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** See Appendix 1*

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Abstract

Misleading results can arise from using the DIFFUSE computer code to model hydrogen uptake and release by metals. This is due to the fact that it explicitly uses the Baskes model for the hydrogen recombination coefficient. These points are illustrated using recent results on the release of deuterium implanted in beryllium as an example. These data are reinterpreted using the PERI computer code, which is less model dependent than DIFFUSE. The new value for the pumping parameter $D/(2k_r)$ (diffusion coefficient over recombination coefficient) is much more in keeping with the values obtained in other experimental investigations.

1. Introduction

Using Be as a plasma facing material in the JET tokamak combines the advantages of metal walls (fast pumping of hydrogen and therefore improved density control) together with the advantages of low Z materials (lower radiation losses and reduced plasma dilution) [1]. The wall pumping effect is due to potential energy barriers at the metal surface and is increased by thin impurity layers which impede the release of hydrogen by recombination but have little or no effect on the uptake of predissociated hydrogen (D^0 & D^+) [2]. Computer codes, such as PERI [3] and DIFFUSE [4], have been used in the past to model laboratory simulations of D^+ pumping by Be [5,6].

Two levels of theory exist when considering hydrogen transport models. On one level, equations for the hydrogen flux are written out in terms of the hydrogen densities (eg. pressure, ion flux, bulk concentration in the metal) and transport parameters (diffusivity, solubility, recombination coefficient). On a second, more detailed level, the individual processes which make up recombination are considered so as to give a value for the recombination coefficient in terms of fundamental constants (eg. heat of solution). In section 2 below, the models and "level of theory" incorporated in each of the DIFFUSE and PERI computer codes are reviewed.

Next (section 3), the phenomenon of deuterium pumping by a plasma-facing beryllium surface is examined. Specifically, we consider recent experimental measurements by Hsu et al [6] on the evolution of gas upon terminating a deuterium plasma discharge between Be disks. These authors used DIFFUSE to arrive at a value of the pumping parameter $D/(2k_r)$ for this system (D = diffusion coefficient, k_r = recombination coefficient). Note that the $2k_r$ used here is the same as the K_r employed by the users of DIFFUSE, and the same as the $2\sigma k_r$ used elsewhere [3,7]. In this paper, PERI is used to reinterpret the same data. We therefore do not question the measurements of Hsu et al, only their interpretation.

2. The Physical Models Behind PERI and DIFFUSE

On the non-detailed level of theory, hydrogen transport in a metal is given by the particle diffusion equation

$$\frac{\partial c(x,t)}{\partial t} = D \frac{\partial^2 c(x,t)}{\partial x^2} \quad (1)$$

where x is the distance into the material from the surface, t is time and c is the hydrogen concentration. This is coupled with recombinative release (i.e. quadratic) boundary conditions of the form

$$\phi_{in}(t) = 2k_r c^2(0,t) - D \frac{\partial c(x,t)}{\partial x} \Big|_{x=0} \quad (2)$$

where the vacuum/solid interface occurs at $x = 0$ in this example. ϕ_{in} is the incoming deuterium flux which penetrates the surface (ions & atoms tend to penetrate or stick with near unity probability while molecules tend to give a negligible contribution unless surfaces have been rigorously cleaned).

The recombination coefficient k_r is a phenomenological rate constant and is not meant to represent a single atomic process. Equations 1 & 2 are solved numerically in both PERI and DIFFUSE, therefore both codes adopt the phenomenological model where the release of gas can be written as $2k_r c^2$.

As far as hydrogen release models are concerned, however, this is as far as PERI goes. The value of k_r in PERI is fully adjustable. It can therefore be used without excluding any of the various models for the value of k_r . The DIFFUSE code, however, imposes a constraint on the allowable values of k_r by incorporating a model by Baskes [8]. The predictions of this and other models for the value of k_r are summarized in Table 1.

Experimentally determined values of k_r [11] show large variability, both in magnitude and activation energy. This is thought to be due to differences in the surface cleanliness among the various investigations. It is clear that a model for k_r which predicts a unique value of the apparent activation energy for recombination can only describe a select portion of the existing database. For this reason, PERI is more flexible since it allows any choice of k_r , while DIFFUSE is incompatible with the known behavior of k_r .

3. Release of Deuterium Implanted in Beryllium

We will now focus on recent experimental results by Hsu et al on the thermal release of deuterium gas following the exposure of Be disks to a deuterium plasma. This situation can be modelled as a semi-infinite wall of beryllium, meaning that for the periods of time t considered, the physical extent of the diffusion ($\sim \sqrt{Dt}$) is small compared to the material thickness. The external deuterium flux is of the form

$$\phi_{in}(t) = \begin{cases} \phi_d & \text{for } 0 < t < t_1 \\ 0 & \text{for } t > t_1 \end{cases} \quad (3)$$

and the initial condition is taken to be

$$c(x,0) = 0. \quad (4)$$

In order to deduce a value of $D/(2k_r)$ from the quantity of released gas and the time dependence of the release rate, we must first deduce the total amount of deuterium pumped (inventory) for a given value of the pumping parameter, and then calculate what fraction of this gas is released over a finite measurement time.

3.1. Relationship between Inventory and Pumping Parameter

When ϕ_{in} is turned on, the bulk concentration just below the surface, $c(0,t)$, gradually increases to a level where the recombining flux, $2k_r c^2(0,t)$ is almost equal to the incoming flux, ϕ_d . The difference between the two is the flux of deuterium diffusing into the material. The time at which the recombining flux reaches half the incoming flux is labelled τ_p (the characteristic time for recycling) and is given by [3]

$$\tau_p = \frac{D}{2k_r \phi_d} \quad (5)$$

For small bombardment times t_1 , the hydrogen inventory at the end of bombardment is approximately equal to the total incident fluence $\phi_d t_1$ since the recombining flux is always small.

For large bombardment times, $c(0,t)$ builds up to a quasi-steady value $\sim \sqrt{\phi_d/(2k_r)}$. Approximating this situation by a step increase in $c(0,t)$, from 0 to $\sqrt{\phi_d/(2k_r)}$, the diffusion equation has an exact analytical solution [12] and the resulting inventory is given by

$$\begin{aligned} \text{inventory} &= \frac{2}{\sqrt{\pi}} c(0,t) \sqrt{Dt} && \text{for } t_1 \gg \tau_p \\ &= \frac{2}{\sqrt{\pi}} \sqrt{\frac{D}{2k_r}} \sqrt{\phi_d t_1} \end{aligned} \quad (6)$$

The relationship between inventory, fluence and pumping parameter is illustrated in Fig.1. Note that in Fig.1, both the inventory and pumping parameter are normalized by the total fluence so that from eqn.(5),

$$\frac{D/(2k_r)}{\text{fluence}} = \frac{D/(2k_r)}{\phi_d t_1} = \frac{\tau_p}{t_1} \quad (7)$$

3.2. Relationship Between the Quantity of Released Gas and the Inventory

Following bombardment, the retained deuterium will be released from the material, and should all be recovered given sufficient time. For the case where bombardment continues for a long period, ($t_1 \gg \tau_p$), the time needed to release most of the particles is of the same order as the bombardment time. For the case where $t_1 \ll \tau_p$, however, the time needed to release the majority of the retained D can be many times greater than t_1 . To be convinced of this, recall that for this case, $k_r c^2(0,t)$ is always $\ll \phi_d$, therefore the total amount released over a period t_1 , $\int_0^{t_1} k_r c^2(0,t) dt \ll \int_0^{t_1} \phi_d dt \sim$ amount retained. Therefore for a release integrated over a time of order t_1 , the fraction of the deuterium recovered from the walls goes from nearly unity for $\tau_p \ll t_1$, i.e. small $D/(2k_r)$, to a value $\ll 1$ as τ_p becomes $\gg t_1$,

i.e. large $D/(2k_r)$. This is illustrated in a plot of fractional deuterium recovery calculated using PERI (Fig.2).

3.3 Calculation of the Pumping Parameter using PERI

By multiplying the quantities in Figs. 1 and 2, one obtains the integrated release as a function of $D/(2k_r)$ (Fig.3). By measuring the amount of gas released over a limited time, two possible values of $D/(2k_r)$ may be inferred. Whether one of the two may be discarded on the basis of being unphysical, or by the time dependence of the release, remains to be seen. The two cases, small and large $D/(2k_r)$, are discussed below and values of $D/(2k_r)$ are estimated from the results of Hsu et al [6].

i) Small $D/(2k_r)$

For the smaller $D/(2k_r)$ solution, the amount of released gas is approximately equal to the inventory given by eqn.6. Using the conditions of Hsu et al's experiment as input parameters, it was estimated from PERI that approximately half the total inventory would be released during the interval measured (see Fig.2). Fig.4 shows the values of $D/(2k_r)$ thus calculated, together with other values of $D/(2k_r)$ for deuterium/beryllium taken from the literature [5,13,14]. Interestingly, the $D/(2k_r)$ evaluated here is very different in temperature dependence ($<0.1\text{eV}$ activation energy as compared to 1eV) than that determined by Hsu et al using the DIFFUSE computer code to model the same data. This is because DIFFUSE is limited to a choice of k_r as given by the Baskes model, while here k_r has been free to assume any value (specifically any activation energy). This translates into a much better fit to the released gas versus temperature data than previously achieved (see Fig.5).

The $D/(2k_r)$ used by Hsu et al as an input parameter for their DIFFUSE code simulation has a very high activation energy (1eV), resulting in a steep slope at high temperatures, where recombination is slow and hence rate-limiting. At low temperatures, recombination is relatively fast and diffusion from the implantation zone (of the order of 10nm depth) to the surface controls the release. This mechanism is not incorporated into PERI, which assumes the deuterium concentration just below the surface is about the same as in the implantation zone. It is doubtful, however, that this mechanism is controlling the release over the entire temperature range in the experiments of Hsu et al. As those authors have noted, this would result in a temperature dependence at odds with experiment.

The spread between Hsu et al's $D/(2k_r)$ (for our small $D/(2k_r)$ interpretation), the results for Be-coated stainless steel surfaces [5] and for the JET walls and limiters [14] is now reduced to 1-2 orders of magnitude (Fig.4). Given the fact that many orders of magnitude can separate $D/(2k_r)$ values [15], these results, done under different experimental conditions, are very much in line with one another.

ii) Large $D/(2k_r)$

Alternatively, one could assume that k_r is very much smaller, meaning large $D/(2k_r)$ and large τ_p . The slow rate of recombination means that i) virtually all the ions and atoms incident on the beryllium contribute to the total inventory, and

ii) that the amount released following bombardment is negligible compared to the inventory for release times of the same order as t_1 . Using the PERI code, it was found that with a value of $D/(2k_r) \sim 10^5 \times$ greater than in the previous section, an equally good fit to the data was possible. Fig.6 shows how the time evolution of release as given by the PERI code compared with the experimental results of Hsu et al at 700K. The values of $D/(2k_r)$ that could explain Hsu et al's data for the large $D/(2k_r)$ scenario are shown in Fig.4.

One obvious way to check if this large $D/(2k_r)$ is unphysical is to calculate the bulk deuterium concentration in the material: it must be less than the metal atom density. For the value of $D/(2k_r)$ at 700K of $6.8 \times 10^{20} \text{cm}^{-2}$, and a diffusivity as given by Jones et al [16], $2k_r$ is $1.8 \times 10^{-30} \text{cm}^4/\text{sec}$. The maximum release rate of $\sim 10^{15} \text{cm}^{-2} \text{s}^{-1}$ (see Fig.6) gives

$$c_{\max} = \sqrt{\frac{\phi_{\max}}{2k_r}} = 7.2 \times 10^{21} \text{cm}^{-3} \sim 0.07 \text{ atoms/Be}$$

which is getting very close to the maximum possible concentration. However, this should be taken as an upper limit since more recent measurements of D [17] suggest a value $\sim \times 10$ higher than in reference [16]. At the moment, there does not seem to be enough evidence to conclusively rule out the possibility of the high $D/(2k_r)$ solution. More experimental work is needed on the deuterium/beryllium system.

4. Updating DIFFUSE

The continued use of the DIFFUSE code in its present form is not in keeping with research on the nature of k_r conducted over the last 10 years. Recent experimental measurements (notably that of Livshits [10] and Wampler [18]) and theoretical work [19] have underlined the inadequacy of the Baskes model.

Although Hsu et al [6] state that they only determine the necessary k_r to explain their data without relating it to the Baskes model, their conclusion that $D/(2k_r)$ must rise sharply with temperature is purely a result of using $k_r = k_{r0} \exp(\frac{+E}{kT})$, with $E = 1\text{eV}$ in their computations. Clearly Hsu et al do not do their data justice since a choice of $E = 0.08\text{eV}$ instead of 1eV (together with their choice of the activation energy for diffusion $E_d = 0$) would have given a much better fit to the data (compare the DIFFUSE and PERI calculations in Fig.5). If one uses the heat of solution deduced by Swansiger, $E_s = 1.0\text{eV}$ [20], with the model of Pick and Sonnenberg [2] (see Table 1), a value for the surface potential barrier $E_c = E_d + 0.96\text{eV/atom}$ would be required to explain an apparent activation energy for $D/(2k_r)$, $E_d + E = 0.08\text{eV}$ (see Fig.4). However, with the Baskes model and the same value of E_s , one is left with $E_d + E = 1.0\text{eV}$. The value of $E = 1.0\text{eV}$, therefore, does not arise from the experimental data, but from the Baskes model and Hsu et al's choice of $E_d = 0$.

Even though DIFFUSE can give misleading results, we do not suggest it be abandoned in favour of, say, PERI. DIFFUSE contains many useful features: it considers the effects of implantation, deep traps for hydrogen, tritium decay and multilayer materials. To resolve the shortcomings of DIFFUSE, one must

simply allow the energy E in the expression for k_r (see Table 1) to become an adjustable parameter with a maximum value of $2E$, [2].

Conclusion

The usefulness of the DIFFUSE computer code in modelling hydrogen transport is severely limited since it explicitly incorporates the Baskes model for the recombination coefficient. This has been illustrated by reinterpreting recent experimental data by Hsu et al [6] using the PERI code, which allows any choice of the recombination coefficient. Modelling the temperature dependence of the quantity of gas released was improved over the original interpretation. The new value for the pumping parameter $D/(2k_r)$ (diffusion coefficient over recombination coefficient) is much more in keeping with the values obtained in other experimental investigations. Improving the DIFFUSE code would be a simple matter of allowing the activation energy for recombination to be an adjustable parameter.

In deducing a value of the pumping parameter $D/(2k_r)$ two possible solutions were found, each describing the observed behaviour equally well.

The lower value of $D/(2k_r)$ represents the case where the majority of the absorbed deuterium is released on a time scale comparable to the bombardment time. This value of $D/(2k_r)$ is much closer to that found in other experimental investigations than that deduced by Hsu et al from the same data. This is a result of relaxing the constraint on allowed values of $D/(2k_r)$ permitted by the Baskes model.

The higher value of the pumping parameter represents the case where essentially all the incident deuterium ions contribute to the inventory, yet only a small fraction is measured to be released. In experiments similar to those of Hsu et al, runs at different bombardment fluences would be needed to determine which of the two values of pumping parameter apply.

Acknowledgements

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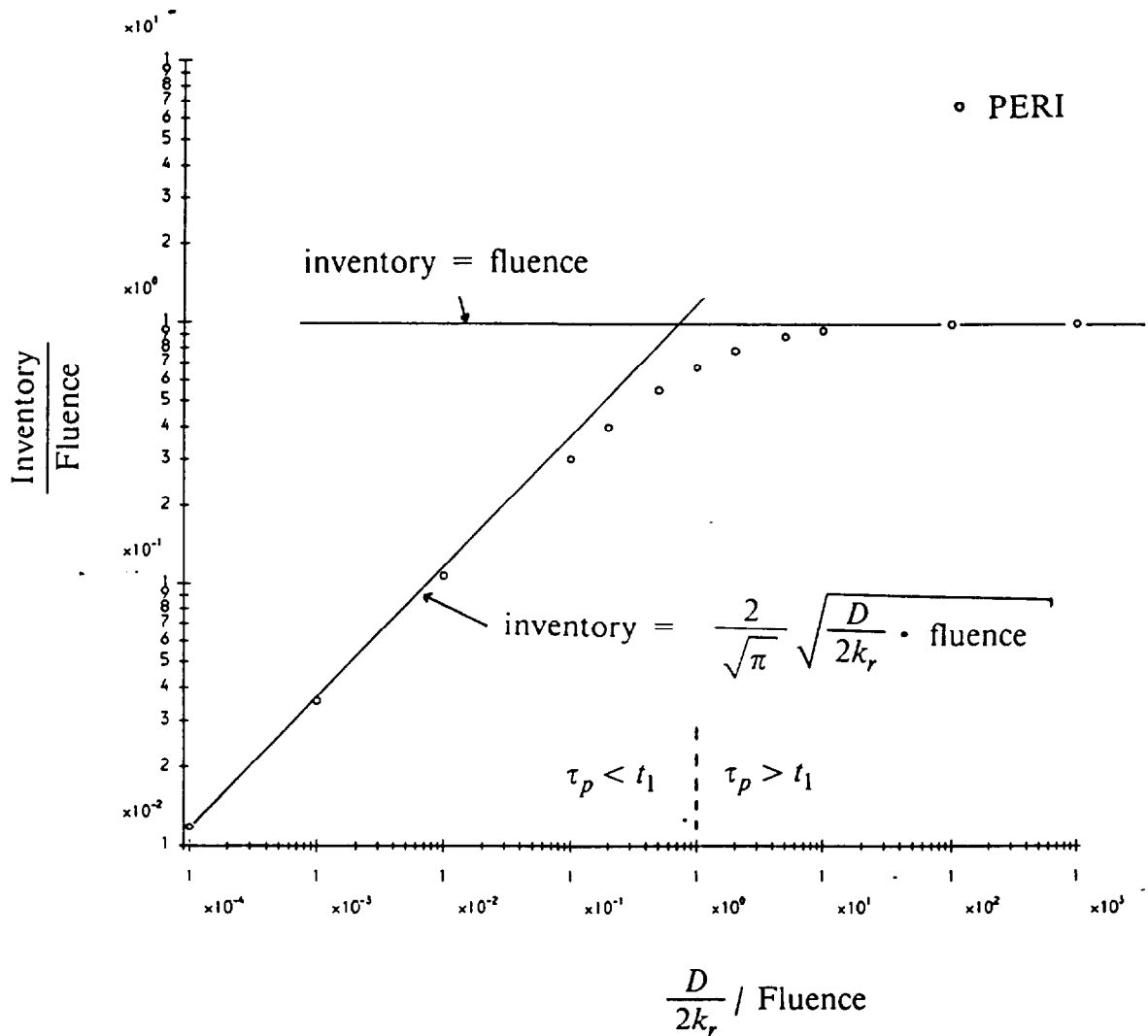
Table 1

Alternative Models for the Recombination Coefficient

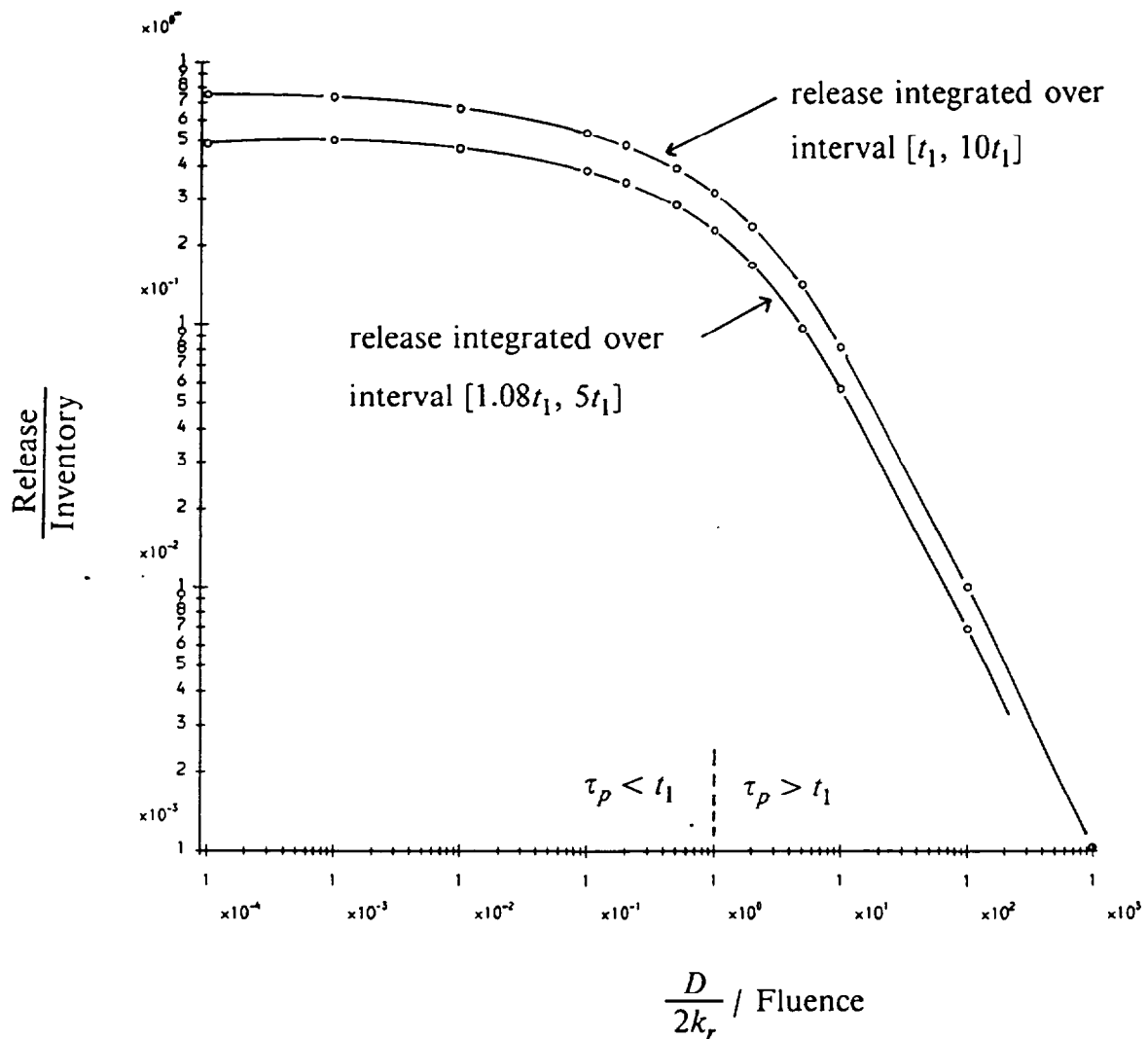
$$k_r = \frac{\alpha_0}{\sqrt{2\pi mkT} S_0^2} \exp\left(\frac{+E}{kT}\right)$$

Model	α_0	E	Underlying Mechanism
Baskes [8]	≤ 1	$E_s - E_d$ for $E_s + E_d > 0$ $2E_s$ for $E_s + E_d < 0$	release is controlled by a process where a hydrogen atom on the surface recombines with one from the bulk
Livshits [9] or Pick and Sonnenberg [2]	≤ 1	$2E_s - 2E_c$, $2E_s$ for clean surfaces	recombination is between two surface atoms, impeded by a potential energy barrier E_c at the surface resulting from contamination by impurities
blocking [10]	≤ 1	$2E_s$	the area available for recombination of surface atoms is reduced by surface impurities

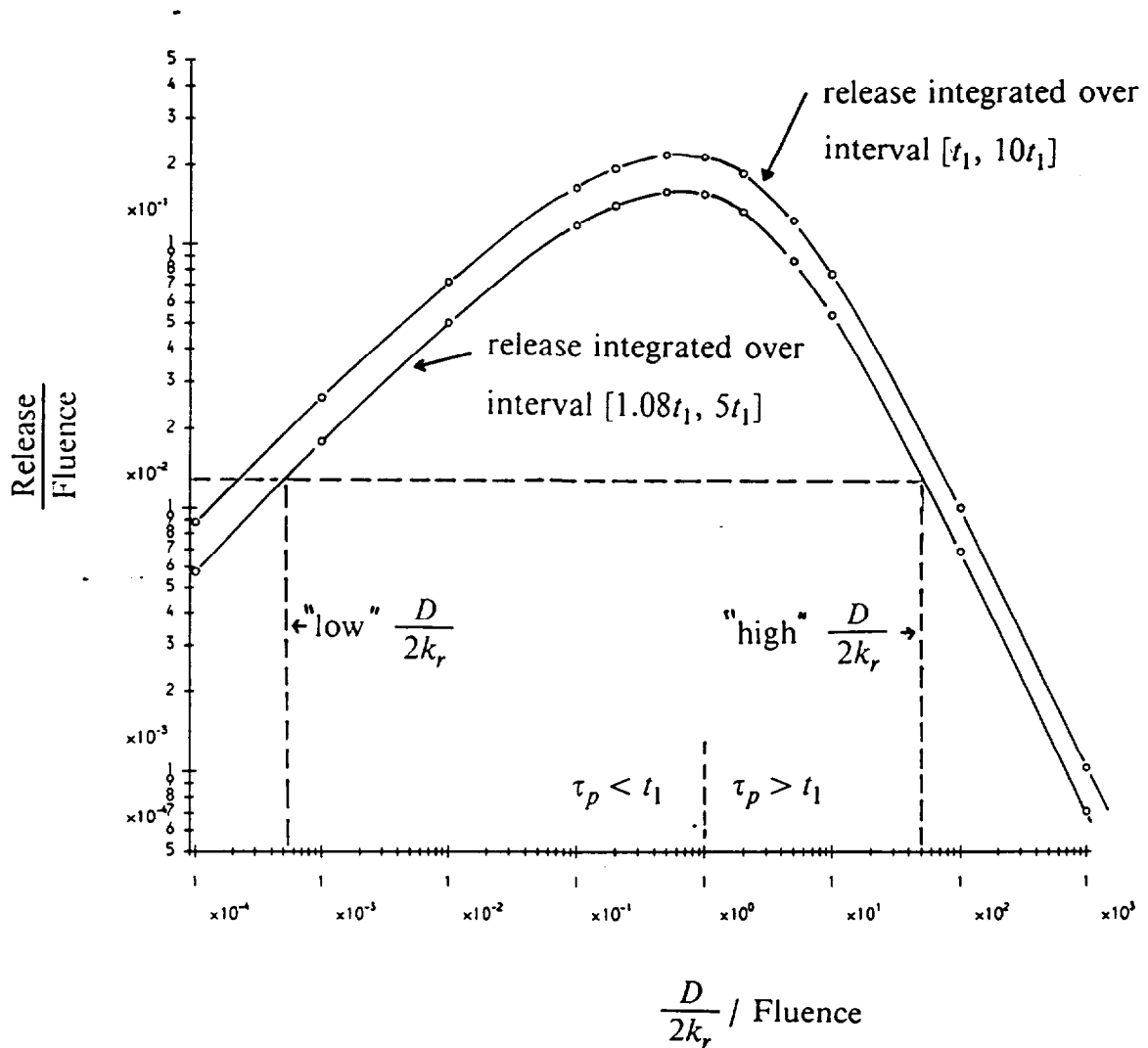
$S = S_0 \exp\left(\frac{-E_s}{kT}\right)$ = solubility, E_s = heat of solution, E_d = activation energy for diffusion



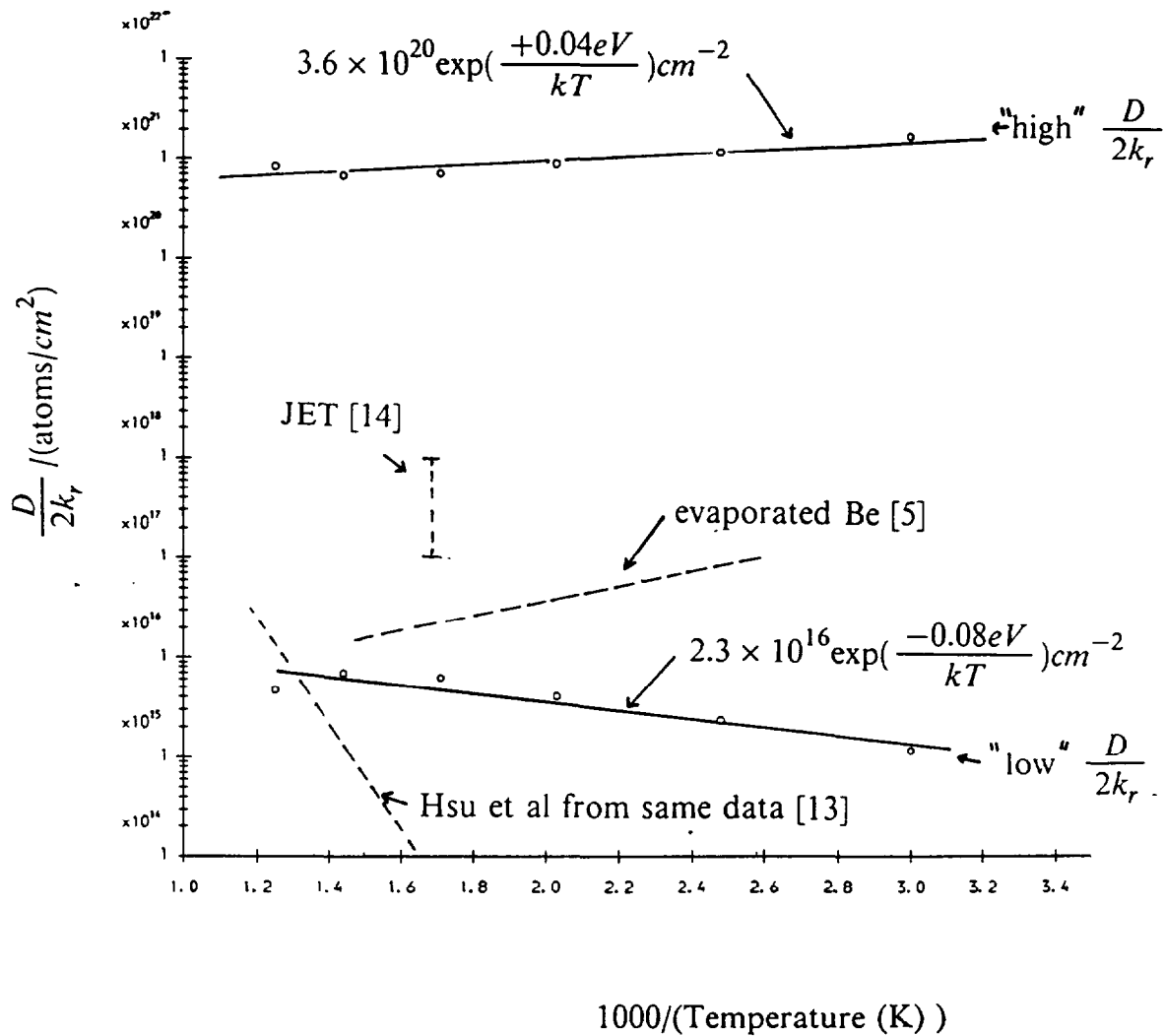
1. The total deuterium inventory as a function of the pumping parameter at the end of bombardment with a constant flux. Both the inventory and pumping parameter, $D/(2k_r)$, are here normalized by the total fluence, $\phi_d t_1$. Open circles are values calculated by PERI, while the solid lines represent analytical approximations to the extreme cases.



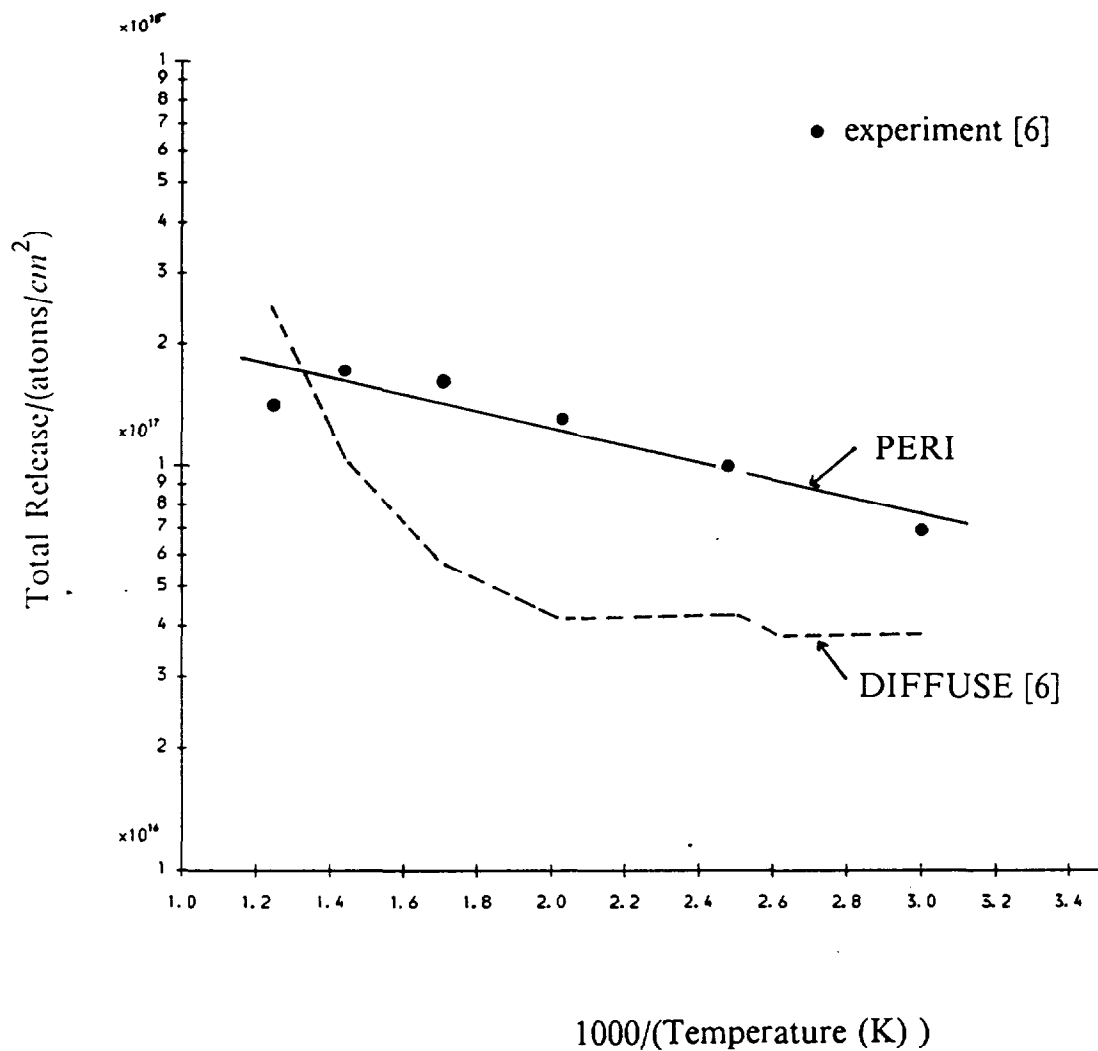
2. Fractional deuterium recovery following bombardment as a function of the pumping parameter (again normalized by the fluence) as calculated by PERI. The upper curve represents release integrated over a period 10x greater than the bombardment time (i.e. over the interval $[t_1, 11t_1]$). The lower curve represents release over a smaller interval $[1.08t_1, 5t_1]$ which is similar to that used in the experiments of Hsu et al.



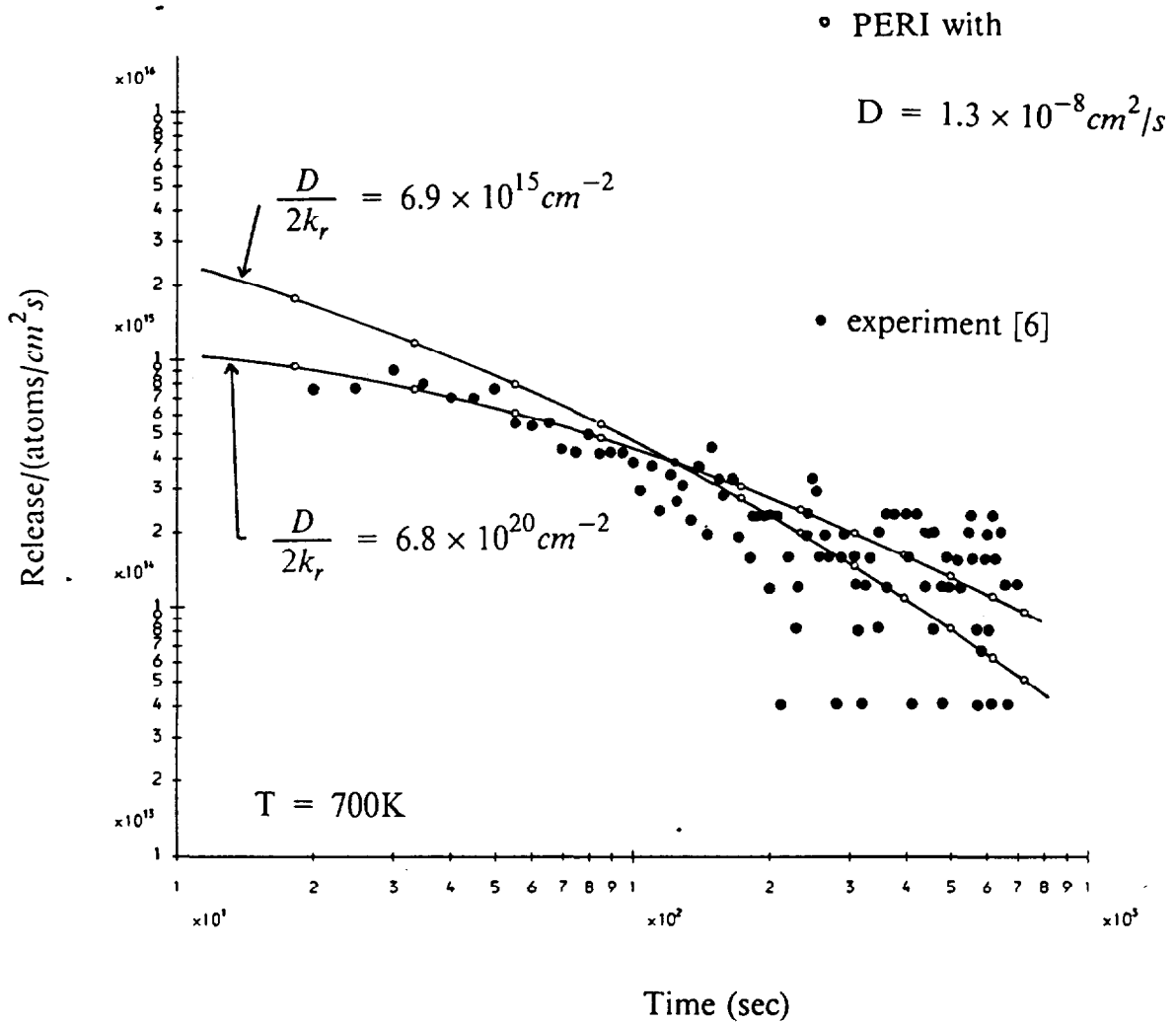
3. Deuterium recovered following bombardment at a constant flux as a function of the pumping parameter. Both quantities are normalized by the fluence. The intervals over which the release is integrated are as in Fig.2. Dashed lines show how a single value of total release deuterium implies one of two values of the pumping parameter.



4. Pumping parameter for deuterium/beryllium versus reciprocal temperature. Open circles represent values taken from the lower curve in Fig.3 given the release/fluence measurements of Hsu et al [6]. Solid lines represent least squares fits to the above, while dashed lines show several values taken from the literature.



5. The total release of deuterium as measured (•) and calculated (----) by Hsu et al [6]. Solid line is the result of the present interpretation using either of the least squares fits in Fig.4 together with the lower curve in Fig.3.



6. Release rate of deuterium following bombardment given by PERI (here, bombardment ceases at time = 0). Results are shown for 2 different values of $D/(2k_r)$, taken from Fig.4. Solid circles represent Hsu et al's data at 700K.

APPENDIX 1.

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