ELSEVIER

Journal of Nuclear Materials 283–287 (2000) 1121–1127

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Changes of composition and microstructure of joint interface of tungsten coated carbon by high heat flux

K. Tokunaga [a,*], T. Matsubara [a], Y. Miyamoto [a], Y. Takao [a], N. Yoshida [a], N. Noda [b], Y. Kubota [b], T. Sogabe [c], T. Kato [d], L. Plöchl [e]

[a] *Research Institute for Applied Mechanics, Kyushu University, 6-1 Kasuga-park, Kasuga, Fukuoka 816-8580, Japan*

[b] *National Institute for Fusion Science, Toki, Gifu 509-5292, Japan*

[c] *Toyo Tanso Co. Ltd., Ohnohara-cho, Mitoyo-gun, Kagawa 769-1612, Japan*

[d] *Nippon Plansee K.K., Chiyoda-ku, Tokyo 102-0083, Japan*

[e] *Plansee Aktiengesellschaft, A-6600 Reutte, Austria*

Abstract

Tungsten coatings of 0.5 and 1 mm thickness were successfully deposited by the vacuum plasma spraying (VPS) 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. The cross-sectional composition and structure of the interface of VPS–W and carbon material samples were investigated. Compositional analyses showed that the Re/W multi-layer acts as diffusion barrier for carbon and suppresses tungsten carbide formation in the VPS–W layer at high temperature about 1300°C. Microstructure of the joint interface of the sample changed in the case of a peak temperature of about 2800°C. The multi-layer structure completely disappeared and compositional distribution was almost uniform in the interface of the sample after melting and resolidification. The diffusion barrier for carbon is not expected to act in this stage.

1. Introduction

Although the utilization of low Z materials like carbon materials for plasma facing components has enabled the improvement in plasma confinement, their high erosion rates at elevated temperatures are now a serious problem. Degradation of thermal conductivity by neutron damage and high tritium retention would be a serious problem in a next generation 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 the next fusion experimental devices.

Disadvantages of tungsten as a plasma facing material are its heavy weight and poor workability. One of the possibilities to overcome these disadvantages is to coat tungsten on light carbon materials, which have shown good heat load resistance in the present plasma confinement devices. Tungsten coatings on graphite by plasma spray (PS) or physical vapor deposition (PVD) were produced and their performance under high heat flux loading has been examined [2,3]. From the viewpoints of thermal conductivity and mechanical strength, it seems that carbon/carbon fiber composites (CFCs) are preferable as a substrate material for high heat flux loading. Thick tungsten coatings on CFC and isotropic fine grained graphite were successfully produced by vacuum plasma spray (VPS) technique and their good thermal and adhesion properties have been confirmed by high heat flux loading tests [4,5]. In addition, thermal response and thermal fatigue properties of the VPS–W coated CFC and isotropic fine grained graphite brazed on OFHC block have been examined under an actively cooling condition [6].

* Corresponding author. Tel.: +81-92 583 7986; fax: +81-92 583 7690.

E-mail address: tokunaga@riam.kyushu-u.ac.jp (K. Tokunaga).

PII: S0022-3115(00)00384-6

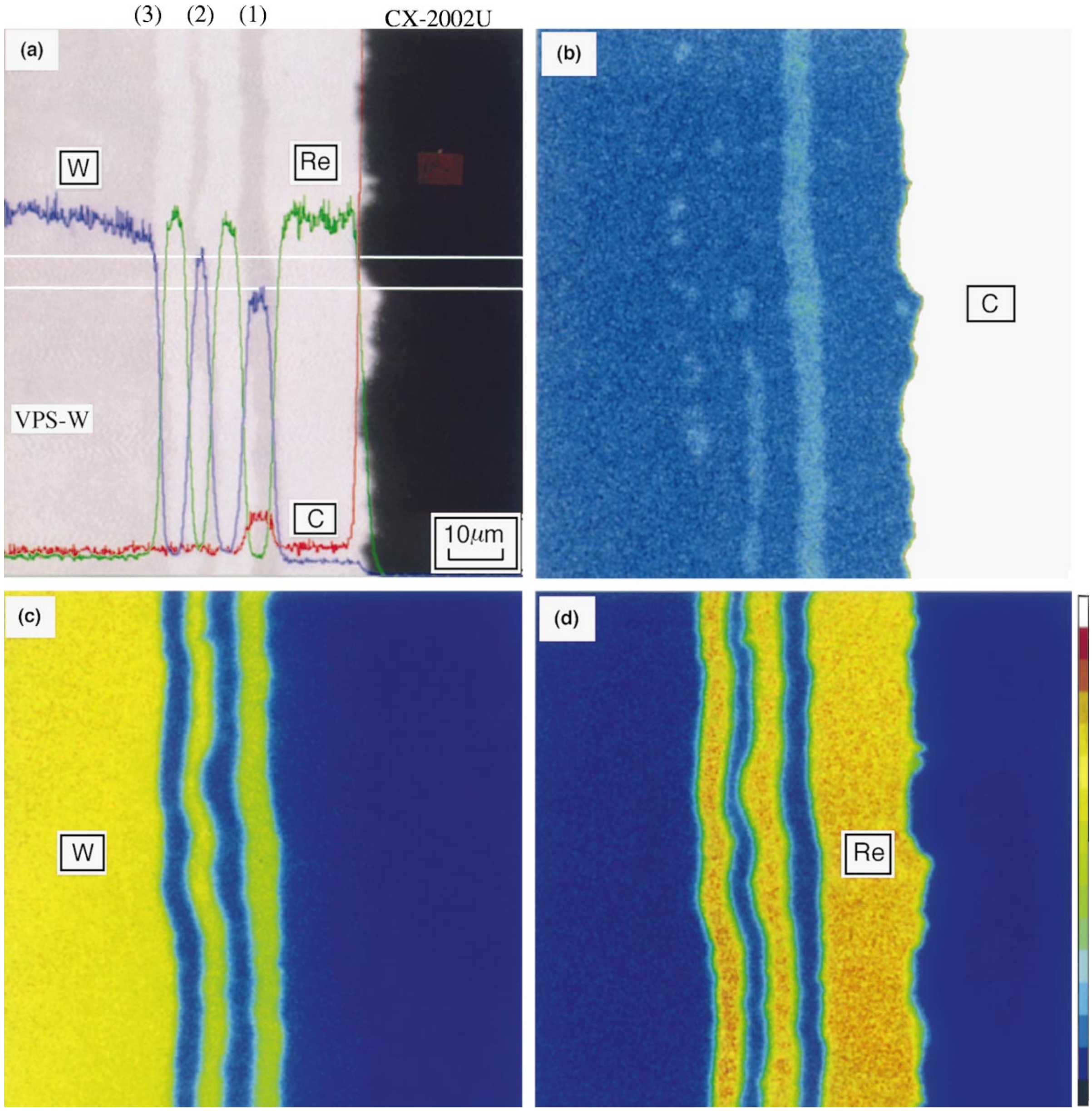

Fig. 1. Composition analyses of VPS–W and carbon material interface of VPS–W (1.0 mm) coated CX-2002U(#6) with an EMPA: (a) backscattered electron image and line analyses of carbon, W and Re; (b) mapping of carbon; (c) mapping of W; (d) mapping of Re.

It is well known that the interface of joint materials is weak from some viewpoints. In the present case, the CFC or the graphite substrates had received PVD multi-layer diffusion barrier layers [1] of rhenium and tungsten prior to the coating in order to inhibit uncontrolled brittle carbide formation [5]. Cracks due to focusing of thermal stress and formation of metallic alloys due to diffusion of atoms at high temperature are expected to occur in joint interface of tungsten and carbon materials exposed to high heat flux. In the present work, details of the microstructure and the composition change of the cross-section of the tungsten and carbon interface before and after heat flux loading were observed to evaluate stability of structure and diffusion of atoms at high temperatures.

[1] PVD Re/W multi-layer diffusion barrier coating is patented by Plansee.

2. Experimental

Tiles (20 mm × 20 mm × 10 mm) of carbon/carbon composite, CX-2002U and isotropic fine grained graphite IG-430U [7] made by Toyo Tanso were coated with tungsten by VPS technique. The carbon or the graphite substrates had received PVD multi-layer diffusion barrier layers of rhenium and tungsten prior to the coating as described above. Heat treatments were performed to stabilize the microstructure of the sample. The thickness of the VPS tungsten layer was 0.5 and 1.0 mm and its density was 92.5% of the theoretical value.

Heat load experiments were carried out with an active cooling teststand (ACT) of National Institute for Fusion Science (NIFS) [8–10]. The samples were placed on a carbon/copper block actively cooled with water. Uniform electron beam at 30 keV was irradiated on the tungsten surface through a beam limiter with an aperture of 30 mm × 30 mm. 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 heat load experiments were performed by increasing the heat flux stepwise. The surface temperature of the central part about 5 mm in diameter was measured with an optical pyrometer (300–3000°C). Details of method of the heat flux experiments were described in the previous paper [5].

Before and after the loading of heat flux, microstructure and composition were examined with a scanning electron microscope (SEM) equipped with an energy dispersion X-ray spectroscope (EDS) and electron probe micro analyzer (EPMA). Micro-Vickers hardness of the W and carbon interface was also measured.

3. Results

3.1. Composition distribution of VPS–W and carbon interface

Figs. 1 show result of the compositional analyses of the joint interface of the cross-section of the VPS–W coated CX-2002U(#6) with a tungsten thickness of 1 mm. Fig. 1(a) shows backscattered electron image and line analyses of carbon, W and Re. Figs. 1(b), (c) and (d) show distribution of carbon, W and Re, respectively. The multi-layer structure of Re and W is seen between VPS–W and CX-2002U. It can be seen that carbon in the multi-layer exists mainly in the first W layer denoted by (1) in Fig. 1(a). On the other hand, the second layer ((2) in Fig. 1(a)) and thick VPS–W layer of tungsten did not include such large amount of carbon. This can be explained that carbon was diffused from CX-2002U to the Re/W multi-layer and trapped mainly in the first tungsten layer during the heat treatment (1300°C) after the coating.

Fig. 2 shows quantitative composition in these tungsten layers. Concentration of carbon decreases with increasing distance from the CX-2002U interface. Taking into account the temperature of the heat treatment and the composition, it is likely that tungsten carbide was formed in the tungsten layers [11].

Fig. 3 shows micro-hardness of the cross-section around the joint interface. Hardness of the first and second layers of tungsten were hard comparing with that of other layer. This indicates that tungsten carbide was formed in these layers. This is coincident with the result of previous discussion. On the other hand, hardness in Re layers did not increase because carbide was not formed in Re.

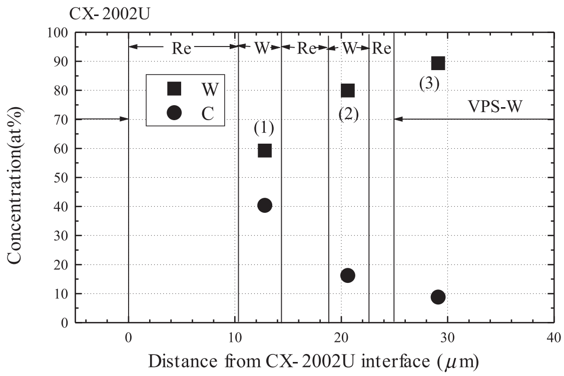

Fig. 2. Composition in tungsten layers of interface of VPS–W and carbon material of VPS–W (1.0 mm) coated CX-2002U(#6).

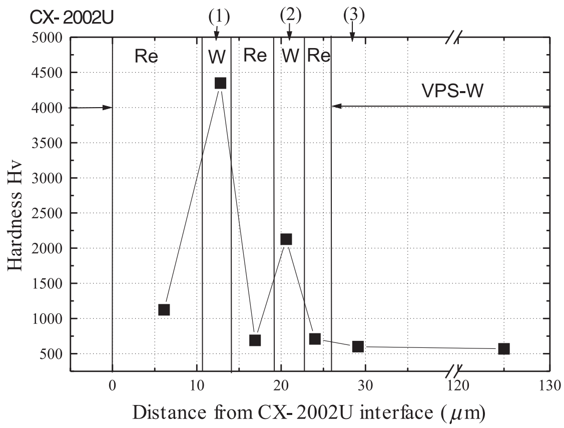

Fig. 3. Hardness as a function of distance of CX-2002U interface of VPS–W (1.0 mm) coated CX-2002U(#6).

3.2. Changes of composition and microstructure of interface by high heat flux

After the electron beam irradiation, the samples were removed from the ACT and observed with the SEM, EDS and EPMA. Figs. 4 show result of the compositional analyses of the joint interface of the cross-section of VPS–W coated IG-430U(#20) with a tungsten thickness of 0.5 mm after loading of heat flux. The maximum surface temperature by loading of heat flux was about 2800°C and the maximum heat flux was 4.5 MW/m^2.

The structure and composition between the IG-430U and the VPS–W before the electron beam irradiation was the same as the CX-2002U case as shown in Fig. 1 because the manufacture process was same. It can be seen that the joint of the IG-430U and Re is good enough but structure of the multi-layer changed in the layer between the VPS–W and the IG-430U. Microcracks and exfoliation were seen in the layers denoted by

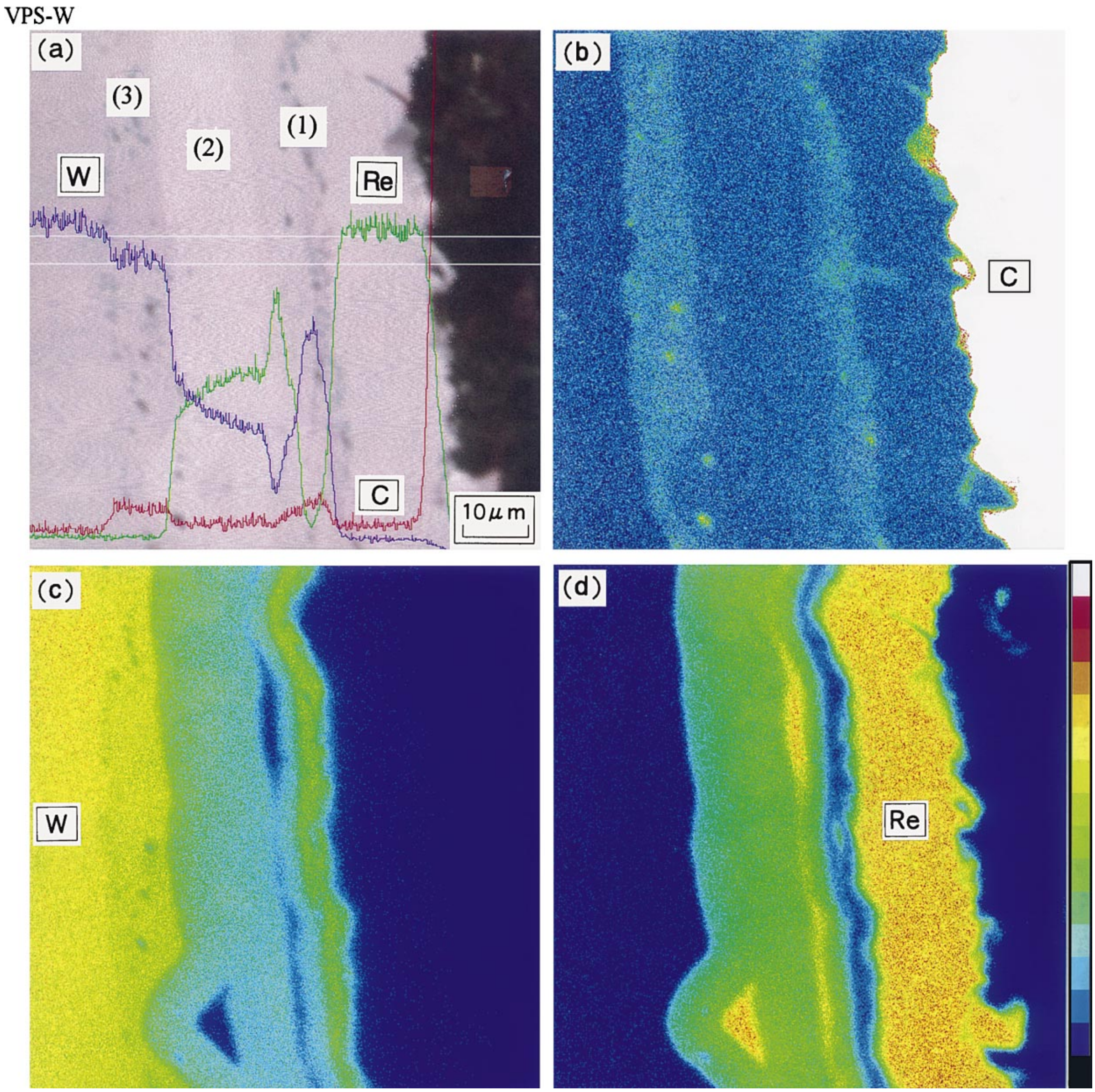

Fig. 4. Composition analyses of VPS–W and carbon material interface of VPS–W (0.5 mm) coated IG-430U(#20)with an EMPA after loading with a maximum heat flux of 4.5 MW/m^2 and peak temperature of surface of 2800°C: (a) backscattered electron image and line analyses of carbon, W and Re; (b) mapping of carbon; (c) mapping of W; (d) mapping of Re.

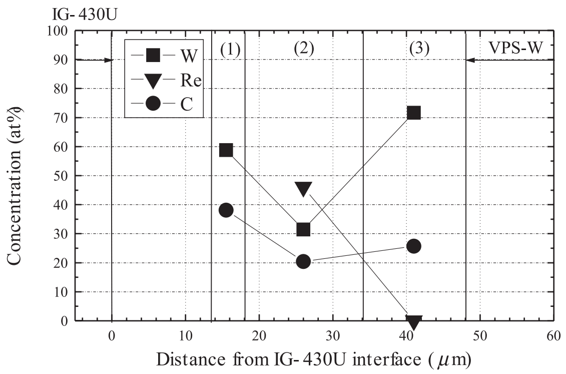

Fig. 5. Composition of newly formed layer of interface of W and carbon material of VPS–W coated IG-430U(#20) after loading with a maximum heat flux of 4.5 MW/m^2 with a peak temperature of surface of 2800°C.

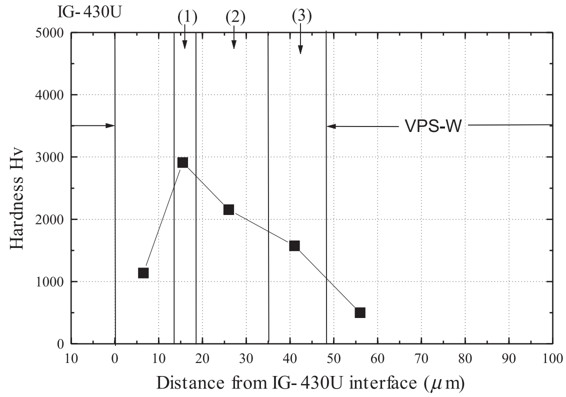

Fig. 6. Hardness as a function of distance of IG-430U interface of VPS–W (0.5 mm) coated IG-430U(#6) after loading with a maximum heat flux of 4.5 MW/m^2 and a peak temperature of surface of 2800°C.

(1) and (3) in Fig. 4(a). Fig. 5 shows quantitative composition of W, Re, carbon and O in areas corresponding with (1), (2) and (3) in Fig. 4(a). These indicate that the composition changes depending on the newly formed layers and that micro-cracks are formed in the layers which include large amount of carbon. Therefore, it is likely that this damage was caused by carbide formation and embrittlement. It may be that this influences degradation due to thermal fatigue. Fig. 6 shows hardness of the cross-section around the joint interface of this sample. This indicates also that hardness depended on the newly formed layers.

Fig. 7 shows result of compositional analyses of VPS–W/CX-2002U(#2) with a thickness of tungsten of 0.5 mm after the loading of heat flux. The maximum heat flux was 5.5 MW/m^2. The surface was melted and many cracks were formed on the surface. It can be seen that the multi-layer structure has completely changed and a new structure was formed. In addition to this, micro-cracks between CX-2002U and metal were observed. Local composition was different but the composition of a large area was almost uniform.

4. Discussion

In this work, thick W was coated by VPS after the coating of multi-layer of Re/W by PVD. Diffusion of carbon in the PVD multi-layer through pores, which have high diffusion performance, hardly occurred because the PVD coating layers are very dense. The diffusion coefficient of carbon in Re is smaller than that in W. [2] As a result, Re layers act as a diffusion barrier. Embrittlement due to formation of carbide is not generated because carbide is not formed in Re. On the other hand, W forms carbide at high temperature. When carbides are formed, the activation energy of diffusion of carbon increases. [2] This means diffusion of carbon becomes more difficult.

The samples used here were heated at 1300°C to stabilize the structure after the coatings. Compositional analyses indicated that carbon was trapped in the first layer of W. This can be explained that a small amount of carbon has diffused through the first Re layer from the carbon material and has reached the W layer, and then accumulated there during the heat treatment. It was found that tungsten carbide was formed in the first W layer by the compositional analyses and the hardness measurement. This also acts a diffusion barrier for carbon because of the high activation energy of diffusion. The tungsten carbide is brittle but, in the case of thin layer like this, strength and ductility of the layer of tungsten carbide are sufficient [12]. It was found that the multi-layer Re/W acts functionally as a diffusion barrier.

Multi-layer structure remained but the microstructure changed in the joint interface of the sample with a peak temperature of 2800°C. Micro-cracks were partly formed. It is likely that exfoliation is formed by repeated loading of heat flux. On the other hand, the multi-layer structure completely disappeared and compositional distribution was almost uniform in the interface of the sample after melting and resolidification. In this stage, it is certain that the diffusion barrier for carbon does not act.

5. Summary

1. Tungsten coatings of 0.5 and 1 mm thickness were successfully deposited by the VPS technique on car-

[2] High temperature compound material property table, Japan–Russian communication company, 1994 (Japanese).

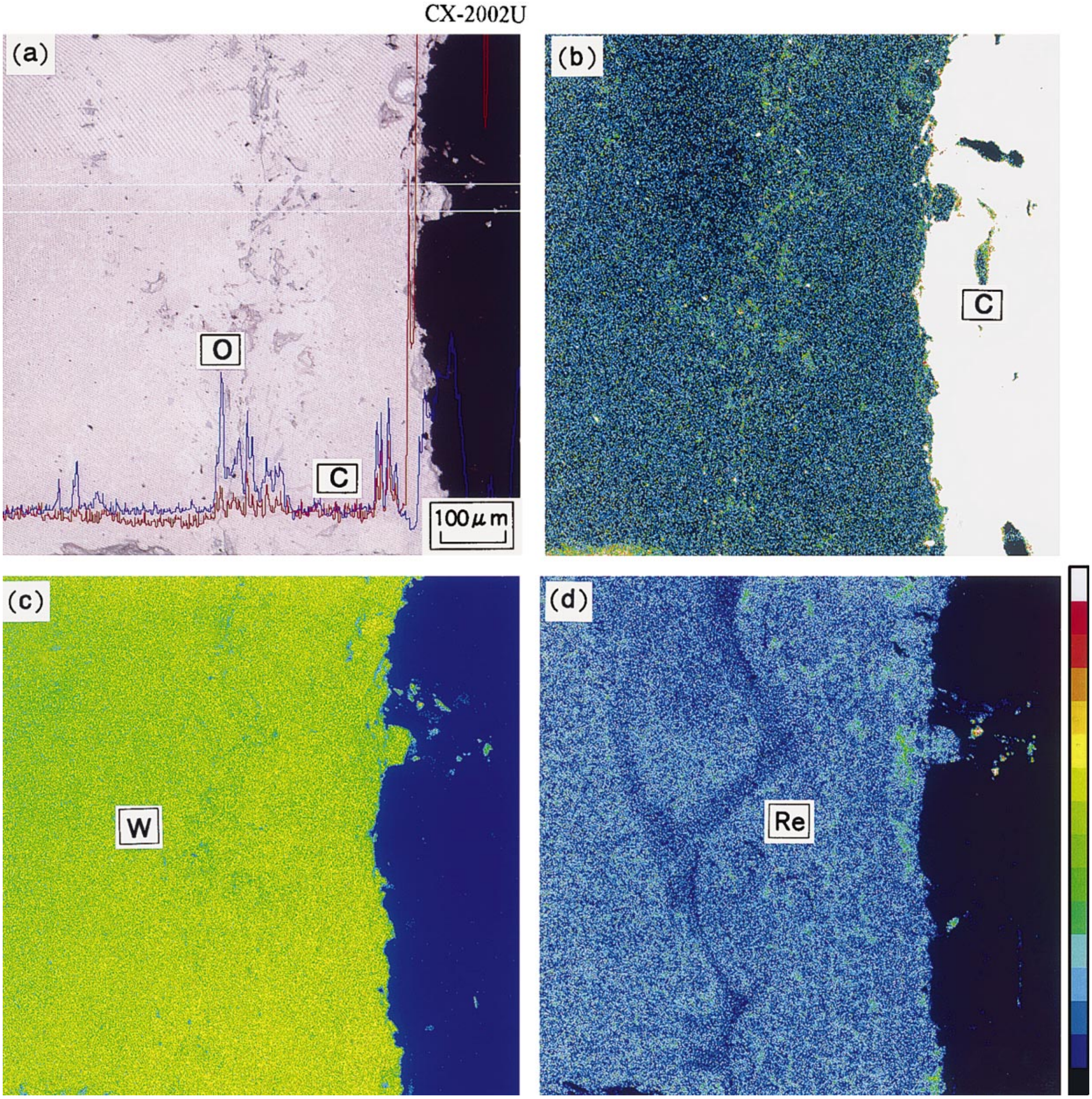

Fig. 7. Composition analyses of VPS–W and carbon material interface of VPS–W (0.5 mm) coated CX-2002U(#2) with an EMPA after loading with a maximum heat flux of 5.5 MW/m^2. Surface was melted and resolidified: (a) backscattered electron image and line analyses of carbon, oxygen; (b) mapping of carbon; (c) mapping of W; (d) mapping of Re.

bon/CFC, CX-2002U and isotropic fine grained graphite, IG-430U.
2. 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.
3. The cross-sectional composition and structure of the interface of the VPS–W and carbon samples were investigated. The compositional analyses showed that the Re/W multi-layer acts as a diffusion barrier for carbon and suppresses tungsten carbide formation in the VPS–W layer at a high temperature of about 1300°C.
4. The microstructure of the joint interface of the sample changed in the case of a peak temperature of about 2800°C.
5. The multi-layer structure completely disappeared and compositional distribution was almost uniform in the

interface of the sample after melting and resolidification. The diffusion barrier for carbon is not expected to act in this stage.

References

[1] W.O. Hofer, J. Roth, Physical Processes of the Interaction of Fusion Plasmas with Solids, Academic, New York, 1996, p. 341.
[2] R. Neu, K. Asmussen, S. Deschka, A. Thoma, M. Bessenrodt-Weberpals, R. Dux, W. Engelhardt, J.C. Fuchs, J. Gaffert, C. Garcia-Rosales, A. Herrmann, K. Krieger, F. Mast, J. Roth, V. Rohde, M. Weinlich, U. Wenzel, J. Nucl. Mater. 241–243 (1997) 678.
[3] C. Garcia-Rosales, S. Deschka, W. Hohenauer, R. Duwe, E. Gauthier, J. Linke, M. Lochter, W. Mallener, L. Plochl, P. Rodhammer, A. Salito and the ASDEX-Upgrade team, Fus. Technol. 32 (1997) 263.
[4] K. Tokunaga, N. Yoshida, N. Noda, T. Sogabe, T. Kato, J. Nucl. Mater. 258–263 (1998) 998.
[5] K. Tokunaga, N. Yoshida, N. Noda, Y. Kubota, S. Inagaki, R. Sakamoto, T. Sogabe, L. Plöchl, J. Nucl. Mater. 266–269 (1999) 1224.
[6] K. Tokunaga, N. Yoshida, Y. Kubota, N. Noda, Y. Imamura, T. Oku, A. Kurumada, T. Sagabe, T. Kato, L. Plöchl, Presented at ISFNT-5, September 1999, Rome, Italy, Fus. Eng. Des., to be published.
[7] T. Matsuda, T. Sogabe, K. Kuroda, in: Proceedings of the Japan–US Workshop P243 on High Heat flux Components and Plasma Surface Interaction for Next Fusion Devices, 1995, p. 366.
[8] I. Fujita, Y. Hirohata, T. Hino, T. Yamashina, Y. Kubota, N. Noda, O. Motojima, T. Sogabe, T. Matsuda, K. Kuroda, J. Nucl. Mater. 241–243 (1997) 1185.
[9] K. Kubota, N. Noda, A. Sagara, R. Sakamoto, O. Motojima, I. Fujita, T. Hino, T. Yamashina, K. Tokunaga, N. Yoshida, Fus. Eng. Des. 39&40 (1998) 247.
[10] K. Tokunaga, N. Yoshida, Y. Kubota, N. Noda, O. Motojima, D.L. Youchison, R.D. Watson, R.E. Nygren, J.M. McDonals, T.D. Marshall, J. Nucl. Mater. 258–263 (1998) 1097.
[11] T.B. Massalski, Binary Alloy Phase Diagrams, American Society for metals, Academic, New York, 1986.
[12] B. Schedler, Plansee, Reutte/Tirol, Austria, private communication.