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TMAP-7 simulation of D_2 thermal release data from Be co-deposited layers

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ABSTRACT

The efficacy of (1) bake-out at 513 K and 623 K, and (2) thermal transient (10 ms) loading to up to 1000 K, is explored for reducing D inventory in 1 μm thick Be–D (D/Be ~0.1) co-deposited layers formed at 323 K for experiment (1) and ~500 K for experiment (2). D release data from co-deposits are obtained by thermal desorption and used to validate a model input into the Tritium Migration & Analysis Program 7 (TMAP). In (1), good agreement with experiment is found for a TMAP model encorporating traps of activation energies, 0.80 eV and 0.98 eV, whereas an additional 2 eV trap was required to model experiment (2). Thermal release is found to be trap limited, but simulations are optimal when surface recombination is taken into account. Results suggest that thick built-up co-deposited layers will hinder ITER inventory control, and that bake periods (~1 day) will be more effective in inventory reduction than transient thermal loading.

1. Introduction

In the ITER and JET 'ITER-like' wall designs, Be forms the largest surface area for plasma–material interaction (PMI). Erosion of the Be by the plasma leads to a Be fraction in the scrape-off layer that deposits on the same or different PMI surfaces in other locations. In net deposition locations, the Be co-deposits with other species in the local plasma. Previous PISCES divertor plasma simulator experiments, reveal that hydrogen isotopes (D) co-deposit readily with Be and can produce a large retained D inventory (D/Be > 0.1) [1] in some instances, particularly at low temperature (<500 K) like in ITER and JET operation. Consequently Be co-deposits will be a driver of in-vessel retention in the ITER design [2] and removal strategies will be necessary to reduce retained T inventory and satisfy the requirements of nuclear safety licensing.

In this article, the efficacy of both long bake-out and thermal transient loading is explored as a means for reducing hydrogen isotope inventory in Be codeposited layers. Experimental data on the release of D from Be–D codeposits are used to validate model input into the Tritium Migration & Analysis Program 7 (TMAP), for the purpose of potential application to the ITER environment.

2. Experiment and modelling

Two types of experiment were carried out:

(1) Slow bake-out studies, where identical co-deposited Be–D layers were held at ITER relevant bake temperatures of 513 K and 623 K for various lengths of time in order to examine the efficacy of an ITER like bake-out procedure.
(2) Transient desorption experiments, where identical Be–D layers were flash desorbed for 10 ms by focused YAG laser radiation (<50 J) at 1064 nm. The heating rate achievable $\sim10^6$ K s^{-1} provides a way to examine the efficacy of thermal release during proposed 'controlled shutdown' procedure.

A magnetron sputter system was used to produce Be–D co-deposits, the details of which [3] are reported elsewhere. In this device, deposition targets are subject to a Be flux of $\sim2\text{–}3 \times 10^{19}$ m^{-2} s^{-1} in ~6 m Torr of 80–20% Ar–D_2 background fill gas. An applied target potential difference (−120 V) facilitates ion bombardment from fill gaseous species. Little detectable Ar is co-deposited. For bake-out studies (1), the co-deposited layers were produced over the surfaces of a batch (up to 30) of 2 mm dia. W spheres, rolled (in a rotating pan kept at below 323 K) to ensure layer uniformity. For transient desorption experiments (2), the co-deposits were produced as 2 mm dia. spots (using a mask) on flat W substrates at ~500 K. All co-deposited Be–D layers were produced to a thickness of 1 μm, as determined by mass gain, and all W spheres and substrates had been previously annealed in-vacuo for 1 h at 1273 K.

The effectiveness of co-deposit inventory reduction in experiments (1) and (2) is measured by thermal desorption mass spectrometry (TDS). During TDS, co-deposits are out-gassed according to a single, or sequential combination of linear heating rates in the range 0.0(bake)–1.0 K s^{-1} up to a maximum temperature of

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923 K. The $m/e = 4$ (D_2) partial pressure–time profile, is concurrently measured and recorded using an SRS RGA-100 residual gas analyzer system. Integration of this profile, when calibrated against a known D_2 leak rate, provides a measure of total retention if the entire D inventory is desorbed.

TDS spectra collected in experiments (1) and (2) were modeled using the TMAP7 hydrogen transport and trapping code [4,5], which numerically solves the one dimensional diffusion equation and accommodates thermal transport, surface recombination, and trapping, with conditions of flux equilibria applied at boundaries. In the modelling of TDS data, all TMAP input associated with D in Be transport was taken from the literature ([6] and Refs. therein) and a Debye frequency (Be) of $4 \times 10^{12}\ \mathrm{s}^{-1}$ used based on the assumption that trapped D is in equilibrium with the Be lattice. The total trap concentration was determined from the overall retention (inferred from TDS data), and in simulation, only trap activation energies and relative trap concentrations were adjusted so as to yield optimal agreement with experiment. Based on the manner by which co-deposits were prepared, it was assumed that the trap distribution and D profile in the layer are initially homogenous, and that the nature of retained D in highly saturated Be–D layers is consistent with the physics of atomic de-trapping and capture (i.e. no D in bubbles or hydrides). In TMAP simulation, traps are immobile and trap concentration (not occupancy) does not vary.

3. Results

Fig. 1 shows D_2 thermal desorption data (symbols) from a single Be–D co-deposit sphere as well as the output from four different TMAP models (lines labeled i–iv). The experimental data shows thermal release features consistent with two low temperature release D trapping states, (labeled α and β with a higher temperature 'tail' (labeled γ) that is reminiscent of second order release (i.e. D concentration dependent). The TMAP output (i, ii, iv) pertain to a single layer Be(1 μm) model with two trapping states, of activation energy 0.80 eV and 0.98 eV, and total trap fraction of ~0.1 consistent with TDS determined D retention (D/Be = 0.1) on full desorption. Curve (i) shows thermal release computed from a Sievert's law equilibrium applied at the release surface. The result simulates experiment well around peaks α and β, where trap release/capture is rate limiting, but does not duplicate the initial or final (γ) stages of experiment. The TMAP output (ii), where the surface atomic to molecular recombination rate was explicitly specified ([6] and Refs. therein), reproduces experiment better in comparison with (i) and, as such, was retained through the rest of this work.

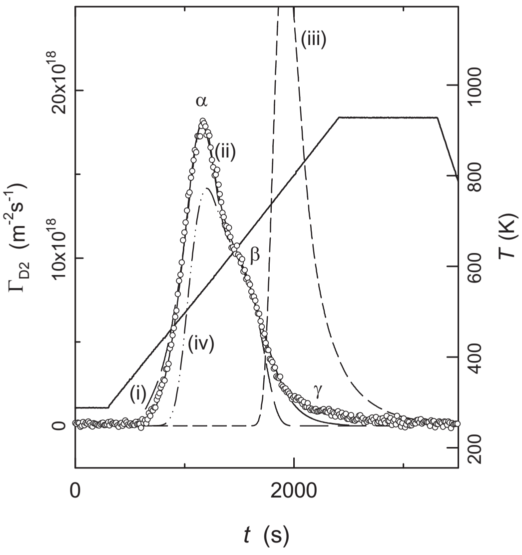

Fig. 1. D_2 thermal desorption data (symbols) from a single Be–D co-deposit sphere as well as the output from four different TMAP7 models (lines labeled i–iv). The full line shows the temperature of the sphere during desorption. Refer to text for description.

Other potential influences on the desorption profile, such as surface oxide formation and the W deposition substrate, were explored in more complicated multilayer TMAP models, but rejected in favor of the single Be layer model. In examining substrate, the computed output from a multilayer Be(1 μm)–W(1 mm) model (W properties taken from validation 2(d) in [5,7]) was found to be almost redundant with the single layer model. Yet an oxidized surface multilayer model BeO(10 nm)–Be(1 μm) (BeO properties taken from validation 2(b) in [5,8]) did not at all match experimental desorption data as shown by curve (iii) in Fig. 1, which depicts desorption though a 10 nm thick BeO surface layer. The remaining curve, (iv) in Fig. 1, is a simulation using the model of (ii), but following a flash heat event to 1123 K at a rate of $\sim 10^{6}\ \mathrm{K\,s}^{-1}$ with hold at 1123 K for 10 ms. Although 1123 K is well beyond the release temperature for peaks α and β, simulation suggests that only a minimal amount is desorbed from the lower temperature release peak.

Fig. 2, shows experimental desorption data for Be–D co-deposited spheres desorbed at different thermal ramp rates of 0.1, 0.3 and $1.0\ \mathrm{K\,s}^{-1}$. Here the consistency of the properties of individual spheres in the batch, and the validity of assumptions made about the co-deposited layer are evident by good agreement with the same single Be(1 μm) layer TMAP model depicted as curve (ii) in Fig. 1, but with different heating rates provided in the computation.

The effectiveness of fixed temperature bake-out on Be–D co-deposits was evaluated by thermally desorbing a selection of individual spheres for varying lengths of bake-out time. Each sphere was heated ($0.3\ \mathrm{K\,s}^{-1}$) to a specific bake-out temperature (513 K or 623 K), held at this temperature for a set length of time ($<10^{-2}$, 1, 5 or 25 h), then further heated (at $0.3\ \mathrm{K\,s}^{-1}$ to 923 K) to desorb any remaining D inventory. An example of such data is shown in Fig. 3 for two separately baked Be–D co-deposited spheres held at 513 K for bakes times, t_b, of 1 h and 5 h respectively. For clarity, the full desorption profile is only shown for the 1 h baked sphere, and only the remaining inventory desorption

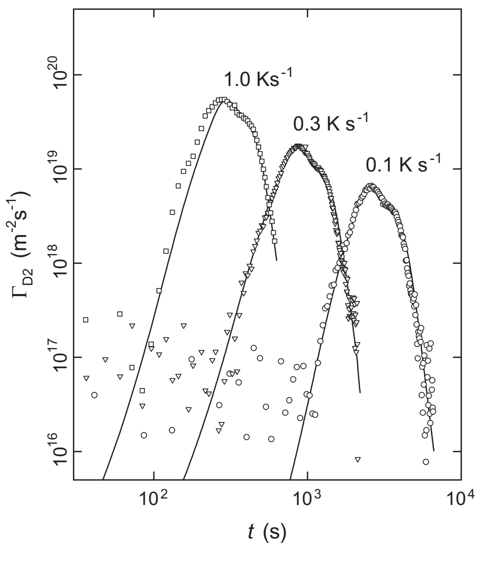

Fig. 2. TDS data for Be co-deposited spheres desorbed at 0.1, 0.3 and $1.0\ \mathrm{K\,s}^{-1}$. The lines are TMAP7 simulations (as in Fig. 1 (ii)) with corresponding rates of heating.

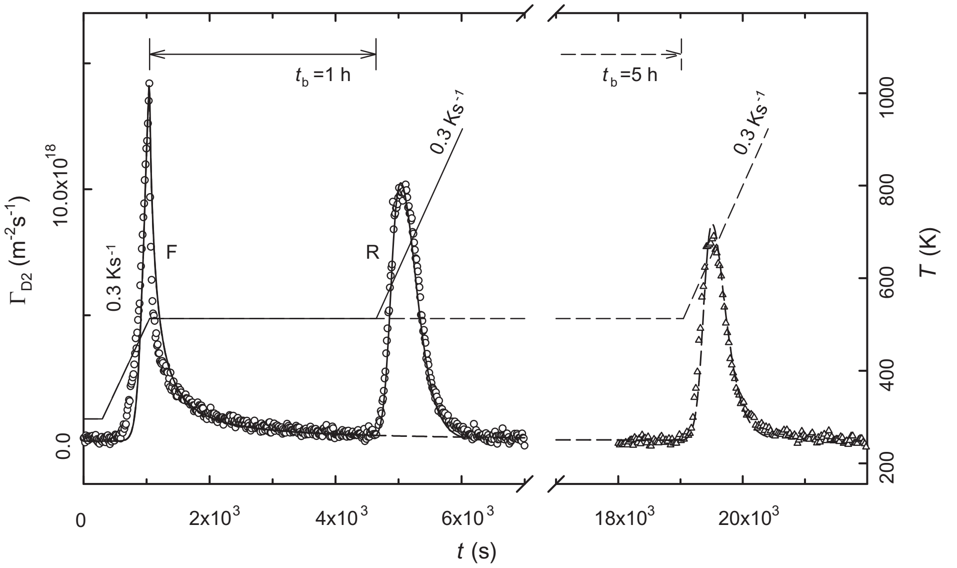

Fig. 3. D_2 thermal desorption data (symbols) for two separately baked Be–D co-deposit spheres held at 513 K for 1 h and 5 h respectively. For clarity, the full profile is given only for the 1 h case; the 5 h case shows only the remaining inventory desorption segment. The lines through the data are TMAP output from the Be(1 μm) single layer model. As in Fig. 1, the temperature profile is also indicated.

segment shown for the 5 h case as the initial features are identical in both. The lines through the data are TMAP output from the Be(1 μm) single layer model including an adjustment to the Arrhenius form of the surface recombination rate k_r, by a factor, $[1 + \exp(c_D/A)]$, where c_D is the dynamically computed D concentration in the near surface, and A is an adjustable constant. This was necessary to allow TMAP simulations to better follow the sharp fall (F in Fig. 3) and rise (R in Fig. 3) in desorption on entering and leaving the fixed temperature bake segments, and is a simplified version of the approach used for modelling D permeation in Be by Longhurst et al. [9]. While largely redundant with simulation (ii) in Fig. 1, the necessity of this modification in specific circumstances, suggests that k_r may depend on surface D concentration in highly saturated Be.

Fig. 4 shows further analysis of the kind of data shown in Fig. 3. The remaining D trapped in the co-deposited layer $\Theta_D(t_b)$, normalized to the initial D inventory $\Theta_D(t_b = 0)$, is plotted against bake time. Both bake-out regimes of 513 K and 623 K are shown and symbols are the remaining inventory in experimentally desorbed Be–D co-deposited spheres. The full lines, which depict the trapped inventory according to the single Be(1 μm) layer TMAP model agree reasonably well with experiment. The dashed lines are similar TMAP output for various thicknesses of the Be-co-deposited layer (0.2, 5 and 10 μm) and suggest that thicker co-deposited layers require increasingly longer time to desorb comparable amounts.

The efficacy of transient thermal loading in reducing co-deposit inventory is explored in Fig. 5 for a further batch of

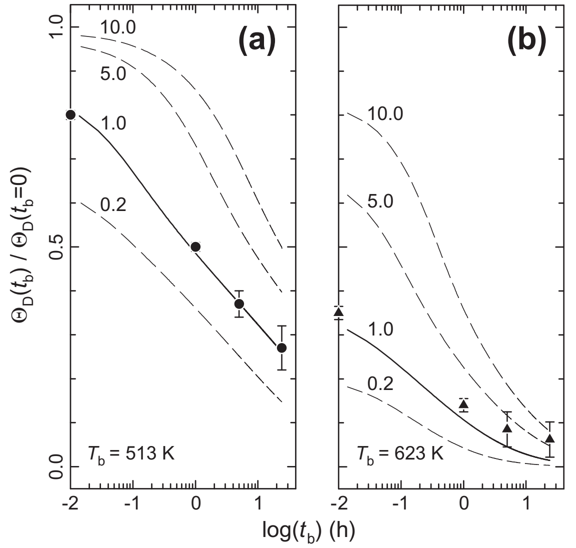

Fig. 4. Plot of remaining D trapped in the co-deposited layer $\Theta_D(t_b)$ normalized to the initial D inventory $\Theta_D(t_b = 0)$, versus bake time for (a) bake-out at 513 K and (b) 623 K. The solid line shows is the single Be(1 μm) layer TMAP model. Dashed lines are similar TMAP output but for different thicknesses of the Be-co-deposited layer of 0.2, 1 and 10 μm.

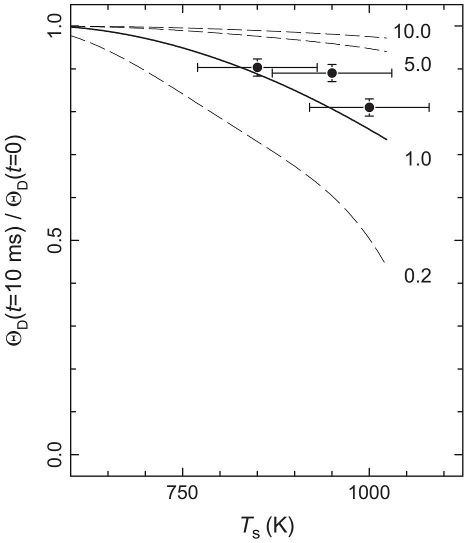

Fig. 5. Plot of remaining D trapped in the co-deposited layer $\Theta_D(t = 10\text{ ms})$ normalized to the initial D inventory $\Theta_D(t = 0)$, versus co-deposit layer temperature achieved in a thermal transient load. The solid line shows is the single Be(1 μm) layer TMAP model. Dashed lines are similar TMAP output but for different thicknesses of the Be-co-deposited layer of 0.2, 1 and 10 μm.

Be–D co-deposited layers formed at the more ITER relevant temperature of ~500 K. As with Fig. 4, normalized remaining D inventory in the co-deposit is plotted against the temperature achieved by the layer during a laser pulse transient lasting 10 ms. Data points are experimentally determined values, while the full line is the trapped D inventory in the layer following the transient according to the single Be(1 μm) layer TMAP model[2]. Again, TMAP simulation agrees reasonably well with experiment. The dashed lines show additional TMAP output for other thicknesses of Be-co-deposited layer (0.2, 5 and 10 μm) and, as with Fig. 4, suggest that thick co-deposits will not efficiently desorb at peak temperatures below 1000 K on the ms time scale. The details of this work are reported by Yu et al. [10].

4. Discussion and conclusion

In a recent article by Reinelt et al. [11], as many as six bound states of deuterium were noted from thermal desorption of Be implanted to saturated hydrogen concentration levels by 1 keV D^+ bombardment. Predominate release peaks and two main regions of thermal release were noted: (I) low temperature ~400–500 K desorption from two binding states when the Be lattice is saturated, and (II) higher temperature desorption ~600–950 K from three bound states associated with implantation induced damage. In the current study, co-deposits are formed under conditions of low ion impact energy, and the layer is highly saturated (D/Be = 0.1). Although the formation temperature (323 K) for experiment (1) is slightly lower than might be expected in an ITER regime, thick (>1 μm) saturated co-deposits are expected in ITER, and correspondingly, a significant level of thermal release is likely to proceed in a low temperature manner similar to that of Fig. 1 and other studies [1,11]. In experiment (1), the low temperature release region is well described by TMAP simulation as due to release from two traps of activation energies, 0.80 eV and 0.98 eV, similar to that reported in [11]. In experiment (2), TMAP simulation (not shown) required the implementation of an addition 2.0 eV trap, also in agreement with [11], and likely due to the higher temperature during deposition [10]. As with other studies [11,12] release is found to be predominately trap limited, but TMAP simulation is optimal when surface recombination is also taken into account. It is also interesting to note that TMAP models did not require the implementation of BeO surface layers in order to match experiment; a result that agrees also with experimental work in Ref. [11] where clean and oxidized surfaces show similar thermal release.

In Figs. 2 and 3, the effectiveness of TMAP in modelling experimental results is shown: Here the basic model of Fig. 1 (ii) is adjusted with thermal histories that reflect variations in experiment, and shows very reasonable agreement to experiment for varied heating rates in the range 0.0(bake)–1.0 K s^{-1}, lasting time scales of minutes to tens of hours desorption time. In Fig. 4, desorption of 1 μm thick co-deposited Be–D layers suggest that up to 50% and 30% extra D can be degassed over a period of ~1 day at fixed temperatures of 513 K and 623 K. Yet, although it remains to be verified by experiment, TMAP predictions (dashed lines in Fig. 4) point to significantly longer bake-out times as necessary for layers of increased thickness. A similar result is also evident with thermal transient studies, as depicted in Fig. 5. Experiment shows that D desorption from a 1 μm thick Be–D co-deposit, arising from flash heating up to 1000 K for 10 ms, is only marginally useful at removal of trapped D inventory, and that again, co-deposit thickness is a critical parameter according to TMAP simulation. Simulation suggests that flash desorption of increasingly thicker co-deposited layers will be increasingly ineffective. Thus, in view of all of the results, ITER strategies that avoid thick co-deposit build up, coupled with as frequent as permitted periods of bake-out, ought to lead to better efficacy in T inventory control.

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