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Testing of beryllium marker coatings in PISCES-B for the JET ITER-like wall

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ABSTRACT

Beryllium has been chosen as the first wall material for ITER. In order to understand the issues of material migration and tritium retention associated with the use of beryllium, a largely beryllium first wall will be installed in JET. As part of the JET ITER-like wall, beryllium tiles with marker coatings are proposed as a diagnostic tool for studying the erosion and deposition of beryllium around the vessel. The nominal structure for these coatings is a ~10 μm beryllium surface layer separated from the beryllium tile by a 2–3 μm metallic inter-layer. Two types of coatings are tested here; one with a nickel inter-layer and one with a copper/beryllium mixed inter-layer. The coating samples were deposited by DC magnetron sputtering at General Atomics and were exposed to deuterium plasma in PISCES-B. The results of this testing show that the beryllium/nickel marker coating would be suitable for installation in JET.

1. Introduction

Beryllium has been chosen as the first wall material for ITER. In order to understand the issues of material migration and tritium retention associated with the use of beryllium more fully, a largely beryllium first wall will be installed in JET. As part of the ITER-like wall (ILW), beryllium (Be) tiles with marker layers are proposed as a diagnostic tool for studying the erosion of Be at locations where Be erosion is generally expected to be of the order of 10 μm or less. For erosion greater than this it will be possible to make direct measurements of the tiles. The nominal coating structure for these marker layers is a ~10 μm beryllium surface layer separated from the beryllium tile by a 2–3 μm metallic inter-layer. The proposed positions of the coatings are on limiter tiles covering locations around a poloidal section of the main chamber wall, as shown in Fig. 1. It is expected that the JET limiters will normally be exposed to power loads of the order of 1–2 $\mathrm{MWm^{-2}}$ for about 10 seconds per pulse and in general the temperature is expected to be below 600 °C; however under certain operating conditions power loads up to 6–7 $\mathrm{MWm^{-2}}$ and temperature excursions above 600 °C are possible, as seen on mushroom limiters [1]. Whilst it is desirable that the marker coatings be able to withstand these conditions, this may not be possible in all locations, where, for example at the mid-plane of the inner wall guard limiter (IWGL), strong erosion occurs during the limiter phase of a JET pulse [2].

The metal to be used as an inter-layer should have a melting temperature above that of Be (1289 °C), should not form inter-metallic phases, should have a linear thermal expansion coefficient similar to that of Be to avoid thermal stresses at the interfaces and should have a low diffusion rate with Be to ensure the inter-layer remains discreet so that the thickness of the Be coating remaining when removed from the vessel can be determined. Based on these criteria, nickel (Ni) was deemed to be the best candidate for the metallic inter-layer. In addition a mixed copper/beryllium (Cu/Be) inter-layer, with a composition $\sim Cu_{0.1}/Be_{0.9}$ and a graded interface to improve adhesion with the top Be layer, was also considered a possibility.

2. Experimental details

Marker coatings were deposited at General Atomics (GA) using DC magnetron sputtering. Be disks 25 mm in diameter and 2 mm thick (compatible with the PISCES-B sample mount) were used as substrates. Two different substrate surface finishes were tested; one was an as-machined surface which was roughened with abrasive paper prior to deposition, referred to as 'mechanically cleaned', the second was the chemical etch treatment that will be used for

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[1] See the Appendix of M.L. Watkins et al., Fusion energy, in: Proceedings of Twenty first International Conference, IAEA, Chengdu, 2006.

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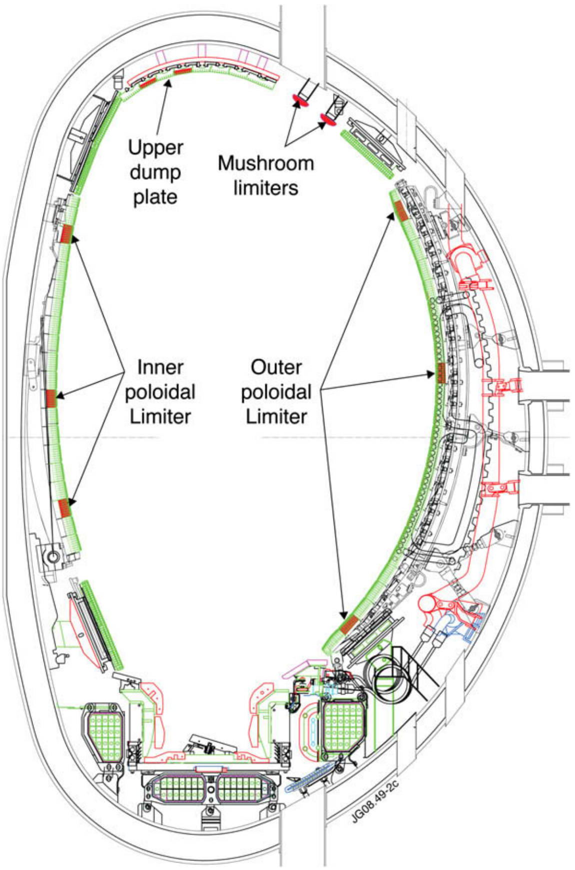

Fig. 1. Poloidal cross section of the JET ILW configuration. The proposed positions of the Be tiles with marker coatings are indicated.

the Be tiles to be installed in JET, followed by an argon ion etch prior to deposition, referred to as 'etched'. The samples were deposited in batches; hence samples with the same number in the sample name were deposited under identical conditions (e.g., JET2A and JET2B). In addition two witness samples, JET4C and JET5C, were deposited and used for comparative studies. Details of the sample preparation and testing conditions described throughout this section are summarised in Table 1.

The marker coating samples were exposed to deuterium plasmas in the linear plasma simulator PISCES-B [3] for ~1200 s, i.e., ~200 s to reach stable plasma conditions and sample temperature and then 1000 s under stable conditions. The target was biased to −100 V resulting in a deuterium ion energy of ~90 eV after into taking account the plasma potential. The sample temperature was achieved by balancing the incident flux (i.e., the power load) and the sample cooling. The marker coatings were tested in the temperature range 400–625 °C, measured by a thermocouple located on the back of the sample. Additional surface temperature measurements were taken using infra red (IR) spectroscopy, however some uncertainty is expected in the IR values determined as only a small percentage (<1%) of light from the hot LaB_6 plasma source in PISCES-B reflected at the sample surface contributes a significant thermal background to the raw data. In addition the varying emissivity and reflectivity due to changes in the sample surfaces adds to the difficulty in satisfactorily deconvolving accurate surface temperatures from the spectral data; thus the IR spectroscopy data was only used to show trends in temperature and make comparisons between samples. In general the surface temperatures from IR spectroscopy were higher and varied more than those measured using the thermocouple on the back of the samples. The variation in temperature from IR results is likely to be due to changes in surface emissivity of the samples during exposure to the plasma.

Power loading is also an important factor for the testing of the coatings; however it was not possible to cool the samples sufficiently in PISCES-B such that temperatures around 600 °C could be maintained with high power loading. Thus the heat fluxes used during these tests were in general an order of magnitude lower than anticipated on the JET main chamber wall.

Two samples (JET3A and JET4A) were heated under vacuum in the temperature range 400–600 °C to test the integrity of the Be marker coatings. One full heating cycle under vacuum involves 0.5 h heating to set temperature, 1 h at temperature (maximum 600 °C) and 0.5 h cooling time. Sample JET3A underwent one heat cycle and sample JET4A underwent three heat cycles; after each heating cycle the sample was cooled and inspected. Sample JET4A was subsequently exposed to deuterium plasma in PISCES-B.

Analysis of the samples was performed using scanning electron microscopy (SEM), including backscattered (BS) imaging and X-ray wavelength dispersive spectroscopy (WDS), and also using proton backscattering with 2.5 MeV H^+ ions.

3. Results

Samples JET4A and JET5A, heated to 490 °C, and samples JET4B and JET5B, heated to 625 °C, all survived exposure in PISCES-B. These samples were all deposited on an etched surface. A mass loss of 0.57 mg was measured for sample JET4A, equivalent to erosion from the surface of the order of 0.8 μm. Based on the ion flux $7 \times 10^{17}\ cm^{-2}\ s^{-1}$ during the 1000 s exposure over a surface area of ~4 cm^2, a sputtering yield of ~1% was calculated for ~90 eV deuterium ions. It was not possible to determine the mass loss of samples JET4B, JET5A and JET5B as the grafoil sheet used to ensure good thermal contact between the sample and the target holder bonded to the back of the substrates.

WDS analysis of series JET4 and JET5 revealed that the interlayer metal had broken through to the surface of the sample in

Table 1
Details of Be marker coatings tested in PISCES-B linear plasma simulator.

Sample	Inter-layer metal	Substrate surface finish	Ion flux ($cm^{-2}\ s^{-1}$)	Heat flux (MWm^{-2})	Temperature sample (°C)	Vacuum oven test
JET2B	Ni	Mechanically cleaned	2.6e18	0.4	590	–
JET2A	Ni	Mechanically cleaned	1.3e18	0.2	400	–
JET3A	Ni	Mechanically cleaned	–	–	–	600 °C 1 h
JET4A	Ni	Etched	0.7e18	0.1	490*	400 °C 1 h, 500 °C 1 h, 600 °C 1 h
JET5A	Cu/Be	Etched	0.7e18	0.1	490*	–
JET4B	Ni	Etched	1.2e18	0.2	625*	–
JET5B	Cu/Be	Etched	1.2e18	0.2	625*	–

* Elevated temperatures were achieved with low ion flux by turning off the water cooling to the sample holder. Sample JET3A was only tested in vacuum oven.

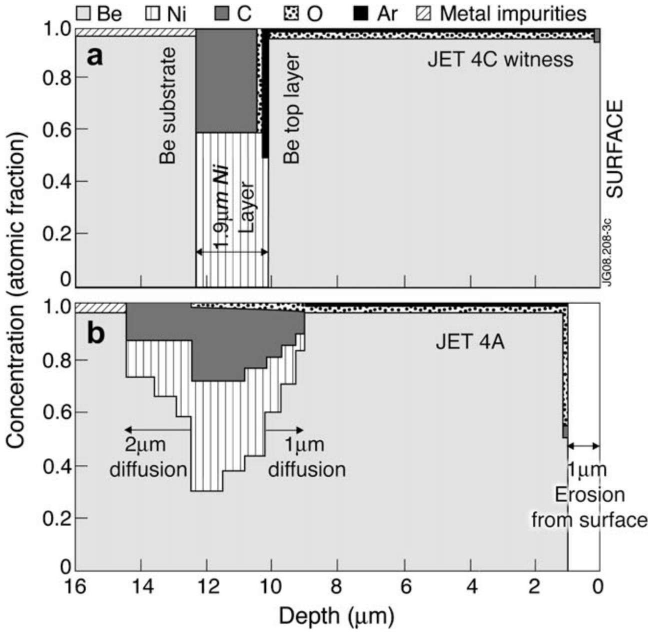

Fig. 2. Results from modelling [4] of proton backscattering data for (a) sample JET4C (witness) and (b) JET4A.

small patches. These patches ranged in size from 10–100 μm. From backscattered SEM images the upper limit of the surface area covered by these ‘break-through’ patches is 0.3%.

Proton backscattering measurements show diffusion at the layer interfaces and erosion from the surfaces of the samples. Fig. 2 shows the integral atomic fraction of the layers in samples JET4C (witness) and JET4A obtained from modelling the proton backscattering data using SIMNRA [4]. For sample JET4C, summarised in Fig. 2(a), the initial structure was shown to be 10 μm-Be/1.9 μm-Ni/Be-substrate with 40 atomic percent (at.%) carbon (C) in the Ni layer. The high C contamination has arisen because the Ni layer was not deposited under UHV conditions, thus C from organic contamination has become incorporated into the background gases of the sputtering chamber during ion etching procedures. An oxide is present at the upper Ni/Be interface, due to exposure to air between the deposition of the Ni layer and the top Be layer. Argon is also present at this interface, due to ion etching before deposition of the top Be layer. The results of modelling for sample JET4A, Fig. 2(b), confirms diffusion at the Ni/Be interfaces, with Ni extending up to 1 μm into the top Be layer and 2 μm into the substrate; Be has also diffused throughout the Ni inter-layer which comprises approximately 25 at.% Be, 45 at.% Ni and 30 at% C. The JET4A model also requires a thinner top Be layer than the JET4C model, indicating erosion from the surface of the order of 1 μm; this corresponds with the estimated erosion due to the mass lost from the sample. A similar level of diffusion and erosion is observed for sample JET4B.

The resolution of the proton backscattering technique was insufficient to enable the graded interface between the Cu/Be inter-layer and the Be top layer of series JET5 to be modelled accurately, hence the samples were assumed to have a Cu/Be mixed inter-layer with an abrupt interface. The best fit for the modelling of the layers on sample JET5C was 10 μm-Be/1.6 μm-$Cu_{0.14}Be_{0.86}$/Be-substrate, which is taken as the as-deposited structure for sample series JET5. Due to the low concentration of Cu in the Cu/Be inter-layer, the signal from the Cu is small, making it more challenging to produce a satisfactory model for the exposed samples JET5A and JET5B that fits well with the raw data. However it was possible to get some indication of the changes to the exposed samples, and as with the Ni inter-layer samples, there is evidence for diffusion at the interfaces and erosion from the surface. The modelling also indicates that the extent of the inter-layer, including the interface regions, has at least doubled following exposure in PISCES-B.

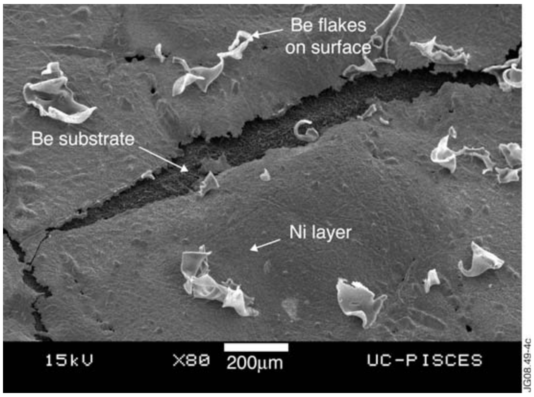

Fig. 3. SEM image of sample JET2B showing delamination of Be and Ni layers.

When samples JET2A and JET2B, deposited on mechanically cleaned surfaces, were removed from PISCES-B, it was found that both coatings had delaminated. An SEM image of the surface of sample JET2B is shown in Fig. 3. The image shows the remains of Be flakes on the sample surface with the Ni layer delaminating from the Be substrate. The mass loss for this sample was 6 mg, equivalent to the loss of an 8 μm Be layer, consistent with the findings from SEM images. During the testing of samples JET2A and JET2B in PISCES-B it was noted that the coatings got sufficiently hot that they glowed, a characteristic that was not observed on samples from series JET4 and JET5. Evidence of this was observed from IR spectroscopy which indicated that the surface temperature of the JET2 samples were higher than the other samples tested. For example the surface temperature of sample JET2A was >300 °C higher during the first 200 s of exposure in PISCES-B, than sample JET4B, exposed to the same heat flux. For the rest of the exposure the surface temperature of JET2A was lower but still remained $\geqslant$100 °C higher than sample JET4B. The surface temperature of sample JET2B was also found to be ~300 °C higher than for the samples from series JET4 and JET5.

The result of the vacuum heating tests showed that the coating on sample JET4A deposited onto an etched surface survived all three heating cycles, to a maximum temperature of 600 °C, whilst the coating on sample JET3A on a mechanically cleaned surface delaminated after one heating cycle to the same temperature.

4. Discussion

The aim of these experiments was to test whether suitable Be marker coatings could be developed to withstand the conditions expected within JET and whether such coatings would provide information on Be erosion by post-mortem analysis. Samples in series JET4 and JET5 deposited on etched Be substrates survived exposure in PISCES-B and heating under vacuum to 600 °C, and on this basis they would make suitable marker coatings for installation in JET, whilst samples from series JET2 and JET3 delaminated during similar testing and are not suitable. These results show that substrate preparation is an important factor in the marker coating deposition as poor surface preparation results in poor adhesion of the layers with the substrate. This is the likely cause of the higher surface temperatures observed for samples JET2 as poor adhesion leads to the poor thermal contact of the coating with the substrate thus preventing the heat in the coating from dissipating. Testing on similar Be marker coatings (produced by a different manufacturer) has been performed at the JUDITH electron beam facility [5]. These coatings also survived heating to 600 °C, but start to fail at temperatures above this.

The 1% yield observed during these tests is of the same order as other sputtering yields for Be surfaces from normal incidence deuterium ions [6,7], it also agrees with the 1.5% Be yield observed for the main wall of JET for the period 1999–2001. Based on the observed yield and the average ion flux, $5 \times 10^{15}\ \mathrm{cm}^{-2}\ \mathrm{s}^{-1}$, to the JET main wall during 2005–2007 [8] it is expected that the Be coating will survive for a period of 25×10^5 s, far in excess of the operating period, $\sim 1.4 \times 10^5$ s, during 2005–2007 [9]. However, enhanced erosion is expected at certain locations during the initial plasma limiter phase, such as at the mid-plane on the IWGL where a 3 μm tungsten coating was completely eroded during 2005–2007 [2]. Clearly the variation in erosion around the main chamber wall makes it difficult to settle on a coating with one nominal thickness which will survive all locations specified in Fig. 1. An obvious solution would be to increase the thickness of the Be layer, however an over-thick layer is also undesirable as it will not be possible to analyse it by proton backscattering using 2.5 MeV H^+, since the protons will not penetrate through the coating into the bulk Be. Furthermore, in regions where gross erosion occurs (i.e., >10 μm) it will be possible to make direct measurements of the tile thickness. Thus taking all factors into consideration a 10 μm Be top layer is the optimum thickness required for these marker coatings.

The diffusion at the interface also presents a challenge if the marker coatings are to provide a useful means of investigating Be erosion. From modelling of backscattering data for sample JET4A it was estimated that Ni had diffused approximately 2 μm into the substrate and 1 μm into the Be top layer. A similar 1 μm layer has been observed at the interface with the top Be layer in samples treated in JUDITH [5], although no diffusion at the substrate layer was reported. This may be due to differences in surface preparation. The reason for the lower diffusion rate at the interface with the top Be layer is attributed to the presence of an oxide. The diffusion coefficient required for ~2 μm diffusion of Ni in Be in 3600 s is $4.5 \times 10^{-13}\ \mathrm{cm}^2\ \mathrm{s}^{-1}$, which would be reached at a temperature of 625 °C, based on the mutual diffusion of Ni and Be [10]. This is comparable with the highest temperature that sample JET4A was exposed to under vacuum. The diffusion during this heating stage dominates as the diffusion coefficient is more than an order of magnitude higher, due to its exponential dependence on temperature, than at any other stage during the testing of JET4A, including exposure in PISCES-B. This implies that provided the temperature of the main chamber wall in JET does not reach 600 °C for more than a few thousand seconds (typically <1% of total operating time), Be/Ni based coatings will be suitable as erosion markers, since the level of diffusion will be low enough that post-mortem analysis will be possible.

The backscattering data for the JET5 series shows that diffusion has occurred at the interfaces; however detailed evaluation has not been presented due to the low concentration of Cu in the inter-layer. Additional analysis at GA of the diffusion of Cu into Be for a similar sample has shown that at 425 °C for 25 h Cu diffuses up to 5 μm into Be. This level of diffusion is an order of magnitude greater than for Ni at the same temperature. As a result of this higher diffusion rate it is concluded that the Cu/Be inter-layer would not make a suitable marker coating.

5. Conclusions

Samples of marker coating which could be used in assessing Be erosion during the ILW operation of JET have been tested in PISCES-B. The results showed that the favoured 10 μm Be top layer with 2 μm Ni inter-layer deposited onto etched Be substrates would survive limited periods of heating to 600 °C, the highest temperature expected on the JET main chamber wall. Diffusion was observed at both the top Ni/Be interface and the interface with the substrate. The diffusion at the upper interface was less than that observed at the substrate; this is likely to be due to the presence of an oxide at the upper interface acting as a diffusion barrier. A diffusion barrier might be considered at the interfaces of the final samples, but this is accompanied by the risk of poor adhesion. The amount of diffusion observed is unlikely to pose a problem for post-mortem analysis. A sputtering yield of 1% was observed for 100 eV deuterium ions. Based on this sputtering yield, the flux to the wall and other evidence of erosion, it is concluded that 10 μm of Be on the top surface is the optimum thickness required for the coatings, particularly as it will be possible to make direct measurements of tile 5 thickness where gross erosion occurs.

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References

[1] P. Andrew, A. Alonso, G. Arnoux, et al., J. Nucl. Mater. 363–365 (2007) 1006.
[2] J.P. Coad et al., Contribution P2-53 at PSI-18, 2008.
[3] R. Doerner, M.J. Baldwin, K. Schmid, Phys. Scr. T111 (2004) 75.
[4] M. Mayer, SIMNRA User's Guide, Report IPP 9/113, Max-Planck-Institut für Plasmaphysik, Garching, Germany, 1997.
[5] T. Hirai et al., Fus. Eng. Des. 83 (7-9) (2008) 1072–1076.
[6] R.P. Doerner et al., J. Nucl. Mater. 275 (1998) 51.
[7] D. Nishijima et al., Contribution P2-14 at PSI-18, 2008.
[8] M.F. Stamp et al., Contribution P1-05 at PSI-18, 2008.
[9] M. Rubel et al., Contribution O-12 at PSI-18, 2008.
[10] V.M. Anan'in et al., translated from Atomnaya Energiya 29 (1970) 220–221.