EFDA–JET–CP(12)05/17

C. Ruset, E. Grigore, D. Falie, M. Gherendi, H. Maier, M. Rasinski,
G. Matthews, V. Zoita and JET EFDA contributors

The Impact of Thermal Fatigue and Carbidisation on the W Coatings Deposited on CFC tiles for the ITER-like Wall Project at JET

The Impact of Thermal Fatigue and Carbidisation on the W Coatings Deposited on CFC tiles for the ITER-like Wall Project at JET

C. Ruset[1], E. Grigore[1], D. Falie[1], M. Gherendi[1], H. Maier[2], M. Rasinski[3], G. Matthews[4], V. Zoita[1,5] and JET EFDA contributors*

JET-EFDA, Culham Science Centre, OX14 3DB, Abingdon, UK

[1]National Institute for Laser, Plasma and Radiation Physics, Bucharest, EURATOM-MEdC Association Romania
[2]Max-Plank Institut für Plasma Physik, EURATOM Association, 85748 Garching, Germany
[3]Faculty of Materials Science and Engineering, Warsaw University of Technology, Warsaw
[4]EURATOM-CCFE Fusion Association, Culham Science Centre, OX14 3DB, Abingdon, OXON, UK
[5]EFDA Close Support Unit, Culham Science Centre, Abingdon, OX14 3DB,UK
** See annex of F. Romanelli et al, "Overview of JET Results",*
(23rd IAEA Fusion Energy Conference, Daejon, Republic of Korea (2010)).

Preprint of Paper to be submitted for publication in Proceedings of the
27th Symposium on Fusion Technology (SOFT), Liege, Belgium
24th September 2012 - 28th September 2012

ABSTRACT

Since August 2011 JET operates with the ITER-like wall comprising bulk Be tiles, bulk W tiles and W coated CFC tiles with a thickness of 10-15μm and 20-25μm. In order to evaluate behavior of the W coatings to a cyclic thermal loading relevant to JET operation, High Heat Flux (HHF) tests have been carried out up to 5,100 pulses with an electron beam facility at peak temperatures of 1000°C, 1250°C and 1450°C. The pulse duration was 24s. Optical inspections of the W layer performed periodically by interrupting the test revealed small delaminations with the size of 50-500μm. The dependence of the delamination percentage on the number of pulses can be seen as a degradation curve for each particular W coating. In this way the thermo-mechanical properties of the W coatings can be characterized quantitatively. Thermal fatigue and carbidization of the tungsten due to the diffusion of the carbon from the substrate have been recognized as mechanisms for degradation of the coatings. Tungsten carbides have been identified by using TEM diffraction analysis on FIB prepared cross-section samples subjected to HHF tests. Nano-pores developed at the CFC-Mo and Mo-W interfaces during the tests might be also responsible for the degradation of the coating.

1. INTRODUCTION

The extensive research carried out in the R&D phase of the ITER-like Wall (ILW) project at JET demonstrated superior thermo-mechanical properties of the W coatings with Mo interlayer deposited by Combined Magnetron Sputtering and Ion Implantation (CMSII) method in comparison with other PVD or CVD techniques [1]. CMSII technology was used for coating of about 1800 CFC tiles with W layers of 10-15μm and 20-25μm for the new ILW of JET [2,3]. Due to the time constraints of the ILW project, only limited tests have been carried out on these coatings. In accordance with the technical specification thermo-mechanical properties were investigated in the hydrogen beam test facility GLADIS at IPP Garching at the following parameters: (i) thermal screening up to 23MW/m^2 – 1.5s and (ii) cycling thermal fatigue at 10.5MW/m^2 – 5s – 200 pulses. The coatings survived these tests without delaminations, but the actual limits of the coatings were not reached. It is very important for the exploitation of the new JET wall to know in advance the limits of the W coatings when they are subjected to a high number of heating pulses (thousands) relevant to JET operation. During normal operation of the tokamak the first wall is subjected simultaneously to long pulse thermal loading (about ten seconds) and high power short pulses (~ 1ms) produced by ELMs. The objective of the present work was to investigate behavior of the W coatings deposited on CFC substrate by CMSII technology to a large number (>3000) of long duration pulses. The research was focused on two aspects: (i) quantitative investigation of the delamination depending on the peak temperature and number of pulses and (ii) micro/nano scale investigation of the effects produced by high temperature cyclic thermal loading on the structure and integrity of the W coatings.

2. EXPERIMENTAL SETUP

Tungsten coated samples were heated by an electron beam and cooled down below the ductile to

brittle transition temperature using the High Temperature Test Facility equipment at MEdC-Romanian Euratom Association. The maximum power of the electron beam is 1.5kW for an accelerating voltage of 15kV. The cross section of the electron beam is an ellipse with axes 18/12mm and an area of ~170mm^2. The electron gun, the diagnostics and the support for the sample to be tested are installed on the top lid of the vacuum chamber (Φ540×640 mm). Five CFC samples (30×30×6mm) have been tested. Two of them were coated with standard W layer of 10μm and the other three with 20μm. A Mo interlayer of 2-3μm was introduced to improve the adhesion between W and CFC substrate. The experimental arrangement is shown in Fig. 1. The water cooled Cu support (1) is positioned by means of the cooling pipes (2) on the axis of the chamber. A special spring system (3) holds the testing sample (4) on the water cooled support. A fine graphite powder layer ensures a good thermal contact between the W coated CFC sample and Cu support. This is essential during the experiments because the CFC sample should be cooled down to 200°C as fast as possible after the electron beam was switched off. The water flow rate was ~ 3 *l*/min. At 3mm below surface a C type thermocouple (TC) (W5%Re/W26%Re) (5) with a length of 150mm and a diameter of 1.5 mm was inserted till the middle of the sample. A set of five stainless steel shields (6) were used to protect the thermocouple connector from the heat coming from the sample. The electron beam is coming from the top lid along the chamber axis.

The surface temperature is monitored in the range 250-2000°C by an IMPAC IGA-5 pyrometer which is sensitive in the wavelength range 1.45…1.8μm. The temperature was measured approximately in the middle of the W coating. The emissivity of the W coatings was set to 0.5. This was calculated starting from the value of 0.63 measured for W coatings at 1,064nm.

3. TESTING PARAMETERS

The testing temperatures and the corresponding power densities are shown in Table 1, where d is the W coating thickness, N* is the number of delaminations at the end of test and PDestim is the estimated power density on the W coating.

For all tests the pulse duration was 24s and the interpulse duration 35-45s. The minimum temperature of the sample during the thermal cycling was adjusted by modifying the interpulse duration. More than 3,100 pulses were applied in each test. The time variation of the temperature for the runs with the peak temperature of 1450°C as recorded by the pyrometer for 10 successive pulses is shown in Fig.2.

4. EXPERIMENTAL RESULTS AND DISCUSSION

The thermal cycling was stopped from time to time and the W coating was inspected with a stereomicroscope with a magnification from 10 to 45. The objective of these inspections was to count the number of the delaminated zones and to estimate their areas. The percentage of the delaminated area was calculated by dividing the total estimated damaged area to the electron beam spot area (~170mm^2). This can be seen on the sample WCL-02 after 542 pulses (Fig.3). The graph

showing the increase of the damaged area of the W coatings as a result of the thermal cycling tests can be seen in Fig.4. This kind of curves can be used for quantitative characterization of the thermo-mechanical properties of the coatings deposited on carbon based materials. The thinner coatings have a better behavior from the thermo-mechanical point of view than thicker ones. This might be associated with the heat transfer from the coating surface to the substrate.

Two SEM images of the W coatings in delaminated area are shown in Fig.5 The delaminations appear as buckling with the size in the range 50-500μm. Sometimes the coating is detached from the CFC and remains under a certain angle with the surface. By loosing the contact with the substrate these small chips become much hotter than the coating during the electron pulse and sometimes are melted. Locally, in the vicinity of these very hot spots the temperature of the W coating could exceed significantly that measured by pyrometer.

The fifth CFC sample coated with 20 μm of W survived 5,100 pulses at a peak temperature of 1,000^{o}C without delaminations.

Analyses carried out by EDX in the delaminated area indicated the presence of Mo, C and some traces of W (Fig.6). It appears that the Mo-W interface was affected during the thermal cycling. In order to investigate this aspect SEM analyses have been carried out on the non-damaged zones at different distances from a delaminated area using Focused Ion Beam (FIB) cutting technique. At 10mm (Fig.7) one can clearly see chains of nano/micro-pores formed at both Mo-C and Mo-W interfaces. TEM analyses demonstrated that the Mo interlayer was transformed into molybdenum carbide and above this layer tungsten carbides started to be formed. It is important to notice that in the right side of the picture, where the Mo-W interface was much more affected the WC layer is thinner. Similar carbides layers were detected at 5mm from the delamination.

At 0.2mm away from the delaminated zone no W carbides have been detected by TEM. As it can be seen in Fig.8 the interfaces and the Mo interlayer were severely damaged in that area. This was probably caused by a higher temperature of the coating produced near the delaminated chip that was melted and re-solidified many times. The structure of that chip contains many pores with dimensions of 10-1000 nm as it can be seen in Fig.9. It seems that the first effect of the thermal cyclic at high temperature is formation of nano-pores especially at the interfaces between CFC, Mo and W.

This might be associated with the difference of thermal expansion coefficients of these materials, but not only because at melting temperature pores in the micron range are formed into the whole volume of the W coating.

The density and dimensions of the pores depend on the temperature and the number of pulses. Initially the pores appear to be produced at the CFC-Mo and Mo-W interfaces. These pores could act as a diffusion barrier for carbon from CFC. The properties of this barrier depend on dimensions and density of the pores, which are time and temperature dependent. This complicates the transfer of the carbon from the CFC and formation of carbides. Kinetics of this process was investigated in other paper by heating the W coatings deposited by CMSII on CFC in a vacuum oven at different

constant temperatures (1200°C and 1350°C) for 2, 5 and 20 hours [5]. For the temperature of 1200°C a total carbide layer of 6.5μm was formed in 2h and the thickness of that layer increased to only 8μm for 20h. This saturation could be associated with the pores diffusion barrier.

On the other hand the thermal stability of the tungsten carbides is not so high. Said El Mrabet, et al. [4] demonstrated that heating a WC1-x/a-C film deposited on Si up to 1100°C the structure of WC1-x changes to W2C and WC between 700°C and 900°C. Over 900°C WC decomposes gradually and at 1200°C the dominant phase is W. If the substrate is a carbon material it can supply the necessary carbon producing a carbide structure at higher temperatures in accordance with the W-C phase diagram. In the case of W/Mo coatings deposited on CFC and subjected to high thermal loads the flux of carbon can be limited by the pores generated at the interfaces or by severe damage of the Mo interlayer. In this way the absence of the carbides near to the delaminated zone might be explained.

At the same time the pores structure limits the heat transfer from the outer surface of the W coating to the substrate and the coating becomes hotter. This leads to an increase of the density and dimensions of the pores and locally small fragments of the coating are finally detached from the substrate.

CONCLUSIONS

- The damage of the W coatings deposited on CFC substrate occurs gradually with increasing the number of the heating pulses.
- 10μm and 20μm W coatings have been tested with more than 3100 pulses at peak temperatures of 1250°C and 1450°C. The pulse duration was 24s. Under these conditions the total delaminated area of the W coating of 20μm was about 1% of the thermal loaded area while for coating of 10μm this percentage was about 0.2% only. By increasing the testing temperature to 1,450°C the percentage of the delaminated area increased to about 6% for 20 μm W coating and 1.2% for 10μm W coating for a similar number of heating pulses.
- The thinner coatings have a better thermo-mechanical behavior than the thicker coatings.
- The damage of the W coatings occurs by buckling with the size of delaminated zones in the range of 50-500μm.
- 20μm W coating survived 5100 pulses at a peak temperature of 1000°C without any delamination.
- The nano-pores structure generated at the CFC-Mo and Mo-W interfaces during the HHF tests could play an important role in W coating degradation, but this subject should be investigated in more detail.

ACKNOWLEDGMENTS

The work was supported by the European Commission and the Romanian Ministry for Education and Research under EURATOM Program EFDA Task Agreement Code: Fusion Technology JW11-FT-4.19 and by LAPLAS Program Code:PN09.39.03.01. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

REFERENCES

[1]. T. Hirai, et al., Fusion Engineering and Design **82** (2007) 1839–1845

[2]. H. Maier et al., Journal of Nuclear Materials **363–365**, 1246–1250 (2007)

[3]. H Maier et al., Physica Scripta **T138** (2009) 014031 (5pp)

[4]. S. El Mrabet, et al., Plasma Process and Polymers, **6**, S444-S449, (2009)

[5]. H. Maier et al., Journal of Nuclear Materials **415**, S310-S312 (2011)

Sample ID	d (μm)	T_{peak} (pyrom.) (°C)	T_{peak} (TC) (°C)	N*	PD_{estim} (MW/m^2)
WCL-01	20	1250±50	1050±50	56	6.9±0.4
WCL-02	10	1250±50	1050±50	12	6.9±0.4
WCL-03	20	1450±50	1250±50	86	8.4±0.4
WCL-04	10	1450±50	1250±50	29	8.4±0.4
WCL-06	20	1000±50	900±50	0	4.2±0.2

Table 1 Testing temperatures and power densities

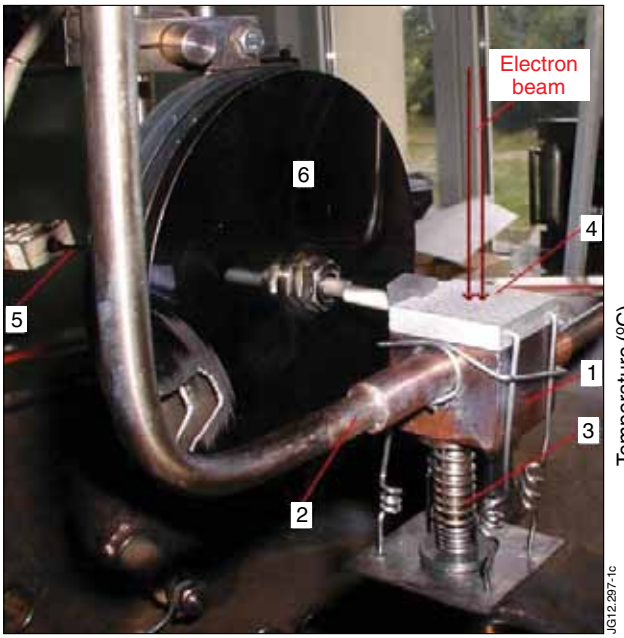

Figure 1: Experimental setup for testing the W coatings 1. Copper support; 2. Cooling pipe; 3. Holding system; 4. W coated sample; 5. Thermocouple; 6. Thermal shields

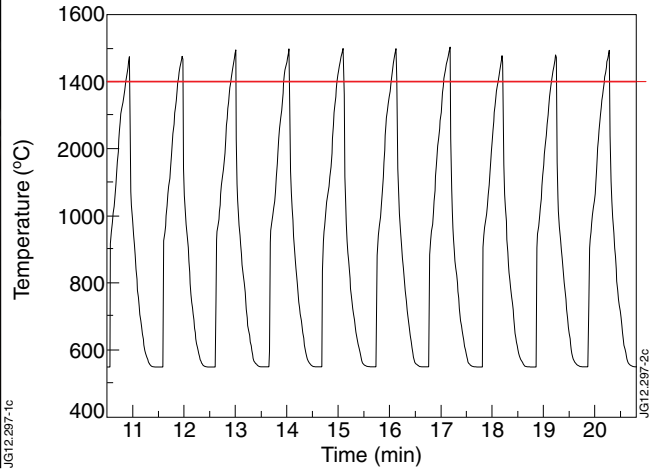

Figure 2: Time variation of the W coating temperature during the thermal cycling at 1450±50°C.

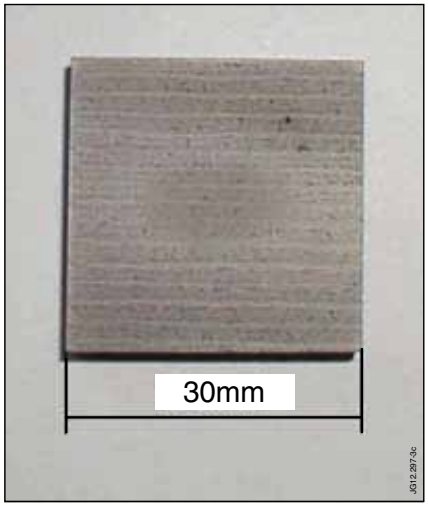

Figure 3: Electron beam spot image on W coated CFC sample.

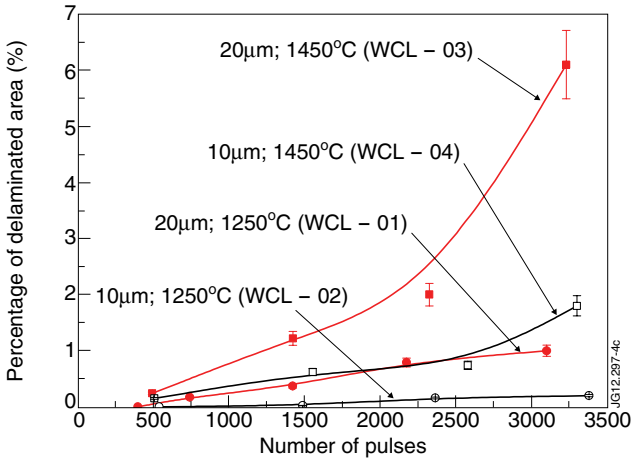

Figure 4: Degradation curves of the W coatings as a result of thermal cycling at 1,250±50°C and 1,450±50°C.

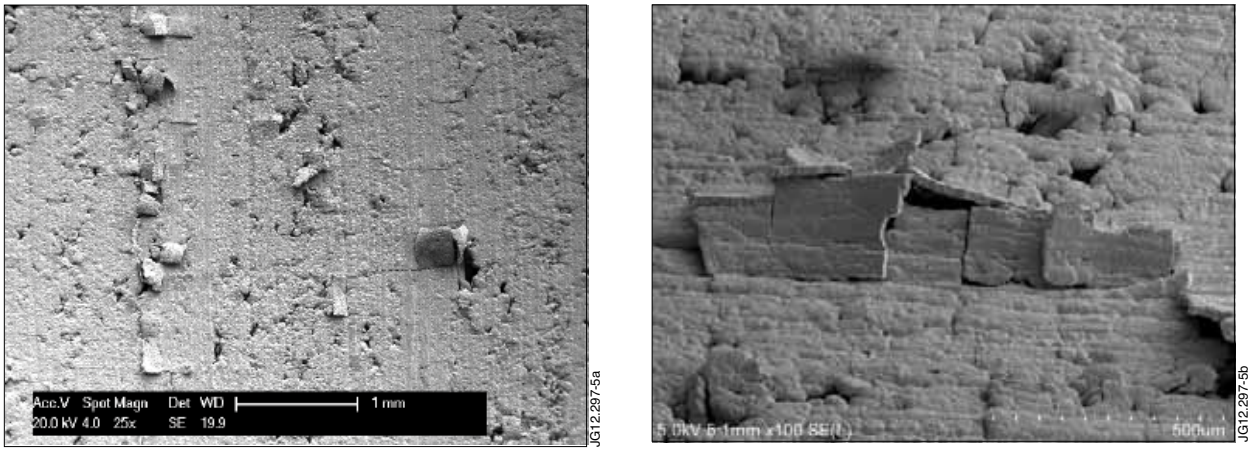

Figure 5: General view of the sample WCL-01 (a) and detailed view of the delaminations on the W coating WCL-03 after thermal cycling at 3200 pulses (b).

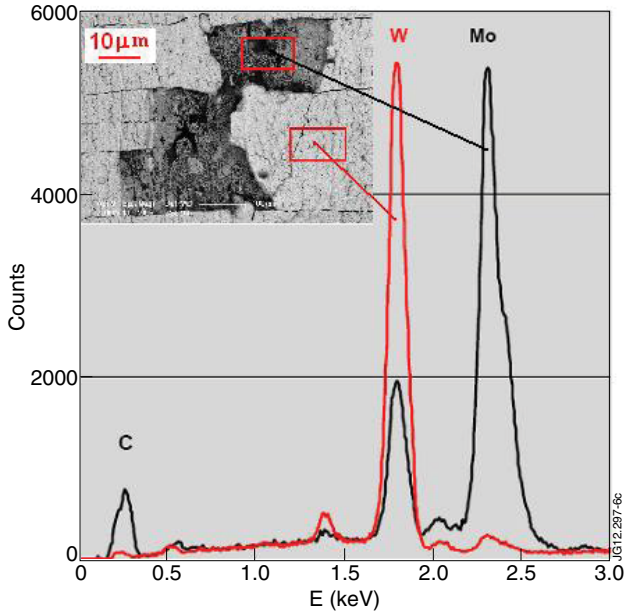

Figure 6: SEM image and the EDX analysis of the composition for delaminated and non-delaminated areas on the sample WCL-03.

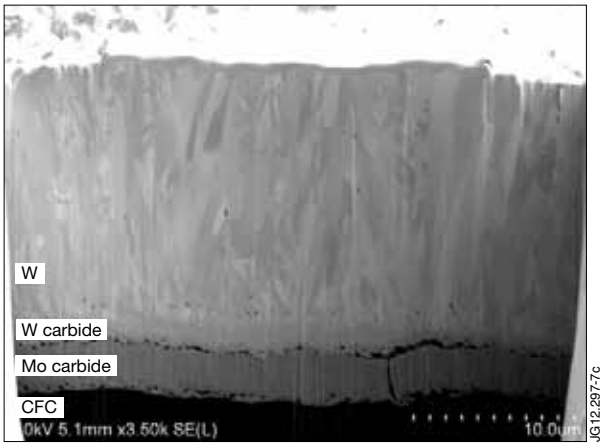

Figure 7: SEM image of a cross section through the sample WCL-03 taken at the end of test in a non-delaminated zone at 10mm away of a delaminated chip.

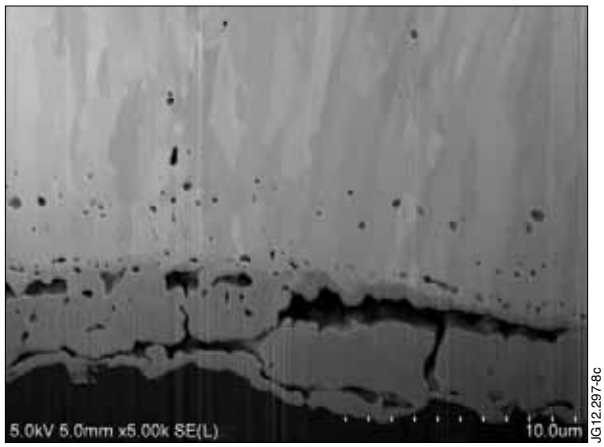

Figure 8: SEM image of a cross section through the sample WCL-03 taken at the end of test in a non-delaminated zone at 0.2 mm away of a delaminated chip.

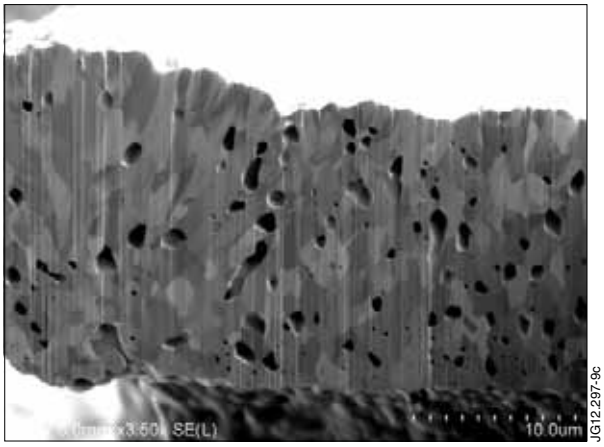

Figure 9: SEM image of a cross section through a delaminated chip melted and re-solidified many times.