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Composition and hydrogen isotope retention analysis of co-deposited C/Be layers

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Abstract

A neutral Be atom source and deposition probe have been installed on PISCES-B to simulate ITER diverter erosion and deposition phenomena where Be impurity concentration in the plasma, up to 10%, is expected due to first wall erosion. Graphite target erosion by deuterium plasma is found to be significantly reduced with as little as ~0.1% Be impurities in the plasma [K. Schmid, M.J. Baldwin, R. Doerner, these Proceedings]. Deposited Be on the target is re-eroded and leads to deposited layers in the target vicinity, that are sampled with the deposition probe. These layers are Be rich and contain only small C concentrations (<3 at.%). The ratio of co-deposited deuterium to deposited beryllium (D/Be) decreases rapidly with increasing probe temperature up to 573 K. D/Be values are systematically lower and O/Be values higher compared to similar experiments in TPE [R.A. Causey, D.S. Walsh, J. Nucl. Mater. 254 (1998) 84]. The difference is likely due to different target-collector geometries and incident hydrogen isotope fluxes during collection.

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1. Introduction

Sputtering of the plasma-facing components in fusion devices leads to eroded impurity fluxes that deposit, forming material layers, in locations where the plasma erosion is less intense. Continual bombardment of these layers by plasma species leads to co-deposited and implanted isotopic hydrogen inventories and other gas impurities. In a device like ITER, the build up of tritium in deposited layers is a serious issue, which for safety reasons, could impact on operational time. For example, Federeci et al. [3] quote an anticipated ITER tritium inventory in deposited carbon films alone to be 0.1–10 g per 1000 s of operational time. Current in-vessel tritium limits of 350 g in co-deposited layers [4] would necessitate tritium removal after every ~10 h of operation in the worst case.

This estimate does not however, take into account the expected beryllium impurity flux of up to 10% that will be present in the ITER plasma flow into the divertor due to physical sputtering of the beryllium first wall. Recent PISCES experiments [1,5] have shown a strong reduction in the chemical and physical erosion of graphite targets

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exposed to deuterium plasmas containing small amounts of ~0.1% beryllium impurities. Extended to an ITER scenario, where the Be impurity flux will be much greater, the possible reduction in the erosion of graphite components leads to a natural conclusion that the build up of deposited carbon layers should correspondingly be reduced. The implications are that the anticipated tritium inventory in deposited films in a device like ITER may need to be re-evaluated.

In the experiments reported here, ATJ graphite targets are exposed to deuterium plasmas with Be impurity fractions ~0.1%. Eroded material that is deposited is sampled in the vicinity of the target using a deposition probe with coupon collector substrates that are subsequently analyzed for elemental composition and deuterium content.

2. Experimental

Fig. 1 shows schematically the experimental arrangement. The UC-San Diego PISCES-B device is used to provide a steady-state, high-flux ($\sim 3\times10^{22}$ D ions $m^{-2}\,s^{-1}$) flowing plasma that simulates the flow of scrape-off-layer plasma into the ITER diverter. ATJ graphite targets are used to simulate the ITER graphite strike points. A Be impurity flux is generated in PISCES-B [5,6] through the use of an evaporative neutral atomic beam source (VEECO/APPLIED EPI Molecular Beam Epitaxy effusion cell). Evaporated Be from the neutral atom source enters the plasma in a cross-field geometry. The Be is entrained uniformly, once it ionizes, in the magnetized-plasma-flow incident on the target. Adjustment of the effusion cell temperature allows independent control of the Be impurity flux in the plasma. The chosen plasma conditions used, $N_e \sim 3\times10^{18}\,m^{-3}$ and T_e = 6–8 eV, are sufficient to fully ionize the injected Be beam [5,6]. The concentration of Be ions in the plasma column is determined from the line intensity of Be II (467 nm) using absolutely calibrated optical emission spectroscopy. For this calculation, a reciprocating double Langmuir probe is used to measure the required plasma electron temperature T_e and density N_e. The rate coefficient for electron impact excitation of Be II, is taken from the ADAS database [7].

Intense ion bombardment of targets is achieved by biasing to −50 V. In PISCES-B, the ion-neutral collisional-free path is much larger than the target sheath so that ions impact on the target normally with almost the full energy of the sheath potential, which is typically $\sim kT_e$ (~6 eV) lower than the bias potential.

Material eroded from targets during plasma exposure is collected on substrate coupons (Ta, Mo and W) through the use of the deposition probe shown in Fig. 1. Coupons are shielded from cross-field plasma transport so that only particle fluxes from sputter erosion and reflection from the target contribute to deposition/co-deposition. The deposition probe can be independently heated in the temperature range 300–700 K and is fully retractable into a vacuum interlock chamber to allow rapid coupon replacement without breaking the PISCES-B vacuum.

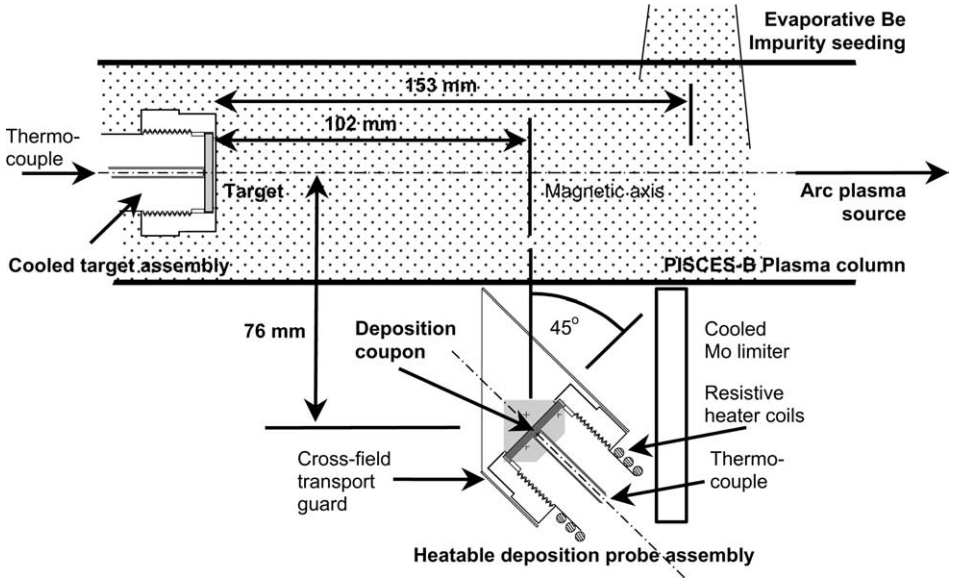

Fig. 1. Arrangement used to collect deposited/codeposited material during plasma exposure of target materials in PISCES-B. Targets and deposition probe coupons are 25 mm in diameter. The plasma column is typically of ~100 mm diameter.

3. Results and discussion

Small Be impurity fractions in the PISCES-B plasma, are found to strongly suppress deuterium plasma erosion of graphite below 1000 K [4], as shown by the emission spectroscopy data of Fig. 2. The emission from eroded material in front of the target for 0.2% Be impurity seeding and no Be are compared. The two target temperature regimes were chosen to demonstrate reductions in both the chemical (~600 K) and physical (~1000 K) erosion mechanisms. At ~600 K Be impurity seeding reduces the CD band structure (~430 nm) in the plasma to almost insignificant levels, and a likewise strong reduction in C I line emission at 941 nm is found for the target exposed ~1000 K. Subsequent weight loss measurements for these targets reveals weight reductions in the Be seeding cases of only a few percentage of the no Be seeding cases. Wavelength dispersive X-ray analysis of the target surfaces reveals high Be surface concentrations ~60–80% in Be seeded cases. A more comprehensive description of the influence of Be impurities on target erosion is given in Refs. [1,5,6].

Consequently, it is not surprising that deposited materials on the deposition probe coupons show little evidence of eroded graphite. X-ray photo-electron spectroscopy (XPS) sputter-depth profiles are shown in Fig. 3 for separate polished Ta deposition-probe coupons used during target exposures at ~600 K and ~1000 K where there was ~0.2% Be impurity seeding in the plasma flux. Both targets were exposed to ion fluences of $2.4 \times 10^{26}\ m^{-2}$ while the Ta coupons were kept at 300 K. The XPS profiles show a reverse history of the plasma exposure of the target. At plasma 'switch on', target erosion leads to a small amount of deposited C on the deposition probe coupons. Chemical erosion of the target at 600 K results in more deposition of C compared to the ~1000 K target exposure. Both deposited layers are ~100 nm thick and almost free of O impurities (1–3 at.%) except at surface interfaces which have native oxides. Strong Be deposition on the deposition probe is indicative of a high Be surface concentration on the target since eroded material fluxes for Be and CD_4 are reionized with similar efficiencies under the present plasma conditions [6]. W coupons kept at ~300 K during collection, but with half the Be seeding (~0.1%), show similar O content and reduced layer thickness ~50 nm. At higher temperature however, the O content in the deposited layers rose to ~30 at.%, presumably due to the deposition probe assembly outgassing. All coupons examined, regardless of temperature, showed low C content throughout the deposited layer (<3 at.%) once target erosion was suppressed by coverage in Be. The data and analysis of eroded material collected in the absence of a Be impurity flux will be published separately.

Deuterium retention in the deposited layers was measured as a function of the deposition probe temperature

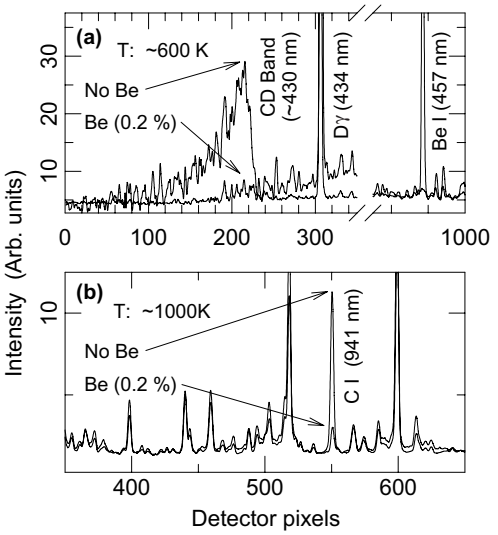

Fig. 2. Compared (No Be and ~0.2% Be impurity seeding) optical emission spectra from a region of plasma in front of the target during plasma exposure. Reduction in chemical ~600 K (a) and physical ~1000 K (b) erosion of ATJ graphite is observed.

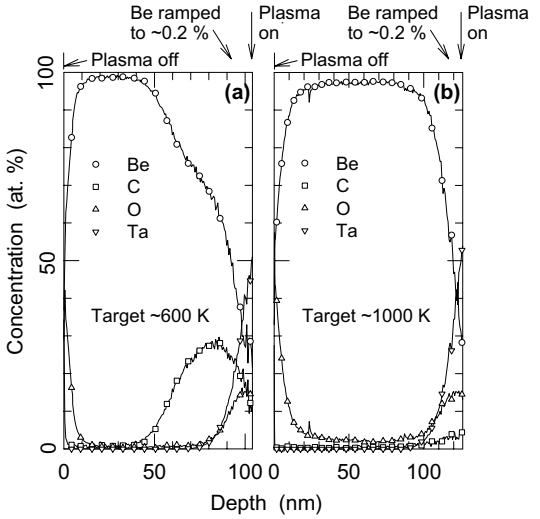

Fig. 3. XPS sputter-depth-profiles for deposited material collected on Ta deposition probe coupons. For each, ATJ graphite targets were exposed to deuterium plasmas with a Be impurity seeding fractions of ~0.2% at ~600 K (a) and ~1000 K (b). The D ion fluence to the targets was $2.4 \times 10^{26}\ m^{-2}$. Increasing depth into the deposited layer reflects the reverse history of the plasma exposure. Labels along the top of each plot are approximate due to the effects of ion beam mixing during sputter profiling.

in the range 300–573 K using W coupons. Two target exposure regimes of ~600 K and ~1000 K were used. In all plasma exposures the conditions were identical and the Be impurity seeding fraction was ~0.1%. Fig. 4 shows calibrated D_2 thermal desorption spectra (TDS) for the deposited layers corresponding to targets eroded at ~1000 K. TDS profiles for the cases where targets were ~600 K show similar features. Retained deuterium in the deposited layers is found to diminish with increased coupon temperature during collection. D_2 ($m/e = 4$) was the most significant deuterium species released and, consequently, the only TDS mass profile used to determine the desorbed deuterium content from the deposited layers. TDS desorbed deuterium contents were found to be in good agreement with nuclear reaction analysis (NRA) measurements on sectioned pieces of the same samples, implying the full removal of deuterium during the TDS procedure. The desorption profiles in Fig. 4 are characterized by a rapid onset that approximately coincides with the coupon temperature during collection. Further, the commonality between desorption profiles is suggestive of a single trapping mechanism, which is unlike that found with either co-deposition with BeO (~900 K) [8] (despite the high O content in the layers collected at 423 K and 523 K), or thermal release from implanted pure beryllium [9].

The D/Be and O/Be ratios for the deposited films are plotted in Fig. 5 and compared with other data in the literature [2,10,11]. The D/Be ratios of the PISCES data, as with the other studies, are found to decrease rapidly as the collection temperature is increased. However,

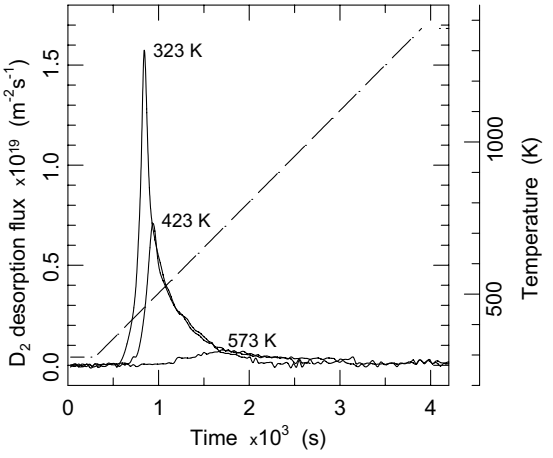

Fig. 4. D_2 TDS profiles for deposited material collected on W, Mo, and Ta deposition probe coupons. Coupons were fixed in temperature (323, 423 and 573 K) during plasma exposure of graphite targets to Be impurity seeded ~0.1% deuterium plasmas. The D ion fluence on target was $2.0 \times 10^{26}\ m^{-2}$ for all exposures. The dashed line shows the heating rate: $1.67\ K\ s^{-1}$.

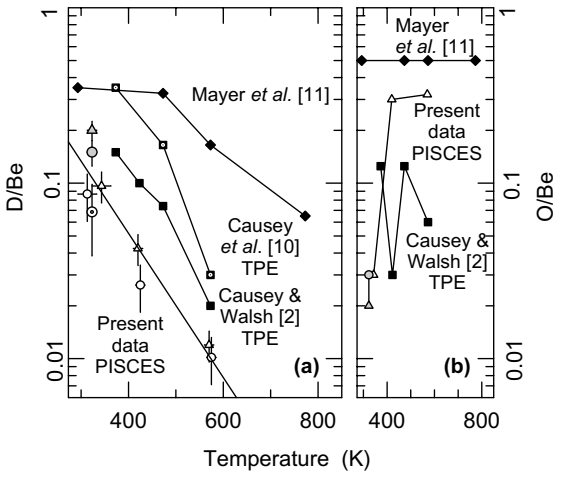

Fig. 5. D/Be (a) and O/Be (b) ratios for deposited material collected on Ta (grey symbols), Mo (dotted symbol) and W (white symbols) deposition probe coupons as a function of coupon temperature. Target temperatures are indicated by triangular (~600 K) and circular symbols (~1000 K).

the PISCES data systematically show the lowest deuterium retention with temperature. Similar D/Be results with different coupon materials suggest deuterium trapping mainly in the deposited layers.

It is interesting to note that the retained hydrogen isotope fractions in the PISCES deposited layers are systematically low despite the rather high O content in the layers at higher temperature. It has been suggested [2,9,12,13] that O content in deposited Be films can have a strong influence on hydrogen isotope retention, effectively increasing the D/Be ratio to values comparable with a:C–H films during room temperature deposition. When compared, the D/Be data of Mayer et al. [11] and Causey et al. [2,10] suggest this: the D/Be ratios are systematically lower in the Causey data where the O content in the films is coincidently also lower. In their experiments, deposited material was collected from sputtered Be targets, and the arrival rates of the various species are certain to be a strong influence [13]. Mayer et al. used an ion beam whereas Causey et al. used the intense plasma in the tritium plasma experiment (TPE). As noted by Anderl [13], the experiments of Mayer et al. are such that the arrival rates of gas impurities (O_2, H_2O, etc.), hydrogen isotopes, and Be are comparable, likely leading to the observed high O and D content in the Mayer films. In the experiments of Causey et al., more intense plasma sputtering of the Be target leads to comparatively higher Be arrival rates at the collector than O.

The current experiment in PISCES is similar to the TPE experiment due to close similarity of the two machines, but the PISCES results show systematically low

D/Be values with temperature regardless of the O content in the film. Comparison of the O/Be plots of Fig. 5(b) suggest a complicated picture where O content in the deposited layer is not necessarily a mechanism which tends to increase the uptake of hydrogen isotopes in the film. However, there are differences between the PISCES and TPE experiments. Deposited material collected in PISCES is shielded from cross-field transport. The arrival rate of deuterium to the deposition probe would therefore have been significantly less in PISCES than TPE. The collector solid angle from the target and target area were also larger in TPE compared to PISCES. This difference in target-collector geometry means that a larger reflected hydrogen isotope flux was incident on the collector in TPE. Further study is warranted to understand these influences.

4. Conclusions

A small Be impurity flux in PISCES-B deuterium plasmas is found to produce a high surface concentration of deposited Be that reduces target erosion on ATJ graphite. In turn, target-surface Be re-erodes and leads to Be rich deposited layers in line-of-sight locations from the target due the high sticking probability for Be metal atoms. The line-of-sight nature of the deposition is more predictable and manageable than a:C–H film growth which occurs in unexpected locations, like pump ducts, because of the poor sticking probabilities of hydrocarbon molecules. Measurements of the deuterium retention in deposited layers reveal a trend in the ratio of D/Be that decreases from ~0.1 at 300 K to ~0.01 at 600 K. The results suggest that co-deposited hydrogen isotope inventories in Be rich film deposits can be substantially reduced by baking or operation at temperatures near 600 K.

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