Behaviour of Redeposited Tungsten Layers with Varying Impurity Content during Thermal and Radiation Loads

L.B. Begrambekov, O.A. Bidlevich, A.V. Grunin, N.A. Puntakov, A.N. Voityuk.
National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe shosse 31, 115409 Moscow, Russia.
lbb@plasma.mephi.ru

Abstract. Tungsten is going to be implemented as a material for plasma-facing divertor tiles in an ITER tokamak. Accumulation of dust particles due to exfoliation of redeposited tungsten layers can severely affect the operation of a fusion devices. Behaviour of tungsten films deposited on tungsten, including layers with added impurities of iron (0.2 at.% and 2 at.% concentration), as well as dust particles with Al impurity, has been studied. It is shown that a film with 2 at.% impurity of iron exhibits exfoliation and blistering when exposed to thermal and radiation loads, while tungsten dust particles with aluminium impurity undergo various structural changes depending on their Al content when heated.

1. Introduction
Tungsten will be used as a plasma-facing material for ITER divertor tiles. Behavior of tungsten during thermal cycling and plasma irradiation in different modes was investigated in a number of works [1-3]. Tungsten atoms sputtered by plasma irradiation would form redeposited layers on divertor which tiles are irradiated with lesser intensity [4,5]. Formation of dust particles might be the result of the formation of films with poor adhesion. Dust accumulation can severely affect the discharge parameters in future thermonuclear facilities and lead to the accumulation of dangerously high amounts of tritium. This work is dedicated to the investigation of processes in deposited layers during thermal cycling and irradiation by hydrogen ion flux of high power density. The experiments on generating tungsten and tungsten-aluminium dust composed of flake particles formed by destruction of layers formed on the walls of a plasma chamber by irradiating tungsten and aluminium targets with plasma ions are also presented.

2. Experimental Setup
Deposition of layers and irradiation by hydrogen ion flux was conducted in a CODMATT (Coating Deposition and MATerial Testing) facility [6]. Tungsten films were formed on polished tungsten plates 15×7.5×1 mm$^3$ in size by atoms sputtered from the surface of a tungsten(99.9 at%) target by argon plasma ions. Deposition rate was 2 μm/hr, layer thickness was 2 μm.

During the investigation of the behavior of redeposited tungsten films in ITER-like conditions, it is important to consider the possibility of impurities added to tungsten during redeposition, such as beryllium and, possibly, stainless steel sputtered from different parts of the facility. Due to safety reasons our laboratory is not permitted to work with beryllium, thus aluminium is used as a beryllium
proxy and an impurity in tungsten films, as well as stainless steel atoms (Fe, Cr, Ni) deposited in a different experiment.

Thermal cycling of samples was conducted in a Multifunctional Research Mass-Spectrometry Analysis Complex (MIKMA) [7]. The samples were heated by radiation from a flat tungsten filament mounted on an opposite side of a sample under investigation and heated by a pulse current in a vacuum of no less than $10^{-6}$ Torr. The sample was heated to the temperatures of up to 1200 °C. Temperature gradient on a sample was no more than $\pm 10$°C at the sample size. The samples were heated at a rate of 5°C/s and cooled for 5 min to the temperature of 300°C. A number of samples were prepared for testing, with a small (0.2 at.%) and large (2 at.%) iron impurity content in the deposited film.

Amorphous tungsten dust was accumulated in a “DEKOR” plasma coating deposition facility [8]. During the discharge, deposited films formed on stainless steel sheets that partially exfoliated as dust particles. Approx. 0.5 g of dust was collected after each discharge. The particles had a form of flakes ranging in size from a few microns up to 500 μm (Fig. 1a) and being 2-6 μm thick. As such, the size of flakes was comparable to those obtained in modern thermonuclear facilities [9]. Particle content measured using Energy-dispersive X-ray spectroscopy (EDS), included W (up to 90 at.%), Al (up to 60%) and O (50 at.%). An addition of a small (<10 at.%) amount of stainless steel impurity (Fe, Cr, Ni) leads to the consequence that oxide layers formed on the plasma chamber wall exfoliated along the tungsten flakes. The flakes were then separated by size using a $35 \times 35 \, \mu m^2$ net.

![Fig. 1. Flake particles: a) overview, b) thickness of a particle.](image1)

Analysis of the deposited layers’ structure and profile before and after testing was conducted using a Tescan Vega 3 Scanning Electron Microscope (SEM), with a cross-section done using a FIE Quanta 200 3D system (scanning electron microscope with gallium ion beam).

3. Experiment Results

3.1. Thermal cycling of deposited coating
After 260 cycles, traces of restructurisation were observed on the surface of deposited coating with a small impurity content – formation of separate globules of 200-500 nm in size. (Fig. 1) No exfoliation or cracking of the coating was observed.
Fig. 2. Deposited tungsten layer with a small impurity content: a) directly after deposition; b) after 260 heating cycles.

Similar testing was conducted on a tungsten sample with a large impurity content (2 at.%). Thermal cycling led to the formation of larger globules 1-3 μm in size (Fig. 3), which is supposedly connected to the recrystallisation processes on the deposited layer. After 250 heating cycles, formation of pores can be observed on the surface. After 360 cycles, pore formation continued. Additionally, exfoliation occurred on some parts of the sample, which may indicate the formation of a large number of pores in the bulk of a deposited layer.

Fig. 3. Deposition layer with a 2 at.% iron impurity content: a) pores on a surface after 260 heating cycles; b) exfoliation after 350 heating cycles.
3.2. Irradiation of deposited films by ions

During the irradiation of a sample with a 2 at.% impurity content by H$_2^+$ ions with a power density of 5 MW/m$^2$ (ion energy 5 keV/at., cyclic irradiation mode, 20 pulses with the duration of 5 seconds each), blisters formed on the surface of the sample. The temperature of the sample during the experiment did not exceed 1200 °C (the sample cooled between the pulses to the temperature of 600 °C). Fig. 4a shows the image of the film after irradiation. No blisters formed on the sample with a low impurity content. Fig. 4b shows the formation of a large number of microscopic gas bubbles near the interface between the layer and the substrate. Accumulation of hydrogen in these bubbles could lead to the formation of strains parallel to the surface in the deposited films and form blisters. It is interesting to note that pores formed in the samples heated without ion irradiation. That can be explained by pores formation leading to the gas trapped during deposition escaping the coating, with strains and blisters not forming as a result.

3.3. Investigation of structural changes in dust particles during heating

During the heating experiments, dust particles were places on a box-shaped heating element made of thin-sheet tungsten mounted between the electrodes in a VUP-2K station plasma chamber. The temperature was controlled using a tungsten-rhenium thermocouple attached to a heater. Dust particles were heated to temperatures of up to 2000 K with an increment of 100 K each time, changes in structure were investigated using SEM and EDS after each heating.

No changes in structure of amorphous particles were observed in a temperature range of up to 1073 K. Starting from 1473 K, which corresponds to the minimal temperature at which the recrystallization process for tungsten occurs, the formation of metal grains and pores at the grain boundaries occurred (Fig. 5a). With further temperature increase, the size of grains increased, and the oxygen content decreased due to the decomposition of oxides in the particles. If a particle included aluminium, then, when the content was high (> 55 at.%), molten aluminium droplets formed and spread on the surface of the heater at 1473 K (Fig. 5b). When the particle consisted of two layers, with differing contents for each layer (85 at.% W and 60 at.% Al, respectively), conical structures appeared when the particle was heated to temperatures of up to 1273 K (Fig. 5c), which can be indicative of the development of internal stress in the flake. For small aluminium contents (<25 at.%), no such effects were observed after heating to above temperatures. It is speculated that aluminium evaporates before forming droplets.

In case of crystal dust, large particles were baked along the edges at temperatures above 1173 K, and small particles forming conglomerates baked together (Fig. 5d, 5e).
4. Conclusion
Thermal cycling and irradiation of tungsten samples with deposited tungsten films with different iron impurity concentration (0.2 at.% and 2 at.%) by hydrogen was conducted. During thermal cycling of tungsten films with impurities, globules formed on the surface of the film, with larger globules (1-3 µm) forming on a layer with a higher impurity concentration (2 at.%). Pores also formed on a layer with higher impurity content, which led to exfoliation of deposited film, which wasn’t observed on a lower impurity (0.2 at.%) sample. During hydrogen ion irradiation, pore formation in coatings with a high impurity content resulted in exfoliation and blister formation in deposited films.
It is shown that, during heating of dust particles, a structural change occurs in them, such as: recrystallization and crystal grain growth, formation of cone-like structures due to internal strains, melting and evaporation of components with a low melting point (Al) in case of amorphous dust particles.

References
[1] Krieger K., et al., J. Nucl. Mater., 313—316(2003)327—332.
[2] Buzi L., et al., J. Nucl. Mater., 455(2014)316—319.
[3] Krasheninnikov S.I., et al., Plasma Phys. Control. Fusion, 2011, vol. 53, № 8, p. 083001.
[4] M. Balden, et al., J. Nucl. Mater., 438(2013)S220-S223.
[5] S. Krat, et al., J. Nucl. Mater., 438(2013)742-S745.
[6] E. Azizov, et al., J. Nucl. Mater., 463(2015)792-795
[7] A. Airapetov, et al., J. Nucl. Mater., 415, Issue 1, Supplement, 1(2011) S1042-S1045.
[8] Begrambekov L.B., Gordeev A. A., Grunin A.V., Evsin A.E., et al. Materials of VII
International School-Conference of young researchers and specialists IHISM’11 (24-28 October, 2011, Zvenigorod), p. 323-340

[9] S.I. Krasheninnikov et al., Plasma Phys. Controlled Fusion 53(2011) 083001.

Acknowledgments

This work was conducted with financial support from the Russian Science Foundation under the grant № 17-12-01575.