Implantation of D and He in W-Coated Refractory Carbides

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The effect of high temperature (700 - 1200 ºC) implantation of deuterium and helium in candidate fusion first wall materials was studied in the University of Wisconsin Inertial Electrostatic Confinement (IEC) device. Tungsten coated TaC and HfC "foam", single crystal tungsten, and high-emissivity tungsten coated "foam" were compared to previous tungsten powder metallurgy samples studied in the IEC device for the High Average Power Laser (HAPL) program. Scanning electron microscopy was performed to evaluate changes in surface morphology for various ion fluences at temperatures comparable to first wall temperatures. Single crystal tungsten was shown to exhibit less damage than polycrystalline samples at a fluence of $4x10^{16}$ He^+/cm^2. It was found that no significant deformations occur with deuterium implantation up to ~10^{18} D^+/cm^2 at 800 ºC on W-coated TaC and HfC foam samples. However, helium fluences in excess of $6x10^{17}$ He^+/cm^2 show extensive pore formation at 800 ºC and higher. These changes may have an impact on the lifetime of tungsten coatings on the first walls of inertial and magnetic confinement fusion reactors.

I. INTRODUCTION

The durability and lifetime of thin tungsten or refractory metal coatings on the first walls of inertial and magnetic confinement fusion reactors is a key issue for the feasibility of such devices. Work as early as 1974 by Thomas and Bauer showed blistering in Vanadium as the result of helium implantation at high temperature.[1] Past studies at UW-Madison and ORNL have indicated that tungsten, when subjected to He^+ fluences in excess of $4x10^{17}$ He^+/cm^2, shows extensive pore formation at 800 ºC.[2, 3] The current study has investigated alternative forms of tungsten for future use in fusion devices.

I.A. High Average Power Laser (HAPL) Program

Scientists from across the country are carrying out a multidisciplinary program to develop technologies for fusion energy and defense applications. This work is part of an ongoing effort to develop fusion chamber first wall technology in conjunction with the HAPL program.[4] HAPL scientists are currently focusing on Tungsten as a potential material to act as a shield to protect the first wall from light ions whose energy ranges from tens of keV to a few MeV. This study examines the lower portion of that spectrum.

I.B. Inertial Electrostatic Confinement (IEC) Device

The University of Wisconsin has been studying the performance of a Inertial Electrostatic Confinement (IEC) fusion device for more than a decade.[5] It has been recently used to test the response of fusion related materials to high temperature bombardment from D and He. As seen in Figure 1, one of the UW IEC devices is an aluminum chamber 65 cm tall and 90 cm in diameter.[5] A base pressure of ~10^{-7} torr is maintained in the device

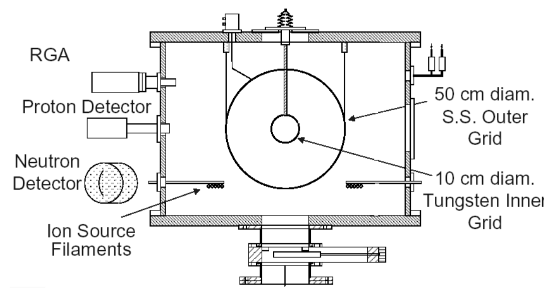

Fig. 1. UW IEC Device[2]

using a 1000 L/s turbo pump. Within this chamber are two highly transparent conducting grids. The outer stainless steel anode grid is 50 cm in diameter and is kept at ground potential. The inner tungsten-rhenium cathode grid is 10 cm in diameter and is connected to a 200 kV power supply with a high voltage feed thru.

During normal operation, deuterium gas is fed into the chamber to produce a background pressure of 2 mtorr. This gas is then ionized using electron bombardment from standard light bulb filaments. Negative voltage is applied to the inner grid anode, and the positively charged ionized gas is attracted to the center of the grid.

The UW-IEC fusion device was modified to conduct this set of experiments. The inner cathode grid was replaced by solid target samples, such as the one shown in Figure 2. The device was then run at a pressure of ~0.5 mtorr to ensure a more uniform ion energy distribution on the samples.

Fig. 2. TaC Sample in IEC

II. EXPERIMENTAL PROCEDURE

II.A. As-Received Samples

Samples have been received both from HAPL scientists and from Ultramet, a manufacturer of tungsten-coated foam. SEM micrographs were taken of all samples both before and after irradiation in the IEC. Figure 3 shows a view of the foam samples. A typical foam sample has a surface area of ~ 4 cm^2, while single crystal tungsten samples have a surface area of 1 cm^2.

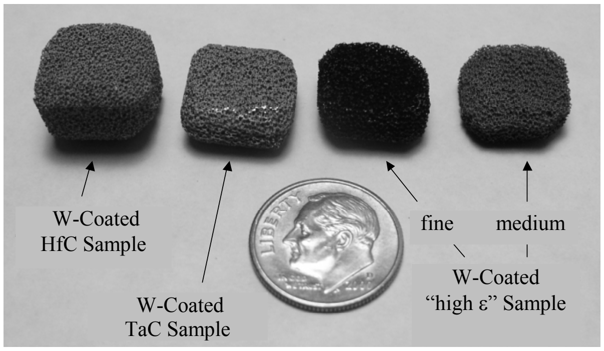

Fig. 3. As-Received Foam Samples

Sample	Ions	Fluence (#/cm^2)	Temp (C)	Energy (kV)
TaC-1	D^+	~10^{18}	800+	up to 50
TaC-2	$^4He^+$	$6x10^{17}$	830	30
TaC-4	---	---	1200	---
HfC-1	$^4He^+$	$6x10^{17}$	775	30
HfC-4	---	---	1200	---
TaC-ε-1	$^4He^+$	$6x10^{17}$	varied	30
TaC-ε-4	---	---	1200	---
Single Crystal	$^4He^+$	$4x10^{16}$	830	30

Table 1. Summary of HAPL Experiments

As shown in Figure 4, the Ultramet foam samples are composed of a hollow TaC or HfC cores with a chemical vapor infiltration technique used to coat the cores with large or medium grain tungsten.[6] The “high ε” foam has an extra layer of fine grain dendritic W particles coating one side of the sample, as seen in Figure 3.

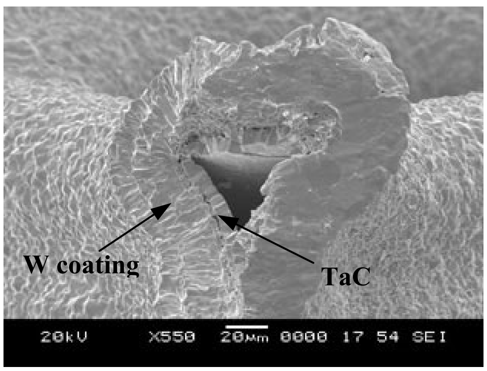

Fig. 4. Cross Section of TaC Sample (Courtesy of Ultramet)

The series of micrographs in Figure 5 of an as-received W-coated TaC sample shows the complex structure of the Ultramet Foam. The three types of foams appear similar at low magnification (the first picture in Figure 5), but can vary significantly at higher magnification, as seen later in the Results section.

Each of the samples was then mounted in the IEC device and irradiated to various fluence levels at temperatures ranging from 700 – 1100 °C. Temperatures were measured using a pyrometer. A summary of the fluence history of the samples used in this set of experiments can be seen in Table 1. Note that many of the samples were not irradiated in the IEC device, but were instead vacuum annealed outside of the device at 1200 °C for 30 minutes. These annealed control samples were used to distinguish damage caused by heating from that due to the irradiation process.

Irradiation times in the IEC device ranged from roughly 20 seconds to 30 minutes. Temperature, pressure, voltage, and current were constantly monitored during the irradiation to ensure constant fluxes.

III. RESULTS AND DISCUSSION

After each sample was irradiated in the IEC device, SEM micrographs were again taken to observe the surface morphology changes that occurred due to ion bombardment.

III.A. Single Crystal Samples

Previous work by Cipiti on the Wisconsin IEC device showed that polycrystalline tungsten samples began to experience pore formation at ion fluences of $4x10^{16}$ He^+/cm^2 at 1100 °C.[3] The pores would then migrate to the grain boundaries or be swept up by the moving grain boundaries during recrystallization, as can be seen in Figure 6. This work provided the motivation to test single crystal samples at similar conditions.

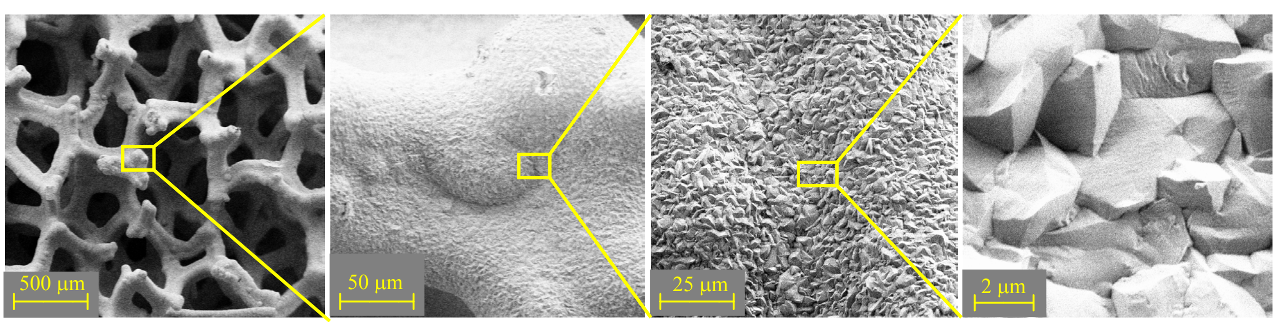

Fig. 5. SEM Micrographs of HfC Foam Samples Coated with W. The final picture reveals the small-scale structure of the tungsten coating.

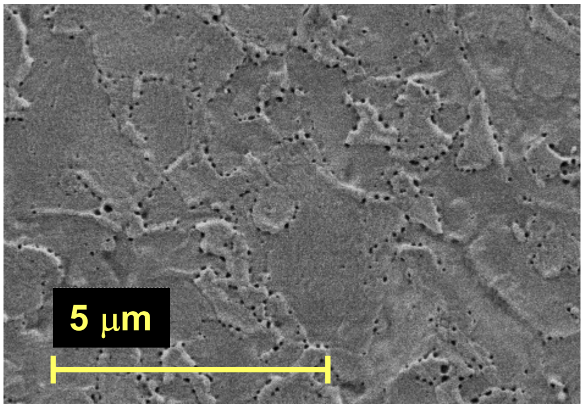

Fig. 6. Irradiated Polycrystalline Tungsten. The sample was irradiated at ~1100 C to 4x10^{16} He^{+}/cm^{2}

The single crystal tungsten sample in this study was irradiated with 30 kV He^{+} ions to a fluence of 4x10^{16} He^{+}/cm^{2} at 830 °C. As can be seen in Figure 7, a dramatically reduced number of pores were formed as compared to the polycrystalline sample. As there were no grain boundaries, there seemed to be no preferential location for pore formation or migration.

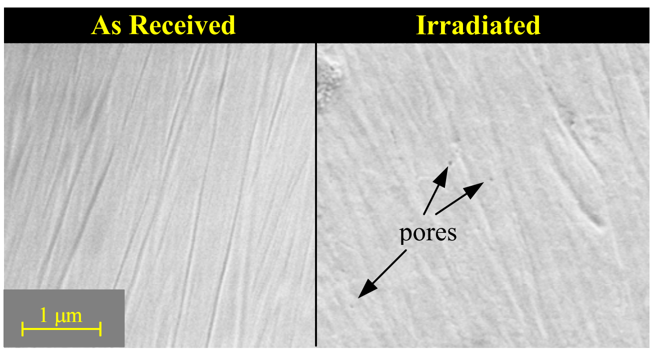

Fig. 7. As-Received and Irradiated Single Crystal. The sample on the right was irradiated at 800 C to 4x10^{16} He^{+}/cm^{2}

III.B. W-coated HfC and TaC Foam Samples

Figure 8 shows SEM micrographs of a large grain W-coated TaC sample before and after being vacuum annealed at 1200 °C for 30 minutes. Note that while the micrographs are of the same sample, they are not of the exact same location. The annealing process had little effect on the surface morphology, aside from a slight roughening of the surface.

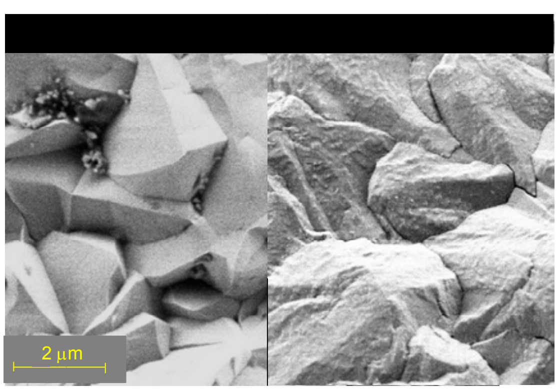

Fig. 8. Large Grain W-coated TaC Sample Before and After Vacuum Annealing. (1200 °C for 30 min)

When subjected to D^{+} and He^{+} fluences in the UW IEC Device, the two types of large grain W-coated foam samples (TaC and HfC) studied were observed to respond very similar to each other. As discussed above, both types of W- coated foam are coated with a relatively thick (30-50 μm) tungsten layer of similar composition. However, the fast ions have a range of less than 100 nm, leaving the carbide substrate unaffected by the irradiation.

The first large grain W-coated sample was irradiated at ~800 °C with approximately a ~10^{18} D^{+}/cm^{2} fluence. As can be seen in Figure 9 below, the surface was significantly smoothed, presumably from the combined high temperature and sputtering environment.

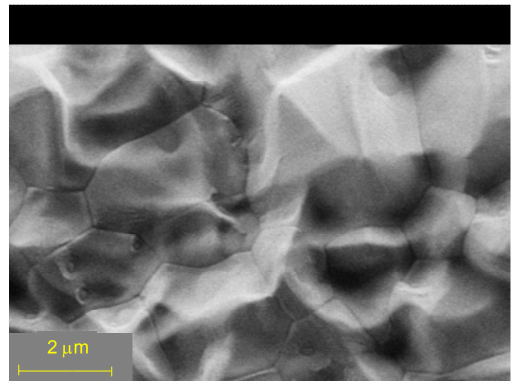

Fig. 9. Large Grain W-coated TaC Sample. This sample was Irradiated at over 800 °C with ~10^{18} D^{+}/cm^{2}

Figure 10 shows SEM micrographs of a W-coated HfC sample before and after being vacuum annealed at 1200 °C for 30 minutes. As in the TaC sample, the annealing process had little effect on the surface morphology.

The W-coated HfC sample was irradiated at 775 °C with a 6x10^{17} He^{+}/cm^{2} fluence. When the foam samples were subjected to this He^{+} fluence, a very different phenomenon was observed than the D^{+} case. Figure 11 shows the dramatic effects on the surface morphology of the target. Not only did this sample experience a smoothing effect much like the D^{+} irradiated sample, but also it showed extensive pore formation at the surface of the sample.

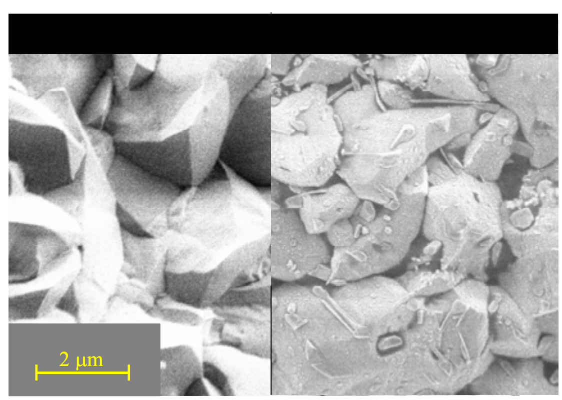

Fig. 10. Large Grain W-coated HfC Sample Before and After Vacuum Annealing (1200 °C for 30 min)

As was noted before, the He^+ ions have a range of roughly 100 nm in tungsten. When the ions become embedded in the material, it is possible their mobility is so low that most are unable to diffuse back out of the bulk. These trapped He atoms could then collect to form bubbles beneath the surface of the material. As material is sputtered from the surface, these bubbles may then be exposed and appear as pores on the tungsten surface. This behavior is very similar to that seen in previous work with polycrystalline samples.[1, 3]

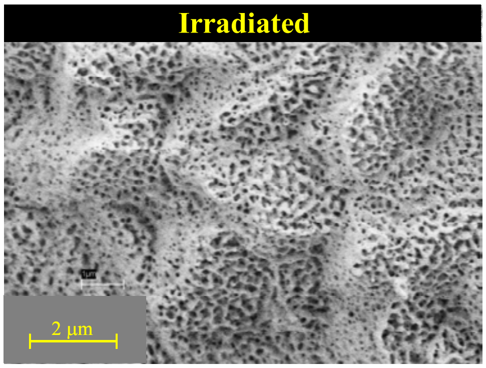

Fig. 11. W-coated HfC Sample. This sample was Irradiated at 775 °C with a 6×10^{17} He^+/cm^2 Fluence

III.B. Medium/Fine Grain W-Coated Foam Samples

The final foam sample that was evaluated had a chemical vapor infiltration medium grain tungsten coating similar to that of the first two samples. However, one side of the sample was then coated with an additional layer of fine grain tungsten dendrites. These small particles act to increase the emissivity of the foam.

SEM micrographs of the medium and fine grain W-coated samples before and after being vacuum annealed at 1200 °C for 30 minutes are shown in Figures 12 and 13. The medium grain side of the sample seemed to react similarly to the large grain W-coated TaC and HfC samples, while the side with the fine grain tungsten appears to have experienced some recrystalization.

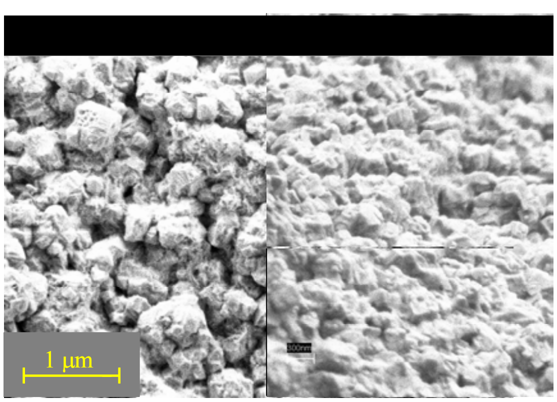

Fig. 12. Medium Grain Side of the "High ε" Sample Before and After Vacuum Annealing (1200 C for 30 min)

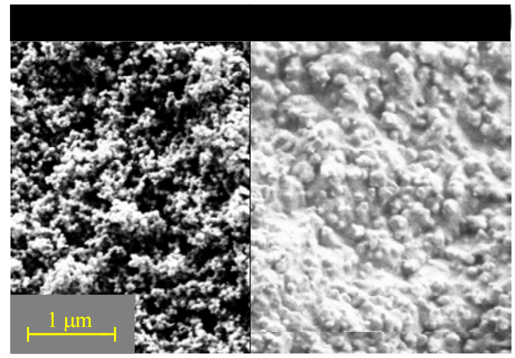

Fig. 13. Fine Grain W Side of the "High ε" Sample Before and After Vacuum Annealing (1200 C for 30 min)

The spherical symmetry of the IEC device provided a relatively uniform 6×10^{17} He^+/cm^2 fluence across both sides of the sample. As seen in Figure 14, the medium grain W side exhibited a uniform pore density similar to those seen in the W-coated TaC and HfC samples.

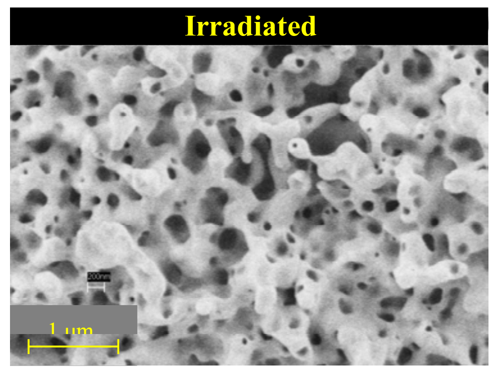

Fig. 14. Medium Grain Side of the "High ε" Sample. Irradiated with a 6×10^{17} He^+/cm^2 Fluence

The fine grain W-coated side of the "high ε" foam experienced a very different phenomenon during irradiation. The "black" tungsten dendrites applied to the foam appeared to grow after irradiation. There also seemed to be a reduced pore density on the tungsten surface.

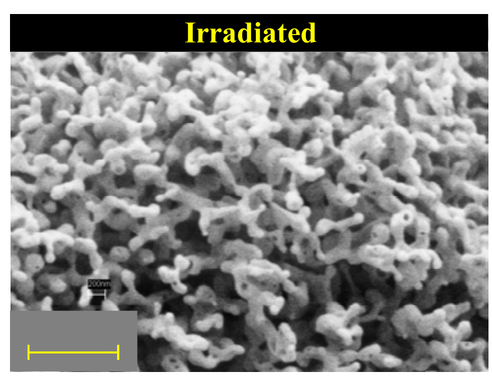

Fig. 15. Fine Grain Side of the "High ε" Sample. Irradiated with a 6×10^{17} He^+/cm^2 Fluence

IV. CONCLUSIONS

IV.A. Single Crystal Samples

The single crystal tungsten sample in this study was irradiated with 30 kV He^+ ions to a fluence of $4x10^{16}$ He^+/cm^2 at 830 °C. SEM micrographs showed a dramatically reduced number of pores were formed as compared to previously studied polycrystalline samples. Future work should extend the range of fluences on single crystal samples to determine the behavior of pore formation.

IV.B. W-Coated HfC and TaC Foam Samples

After vacuum annealing and when subjected to D^+ and He^+ fluences in the UW IEC Device, the two types of large grain W-coated foam samples (TaC and HfC) studied were seen to react very similar to each other. The 1200 °C vacuum annealing process had little effect on the surface morphology, aside from a slight roughening. When a W-coated foam sample was irradiated by D^+ at over 800 °C with a ~10^{18} D^+/cm^2 fluence, it experienced a significant smoothing presumably from the combined high temperature and sputtering environment, but exhibited no pore formation. However, when subjected to the same fluence of He^+ particles at similar temperatures, the samples were seen to undergo extensive pore formation comparable to that seen in previous work with polycrystalline Tungsten.

IV.C. Medium/Fine Grain W-Coated Foam Samples

The medium grain side of the sample seemed to react similarly to the large grain W-coated TaC and HfC samples to both vacuum annealing and irradiation processes. The side coated with the "black" fine grain tungsten appears to have experienced some recrystalization during the vacuum annealing process. When subjected to a $6x10^{17}$ He^+/cm^2 fluence at 800 °C, the fine grain dendrites appeared to grow and showed a reduced pore density as compared to the medium grain side.

IV.C. First Wall Applications

The creation of micro-pores, as well as the changes in surface morphology, in the tungsten surfaces may have an impact on the lifetime of W coatings on the first walls of inertial and magnetic confinement fusion reactors. The high density of small pores could act as nucleation sites for cracking under the repeated shock environment of ICF operation. The interconnection of pores could also produce micron-size radioactive "dust" particles. Finally, the pores may act as release sites for He in the tungsten surface layer, potentially increasing the life of the material. Further study, both to extend the ion energy range and evaluate the thermo-mechanical properties of these materials, will be required to evaluate the feasibility of using tungsten-coated foams for first wall use.

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