Beryllium containing plasma interactions with ITER materials

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Abstract. Beryllium-seeded deuterium plasma is used in PISCES-B to investigate mixed-material erosion and redeposition properties of ITER relevant divertor materials. The beryllium containing plasma simulates the erosion of first wall material into the ITER sol plasma and its subsequent flow toward the carbon divertor plates. The experiments are designed to quantify the behavior of plasma created mixed Be/C and Be/W surfaces. Developing an understanding of the mixed material surface behavior is crucial to accurately predicting the tritium accumulation rate within the ITER vacuum vessel. The temporal evolution of the plasma interactions with the various mixed surfaces are examined to better understand the fundamental mechanisms in play at the surface and to allow scaling of these results to the conditions expected in the ITER divertor. A new periodic heat pulse deposition system is also installed on PISCES-B to simulate the transient temperature excursions of surfaces expected to occur in the ITER divertor during ELMs and other off-normal events. These periodically applied heat pulses allow us to study the effects of transient power loading on the formation, stability and tritium content of mixed-material surfaces that are created during the experiments.

1. Introduction

A bilateral US(DOE)-EU(EFDA) collaboration, focused on experiments performed in the PISCES-B linear divertor simulator [1,2], utilizes a beryllium-seeded deuterium plasma to investigate mixed-material erosion and codeposition properties of ITER relevant divertor materials. The experimental program is designed to reduce uncertainties in the prediction of tritium retention in codeposited mixed-materials in ITER and other future burning plasma devices. The beryllium-containing plasma simulates the erosion of first wall material into the ITER sol plasma and its subsequent flow toward the carbon and tungsten material located in the ITER divertor region.

This paper will summarize the primary, significant results that this collaboration has produced. It will relate these results to the operating conditions expected in ITER and attempt to predict what the consequences of mixed materials on the ITER plasma-facing surfaces might be.

The first, most dramatic result obtained from this collaboration was the suppression of carbon erosion, both physical sputtering and chemical erosion, at very low levels (~0.1%) of incident beryllium impurity ions [3,4]. Typical deuterium plasma parameters in PISCES-B are: ion flux ~ 4 x 10^{22} $m^{-2}s^{-1}$, incident ion energy ~ 50 eV and surface temperature ranging from 100-1000°C. This result was confirmed using both spectroscopic as well as weight loss measurements. These initial measurements focused on the equilibrium state of the plasma-exposed surface. In other words, long fluence plasma exposures were performed, after which the surfaces of the plasma bombarded targets were examined. In addition, during these large fluence exposures material collectors (referred to as witness plates) were positioned outside the plasma column to collect eroded material [5].

Subsequent experiments have documented the temporal evolution of the mixed beryllium-carbon plasma-created surface [6,7]. Through focusing on the time-dependent evolution of the surface and its associated plasma-material interactions, we believe we have been able to identify the fundamental mechanisms responsible for the mitigation of carbon erosion from the mixed-material surface. An ongoing series of experiments is examining the response of the mixed-material surface to periodically applied thermal transients [8] with the goal of understanding how such thin surface layers might react to off-normal events in ITER.

In addition to mixed Be/C studies, the issue of alloy formation between beryllium and tungsten is under investigation [9]. Various beryllide (Be_xW) alloys have been observed to form during different plasma exposure conditions. Again by understanding the formation conditions of these alloys, it will increase the confidence in predictions regarding their importance to the ITER design.

2. Present understanding of mixed Be/C surfaces

Our present understanding of mixed Be/C surface formation and implications is as follows:

- Be ions incident in the bombarding plasma become implanted into a carbon surface and due to the energy of the incoming particles, the implanted berrylium will tend to bond with carbon atoms to form beryllium carbide (Be_2C)
- The formation of Be_2C in the surface acts to inhibit the reaction chain responsible for chemical erosion of carbon and also reduces physical sputtering of carbon atoms from the surface (by diluting the surface concentration of C atoms exposed to plasma bombardment)
- The Be_2C surface layer thickness saturates after a time that depends on the plasma interaction conditions
- Once a protective Be_2C layer forms on the surface, subsequent beryllium ion bombardment will produce an enrichment of the surface with beryllium that will be easier to erode (compared to beryllium bonded as the carbide)
- After the Be_2C layer forms, the primary species eroding from the plasma bombarded mixed-material surface, and codepositing with hydrogenic species on non-plasma-exposed surfaces, is beryllium.

In PISCES-B one can investigate the bonding patterns of atoms in plasma-exposed surface using in-situ X-ray Photoelectron Spectroscopy (XPS). Figure 1 shows the C1s and Be1s signals from a sample exposed to beryllium-containing deuterium plasma at 450°C. Also included in the figure are similar XPS signals from ‘standard’ carbon and beryllium samples exposed to deuterium only plasmas. The figure shows a clear shift in bonding toward the carbidic bonds, with essentially all carbon in the surface being bound as carbides after the higher temperature exposures.

The effect of carbidic bonding of impurities in graphite has been shown to reduce the rate of chemical erosion in a variety of doped graphites [11]. The difference in these PISCES experiments is that the doping of the graphite with beryllium impurities occurs as a consequence of having metallic impurities entrained in the plasma flow toward the graphite target. A similar effect might be expected in the divertor of ITER. Measurements of the effects of beryllium doping of graphite show similarities to those exhibited by boron doped graphite, namely an increase in the hydrogenic species retained by the doped material and a shift in the release patterns of the retained deuterium to lower temperature [10]. This behavior, in an indirect way,

shows the impact of beryllium doping of graphite. Direct measurements of the amount of chemical erosion of carbon from a graphite surface are also obtained in PISCES-B by monitoring the CD band emission from directly in front of the sample surface. Figure 2 shows the variation in the CD band intensity with the fraction of beryllium present in the surface of the sample.

Rutherford Backscattering (RBS) data have been obtained at the University of Wisconsin on targets that were exposed to beryllium-seeded plasma in PISCES [10]. This data reveals the saturation in the thickness of beryllium on the surface of two samples exposed for different times to identical plasma conditions. In the case of a sample exposed to a beryllium ion fluence of 3 x 10^{22} m^{-2}, virtually all of the beryllium incident on the sample from the plasma can be accounted for in the surface layer. When the exposure time, under identical plasma conditions, is increased on a different sample (to 1 x 10^{23} m^{-2}), the same amount of beryllium is found in the surface, indicating that the subsequent beryllium deposited on the sample has been eroded. These stationary Be depth profiles (with increasing fluence) are similar to earlier measurements made of carbon-tungsten mixed material surface layers [12] and indicate the formation of stable mixed-material surface layers.

The beryllium eroded from the sample surface can be observed by monitoring the neutral beryllium line emission (at 457.3 nm) from directly in front of the exposed target [13]. By examining, in detail, the temporal evolution of a typical beryllium seeded plasma exposure in PISCES-B, one can clearly see the increasing beryllium erosion from the sample surface. Figure 3 shows this behavior along with the temporal behavior of the chemical erosion decay (measured by the CD band emission from directly in front of the target surface). Although the mitigation time of the chemical erosion of a surface depends on the plasma conditions during the exposure [6], in this particular exposure the CD band is measured to decay in approximately the first 60 seconds of the exposure after the beryllium oven shutter has been opened (t = 0 sec.). A small amount of neutral beryllium line radiation is detected within the line of sight of the spectrometer when the oven shutter is opened at t = 0 sec. In fact, one observes a slight drop in this initial intensity during the first 10 seconds, or so, of the exposure due to the oven temperature re-equilibrating when the shutter is initially opened. After the beryllium oven temperature has stabilized, the beryllium line radiation remains constant throughout the decay of the CD band intensity, indicating that the beryllium erosion from the target surface is not changing. Once the chemical erosion from the sample has been suppressed, the beryllium erosion from the target is

observed to increase until at about 200 sec. into the discharge (for these conditions) the beryllium erosion from the surface reaches equilibrium with the incoming Be ion flux from the plasma.

Monitoring the D_{gamma} line radiation from the plasma is used to normalize the two other signals, thereby accounting for any possible changes in the optical properties of the detection system, for example coating of the optical windows. Figure 3 also shows the D_{gamma} signal (labeled window transmission in the figure) as a function of time throughout the plasma discharge. During the first 50 seconds of the discharge the D_{gamma} signal does not change (within the scatter in the experimental measurement), however once the beryllium erosion from the sample begins to increase (after 50 sec), the D_{gamma} signal is seen to decrease monotonically throughout the remainder of the exposure. In other words, the optical windows in the PISCES-B vacuum system coat up due to the beryllium erosion of the incident beryllium plasma ion flux.

This interpretation of the D_{gamma} signal is corroborated by the analysis of witness plate samples located outside the plasma column during the discharge [5]. Typical depth profiles of material collected on such witness plates are shown in Figures 4a and 4b. In these cases the collector plate was tantalum and is covered by approximately 100 nm of codeposited material collected during the plasma exposure. In both cases, beryllium is the predominant material collected. The depth profile of material on the witness plate samples can then be interpreted as the temporal dependence of erosion from the plasma-bombarded target.

As has been previously reported, the suppression time of chemical erosion is much shorter when the temperature of the exposed target is higher [6] (at higher surface temperature the beryllium and carbon atoms in the surface react more readily to form beryllium carbide, thereby reducing chemical erosion). The material collected initially during low-temperature carbon target exposure (300°C), Figure 4a, between 70 – 100 nm into the witness-plate layers, contains a significant amount of carbon. As the exposure continues in time, the carbon material collected on the witness plate decreases (50 –70 nm into the layer), until eventually all the material eroding from the target is beryllium (0 – 50 nm into the layer). During a high-temperature target exposure (700°C), Figure 4b, the suppression of chemical erosion is much more rapid [6] and virtually no carbon is detected throughout the layer of collected material on the witness plate.

3. Transient temperature excursion experiments

An important issue related to the formation of mixed-material surfaces in the ITER environment will be their response to off-normal events, such as ELMs and possibly disruptions. Although these events do not occur in the PISCES devices, certain aspects of their occurrence can be simulated. The impact of periodic surface temperature excursions is being investigated during beryllium-seeded plasma discharges by applying a short-duration positive bias to the targets exposed in PISCES-B [8]. Typically, targets are biased negatively in PISCES to provide the ion bombarding energy, however a positive pulsed bias can be applied to collect electron current, and thereby heat, to the targets repetitively during the plasma exposure. Varying the voltage of the positive bias controls the power flux to the target.

Although these systematic experiments are just beginning, a few observations have already been documented. First, during power fluxes that vary the target temperature from the steady-state value of 500°C, up to 1200°C during the 100 msec heat pulse (with a heat pulse duty cycle of 1%), the mitigation time of the chemical erosion of the target is seen to decrease dramatically [8]. This behavior is at least qualitatively understandable from the mitigation time scaling studies [6] mentioned above. The second result is that the amount of deuterium retained in the targets increases, by roughly 50%, during heat pulse experiments when compared to identical exposures without the application of the positive bias heat pulses [8]. This indicates that the high temperature excursions of the surface are acting to increase the diffusivity of deuterium into the sample, rather than increasing the recombination and release of deuterium from the target surface. The pulsed-power supply system has recently been upgraded to allow the application of higher positive bias voltage pulses to the targets.

4. Be/W Mixed-Material Studies

The other mixed-material system relevant to the ITER device is the Be/W binary mixture. The tungsten beryllide system [14] consists of three alloys (Be_2W, $Be_{12}W$ and $Be_{22}W$) all of which have a lower melting temperature than pure tungsten. The importance of beryllide alloy formation was initially observed in PISCES-B due to the unexpected deterioration of the tungsten crucible used in the beryllium oven during the seeding experiments [15]. Subsequent studies of beryllide formation in tungsten samples exposed to beryllium-seeded deuterium

discharges have identified the region of plasma conditions necessary for the alloys to form and grow [9].

In the plasma environment, beryllides begin forming in tungsten surfaces when the temperature reaches about 600°C [9]. However, the diffusion rate of beryllium into tungsten only becomes significant above about 800°C [16]. It has also been demonstrated that the growth rate of the beryllide alloy is restricted when insufficient beryllium exists in the surface [9].

A simple particle balance model has been developed and does a remarkably good job of predicting conditions where the beryllide alloys are likely to be a concern. This model compares the arrival rate of beryllium from the plasma, corrected for reflection of incoming Be ions from the tungsten surface, to the loss rate of beryllium from the surface due to sputtering and sublimation, taking into account redeposition of the material being lost from the surface [9]. The additional loss term accounting for diffusion of surface beryllium into the tungsten bulk is neglected. Neglecting the beryllium diffusion into the bulk allows us to predict the plasma conditions that will, by itself, not result in a beryllium layer forming on the tungsten surface. When beryllium does not form a surface layer on plasma exposed tungsten surface, then the growth rate of the breyllide alloy is restricted. In effect, this model predicts the plasma conditions where the alloy growth rate will not be restricted by the available beryllium.

Figure 5 graphically displays the plasma conditions where beryllium layers are expected to form under conditions found in PISCES-B, and under conditions expected in ITER. Conditions corresponding to regions to the left (lower surface temperature) and above (higher plasma beryllium content) the lines are plasma conditions that result in beryllium covered tungsten surfaces. Regions to the right and below the lines are conditions where sublimation and sputtering, respectively, are expected to keep the tungsten surface effectively free of beryllium. The higher incident particle flux expected in ITER acts to shift the surface temperature and/or incident ion energy to higher values to maintain a beryllium free surface (as shown by the dashed lines in Figure 5).

5. Discussion and summary

The understanding of plasma-created mixed-material surfaces has increased dramatically due to this US-EU collaboration. The fundamental mechanisms responsible for the mitigation of

chemical erosion from graphite surfaces exposed to beryllium containing deuterium plasma have been identified. A scaling law for the mitigation time of chemical erosion, based on data from PISCES-B, has been developed to extrapolate these results to the time needed to suppress chemical erosion of the graphite dump plates in the ITER divertor. The scaling law predicts the chemical erosion in the ITER divertor suppression time to be approximately 10 msec. [6], based on typical ITER divertor plasma parameters [17] between ELMs. This indicates that the beryllium carbide layer should form readily between expected ELMs in ITER. Experiments are currently underway to understand the effects of surface temperature transients on the mixed-material surface during conditions similar to those expected during ITER ELMs. The consequence of beryllium carbide formation on the graphite dump plates in ITER is that beryllium based codeposited material should be expected, rather than carbon based codeposits.

A particle balance model has been developed to explain PISCES Be/W results and to predict various plasma-surface interaction conditions where beryllium-rich surface layers form. This model, coupled with experimental evidence that the beryllide alloy growth rate only becomes significant at temperatures around 800°C, can be used to predict locations in confinement devices where beryllide alloys may be a concern.

It is possible to examine the present ITER design [18] with both the Be/C and Be/W mixed-material studies in mind. First, beryllium carbides tend to form more readily on high temperature carbon surfaces. The graphite dump plates in ITER operate at high temperature where their chemical erosion should be mitigated quickly. If the baffles in ITER were made of graphite, where the surface temperature during operation is much lower, then one would expect a slower mitigation of their chemical erosion and hence the problem of tritium retention with the eroded carbon would be much more worrisome.

The present ITER design uses tungsten as the baffle material and the surface temperature in this region is only in the 400-600°C range. Even if a beryllium layer forms on the tungsten surface at this temperature, one expects little beryllide formation and a very small diffusion rate for Be in W at this temperature. If, however, ITER were to switch to an all tungsten divertor, the higher surface temperature expected on the dump plates would lead to significant concerns about beryllide formation and growth in these surfaces. It, therefore, appears that the choice of graphite dump plates and tungsten baffles, in conjunction with a beryllium first wall, may well be the best choice for the divertor design from a mixed-material perspective.

Acknowledgements

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References

[1] Goebel, D. M., et al., J. Nucl. Mater. 121 (1984) 277.
[2] Doerner, R. P., et al., Physica Scripta T111 (2004) 75.
[3] Schmid, K., et al., Nucl. Fusion 44 (2004) 815.
[4] Schmid, K., et al., J. Nucl. Mater. 337-339 (2005) 862.
[5] Baldwin, M. J., et al., J. Nucl. Mater. 337-339 (2005) 590.
[6] Nishijima, D., et al., J. Nucl. Mater. (in press, PSI-17) 2006.
[7] Baldwin, M.J., et al., Nucl. Fusion 46 (2006) 444.
[8] Pugno, R., et al., J. Nucl. Mater. (in press, PSI-17) 2006.
[9] Baldwin, M.J., et al., J. Nucl. Mater. (in press, PSI-17) 2006.
[10] Baldwin, M.J., et al., J. Nucl. Mater. available on-line 28 August 2006.
[11] Roth, J., J. Nucl. Mater. 266-269 (1999) 51.
[12] W. Eckstein and J. Roth, Nucl. Instr. & Methods in Phys.Res. B53(1991)279.
[13] Doerner, R. P., et al., J. Appl. Phys. 93 (2003) 5816.
[14] H. Okamoto and L. E. Tanner, in “Phase Diagrams of Binary Tungsten Alloys”, Editors S. V. Nagendra Naidu and P. Ramo Rao, Indian Institute of Metals, Calcutta, 1991.
[15] Doerner, R. P., et al., J. Nucl. Mater. 342 (2005) 63.
[16] Linsmeier, Ch., et al., J. Nucl. Mater. (in press, PSI-17) 2006.
[17] Federici, G., et al., J. Nucl. Mater 266-269 (1999) 14.
[18] Technical Basis for the ITER-FEAT Outline Design, ITER EDA Documentation Series No. 19, IAEA Vienna (2000).

Figure Captions

Figure 1 – XPS analysis of beryllium and carbon binding on the surface of carbon target exposed to a beryllium containing plasma at 450°C. The Be-C bond is seen to be primarily carbidic in nature, suggesting the formation of a beryllium carbide surface layer.

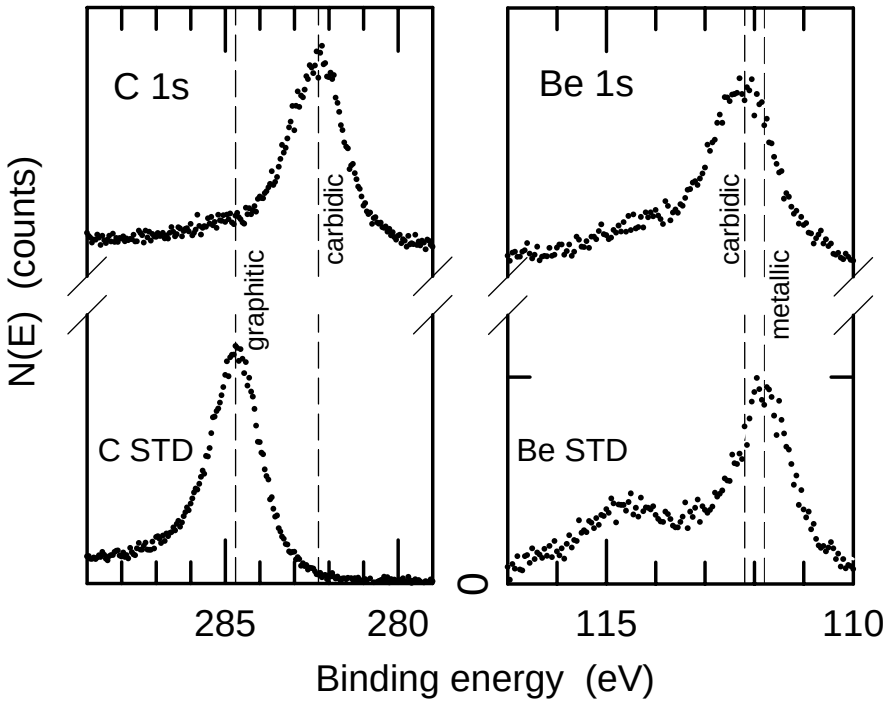
Figure 2 – Variation of normalized chemical erosion rate (CD band intensity) with beryllium surface coverage (from Auger Electron Spectroscopy) of mixed Be/C surface.

Figure 3 – Temporal evolution of the chemical erosion (CD/Dgamma), beryllium erosion (BeI/Dgamma) and the optical window transmission (Dgamma intensity) from a graphite sample exposed to a beryllium-seeded deuterium plasma. The top figure is an expansion of the first 500 seconds of the exposure for clarity. The beryllium oven opens at t = 0 sec.

Figure 4a – Elemental depth profile of the codeposited material collected on a witness plate coupon during the exposure of a graphite target to beryllium-seeded deuterium plasma at 300°C.

Figure 4b – Elemental depth profile of the codeposited material collected on a witness plate coupon during the exposure of a graphite target to beryllium-seeded deuterium plasma at 700°C.

Figure 5 – Results from a plasma-based particle balance model showing regions where beryllium covered surfaces are expected to form on tungsten surfaces, as function of surface temperature and Be concentration in the incident plasma for several different incident plasma ion energies. Solid lines are based on PISCES-B data. Dashed lines represent conditions expected in ITER.



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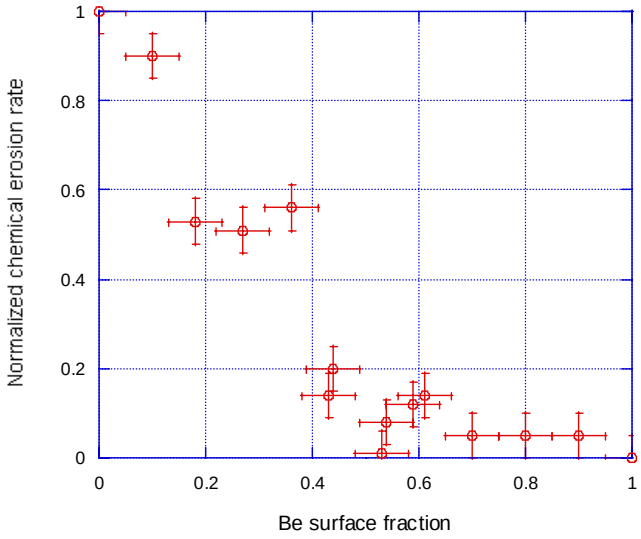

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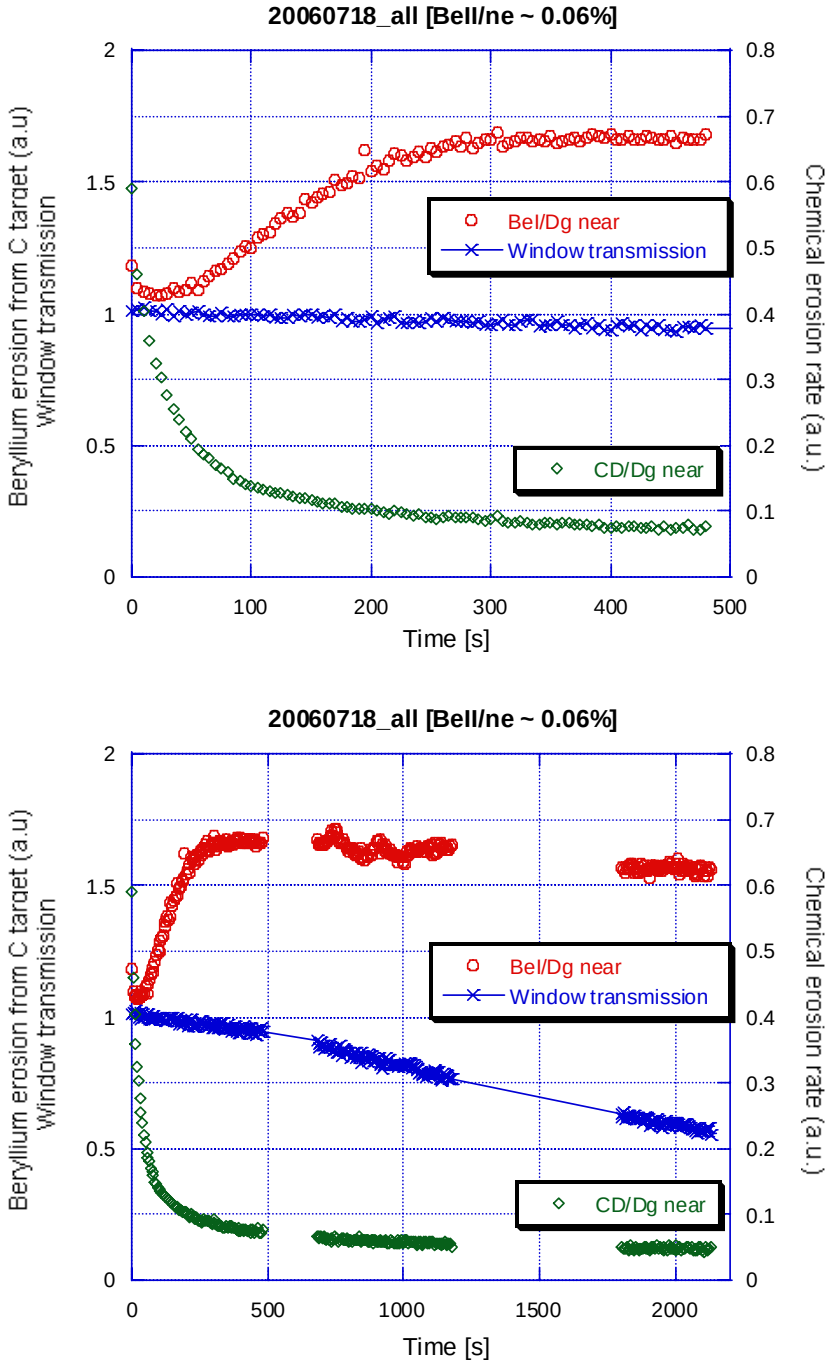

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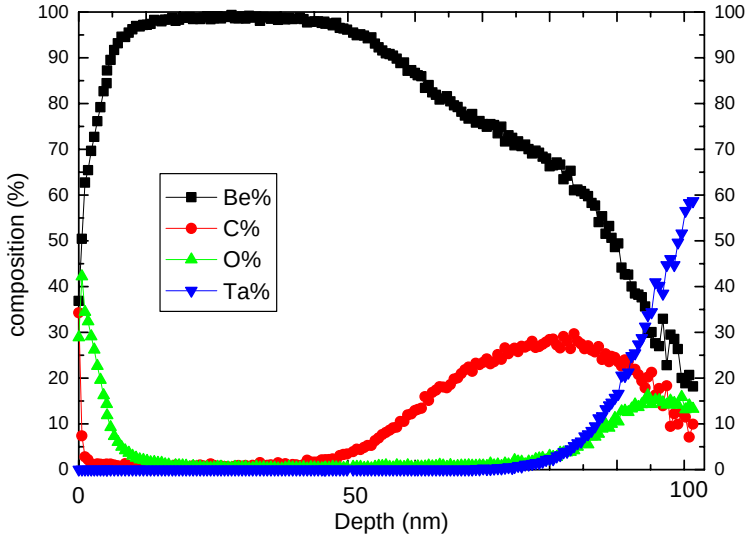

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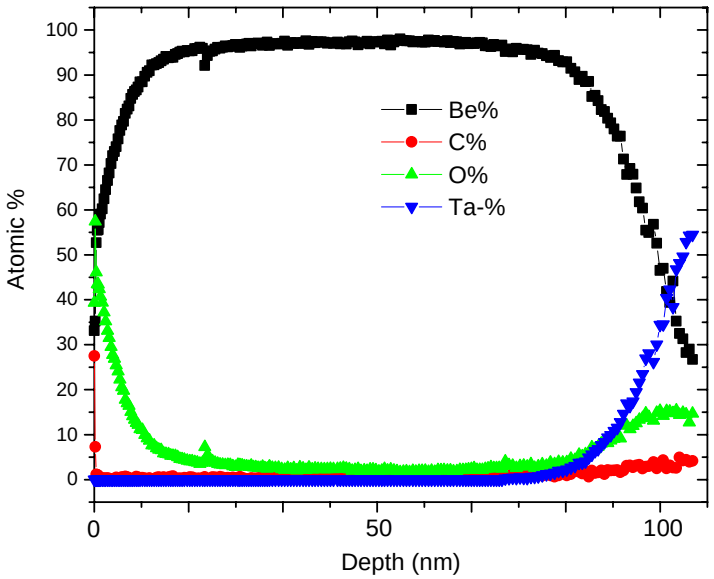
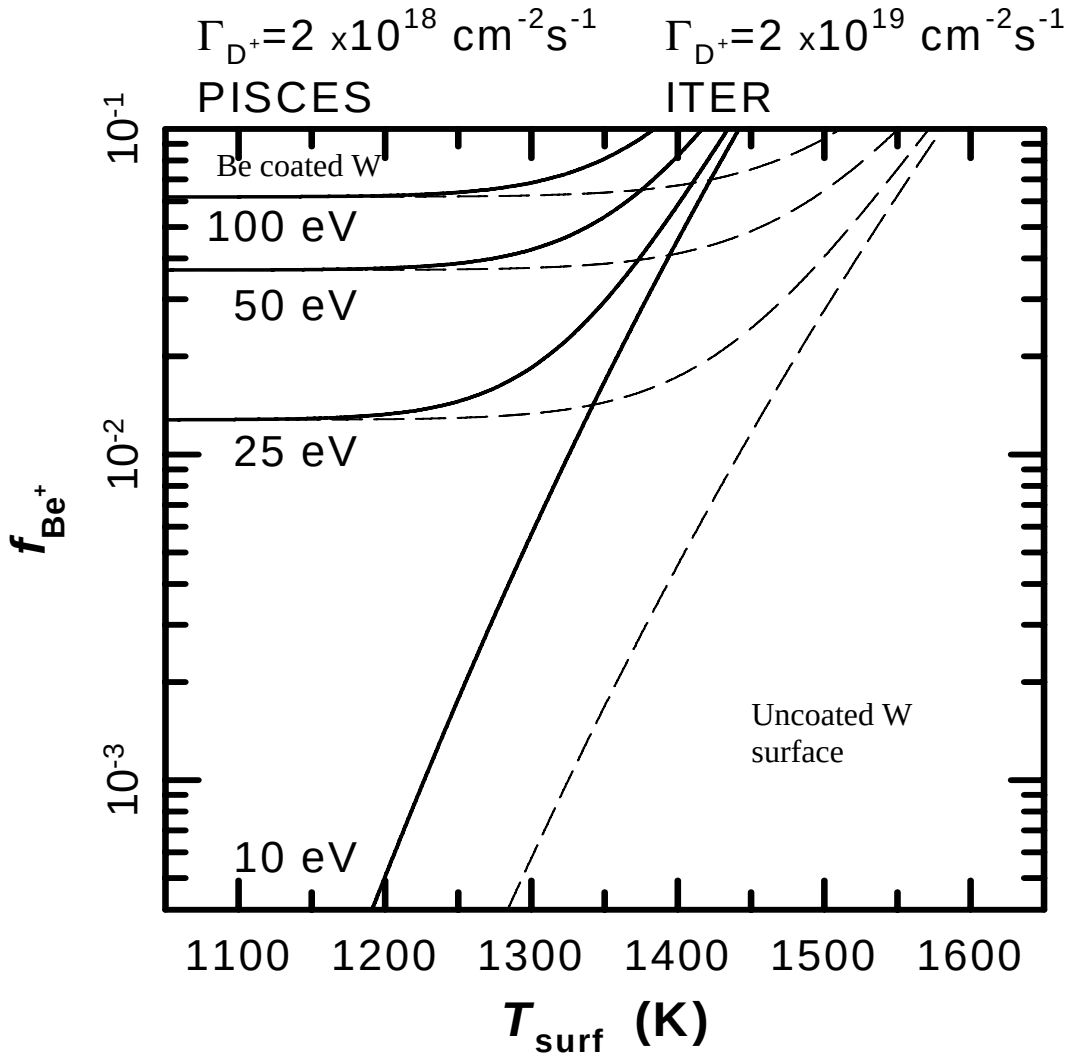


Figure 4b – Elemental depth profile of the codeposited material collected on a witness plate coupon during the exposure of a graphite target to beryllium-seeded deuterium plasma at 700 ℃.



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