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Behavior of plasma-sprayed tungsten coatings on CFC and graphite under high heat load

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Abstract

Tungsten coatings of 0.5 and 1 mm thickness were successfully deposited by the vacuum plasma spraying technique on carbon/carbon fiber composite (CFC), CX-2002U, and isotropic fine grained graphite, IG-430U. High heat flux experiments by irradiation of electron beam with uniform profile were performed on the coated samples in order to prove the suitability and load limit of such coating materials. Heat load properties, gases emission, surface modification and structure changes of cross-section of the samples were investigated. Cracks on the surface and exfoliation between the joint interface of the samples were not formed below the melting point. These results indicated that the thermal and adhesion properties between the substrate and coatings were good under high heat flux. Microstructure of the joint interface of the sample was changed in the case of a peak temperature at about 2800°C. Many cracks and traces of melted tungsten flow were observed on the surface after melting and solidification. Large cavities were also formed inside the resolidified tungsten layer.

Keywords: Carbon based materials; Coating; Divertor armor tiles; Heat flux; Heat load; High Z material; Limiter materials; Tungsten

1. Introduction

Although the utilization of low Z material like carbon materials for plasma-facing components has enabled the improvement in plasma confinement, their high erosion rates at elevated temperatures is now a serious problem. Degradation of thermal conductivity by neutron damage and high tritium retention would be a serious problem in the next generation of D–T fusion experimental reactor [1]. Owing to its low sputtering yield and good thermal properties, tungsten seems a promising candidate material for plasma-facing components in next fusion experimental devices. But tungsten is not easy to machine and weld.

For a technical realization of tungsten material, tungsten-coated carbon tiles can be envisaged. Tungsten coatings on graphite by plasma spray or physical vapor deposition (PVD) were produced. High heat flux experiments were performed on the coating tiles [2,3]. It is preferable to use carbon fibre composite, (CFC) as a substrate for high heat flux components owing to its good thermal conductivity and mechanical strength.

Thick tungsten coatings on carbon/CFC as well as isotropic fine grained graphite were successfully produced [4]. High heat flux experiments have been performed on the coated samples in order to prove the suitability and load limit of such coated materials. In Ref. [4], we reported results of heat flux experiments by irradiation of electron beam with spot-like profile. In this present work, heat load experiments using electron beam with uniform profile were performed. Microstructural and compositional change of cross- section as

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well as surface modification of the samples before and after the irradiation was observed.

2. Experimental

Tiles, 20 mm × 20 mm × 10 mm, were coated by the vacuum plasma spraying technique (VPS). The substrate materials were carbon/carbon composite CX-2002U and isotropic fine grained graphite IG-430U made by Toyo Tanso. The CX-2002U and IG-430U received PVD multilayer diffusion barrier layers [1] of rhenium and tungsten prior to the VPS tungsten coating in order to inhibit uncontrolled brittle carbide formation. Heat treatments were performed to stabilize the microstructure of the sample. The thickness of the tungsten coating layer was 0.5 and 1.0 mm. The density of VPS tungsten (VPS-W) was 92.5% of the theoretical density. In the present experiments, the samples with 0.5 mm thickness of tungsten were used.

The facility used in these experiments was an active cooling teststand (ACT) [5] of National Institute for Fusion Science. The samples were placed on a carbon/copper block, actively cooled in water. The electron beam energy used was 30 keV. The electron beam was irradiated on the sample surface through a beam limiter with an aperture of size 30 mm × 30 mm leading to an almost uniform beam irradiating the sample. The exposure time of the sample was controlled using a beam shift system of the ACT. In this experiment, the duration of the beam was 20 s. The net current was measured by applying a bias voltage (+90 V) to the sample to suppress the secondary electron induced by the electron beam irradiation. The thermal electron from the heated sample surface was also suppressed by the bias voltage. The current was controlled to provide a preselected heat flux by a current controller, which changed the filament current of a electron beam source. The heat load experiments were performed stepwise by increasing the heat flux. The surface temperature of the center region with a diameter of 5 mm of the sample was measured with an optical pyrometer (300°C–3000°C). A calibration curve between the thermocouple temperature and the signal intensity of the pyrometer was obtained in the temperature range from 300°C to 700°C before the experiment. The calibration curve was extrapolated at more than 700°C in the temperature measurement using the pyrometer. Gases emitted from the heated sample surface were detected with a quadrupole mass spectrometer (QMS). Before and after the irradiation, the sample surface was observed with a scanning electron microscope (SEM). Microstructural and compositional change before and after the irradiation was also examined with a SEM equipped with an energy dispersion X-ray spectroscope (EDS).

[1] PVD W-Re multilayer diffusion barrier coating is patented by Plansee.

3. Results

3.1. Observation of VPS-W coatings

SEM observation of the surface of the VPS-W coatings showed that the spherical particles were melted or partially melted, joined with each other and pores were formed on the coatings [4]. Fig. 1 shows backscattered electron image (BEI) of joint interface of cross-section of VPS-W coated CX-2002U(#6) with a tungsten thickness of 1 mm. Tungsten was coated by the VPS after the deposit of multilayer of Re and W. It can be seen that the color of the first layer of the tungsten is darker than that of the other tungsten layers. This indicates that the composition was different from that of the other tungsten layers. Compositional analyses showed that the first tungsten layer includes about 34 at.% carbon. Owing to the temperature of the heat treatment after coating, tungsten carbide is expected to be formed in the first tungsten layer. On the other hand, the second layer and the thick VPS-W layer of tungsten did not include such large amount of carbon. This shows that the W/Re multilayer acts as a diffusion barrier for carbon and suppresses the tungsten brittle carbide formation of the main tungsten layer (VPS-W). No sign of delamination is seen.

3.2. Behavior of VPS-W coating under heat flux

Fig. 2 shows the time evolution of the electric current of the sample (a), surface temperature (b) and pressure of vacuum chamber (c). The sample was VPS-W coated

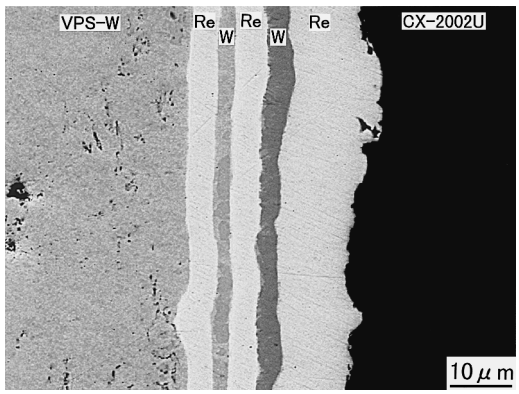

Fig. 1. BEI (backscattered electron image) of the interface of cross-section of VPS-W coated CX-2002U(#6) with a tungsten thickness of 1 mm.

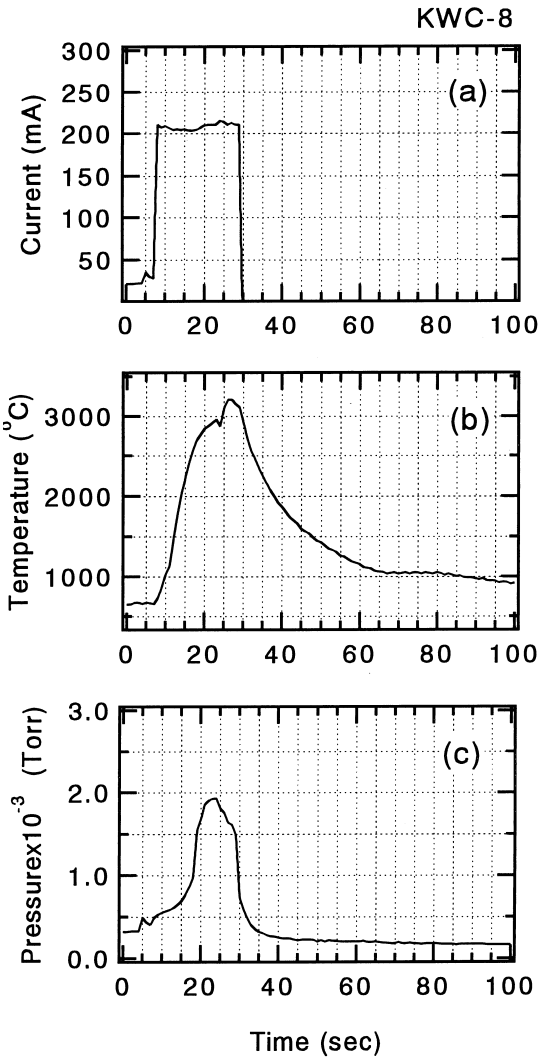

Fig. 2. The time evolution of the electric current of the sample (a), surface temperature (b) and pressure of vacuum chamber (c). Sample was VPS-W coated CX-2002U(#2) with a thickness of tungsten of 0.5 mm. The heat flux was 5.5 MW/m^2.

CX-2002U(#2) with a thickness of tungsten of 0.5 mm and the heat flux was 5.5 MW/m^2. In this case, the surface was melted. The electric current started to increase at the same time as the irradiation started and was almost constant during irradiation. The surface temperature gradually increased and reached about 3200°C and started to decrease when the irradiation ended. Slope of the increased temperature decreased at 2000°C. This is expected to be due to radiation loss from the heated surface. The pressure of vacuum chamber gradually increased and decreased during irradiation.

QMS spectra before and during irradiation showed that the emitted gases were mainly H_2, H_2O, CO and CO_2. This result indicated that the kind of the emitted gases was almost the same as that of the powder metallurgy tungsten and the CX-2002U [6]. Fig. 2(b) shows that when the temperature reached about 2900°C, temperature decreased slightly. Calibration of the temperature measured with the optical pyrometer at high-temperature was not performed, but owing to the temperature evolution, it seems melting occurred when the temperature of signal intensity of the pyrometer was about 2900°C. In addition to this, the time evolution of $m/e = 2$ (H_2), 18 (H_2O) measured with the QMS started to decrease at the same time. It is expected that this was due to gettering effect by tungsten evaporation. This result corresponds to the pressure change as shown in Fig. 2(c).

Fig. 3 shows surface temperature increase of the samples as a function of the heat flux. It can be seen from this figure that the temperature almost linearly increased with increasing heat flux and that there was little difference among the samples. In the case of 5.5 MW/m^2, tungsten of the VPS-W/CX-2002U was melted. In the other cases, cracks on the sample surface and exfoliation between joint interface of the samples were not formed. This indicates that the thermal and adhesion properties at high-temperature were good enough.

3.3. Modification of VPS-W coatings due to heat flux

After the irradiation experiments, the samples were removed from the ACT and observed with the SEM and the EDS. In the case of the VPS-W/IG-430U(#20), maximum surface temperature was about 2800°C. Very small fragments (~0.1 μm) on the surface, which were observed before irradiation [4], disappeared. However,

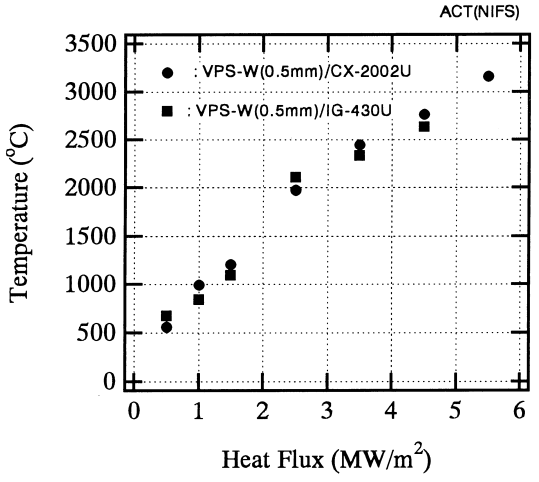

Fig. 3. Surface temperature increase of the samples as a function of the heat flux. The samples were VPS-W/CX2002U(#2) and VPS-W/IG-430U(#20) with a thickness of tungsten of 0.5 mm. The duration of beam was 20 s.

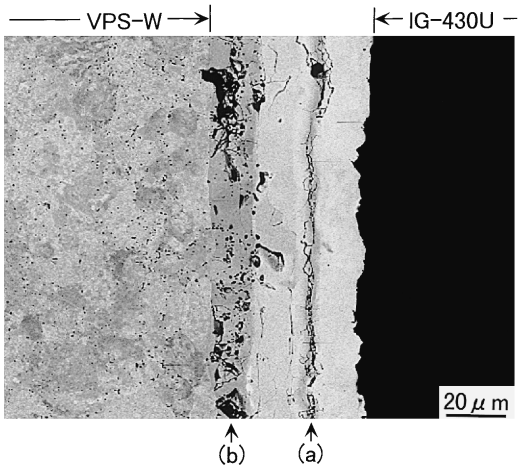

Fig. 4. BEI (backscattered electron image) of joint interface of the cross-section of the VPS-W/IG-430U(#20) after the irradiation experiments. Maximum surface temperature was about 2800°C.

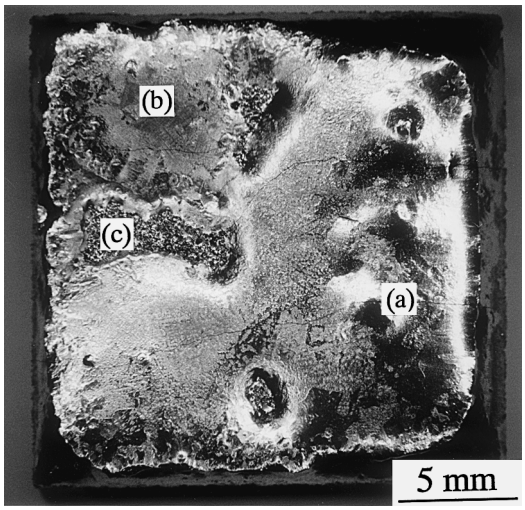

Fig. 5. Photograph of the surface of VPS-W/CX-2002U after the irradiation experiments. The sample was VPS-W coated CX-2002U(#2) with a thickness of tungsten of 0.5 mm. Surface was melted during the irradiation with a heat flux of 5.5 MW/m^2.

no other changes were observed. Cracks due to grain growth were not formed. Cross-sectional observation showed that cracks were also not formed inside the layer of the VPS-W. Fig. 4 shows BEI of the joint interface of the cross-section of the VPS-W/IG-430U(#20) after the irradiation experiments. It can be seen that the joint of the IG-430U and Re was good enough, but structure of the multilayer was changed in the layer between the VPS-W and the first deposited Re. Microcracks and exfoliation are seen in the layers (Fig. 4(a) and (b)). Compositional analyses showed that the layers include carbon. This indicates that these damages were expected to be formed by carbide formation and embrittlement.

Fig. 5 shows a photograph of the surface of the VPS-W/CX-2002U(#2) after the irradiation experiments. The surface was melted and many cracks were formed on the surface. In this case, weight loss was 33 mg. Fig. 6 shows SEM images of the sample. Fig. 6(a) shows comparatively, plainly resolidified area. The shape of the accumulation of spherical particles of tungsten after the VPS coating remains after the melting. A crack of width about 30 μm was observed. Smaller cracks were also observed. Fig. 6(b) shows another type of resolidification formation on a different part of the surface. A dendrite structure was formed. Fig. 6(c) shows an area, in which the mass loss was large. It can be seen that the resolidification structure was influenced by carbon fiber and pyloric graphite of the CX-2002U. Fig. 6(d) shows a cross-sectional view of the sample. It can be seen that the crack reached the interface of the base material and large cavities were also formed in the resolidified layer of the VPS-W. These results indicate that the structure of the melted and resolidified VPS-W drastically changes compared to that of the VPS-W.

Fig. 7 shows BEI of the joint interface of this sample. The multilayer structure was completely changed and a new structure was formed. In addition to this, microcracks between CX-2002U and metal were also observed.

4. Discussion

Results from high heat flux experiments of the same samples by irradiation of electron beam of spot-like profile ($\phi = 8$ mm) showed that cracks were formed at high temperature before the sample surface was melted [4]. In this experiment, on the other hand, irradiation of uniform electron beam did not cause crack formation on the surface. Therefore, it is expected that the cracks on the surface in the previous paper were formed due to local thermal stress by the irradiation of spot-like electron beams. These results indicate that it is necessary to evaluate the beam profile as well as heat flux.

The PVD multilayer diffusion barrier coating was applied to the tungsten coating on carbon materials. The results from this heat flux experiments show the Re layer acting as a diffusion barrier for carbon and suppressing tungsten carbide formation. The coefficient of thermal expansion of the IG-430U (4.1×10^{-6}/K) is almost the same as that of the tungsten (4.5×10^{-6}/K). On the other hand, the coefficient of thermal expansion of the CX-2002U [1.9×10^{-6}/K (X, Y), 6.0×10^{-6} (Z)] is different from that of the tungsten. Re may play a role in suppression of strain between W and CX-2002U. Next step is to apply for bigger parts, which are used in fusion devices.

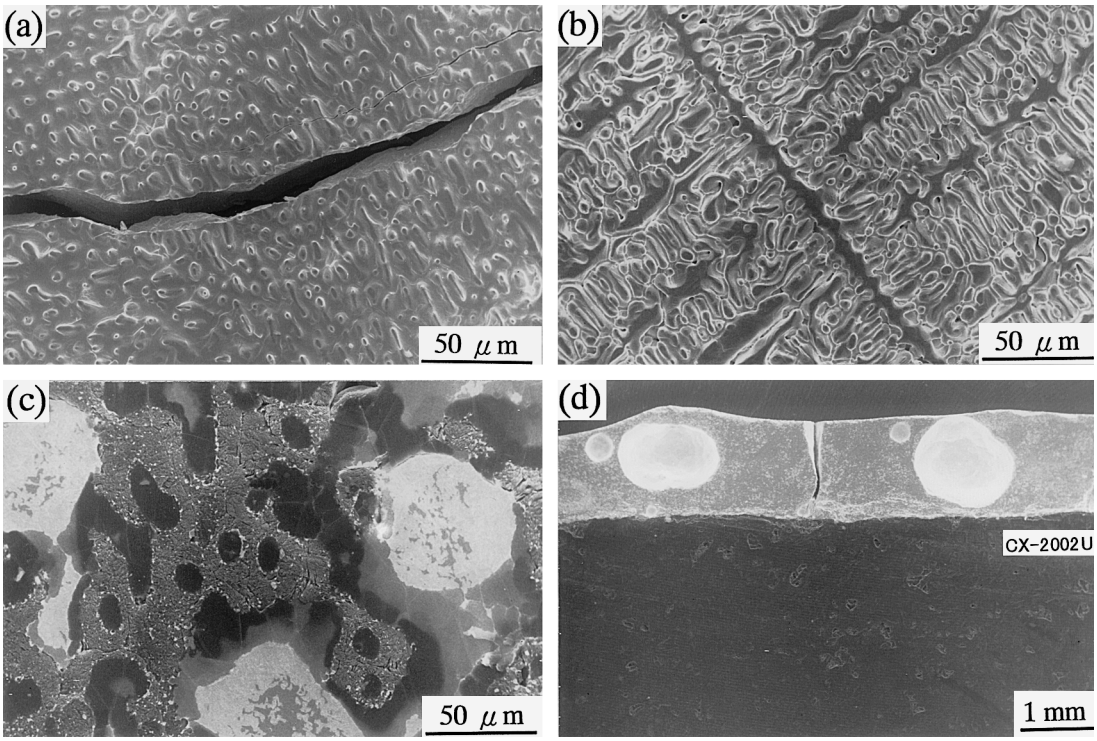

Fig. 6. SEM images of the sample of VPS-W coated CX-2002U(#2) with a thickness of tungsten of 0.5 mm after the electron beam irradiation experiments. (a), (b) and (c) correspond to the area indicated in Fig. 6. (d) A cross-sectional view.

5. Summary

Tungsten coatings of 0.5 and 1 mm thickness were successfully deposited by the VPS on carbon/CFC, CX-2002U, and isotropic fine grained graphite, IG-430U. High heat flux experiments by irradiation of electron beam with uniform profile were performed on the coated samples in order to prove the suitability and load limit of such coating materials. Heat load properties, gas emission, surface modification and structure changes of cross-section of the samples were investigated. Cracks on the surface and exfoliation between the joint interface of the sample were not formed below the melting point. These results indicate that thermal and adhesion properties between the substrate and coatings were good under high heat flux. The compositional analyses show that a Re layer acts as a diffusion barrier for carbon and suppresses tungsten carbide formation. Microstructure of the joint interface of the sample was changed in the case of a peak temperature at about 2800°C. Many cracks and traces of melted tungsten flow were observed on the surface after melting and solidification. Large cavities were also formed inside the resolidified tungsten layer.

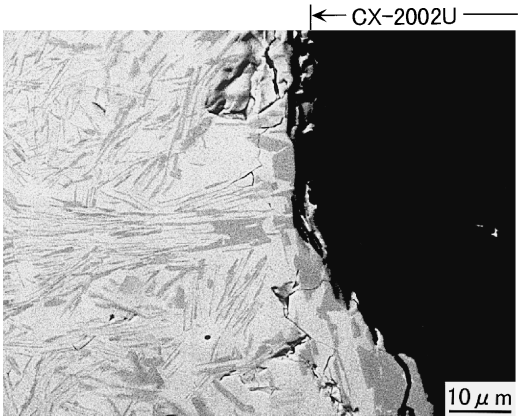

Fig. 7. BEI of the joint interface of VPS-W/CX-2002U(#2) with a thickness of tungsten of 0.5 mm after electron beam irradiation experiments.

Acknowledgements

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