EFDA–JET–CP(10)07/26

C. Ruset, E. Grigore, H. Maier, R. Neu, H. Greuner, M. Mayer,
G. Matthews and JET EFDA contributors

Development of W Coatings for Fusion Applications

Development of W Coatings for Fusion Applications

C. Ruset[1], E. Grigore[1], H. Maier[2], R. Neu[2], H. Greuner[2], M. Mayer[2], G. Matthews[3] 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]EURATOM-CCFE Fusion Association, Culham Science Centre, OX14 3DB, Abingdon, OXON, UK
** See annex of F. Romanelli et al, "Overview of JET Results",*
(Proc. 22nd IAEA Fusion Energy Conference, Geneva, Switzerland (2008)).

Preprint of Paper to be submitted for publication in Proceedings of the
6th Symposium on Fusion Technology (SOFT), Porto, Portugal
27th September 2010 - 1st October 2010

ABSTRACT

The paper gives a short overview on tungsten (W) coatings deposited by various methods on carbon materials (carbon fibre composite - CFC and Fine Grain Graphite - FGG). Vacuum Plasma Spray (VPS), Chemical Vapor Deposition (CVD) and Physical Vapor Deposition (PVD) techniques are analyzed in respect with the characteristics and performances of the W coatings.

A particular attention is paid to the Combined Magnetron Sputtering and Ion Implantation (CMSII) technique, which was developed during the last 4 years from laboratory to industrial scale and it is successfully applied for W coating (10–15μm and 20–25μm) of more than 2,500 tiles for the ITER-like Wall project at JET and ASDEX Upgrade. This technique involves simultaneously magnetron sputtering and high energy (tens of keV) ion implantation. Due to the ion bombardment a stress relief occurs within the coating enabling its growth without delamination to a relatively large thickness. In addition, in order to adjust the thermal expansion mismatch between CFC and W, a Mo interlayer of 2–3μm is currently used. Experimentally, W/Mo coatings with a thickness up 50μm were produced and successfully tested in the GLADIS ion beam facility up to 23MW/m^2.

1. INTRODUCTION

In comparison with CFC or FGG as a plasma facing material in fusion devices, tungsten has some advantages, but disadvantages as well. The main advantages can be summarized as follows: high melting temperature, good thermal conductivity, low tritium retention, low sputtering rate. On the other hand bulk tungsten has a higher electrical conductivity which may cause problems with eddy currents. Difficulties in machining due to its hardness and brittleness and heaviness can be considered as other technical disadvantages. Moreover, the use of W requires that the plasma impurity content be kept lower than that required for low Z impurities in order to avoid increased core radiation losses.

A solution to overcome some of these problems could be W coating of carbon based materials. By this way one can benefit of the high thermal conductivity and relative low electrical conductivity of CFC or FGG having at the same time low tritium retention and sputtering rate. But this idea is not easy to be applied due to the adhesion problems between W coating and carbon materials. The adhesion is particularly important for the resistance to the high heat load at which it is subjected to during the operation of the tokamak. Any loss of the contact between coating and substrate leads to overheating and melting of the W coating. The thermo-mechanical properties and the service lifetime of the W coatings are influenced by the thermal cyclic fatigue and the carbidisation of the W due to the carbon diffusion from the substrate at elevated temperatures. These elements have to be taking into account when the coating performance is evaluated. The necessary thickness of the W coating depends on the erosion rate for each particular position of the coated tile at the tokamak first wall and on its designed lifetime.

2. TUNGSTEN COATING TECHNIQUES

The history of the W coatings for fusion applications is mainly connected with ASDEX Upgrade, which was the first tokamak in the world operated with a full W wall. Various VPS, PVD and CVD

techniques have been developed with the aim to produce W coatings on carbon based materials with the best thermo-mechanical properties in terms of resistance to the high heat fluxes. The first investigations appear to be carried out in 1996 with W coatings deposited on FGG by VPS (100–550 μm) and PVD (30–200μm) [1]. The best results were produced by VPS coatings which were able to withstanding heat loads up to 15MW/m^2 at 2 s pulse length without any structural changes, and cyclic loading with 1000 cycles at 10MW/m^2. This technique has been further developed both in Japan and Europe. The coating thickness was usually in the range of 0.2–1.0mm. A multilayer Re/W was deposited by PVD prior to the VPS coating with the aim to prevent the carbon diffusion from the substrate and formation of brittle tungsten carbides. As the experiments have shown the rhenium, as non-carbide former, is efficient up to 1600°C. For higher temperature the Re/W multilayer structure disappeared and tungsten carbides were formed [2]. Tungsten coatings of 0.5–1.0mm applied by VPS on small samples (20×20×10mm) of CFC (type CX-2002U) and FGG (type IG-430U) were successfully tested up to 18.6MW/m^2 (Tmax ≤2200°C) [3] and at 100 cycles of heat loads at 10MW/m2 - steady state [4].

The big problem with VPS tungsten coatings appears to be the scaling up from small samples to real tiles. A complete divertor with W–VPS coatings of 500μm was installed in ASDEX Upgrade during 1995/1996 [5]. After 800 discharges with power densities on the tile surface of 7–9MW/m^2 cracks have been detected on almost all tiles (approx. 200) [6]. Again in 2007 W-VPS (200 μm) were used in AUG in a new divertor geometry with large tiles. Dramatic failure of W coating occurred on some tiles at power densities above 10MW/m^2 [7].

As alternatives to VPS, PVD and CVD techniques have been developed for W coating of carbon based materials. A comparative investigation of the W coatings deposited by various PVD techniques on FGG was carried out by H. Maier, et al. [8] in 2000-2001. The results can be summarized as follows: (i) Magnetron sputtering could produce W coatings with a maximum thickness of 3 μm which fail at 11MW/m^2 due to high stress; (ii) By E-beam evaporation, the maximum achievable thickness was 0.2-0.5μm and the coating survived up to melting; (iii) Up to 10μm W coatings were produced by cathodic arc evaporation and these coatings survived up to melting. CVD technology using tungsten-hexafluoride (WF6) has been also applied at the laboratory scale to produce W coatings of 1-2 μm on copper, stainless steel, graphite and CFC [9]. The idea of these experiments was to use CVD technique for in situ W coating of the Tore Supra chamber.

Another attempt to identify the best W coating technology for CFC has been made in 2005-2006 in the R&D phase of the ITER-like Wall Project at JET [10]. Ten different PVD and CVD techniques and processing parameters were applied to produce W coatings of 4μm and 10μm thickness on CFC samples (80 x 80 x 40 mm) both perpendicular and parallel to the fiber planes. Four VPS technologies were applied for the same type of samples to produce W coatings of 200μm. All coated samples were analyzed (coating thickness, impurities, adhesion, internal stress) and subjected to high heat flux (HHF) tests in GLADIS ion beam facility at Max-Plank Institute for Plasma Physics (IPP), Garching. The HHF test program included the following three steps: (i) thermal screening from 6MW/m^2 / 6s up to 23.5MW/m^2/1.5s (T_{max} ~ 2200°C), (ii) cycling thermal fatigue at 10.5MWm^2 /5s – 200 pulses and (iii)

ELM-like loading at 0.35 GW/m2 / 1 ms – 1000 pulses on a surface area of 8x8 mm2 in the JUDITH electron beam facility at FZ Juelich. From the coatings deposited by PVD or CVD techniques only those produced by CMSII survived to the first two steps. Small delaminations appeared in the third test. Thick coatings (200 μm) deposited by VPS survived the tests as well [10].

Later, in 2009, due to the technical problems associated with the up-scaling of the VPS technology from small samples to real tiles [11] and as a result of an analysis concerning the tungsten erosion in the divertor area, the W coating thickness for divertor was reduced from 200 μm to 20–25μm. At the same time CMSII technology was extended from 10μm to 20–25μm and it has been applied for all W coated tiles in the new JET wall including divertor.

3. COMBINED MAGNETRON SPUTTERING AND ION IMPLANTATION METHOD

The development from laboratory to industrial scale (design, manufacturing and commissioning) of a new coating unit took approx. 18 months. Qualification and production of the W coating for all type of tiles have been carried out in about 2 years. The CMSII technique is in principle a Plasma Immersion Ion Implantation process where the plasma source is the magnetron discharge. Of course during the ion implantation a deposition process occurs as well. On the other hand it can be seen as a high energy ion assisted deposition technique because energetic ions (tens of keV) strike periodically (pulse duration-20μs, frequency-25Hz) the coating during its growth. Detailed description of the CMSII equipment and technology is given elsewhere [12], but some new relevant aspects will be further discussed here.

One of the problems with the Dunlop DMS 780-CFC material is its anisotropic thermal expansion. The thermal expansion coefficient is $10–12\times10^{-6}\,K^{-1}$ perpendicular to the fibre plane and $0–1\times10^{-6}\,K^{-1}$ parallel to the fibre plane, while for W it is $4–5\times10^{-6}\,K^{-1}$. This mismatch in thermal expansion properties of CFC and W leads to a poor adhesion which is particularly revealed at HHF tests. This problem was solved by introducing a Mo interlayer of 2–3μm between W and CFC. The thermal expansion coefficient of Mo is $7.2\times10^{-6}\,K^{-1}$.

In order to investigate the influence of the Mo interlayer on the global stress induced into the coating during its growth, the cantilever method was used in different coating regimes with and without Mo. The cantilevers were made of fine grain graphite with the following dimensions: 32×5×0.83mm. They have been installed in a special device protecting the back side from coating (Fig.1). The Stoney formula (1) gives the global stress (σf) into a coating deposited on a cantilever depending on the coating and substrate characteristics.

$$\sigma_f = \frac{F_f}{d_f w} = \frac{1}{6R} \cdot \frac{E_s d_s^2}{(1-\nu_s)\, d_f} \qquad (1)$$

E_s and d_s are the Young's modulus and the thickness of the substrate, d_f is the coating thickness, ν_s is Poisson coefficient for the substrate, w is the width of the cantilever, F_f is the force acting on the coating and R is the curvature radius of the coated cantilever. The formula (2) gives a good approximation for the curvature radius R, where one end of the cantilever is fixed.

$$R = \frac{l^2}{2\delta} \tag{2}$$

l is the length of the cantilever and δ is the deviation from the initial, horizontal, position. For particular application of W coating this formula becomes:

$$\sigma_f(MPa) = 2245.5 \cdot \frac{E_s^2 d_s}{(1-\nu_s)\cdot d_f(\mu m)} \tag{3}$$

The following values were taken into account: $l = 32mm$, $d_s = 0.83mm$, $E_s = 10GPa$, $\nu_s = 0.22$. The results, shown in Table 1, can be summarized as follows:

- By introducing the Mo interlayer, although the coating thickness increases from 12μm to 16μm, the global stress decreases by ~ 40%.
- The high voltage pulse discharge reduces the global stress by ~ 30% although the coating thickness increased from 14μm to 16μm.

This is confirmed independently by X-ray diffraction stress measurements using the sin2Θ-method on tungsten coatings deposited with and without high voltage pulses.

In the near-surface layer compressive stress values of (–0.8 ± 0.1) GPa and (–1.2 ± 0.1) GPa were obtained on coatings with and without pulses, respectively.

In order to emphasize the beneficial role of the Mo interlayer, GLADIS tests were carried out on four dedicated CFC samples (80×80×40mm) coated with and without Mo as well as with and without high voltage pulses. The loading program included 200 pulses at 16MW/m² / 2.0s (Tsurface ≈ 1500°C) and 100 pulses at 20 MW/m^2/1.8s ($T_{surface}$ ≈ 1800°C). SEM examinations have been carried out after each 100 pulses. No defects appeared on all samples after 100 pulses. At the end of the heat loading program the coatings without Mo showed a large number of small buckling defects while on the coatings with Mo interlayer no delamination was detected. This is a clear demonstration of the positive role of Mo for mitigating the thermal expansion mismatch between W and CFC.

The compact, pore free structure of the W coating deposited by CMSII is shown in Fig.2a. In the region of the carbon fibers the coating surrounds the ends of the fibers penetrating between them for tens of microns (Fig.2b). In this way, the adhesion is improved.

However, it is well known that CFC is a porous material. W coating can close small pores, but the large pores with dimensions of tens of microns remain open to the atmosphere.

4. APPLICATIONS OF CMSII TECHNOLOGY IN FUSION

The main application of the CMSII technology is the W coating of the JET tiles for the new ITER-like wall. There are two kinds of coatings: 10-15μm for the main chamber and inner divertor and 20–25μm for the outer divertor.

Production has been carried out in accordance with a well established quality control program. A load of G6 and G7 W coated divertor tiles are shown in Fig.3. The outer diameter of the load is ~400mm. Coating thickness and impurity concentration has been checked for each run on witness

samples by Glow Discharge Optical Spectrometry (GDOS). Approx. 10% of the coated tiles were subjected to HHF tests in GLADIS.

The same technology was applied for more than 500 FGG tiles for ASDEX Upgrade tokamak. Approx. 300 of these tiles including divertor tiles were successfully tested during an 8 month campaign (~ 5,000 s of plasma).

CMSII technology was also applied to produce W/Mo markers on 34 tiles for investigation of the erosion in the JET divertor. The structure of these markers consisting of 2-3 μm Mo + 12-14μm W + 3−4μm Mo + 3−4 μm W can be seen in Fig.4. The W and Mo depth profiles were analyzed at IPP Garching by Nuclear Reaction Analysis before the installation in JET. The same analyses will be carried out after the first campaign with the new wall. In this way the erosion rate in different divertor areas will be deduced.

In order to achieve thicknesses and respectively lifetimes comparable to those of VPS coatings, the CMSII technology was extended to ~50μm by doubling the standard 20−25μm. These coatings were successfully tested in GLADIS ion beam facility up to 23MW/m^2 in screening regime followed by cycling at 10.5MW/m^2/5s/150 pulses. No delamination was detected.

CONCLUSIONS

During the last 15 years many VPS, PVD and CVD technologies were developed and applied for W coating of carbon based materials for the first wall in fusion devices. These coatings were tested more or less successfully under different thermal loading conditions with electron or ion beams as well with fusion relevant plasmas. The extension of these technologies from laboratory to industrial scale is not trivial for high performance W coatings.

At present the CMSII technology has been successfully applied at an industrial scale for the W coating of more than 2,500 tiles including divertor tiles for the ITER-like Wall project at JET and for ASDEX Upgrade. The applied coating thicknesses are in the range of 10−15μm or 20−25μm depending on the tile position at the first wall.

Exploratively, a coating with a thickness of ~50μm was produced and successfully tested in GLADIS up to 23MW/m^2.

ACKNOWLEDGMENTS

The work was supported by the European Commission and the Romanian Ministry for Education and Research under EURATOM Program EFDA Task Agreement Code: JW6-TA-EP2-ILC-06 and by CNCSIS –UEFISCSU, project PNII – IDEI No. 141/2008.

The authors wish to thank to Dr. C. Luculescu for his assistance with SEM analyses. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

REFERENCES

[1]. S. Deschka et al., Journal of Nuclear Materials, **233-237**, 645-649 (1996)

[2]. S. Tamura et al., Journal of Nuclear Materials **329–333**, 711–716 (2004)
[3]. K. Tokunaga et al., Journal of Nuclear Materials, **258-263**, 998-1004 (1998)
[4]. K. Tokunaga et al., Fusion Engineering and Design, **81**, 133–138 (2006)
[5]. R. Neu et al., Plasma Phys. Control Fusion **38**, (1996), A165 doi: 10.1088/0741-3335/38/12A/013
[6]. H. Maier et al., Journal of Nuclear Materials **258-263**, 921-926 (1998)
[7]. R. Neu et al., Phys. Scr. 2009 014038 doi: 10.1088/0031-8949/2009/T138/014038
[8]. H. Maier, et al., Surface and Coatings Technology, **142-144**, 733-737 (2001)
[9]. A. Cambe, et al., Fusion Engineering and Design **56–57**, 331–336 (2001)
[10]. H. Maier, et al., Journal of Nuclear Materials **363–365**, 1246–1250 (2007)
[11]. H Maier et al., Physica Scripta T**138** (2009) 014031 doi: 10.1088/0031-8949/2009/T138/014031
[12]. C. Ruset et al., Fusion Engineering and Design **84**, 1662–1665 (2009)

Conditions	**Thickness (µm)**	**Global stress (MPa)**
W + Mo + HV	16	194 ± 16
W + Mo	14	277 ± 28
W + HV	12	337 ± 30

Table 1: The influence of coating conditions on the global stress induced into the coating

Figure 1: Experimental device for investigation of the global stress.

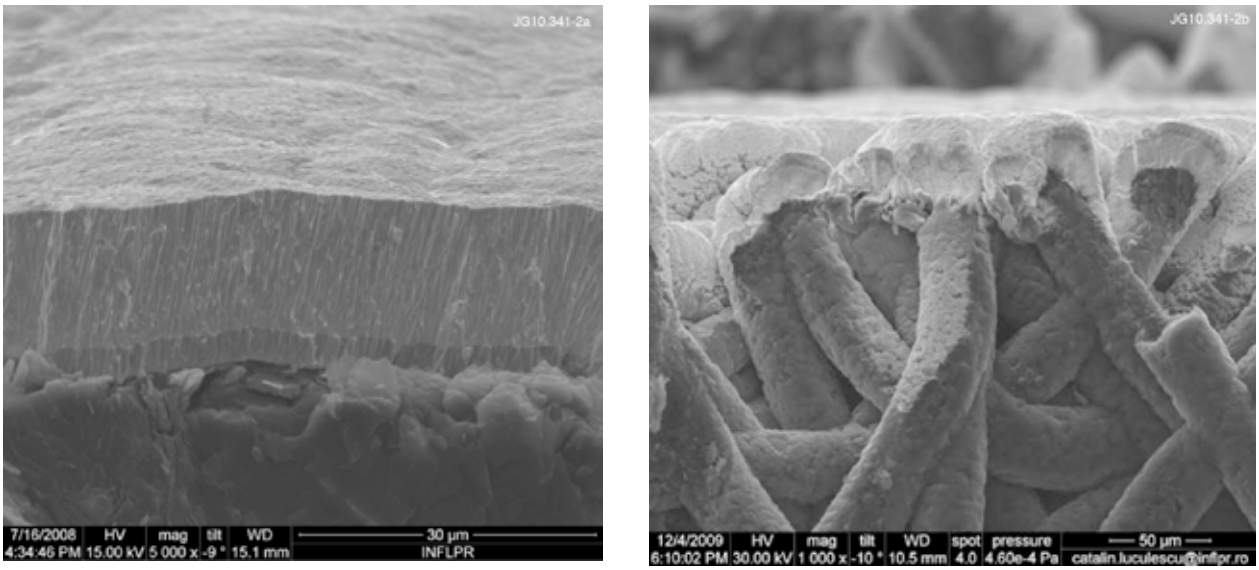

Figure 2: The fracture SEM image of the W/Mo coating deposited on carbon (a) and on carbon fibres (b).

Figure 3: G 6 and G7 divertor tiles coated with 20-25µm W in series production.

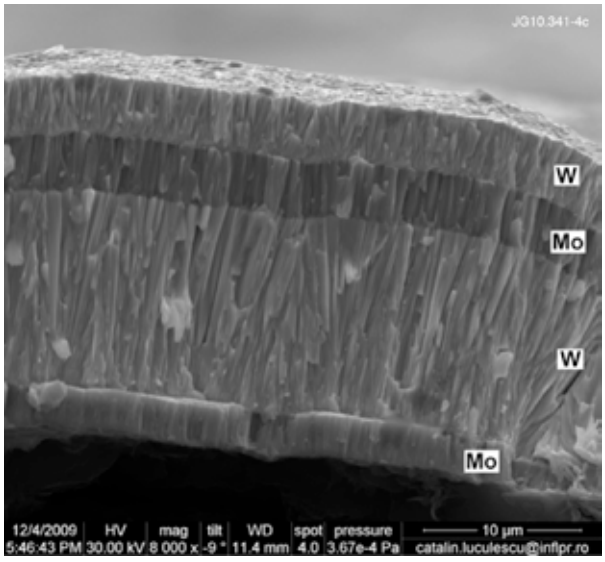

Figure 4: Mo/W markers for investigation of the erosion in JET divertor.