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Letter to the Editors

Void migration in fusion materials

G.A. Cottrell *

EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon, Oxon OX143DB, UK

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Abstract

Neutron irradiation in a fusion power plant will cause helium bubbles and voids to form in the armour and blanket structural materials. If sufficiently large densities of such defects accumulate on the grain boundaries of the materials, the strength and the lifetimes of the metals will be reduced by helium embrittlement and grain boundary failure. This Letter discusses void migration in metals, both by random Brownian motion and by biassed flow in temperature gradients. In the assumed five-year blanket replacement time of a fusion power plant, approximate calculations show that the metals most resilient to failure are tungsten and molybdenum, and marginally vanadium. Helium embrittlement and grain boundary failure is expected to be more severe in steel and beryllium.

1. Introduction

The lifetimes of both plasma-facing armour and structural components under 14 MeV neutron irradiation and particle surface fluxes expected in a commercial fusion power plant will depend critically on the response of the chosen constructional materials to both displacement damage and the presence and behaviour of gas bubbles and voids. Direct ion implantation in plasma-facing surfaces and the neutron-induced (n,α) transmutation reactions in the structure cause both hydrogenic and helium atoms to build-up in the materials, leading to the formation of gas-filled bubbles. For helium, its high heat of solution means that it is insoluble in metals at typical operational temperatures ($\approx$1000 K) causing it to precipitate out and nucleate atomic clusters in the metal matrix. These clusters can coalesce with each other to form larger and more mobile pores and bubbles. The fracture strength of metals containing helium bubbles and voids will be compromised [1] when these defects reach the grain boundaries of the material. If sufficient numbers of helium bubbles migrate to and congregate on the grain boundaries of the metal, then the loss of material cross-section there can reduce both the strength of the metal and its thermal conductivity leading to high material temperatures and temperature gradients.

The purpose of this Letter is twofold. First, it is to review the simple kinetic migrations of helium bubbles and voids in typical candidate fusion materials. Second, these results are used to determine whether such movements are able to impair significantly the performance of the materials during the lifetime of the power plant.

2. The motion of voids

For a metal at constant temperature T, irradiation-produced helium-filled bubbles and empty voids can migrate randomly through thermal diffusion. This 'Brownian motion' occurs because an atom on the interior surface of a void can hop to a neighbouring surface site, a distance of one lattice spacing a away. The atomic surface diffusion coefficient is given by

$$D_s = \frac{a^2}{4} f = \frac{a^2}{4} \nu \exp\left[-\frac{E_A}{kT}\right], \quad (1)$$

where f is the hopping frequency, E_A the activation energy required by the atom to jump to a neighbouring

* Tel.: +44-1235 466 426; fax: +44-1235 466 435.
E-mail address: geoff.cottrell@ukaea.org.uk (G.A. Cottrell).

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site and v the atomic vibration frequency. At any instant only a few surface atoms are in a position – i.e. sitting on, rather than in, the void surface – to make a jump between on-surface sites. This is taken into account by using an E_A measured from bulk changes of surface shapes.

Strictly, we treat here the motion of voids since the collisional effect of the inert gas atoms on the surface diffusivity of the metal atoms is neglected. When the helium pressure is large, as expected in small bubbles, the diffusion coefficient of the surface atoms will be reduced below the value predicted by Eq. (1) as a result of collisions with the close-packed helium atoms. This effect can significantly reduce the mobility of small bubbles [2]. Small high-pressure bubbles are, however, able to grow in size by absorbing both thermal and irradiation-produced vacancies. As this happens, the gas pressure decreases towards, and perhaps eventually below, its equilibrium value. At this stage the atomic surface diffusion coefficient will be described by Eq. (1). Other modes of void mobility include vapour transport (within a void) and, in the case of large faceted voids, the nucleation of atomic ledges plays a key role [2].

A central idea in understanding the random motion of voids in a material is the treatment of an entire void as a single 'particle'. Voids in an isothermal and homogeneous material will perform three-dimensional Brownian migrations. Consider the single jump of an atom on the surface of a spherical void of radius r onto a neighbouring site. The jump shifts the centre of gravity of the void by some distance b, where $b \ll a$. The void thus has a jump distance b for its Brownian migration. Standard theory gives the void diffusion coefficient D_V as

$$D_V = \frac{1}{6} b^2 f_V, \tag{2}$$

where the void jump frequency is

$$f_V = \frac{4\pi r^2}{a^2} f \tag{3}$$

and the void jump distance is

$$b \approx \frac{3a^4}{4\pi r^3}. \tag{4}$$

The effective void diffusion coefficient is given by

$$D_V = \frac{3}{2\pi} \frac{\Omega^{\frac{4}{3}}}{r^4} D_s, \tag{5}$$

where Ω ($\equiv a^3$) is the atomic volume, in agreement with [3]. After an elapsed time t, the void migrates a root-mean-square distance

$$x_{rms} = \sqrt{6 D_V t}, \tag{6}$$

from its starting point.

Few fusion materials will however experience isothermal conditions. In particular, significant temperature gradients are likely to exist in fusion power plant blanket and plasma-facing components, particularly in the vicinity of coolant channels and plasma-facing surfaces. Therefore we consider next how the random void motion is altered by the presence of a macroscopic temperature gradient. To describe the motion of voids in a gradient

$$\frac{dT}{dx} = \frac{\Delta T}{a} > 0, \tag{7}$$

we need to calculate the probability, per unit atomic oscillation period, of one-dimensional atomic jumps on the interior void surface. This is given by the exponential factor in Eq. (1). In one such period, the *net* probability of atomic jumps in the positive x direction is the difference of two exponential functions

$$\exp\left[-\frac{E_A}{k(T+\Delta T)}\right] - \exp\left[-\frac{E_A}{kT}\right] \approx \frac{E_A a}{kT^2} \frac{dT}{dx} \exp\left[-\frac{E_A}{kT}\right]. \tag{8}$$

The multiplying term before the exponential on the right-hand side is the *bias factor*

$$F_b = \frac{E_A a}{kT^2} \frac{dT}{dx}, \tag{9}$$

due to the temperature gradient and is equal to the fraction of all atomic jumps which are thus biased. The net atomic flux per surface atom per unit time is

$$\Gamma = F_b v \exp\left[-\frac{E_A}{kT}\right]. \tag{10}$$

We can obtain an expression for the drift velocity of the void, v_V, moving under the temperature gradient force. The drift arises because there is a flux of atoms departing from the hot, 'leading' side of the void and settling on the cool, 'trailing' side. The net atomic flux on a two-dimensional surface is 1/2 of the value given for the one-dimensional case given above because of the extra degree of freedom. The atomic flux passing through the void is

$$\Gamma_V = \frac{\Gamma}{2} \frac{2\pi r}{a} \tag{11}$$

and the void moves in the direction of increasing temperature, sweeping out a volume containing

$$\frac{\pi r^2 v_V}{\Omega} \tag{12}$$

atoms per unit time. Equating this quantity to the flux derived above, we obtain the drift velocity

$$v_V = \frac{E_A \Omega v}{kT^2} \exp\left[-\frac{E_A}{kT}\right] \frac{dT}{dx} \frac{1}{r}, \quad (13)$$

which is inversely proportional to the void radius, in agreement with [4,5].

Both random and temperature-gradient-driven motion of voids can coexist in a fusion material. In the case of plasma-facing tungsten the bias factor is $\simeq 8 \times 10^{-8}$. Because the bias induced in the individual jumps is small, then in the early stages of migration, the distance x moved by the void will be determined mainly by random motion, i.e. $x = x_{rms}$ (Eq. (6)). But, because the distance migrated due to this increases only as the square root of the number of jumps, $N^{1/2}$ whereas the migration distance due to directed bias increases as N, the latter process takes over and predominates in the later stages. In this regime $x = bF_B f_V t$. We next estimate when the changeover occurs.

In the case of random migration, after a void has made N jumps then, on average, it migrates by these a distance $\approx bN^{1/2}$ At this stage, the fraction of these jumps which give migration away from the starting position is $\approx N^{1/2}/N = N^{-1/2}$ The changeover from random to directed flow occurs at a critical number of jumps

$$N_{crit} = \frac{1}{F_b^2}. \quad (14)$$

The critical time taken to get to the changeover point is just $t_{crit} = N_{crit}/f_V$.

3. Evaluation of candidate fusion materials

To evaluate the magnitudes of these migratory effects, we have used a set of parameters broadly representative of fusion materials (Table 1). A range of first wall and armour materials is considered in the light of these considerations. These are included in Table 2. The results depend critically on the value of the activation energy for each metal. We have used the measured value of the surface activation energy for tungsten, $E_A = 2.35$ eV. In the absence of suitable measured values for the other metals, a pro-rata estimate is given here based on the relative absolute melting points of tungsten and these other metals. The activation energy (Table 2, column 3) should vary roughly in proportion with the melting temperature ratios.

Table 1
Parameters of fusion material properties assumed in the calculation of bubble migrations

Quantity	Value
Atomic lattice spacing, a	3×10^{-10} m
Bubble radius, r	10^{-8} m
Grain size, L	10^{-5} m
Temperature, T	$\approx$1000 K
Macroscopic temperature gradient [a], dT/dx	$\approx 10^4$ K m^{-1}
Atomic vibration frequency, v	10^{12} s^{-1}
Lifetime of fusion armour wall component	$\approx 7 \times 10^7$s($\approx$2yr)
Fusion blanket replacement lifetime	1.8×10^8 s ($\approx$5 y)

[a] Deduced from data presented in [6] for a water-cooled Pb–17Li blanket.

Tungsten is currently under active consideration as a candidate plasma-facing armour material. The results of Table 2 suggest that even if tungsten armour were required to survive for five years in a power plant, then the voids would only have migrated a distance of about one-half of a void radius, many times smaller than a metal grain size. Migration on the timescale considered would be dominated by random motion. We would expect no significant numbers of voids to have migrated to grain boundaries and few void coalescences to have occurred in this time. This conclusion is even stronger for the minimum suggested [8] replacement lifetime (two years) for the inner wall armour. For molybdenum, which has been discussed as an alternative plasma-facing armour material, few voids are expected to reach the grain boundaries in either of the two lifetimes considered. The critical time for this material is of the same order as that of the shorter lifetime and so voids would move approximately equal amounts by random and biased drift. However, the voids in molybdenum migrate by distances

Table 2
Measured and calculated properties for various armour and first-wall metals for the void parameters shown in Table 1 at $T = 1000$ K

Metal	Melting temperature, T_m (K)	Activation energy, E_A (eV)	Diffusion coefficient, D_s (m^2 s^{-1})	Critical time, t_{crit}	Distance x (m), moved in 2 yr	Distance x (m), moved in 5 yr
W	3660	2.35[a]	3.3×10^{-20}	230 yr	2×10^{-9}	2×10^{-9}
Mo	2893	1.88	7.2×10^{-18}	1.5 yr	5×10^{-8}	10^{-7}
V	2193	1.41	1.7×10^{-15}	4.4 d	7×10^{-6}	2×10^{-5}
Fe	1813	1.18	2.5×10^{-14}	10 h	9×10^{-5}	2×10^{-4}
Be	1558	1.0	2.0×10^{-13}	2 h	6×10^{-4}	2×10^{-3}

[a] Deduced from Figs. 6–18 in [7].

large compared with the void radii and so we would expect some coalescences to occur with clearly a larger number at the longer lifetime considered. With vanadium as a structural material the motion of voids is dominated by drift motion on all practical timescales. The voids in this material will reach the grain boundaries. In the case of structural steel, void drift mobility is approximately an order of magnitude higher than in vanadium and so the same conclusions apply. In the case of beryllium, we see a further increase in the void mobility and would expect significant accumulation of voids at grain boundaries.

These conclusions are for pure elements, but in practice alloys may be used. However, we are concerned here with the bulk migration of entire voids, which can occur only if all the surface atoms move from the leading side to the trailing one, so that the bulk migration is controlled by an average of the individual alloy mobilities.

4. Conclusions

The random migrations of small voids or low-density He bubbles, due to the movements of their surface atoms from one side of the void to the other, can lead to their coalescence into larger voids, when they meet. In principle, the steep temperature gradients in a fusion reactor may aggravate this effect by directing the otherwise random migrations into a steady drift velocity up the temperature gradient. For the fusion power plant conditions considered the void migration behaviour is strongly dominated by the activation energy for surface atomic mobility, and hence by the cohesive strength of the material. For W, and to a lesser extent Mo, the bubble migrations are small and the effect of the thermal gradient drift is insignificant. For V, steel and Be, respectively, the voids are increasingly more mobile and the drift process becomes the major one.

There are, inevitably, other factors which determine the extent to which bubbles, He atoms and knock-on defects reach the grain boundaries. Examples of these include the retardation of vacancy migrations by He atoms and trapping of He on lattice precipitates, in addition to the migrations and coalescences of voids which have been the subject of this letter. And, of course, there are other factors, besides swelling, which have to be taken into account in the total evaluation of the effectiveness of candidate fusion materials; for example, embrittlement and sputtering.

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