Tritium Containment in the Dust and Debris of Plasma-Facing Materials Produced During Operations

I. Konkashbaev and Ju. Grebenshikov
Troitsk Institute for Innovation and Fusion Research
Troitsk, Russia

A. Hassanein
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439 USA

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I. Konkashbaev[1], A. Hassanein[2], and Ju. Grebenshikov[1]

[1]Troitsk Institute for Innovation and Fusion Research, Troitsk, Russia
[2]Argonne National Laboratory, 9700 South Cass Ave., Argonne, IL 60439, USA

Tritium behavior in plasma-facing components of future tokamak reactors such as ITER is an essential factor in evaluating and choosing a successful candidate for a plasma-facing material (PFM). One important parameter that influence tritium build-up and release in the generated dust of PFMs is the effect of material porosity on tritium behavior. Diffusion in porous materials, for example, consists of three different diffusion processes: along grain boundaries, along micro-crystallites, and diffusion in pure structure crystallites. A model is developed to evaluate and assess the sensitivity of tritium accumulation and permeation of candidate materials due to porosity. Specific laboratory experiments relevant to reactor conditions, in currently existing and available facilities, are required to help in selecting the best candidate material.

1. INTRODUCTION

Carbon-based and beryllium materials are regarded as candidate materials for the divertor plate and component tools (for the closed divertor concept). It is well known that carbon materials can absorb rather large amounts of tritium. In this paper the problems of tritium inventory and release are more fundamentally discussed. These problems are important because of two main reasons. First, large tritium inventory would result in significant increase of tritium recycling. Second, a safety related issue, due to erosion both during normal operation such as physical and chemical sputtering and during abnormal events such as disruption, power transients, and edge-localized modes (ELMs) which cause vaporization, micro-cracking, and spallation. The redeposited material will have rather different physical properties than the original one due to both strong radiation damage and different structure.

Natural and pyrolytic graphites are anisotropic with very low porosity, but artificial graphites are more porous, with randomly oriented grains with size $L_g \approx 10$ μm which results in more isotropy. Such grains consist of small crystallites with size $L_c \approx 100$ Å as shown in Fig. 1 [1].

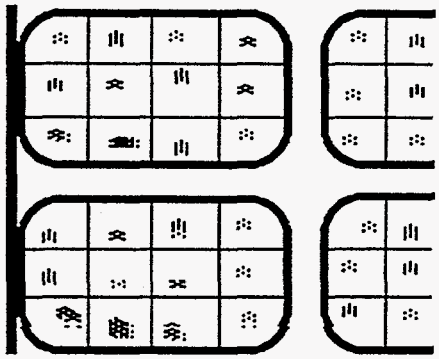

Figure 1. Schematic view of graphite structure.

When a graphite-based material is damaged by radiation (neutrons, energetic atoms, and ions), the crystallites become smaller in size resulting in a significant increase of total crystallites surface area. This has the effect of increasing the number of traps and the diffusivity would also be affected by the radiation. At least 3% of the total volume is occupied by micropores (about 0.5 nm in diameter) which would facilitate the diffusion of molecular hydrogen [2]. The study of physical properties of the redeposited carbon material has only just started. One can expect that such properties as porosity (open and closed) and diffusion coefficients in crystallites as well as in boundaries between

crystallites and between grains will be quite different from the original material properties. In reality, these redeposited layers should not be called graphites because they do not behave like graphite i.e., they are new materials with poor database.

The aim of this report is to discuss and critically assess one major area of concern regarding tritium diffusion and release in these newly-formed materials: the role of porosity.

As stated in Ref. [3], the porosity plays a more important role than previously thought. The porosity allows the tritium gas to enter rapidly into the graphite, effectively exposing all of the inner porosity to an equal gas pressure. Moreover, some models assume each of the grains to be surrounded by the gas which gives results more closely to the experimental data.

2. ANALYSIS

There are number of models describing tritium transport (see, for example, Ref. [4]). Similar problems for fission reactor materials are studied in more details. As it follows from these investigations, most gas between grains exists in the form of gas bubbles. According to current existing opinion, bubbles form initially on the grain junctions due to radiation (Fig. 2a). After that bubbles form on ribs. Above a certain critical volume of these bubbles, ξ_{crit}, the bubbles collapse and connect with each other resulting in percolation clusters, i.e., an open porosity structure (see Figs. 2b and 2c). The value of ξ_{crit} is about 5% according to experimental data [5] or up to 10% according to theory [6]. Above that volume $\xi > \xi_{crit}$, the open porosity is sustained and gas inventory is kept on this critical level. Microcracking due to surplus of bubble gas-pressure will also result in more opening of the porosity. The maximum size of the gas bubble is defined by surface tension and is in the order of $R_b \approx 1$ nm and each bubble contains ≈ 50 atoms/bubble. The total amount of gas is therefore proportional to bubbles surface area. As it follows from this consideration, the redeposited layer exposed to radiation consists of blocks with boundaries that contain open porosity. Inside these blocks, however, close porosity exists. Along the open pores diffusion is described by Knudsen flow (and at T≈1200 K):

$$D_p = \frac{2}{3} r_p \sqrt{\frac{8RT}{\pi\rho}} \approx 1 \text{ cm}^2/\text{s} .$$

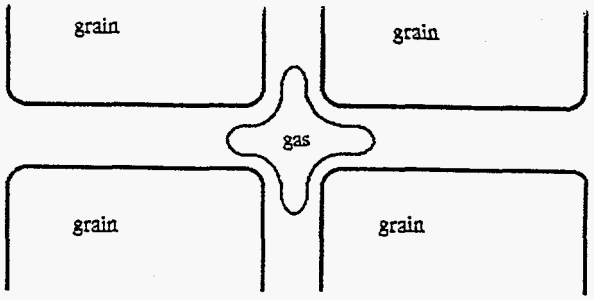

Figure 2a. Gas bubbles form on grain junctions.

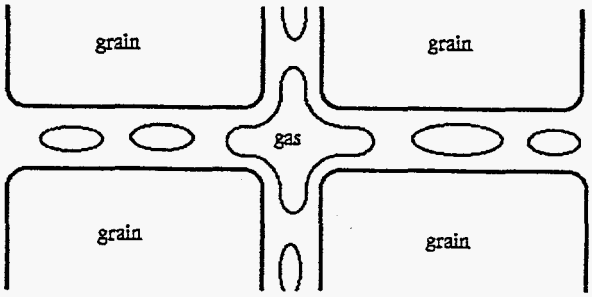

Figure 2b. Bubbles collapsing.

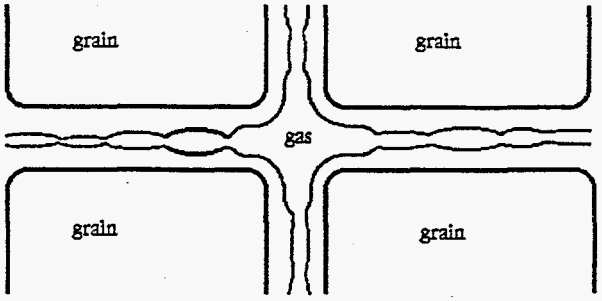

Figure 2c. Formation of open porosity structure.

The gas penetrates rapidly along the open pores. Diffusion is then many orders of magnitude higher than the usual diffusion coefficients in solid, i.e.,

$$D_{solid} \approx 10^{-12} \text{ cm}^2/\text{s} .$$

Therefore, it is reasonable to treat diffusion inside the blocks assuming a closed porosity structure. The total time required to fill the redeposited layer becomes less than that corresponding to the above mentioned results due to the fast gas filling [3].

Let us regard diffusion of tritium into a block with size l_b of grains. A schematic view of such a block is shown in Fig. 3. The block is partitioned into grains separated by boundary layers with width d_c and length l_c. Diffusion coefficients of crystallites D_0, grains boundary layers (pores) D_g, and crystallites boundary layers D_c can be represented by the formula

$$D_\alpha = A_\alpha \exp\left(-\frac{E_\alpha}{T}\right) \text{ cm}^2/\text{s} .$$

For pure crystal [7]:

$$A_0 = 0.93, \qquad E_0 = 2.8 \text{ eV} .$$

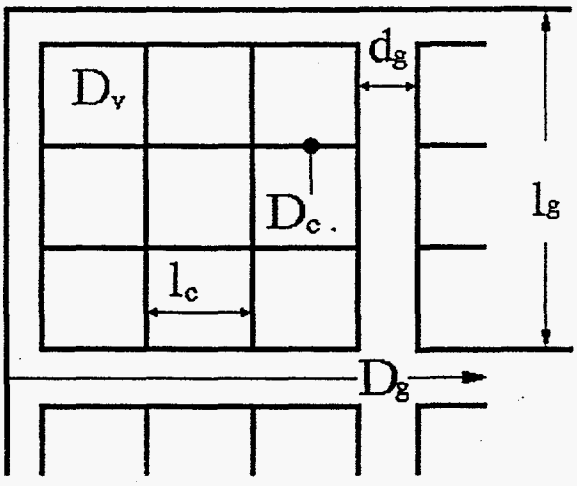

Figure 3. Numerical model of two-level structure.

For the pores, diffusion coefficients can be estimated by the fitting formula [8], where

$$A_g = 6.75, \qquad E_g = 0.7 \text{ eV} .$$

For $T \approx 1160$ K (0.1 eV)

$$D_0 \approx 6.4 \cdot 10^{-13} \text{ cm}^2/\text{s}, \; D_g \approx 6.2 \cdot 10^{-8} \text{ cm}^2/\text{s} .$$

The value of D_c is unknown. However, its value is an intermediate between D_0 and D_c. Let us therefore assume that

$$D_c = \sqrt{D_g D_0} \approx 2 \cdot 10^{-10} \text{ cm}^2/\text{s} .$$

According to these values one can estimate characteristic times of diffusion: τ_0 - into crystallites, τ_c - along crystallites boundary layers, τ_g - along grains boundary layers. These characteristic sizes are estimated to be

$$l_0 \approx 10^{-6} \text{ cm}, \; l_0 \approx 10^{-3} \text{ cm}, \; l_0 \approx 0.1 \text{ cm}$$

$$\tau_0 = \frac{l_0^2}{2D_0} \approx 1\text{s}, \; \tau_c = \frac{l_g^2}{2D_c} \approx 2.5 \cdot 10^4 \text{ s}, \text{ and}$$

$$\tau_g = \frac{l_b^2}{2D_g} \approx 10^5 \text{ s} .$$

Therefore, a hierarchy of processes takes place. For a large time, τ_g, gas diffuses along the grain boundaries and for an intermediate time, τ_c, gas diffuses along crystallites boundaries, and for a short time the gas diffuses into crystallites. These times are much shorter than the time of diffusion into a pure crystal with size l_b

$$\tau_g = \frac{l_b^2}{2D_0} \approx 10^{10} \text{ s} .$$

As one can expect that the redeposited layer has a different and loose structure and can be filled up to equilibrium concentration of tritium for a relatively short time, up to a few days. Actually, the process of gas filling has a more complicated nature because it is necessary to take into account the relation between volumes of grains boundary layers, crystallite boundary layers, and crystallites.

As shown in Fig. 1, the process of gas filling has sufficiently two (or three) dimension behavior. For illustration of such processes, the following simple numerical model was solved. Fig. 3 shows a limited number of grains separated into a limited number of crystallites. Because we are interested in a qualitative picture, different values of D_a, l_a, h_a are used.

The gas diffuses from the surface (y = 0) where a constant density, n_o, is kept. First, the gas diffuses along large channels (pores) in y-direction for distance y according to the classic formula

$$y^2 = 2D_g t$$

with gas flux:

$$S_y = D_g \frac{\partial n}{\partial y} \approx D_g \frac{n_0}{\sqrt{2D_g t}} .$$

For a long time one can consider the average losses along horizontal channels (x-direction) which can be estimated as

$$S_x = D_g \frac{\partial n(y;x)}{\partial x} \frac{2h_g}{L_g}$$

$$S_x \equiv D_g \frac{n_0}{0.5L_g} \frac{2h_g}{L_g} \approx D_g n_0 \frac{4h_g}{L_g^2} .$$

And S_x, approaches S_y after time t*

$$S_x = D_g n_0 \frac{4h_g}{L_g^2} > D_g \frac{n_0}{\sqrt{2D_g t}} ,$$

$$t^* > \frac{L_g^4}{32h_g^2 D_g} .$$

These losses along the x-direction decrease the diffusion flux along the y-direction. Only after filling up the horizontal channels, gas can diffuses forward.

Similar situation exists for diffusion along crystallites boundary layers, but additionally it is necessary to take into account diffusion into crystallites with trapping and detrapping. Therefore, a system of equations qualitatively describing diffusion into a porous system with certain trap density and distribution is solved [9]. Qualitative results of such modelling indicated that three distinctive regimes were seen with three effective diffusion coefficients D_1, D_2, D_3 corresponding to diffusion along grains boundaries, diffusion along crystallites boundaries, and into crystallites properly with taking into account the 2-dimensional structure of channels. The analysis indicates strong dependence of the porous structure parameters and dimensions [9].

3. CONCLUSIONS

The effect of porosity of redeposited dust and debris materials on tritium diffusion and inventory in such materials are quite important. The 2-and 3-dimensional effects of the porous structure of the redeposited dust and debris of PFMs (carbon, beryllium...) must be taken into account in the numerical models describing gas diffusion and gas release in porous two level structural materials. Actual values of various diffusion stages and structure i.e., D_g, D_c, h_g, h_c are currently known with large uncertainties (especially D_c). Laboratory experiments specifically designed in which the dust and debris of candidate PFMs are produced and studied are required to make a correct prediction regarding tritium behavior in these materials.

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