surface temperature of 1500 K. The incident energy of Ne^+ is E_{Ne} = 45 eV. Its particle flux is $\Gamma_i = 3.6\times10^{21}$ $m^{-2}\cdot s^{-1}$ and the fluence is $F = 2.6\times10^{25}$ m^{-2}.

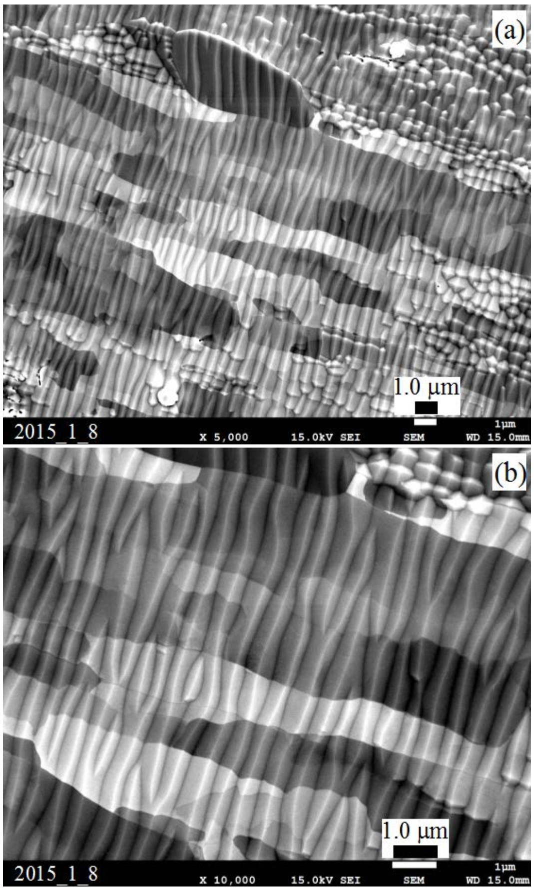

Fig.2 Detailed view of Fig.1. (a): ×5,000 and (b): ×20,000. Clear microwrinkle structure is seen especially on (b).

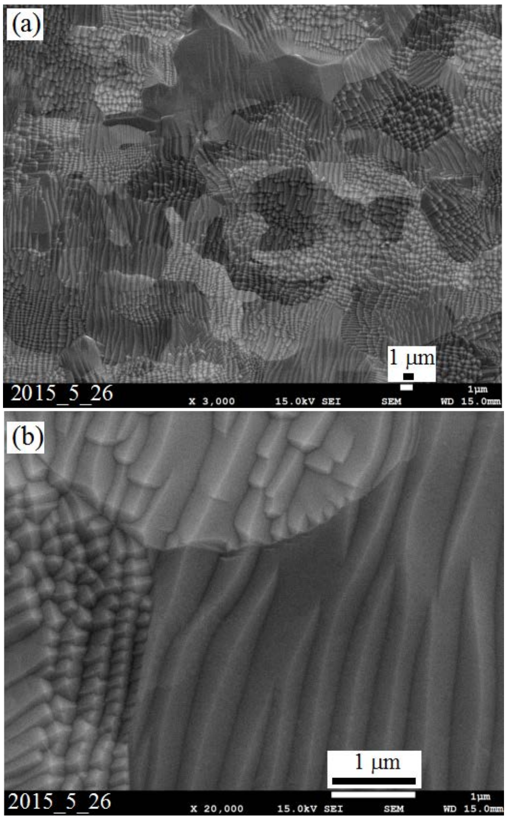

Fig.3 Similar but different microwrinkle structure of PM-W (15×15×0.1 mm) at the surface temperature of 1380 K. E_{Ne} = 50 eV, $\Gamma_i=2.6\times10^{21} m^{-2}\cdot s^{-1}$, $F= 1.9\times10^{25}$ m^{-2}, (a): ×3,000 and (b): ×20,000. The direction of wrinkle and wrinkle detail seem to depend on grain.

A relatively low temperature of 1290 K gives the wrinkle pitch on PM-W surface of about 200 nm as shown in Fig.4. Therefore, the pitch is very sensitive to the surface temperature. It seems that the pitch size increases as the surface temperature increases. Again the broad FE-SEM view like Fig.4(a) suggests the wrinkle orientation is not sensitive to grains in this case.

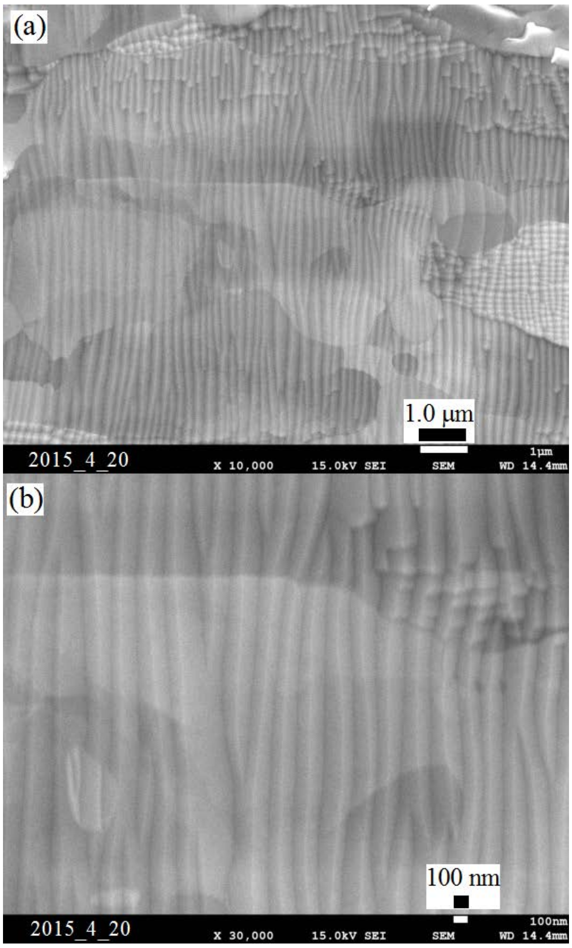

Fig.4 Microwrinkle structure of PM-W (15×15×0.1 mm) at relatively low surface temperature of 1290 K. E_{Ne} =45 eV, Γ_i =2.9x10^{21} $m^{-2}\cdot s^{-1}$ and F = 2.0×10^{25} m^{-2}. (a): ×10,000 and (b):×30,000. The pitch of wrinkle seems to be smaller than that at higher surface temperature.

Ups and downs of the pitch can be seen by cutting the substrate perpendicularly to the surface. Some fractured surfaces are shown in Fig.5 where ups and downs are recognized to be about a few tens nanometer.

2.3 Tungsten Substrate produced by Different Manufacturing Processes

Above FE-SEM images were obtained using PM-W. On the other hand, ITER grade-W is fabricated basically in the same way as PM-W. But the plasma-facing surface is perpendicular to that of PM-W, and has edge of laminating layers. Ne irradiation result is shown in Fig.6 where (a) shows an irradiated surface with several localizations of wrinkle structure. Oblique straight lines show laminating surfaces. Figure 6(b) shows the enlarged view indicating areas containing local wrinkle structures with yellow lines. Wrinkles seem to be localized between laminating interfaces, but do not extend across the interfaces.

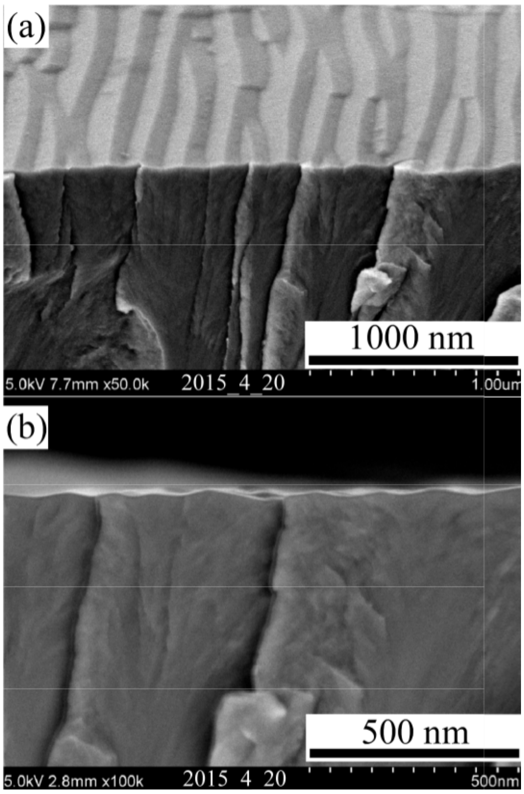

Fig.5 FE-SEM observation of fractured edge. Ups and downs due to wrinkle can be seen. Irradiated W specimen is the same as that in Fig.4. (a) oblique view, and (b) cross-section one.

TFGR-W-1.1%TiC/H (Toughing, Fine-Grained, Recrystalized Tungsten with TiC dispersoid) has a grain size of typically 1 μm and contains TiC as an additive dispersoid by a fraction of 1.1 %. It is fabricated by hot plastic worked compacts in a purified H_2 atmosphere and by hot isostatic pressing, so called HIPS, in order to have very high fracture strength and an appreciable ductility at room temperature$^{6)}$.

Irradiation of Ne plasma on TFGR-W surface gave the surface view by FE-SEM as shown in Fig.7. The surface roughness seems to be fairly large and any wrinkle-like structures were not found due to probably small-sized grains and cracking. However, we need further investigations.

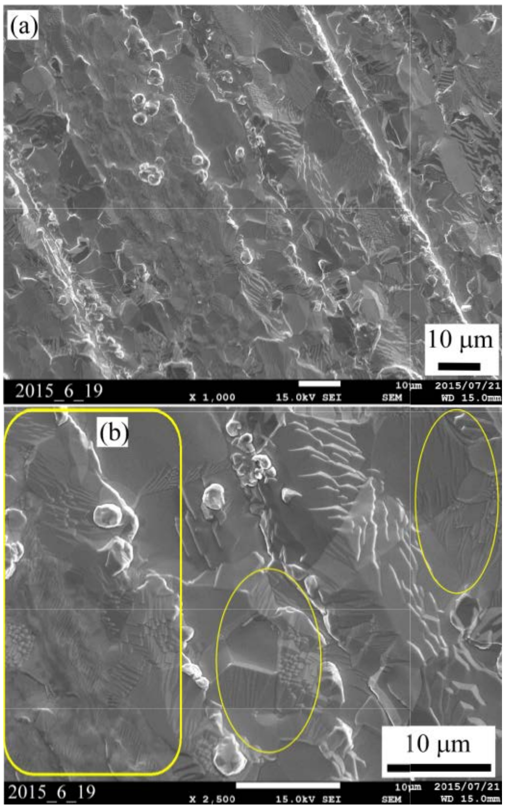

Fig.6 Microwrinkle structure seen on the surface of ITER grade–W (10 × 10 × 0.1 mm) at the irradiation surface temperature of 1260 K. E_{Ne} = 45 eV. (a): ×1,000 and (b): ×2,500 where yellow lines show areas in which wrinkle are found.

Finally we have to touch upon the effect of physical sputtering on surface morphologies. So far, we chose Ne^+ ion energy slightly higher than the sputtering threshold (~ 35 eV) to minimize the physical sputtering and to ensure the injection of Ne atoms into tungsten bulk. If the sputtering is large, then this may modify the surface microstructure because of surface flattening or appearance of sputtering cones which may destroy microwrinkle structure. The case with a relatively strong sputtering at E_{Ne} ~ 65 eV with the yield of 0.006 gives almost unchanged microwrinkle structures on PM-W surface as shown in Fig.8 so that the current understanding contains no importance of physical sputtering with respect to microwrinkle formation. It means that an increase in ion energy makes some defects of crystal atomic network in deep layer, or that the wrinkle structure may be produced after Ne plasma irradiation, that is, during cooling stage after shut-off of

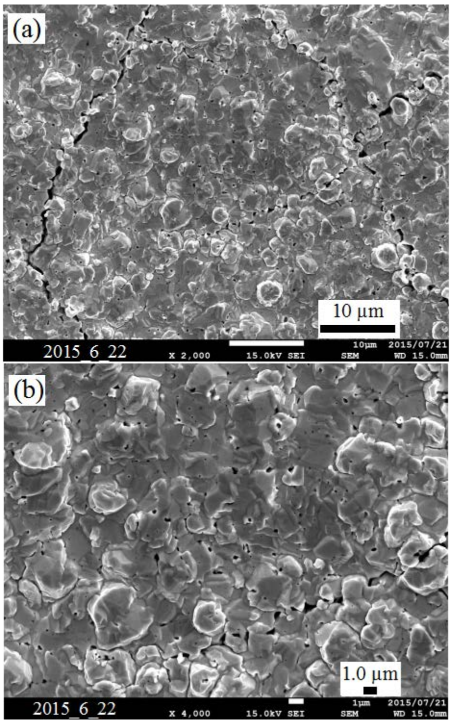

Fig.7 Surfaces of TFGR-W (S = 157 mm^2, thickness = 0.5 mm) at the irradiation temperature of 1380 K. E_{Ne} = 50 eV, $\Gamma_i = 4.4 \times 10^{21}$ $m^{-2} \cdot s^{-1}$ and $F = 3.1 \times 10^{25}$ m^{-2}. (a): ×2,000 and (b): ×4,000.

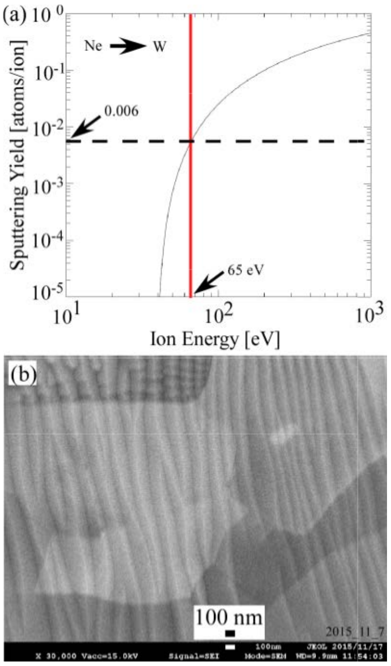

Fig.8 Surface morphologies of PM-W (10×10×0.1 mm) at the temperature of 1260 K with relatively large ion incident energy of 65 eV. (a) sputtering yield of W vs. ion energy. E_{Ne} = 65eV makes the yield of 0.006 compared with less than 10^{-3} at E_{Ne} <50 eV, (b) FE-SEM wrinkle structure.

plasma irradiation.

3. Comparison with Surface Buckling Model

As already mentioned above, the wrinkle pitch strongly depends on the surface temperature. The temperature dependence is shown in Fig.9, indicating higher the temperature, larger the pitch size. According to surface buckling model by T. Ohzono(7) where wrinkle structure is produced when the sample containing thin hard surface layer on soft substrate is compressed laterally as shown in the insert of Fig.10.

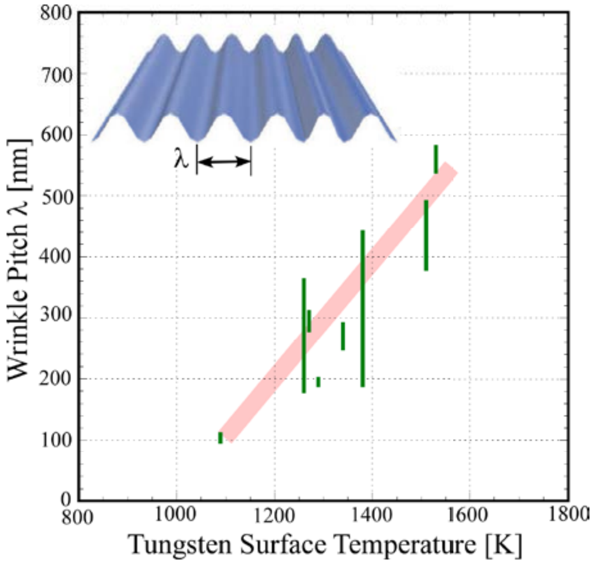

Fig.9 Surface temperature T dependence of wrinkle pitch λ.

T. Ohzono gives the pitch size of wrinkle λ, the spatial wavelength, $\lambda \approx h\left(E_f/E_s\right)^{1/3}$, where h is the thickness of hard surface layer and E_f and E_s are the Young's modulus for hard layer and the soft substrate, respectively. Due to the small exponent, Young's modulus does not affect so much on the wavelength. The important parameter is the thickness of hard layer, h. In the present case, the question is how h is determined if the surface buckling model is applicable. We assume that h would correspond to the layer in which tungsten lattice is defected by the presence of injected Ne atoms, and that invaded Ne atoms migrate from the surface layer by thermal diffusion. The diffusion coefficient would be described by

$$D = D_0(\mathrm{T})\exp(-E_D/T), \tag{1}$$

where D_0 is the pre-exponential factor corresponding to the diffusion coefficient at high extreme temperature and E_D is the characteristic energy for migration. The characteristic penetration depth by thermal diffusion would be described by $h = 2\sqrt{Dt}$ where t is the exposure time of Ne plasma. Then, we obtained the following relation between lnλ and T^{-1},

$$\ln\lambda = \ln\left\{2\sqrt{D_0 t}\right\} - E_D/(2T). \tag{2}$$

The exposure time is always 7200 s. Neglecting rather weak temperature dependence of D_0, we would obtain the linear relation between lnλ and T^{-1} if the diffusion process is valid. Figure 10 shows the logarithmic value of pitch size lnλ as a function of inverse surface temperature, T^{-1}.

The linear relation shown by a thick straight line gives the migration energy of about 1.0 eV which seems to be too large compared with the standard characteristic migration energy as interstitials.

The ductility may come from the propagation of dislocation of crystal structure. The presence of interstitial atoms plays a pinning role against dislocation propagation so that it may bring a hardness of materials.

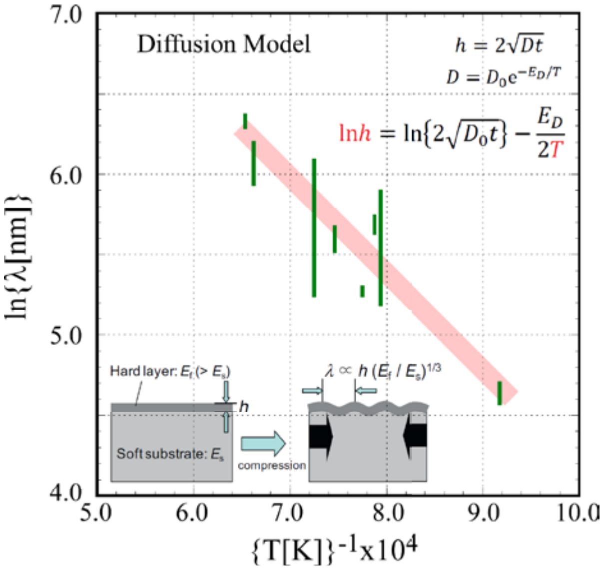

Fig.10 Inverse temperature T^{-1} dependence of logarithm of wrinkle pitch lnλ. Thick straight line is fitted to data bars by this line whose slope gives the activation energy for Ne migration through tungsten atoms. Insertion shows a theoretical formula for wrinkle pitch obtained by a hard layer thickness and the ratio of Young's modulus between hard layer and soft bulk.

愛知工業大学研究報告, 第 51 号, 平成 28 年, Vol. 51, Mar, 2016

4. Discussions on Wrinkle Formation Mechanism

Finding of microwrinkle structure on the surfaces of plasma-facing materials like W is new so that no one discusses the physical and mechanical reasons for the formation of such a structure. In the case of He plasma irradiation on hot tungsten surface at more than 1800 K, we have some preliminary FE-SEM images suggesting wrinkle structures. Molybdenum surfaces irradiated by Ne plasmas similar to the condition for W were confirmed to show microwrinkle structures locally. These are not shown here, and we need more comprehensive data to be discussed in near future. Therefore, wrinkle structure formation on W surfaces due to noble gas plasmas irradiation may have a general importance.

The largest question is how for such microwrinkle structure to be formed on W surfaces exposed by Ne plasmas. Although the below is the final answer, we try to employ a buckling process of hard surface layer on soft bulk substrate compressed laterally as described in the previous section for elastomer surface. Under the irradiation of Ne ions with a few tens eV slightly above threshold energy for physical sputtering on W surface, injected Ne ions are deposited on the near-surface without any channeling effect in this energy range[8).

Noble gas atoms injected into W surface make some defects and diffuse into deep interior thermally. The penetration depth increases as the temperature increases. Interstitial solid solution or substitutional solid solution thus produced plays a resistive role against the propagation of dislocation, that is, an decrease in ductility. Ne atoms make a kind of pinning effect against dislocation transfer.

At the moment we do not have any clear evidence that the surface layer is harder than the substrate bulk. It should be confirmed by using nanoindentation technique. Another important question is how to have a lateral stress to produce buckling. We have the following several possibilities:

(1) Thermal shrinkage stress

Ne plasma irradiation in the present case makes W substrates hot, more than T = 1000 K. On the way to cooling stage a shrinkage produces the thermal stress.

(2) Difference in thermal expansion coefficient between Ne defected surface layer and bulk substrate.

(3) Temperature difference between the surface and the bulk on the way to cooling stage.

We may have other possibilities. Further investigation, such as a temperature increase without any Ne ion injection or a temperature increase with Ne ion energy below the surface barrier potential energy of about 12 eV for Ne-W system[9) and a fairly large incident energy of Ne ions ensuring a channeling penetration and sputtering effect.

5. Summary

First of all, the importance of interactions between noble gas ions and plasma-facing materials, mainly W, has been pointed out in terms of research and development of fusion reactor, and we obtain the first finding of microwrinkle formation on W material surfaces under Ne plasma irradiation. Such structure would bring surface defect and fatigue influencing on the life time of plasma-facing component.

It has been experimentally observed that the pitch size or wavelength of microwrinkle increases as the surface temperature increases. It seems that the physical sputtering of Ne ions on W surface would not give any clear influence on the spatial amplitude of wrinkle. Orientation along wrinkle may either cross the grain boundaries or depends on each grain. Key factors for such different formation are open so far to be understood.

ITER grade-W produces also microwrinkle locally although a fairly broad formation has been observed for PM-W.

We cannot distinguish any clear microwrinkle on the surfaces of TFGR-W, probably because of fine grain property.

Although we have proposed a buckling model to explain wrinkle formation on W surface, that is not a decisive mechanism at the moment. Further investigation would be required for the identification of physical mechanism of microwrinkle formation.

Acknowledgements

The work is supported by the Japan Society for the promotion of Science, Grant-in-Aid for Scientific Research (C) 26420855.

One of the authors (S.T.) would like to thank Dr. A. Ito of NIFS and Dr. S. Saito of NIT, Kushiro College for their discussions, Prof. M. Nagata, Dr. Y. Kikuchi and Graduate student I. Sakuma of Univ. of Hyogo for their helps on manipulating FE-SEM.

References

1) S. Takamura et al., “Formation of Nanostructured Tungsten with Arborescent Shape due to Helium Plasma Irradiation”, Plasma Fusion Res. Vol.1, pp.051 (2 pages), 2006.

2) S. Takamura and Y. Uesugi, “Experimental identification for physical mechanism of fiber-form nanostructure growth on metal surfaces with helium plasma irradiation”, Appl. Surf. Sci. Vol.356, pp.888-897, 2015.

3) K. Tobita, S. Nishio, M. Enoeda, H. Kawashima, G. Kurita, et al., “Compact DENO, SlimCS design progress and issues”, Nucl. Fusion Vol.49, pp.075029 (10 pages), 2009.

4) H. Kawashima et al., “Simulation study for divertor design to handle huge exhaust power in the SlimCS DEMO reactor”, Nucl. Fusion Vol.49, pp.065007 (7 pages), 2009.

5) M. Yajima, M. Yamagiwa, S. Kajita, N. Ohno, N. Tokitani, A. Takayama, S. Saito, A.M. Ito et al., “Comparison of Damages on Tungsten Surface Exposed to Noble Gas Plasmas”, Plasma Sci. Technol. Vol.15, pp.282-286, 2013.

6) H. Kurishita, S. Matsuo, H. Arakawa, T. Sakamoto, S. Kobayashi et al., “Development of re-crystalized W-1.1%TiC with enhanced room temperature ductility and radiation performance”, J. Nucl. Mater. Vol.398, pp.87-92, 2010.

7) T. Ohzono and H. Manobe, “Microwrinkles: Shape tunability and applications”, J. Colloid. Interface Sci. Vol.368, pp.1-8, 2012.

8) S. Saito, A. Takayama, A.M. Ito and H. Nakamura, “Binary-collision-approximation-based simulation of noble gas irradiation to tungsten materials”, J. Nucl. Mater. Vol.488, pp.S895-S898, 2014.

9) T. Tamura, R. Kobayashi, S. Ogata and A.M. Ito, “First-principles investigation of possible clustering of noble gas atoms implanted in bcc tungsten”, Modelling Simul. Mater. Sci. Eng. Vol.22, pp.015002 (12 pages), 2014.

（受理 平成 28 年 3 月 19 日）