DEVELOPMENT OF REFRACTORY ARMORED SILICON CARBIDE - T. Hinoki, L.L. Snead, C.A. Blue, M.L. Santella, D.C. Harper and N. Hashimoto (Oak Ridge National Laboratory), H. Kishimoto (Kyoto University)

OBJECTIVE

The objective of this work is to develop refractory armor on silicon carbide for fusion applications.

SUMMARY

Tungsten (W) and molybdenum (Mo) were coated on silicon carbide (SiC) as refractory armor using high power plasma arc lamp at powers up to 23.5 MW/m^2. Both W powder and Mo powder were melted and formed coating layers on silicon carbide. The effect of pretreatment (vapor deposition of titanium (Ti), W and Mo and annealing) and sample heating conditions on microstructure of the coating and coating/substrate interface were investigated. The microstructure was observed by scanning electron microscopy (SEM) and optical microscopy (OM).

A uniform strong W coating was successfully formed. Tungsten vapor deposition and pre-heating at 5.2 MW/m^2 made for a refractory layer containing no cracks which propagated into the SiC substrate. This layer was formed without the thick reaction layer (WC and W_5Si_3) reported in previous studies. Moreover the thinner interface transition layer and armor avoid coefficient of thermal expansion (CTE) mismatch driven failure previously reported. For this study, small W_xC_y grains were observed adjacent to interface. Silicon carbide grains and W_xSi_y grains were observed within W coating. By contrast, Mo was not successfully formed. Most of Mo coating debonded at reaction layer due to too much absorbed energy for Mo and larger CTE mismatch than that of W and SiC. Further study at lower absorbed powers would be required to carry the Mo coating further.

PROGRESS AND STATUS

Introduction

Silicon carbide can be used in extremely harsh environment due to its excellent thermal, mechanical and chemical stability. Silicon carbide also provides exceptionally low radioactivity following neutron

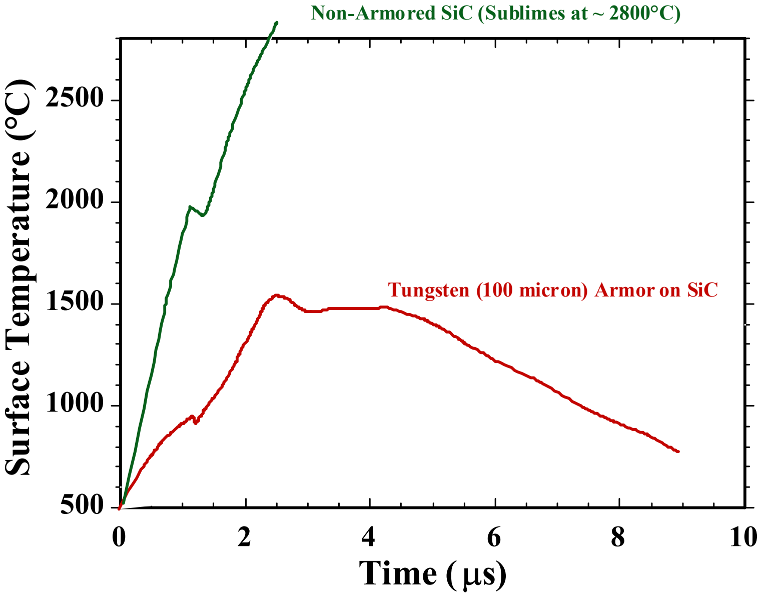

Figure 1: Effect of W armor on the calculated surface temperature of SiC*

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* Data provided by R. Raffray, UCSD, 2002.

irradiation [1]. The intrinsic features of SiC make SiC fiber reinforced SiC matrix composites (SiC/SiC composites) significantly attractive structural material for nuclear application [2].

Refractory armored materials (RAM's) have been previously considered in the magnetic fusion energy community to reduce the introduction of power-sapping impurities into the plasma [3], and to reduce erosion. Recently RAM's have been considered for inertial fusion energy systems as a means to absorb and handle the very high impulse heat loading expected. Figure 1 shows the effect of W armor on the calculated surface temperature of non-armored SiC and W armored SiC.

To select the refractory armor material for SiC, CTE is one key to obtain a strong bond between substrate and refractory armor, since CTE mismatch causes residual stress at the interface. To reduce the CTE mismatch, W and Mo were selected as the refractory armor materials for SiC. Table 1 shows the thermal properties of SiC used, W and Mo.

Table 1: Thermal properties of substrate and coating materials

Material	CTE (RT, 10^{-6}/K)	Melting point (°C)	Thermal conductivity (RT, W/mK)
Hexoloy SA SiC	4.02	2797 (α-SiC)	125.6
W	4.4	3370	163.3
Mo	5.35	2617	138

There are many techniques to apply refractory metals to ceramic substrates including plasma spraying, chemical and plasma vapor deposition. However, the chemical bonding, hence mechanical strength of layers produced with those techniques is low. A novel approach to forming this refractory layer using the Oak Ridge National Laboratory (ORNL) 300 kW infrared plasma arc lamp (IR processing) [4] was used. This facility can apply up to 35 MW/m^2 over an area of 35 cm by 3.175 cm in transient or continuous mode. The assumption driving application of this system for SiC/W was that by applying transient infrared power a layer of W powder could melt and react with the surface of the SiC while keeping the bulk SiC below its sublimation temperature.

Experimental

The substrate material for this study was Hexoloy® SA SiC (sintered α-SiC). Substrates with dimension 25 mm (long) × 15 mm (wide) × 3 mm (thick) were machined from plate. As refractory armor material, W and Mo powder were brushed or sprayed on the SiC to an approximate thickness of 100 µm. Several kinds of pretreatments including vapor deposition of Ti, W and Mo, and pre-melt substrate annealing were applied prior to forming the armor. The thickness of vapor deposition of Ti, W and Mo was 0.20 µm, 2.00 µm and 1.18 µm, respectively. The annealing temperatures after the vapor deposition were 1300 °C or 1500 °C for 72 hours in a vacuum. The conditions of the pretreatments are summarized in Table 2.

Table 2: Pretreatment conditions

Vapor deposition (µm)	Annealing (°C)
W (2.00)	
Ti (0.20) + W (2.00) or Mo (1.18)	
W (2.00)	1300 (72 hours)
Ti (0.20) + W (2.00) or Mo (1.18)	1300 (72 hours)
W (2.00)	1500 (72 hours)
Ti (0.20) + W (2.00) or Mo (1.18)	1500 (72 hours)

The plasma arc lamp used by IR processing is mounted on a Cincinnati-Milacron model T3-776 robotic arm. Using the robotic arm in conjunction with the Plasma Arc Lamp allows for a variety of experiments to be performed in a short amount of time. The powder on SiC was heated by the lamp with 23.5 MW/m^2 for 2~5 sec at argon flow environment. Pre-heating and post-heating at lower power were also applied to optimized IR processing conditions.

The refractory armored SiC was sliced, and the coating and interface were observed by OM and SEM with EDS.

Result and discussion

Both W and Mo on SiC melted in a couple of seconds by IR processing and formed coating on SiC. It is most likely that this SiC was removed by sublimation of the surface of the SiC prior to, ordering the W powder melt. An OM image is given if Figure 2, along with a micrograph of the as-machined substrate to allow comparison of the surface roughness. No diffusion of W into SiC was observed.

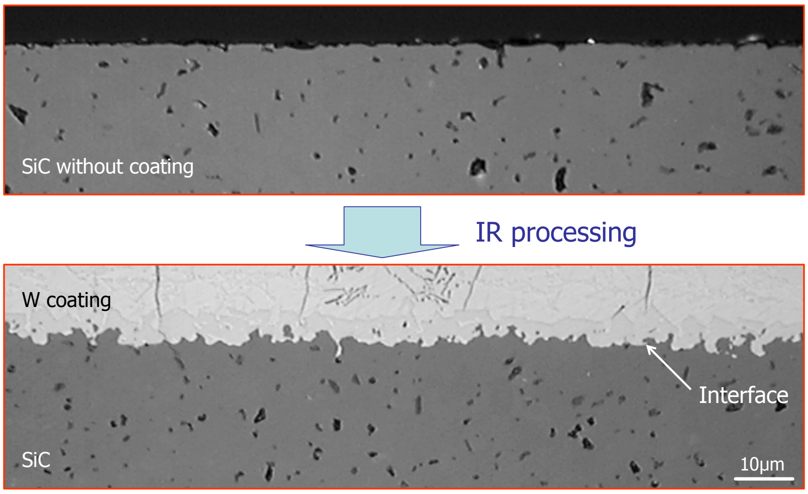


Figure 2: Effect of IR processing on SiC surface roughness (OM images)

All W coated specimens with pretreatments were stable following IR processing, while most of specimens without the pretreatments broke following IR processing. In the "pretreated" cases, the critical ingredient appears to be the vapor deposited W layer. The addition of very thin Ti layer under the vapor deposited W was used in the hope of aiding the wetting of W to SiC. It has been reported that reaction between W and SiC occurs above 1200 °C leading to a bulk diffusion path $W/W_5Si_3/WC/SiC$ [5]. The thickness of those regions depends on temperature and time. In this work it was assumed that a reaction layer several microns thick was formed at interface. However, given the apparent removal of surface SiC through sublimation and the absence of Ti (using EDS) it is felt that the use of Ti and the annealing were unnecessary.

IR processing conditions were optimized using a series of specimens without pretreatments. Maximum power to melt powder was fixed to 23.5 MW/m^2 for 3 seconds. Pre-heating and post-heating at reduced power was used to decrease thermal shock. Post-heating did not improve stability of specimens following IR processing with all samples fracturing. In contrast, pre-heating at 5.2 MW/m^2 for 20 seconds improved stability of specimens, and the specimens with the pre-heating was stable following IR processing. Figures 3 are OM (Nomarski) images showing the effect of W vapor deposition and pre-heating on crack propagation into SiC. In the specimen without W deposition and pre-heating, large cracks induced from W surface propagated into substrate SiC (a), while cracks induced from W surface did not reach the W/SiC interface in the specimens with W vapor deposition (b) and pre-heating (c). It is considered that W powder was porous prior to IR processing allowing part of the energy to be absorbed by the SiC directly, and the energy caused expansion of SiC near interface with crack propagation at cooling. Slowing down the cool down by slowing down the lamp power did not reduce the armor and substrate cracking. However if the whole SiC substrate is allowed to absorb energy at low power not to melt W powder prior to W coating formation, it will expand with temperature increasing. Relative expansion of SiC near surface caused by high energy to melt W was reduced by pre-heating. As a result pre-heating reduced the crack into SiC. By adding the thin W layer by vapor deposition, it is thought that IR power was not effectively coupled to the SiC substrate reducing expansion of SiC near surface.

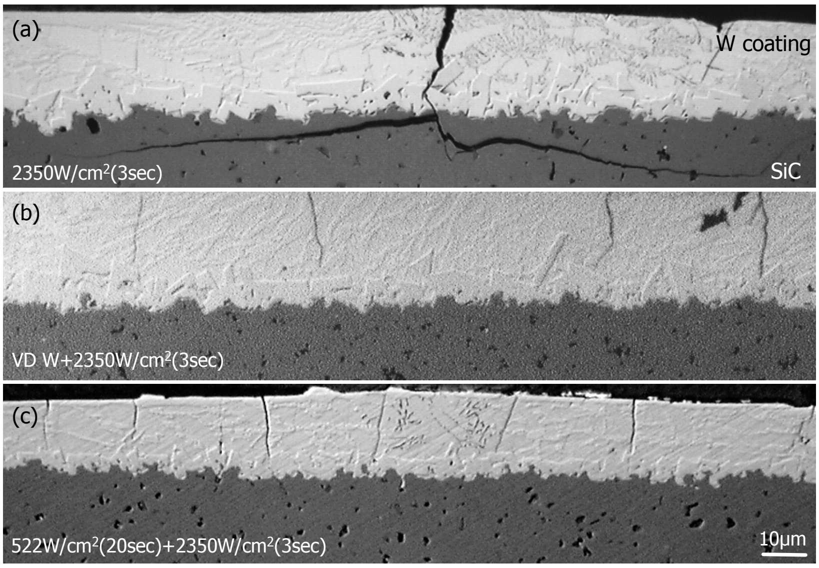


Figure 3: Effect of vapor deposited W and pre-heating on crack propagation into SiC

Figure 4 shows back scattering (composition) electron image of a specimen with pre-heating (5.2 MW/m^2 + 23.5 MW/m^2). SEM observation reveals a complicated microstructure within the W coating. Through EDS analysis, grains it was found that grains adjacent to W/SiC interface within W coating was W_xC_y. The W_xSi_y grains were found near surface. SiC grains were also found within W coating. It is considered that the sublimated SiC at interface decomposed and reacted within the melted W. It has been reported that W was coated on SiC successfully by other methods. However those W coatings included relatively thick reaction layers of WC and W_5Si_3 [5]. Even with strong bonding at room temperature, those reaction layers cause fracture at the interface by stress concentration attributed to mismatch of CTE at high temperature. For this reason W coating with thin reaction layers are preferred. By IR processing the reaction layer was not formed at W/SiC interface, although relatively small reacted grains were observed. This grain structure can reduce the crack initiation by CTE mismatch compared with the reaction layers, since the stress caused by CTE mismatch distributes to each grain boundary.

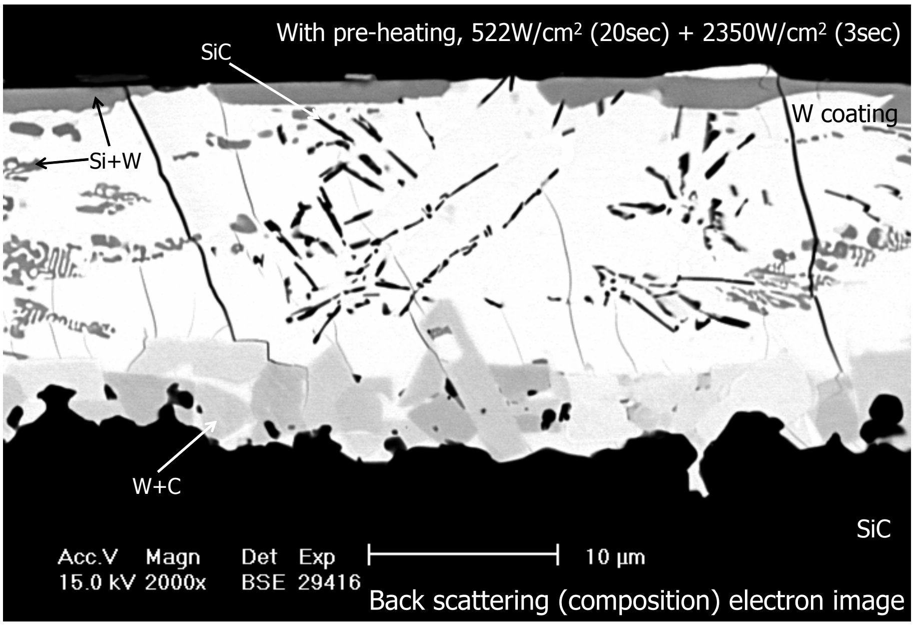
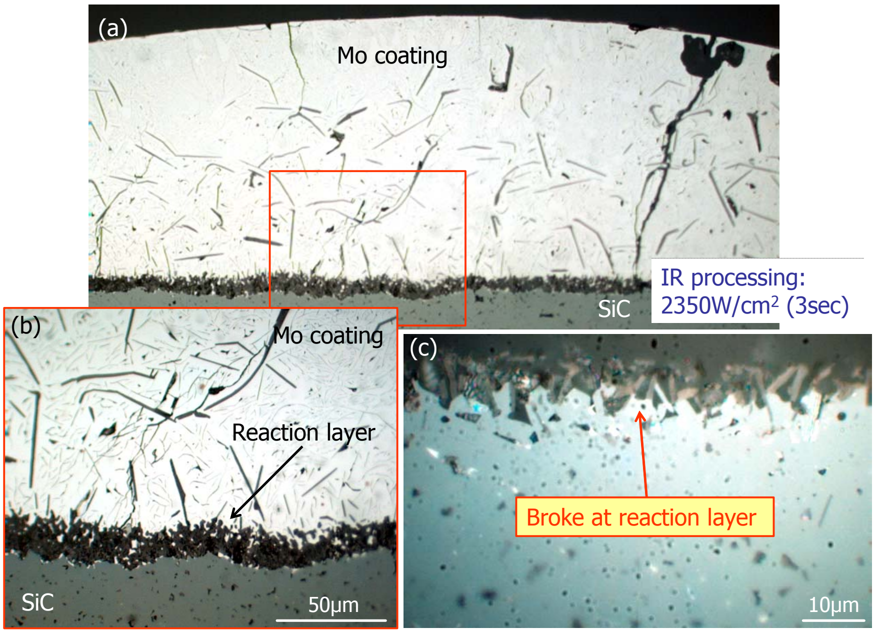


Figure 4: SEM image (back scattering electron image) of W coating on SiC

Molybdenum coating was applied by IR processing at the same power used for W coating. Most of Mo coating disappeared due to application of too high a power density for Mo. Figures. 5 show OM images of the remaining Mo coating (a, b) and debonded interface (c). The reaction layer was formed at Mo/SiC interface. It was found that debonded substrate surface did not have the reaction layer and most of Mo coating debonded at the reaction layer. The difference of CTE of Mo and SiC is larger than that of W and SiC. Even with optimization of IR processing, it might be difficult to obtain strong bonding with Mo coating compared with W coating.



Figures 5: Mo coating on SiC (a,b) and debonded surface of SiC (c)

CONCLUSION

1. Tungsten coating was formed successfully on a SiC substrate. The SiC substrates without pretreatments or pre-heating were broken by IR processing. It was found that vapor deposition of W and pre-heating significantly reduced cracks within the silicon carbide. The W_xC_y grains were formed near interface within W coating in all specimens, while SiC grains and the W_xSi_y grains were observed in middle and surface of W coating. It was found that a thick reaction layer which causes fracture due to CTE mismatch was not formed by the coating method used in this work.
2. Most of Mo coating disappeared due to application of too high a power density. Furthermore a reaction layer which may have enhanced crack propagation was observed.

ACKOWLEDGEMENT

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