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Does pulsing in spallation neutron sources affect radiation damage?

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Abstract

Modern neutron sources, such as the planned European Spallation Source (ESS), will operate in a pulsed mode with very short pulses at relatively high frequencies. Presently, the judgement of radiation damage resulting under such conditions depends on the available experimental experience with radiation damage caused by comparable continuous irradiation and on modelling for extending this experience to pulsed irradiation. In the present contribution, this combination of available experimental and theoretical experience is used to judge possible pulsing effects on radiation damage and related macroscopic phenomena in metallic materials. Significant pulsing effects require: (1) a proper relationship of the lifetimes of mobile defects to the time scales of pulsing, and (2) sufficiently large pulsing-induced fluctuations of defect fluxes to relevant sinks. It is shown that the first condition, quantified by a 'relative dynamic bias', is fulfilled under ESS conditions. Application of the second condition, quantified by an 'absolute dynamic bias', to pulsing sensitive processes such as cavity nucleation and creep by climb controlled dislocation glide shows, however, that the damage yield of one pulse in ESS and similar facilities is much too small to allow any significant pulsing effect.

1. Introduction

The overwhelming majority of experimental and theoretical radiation damage studies consider the damage-generating irradiation to be homogeneous and continuous, or more precisely to be constant in space and time over spatial and temporal scales large compared to those of the microstructure, respectively. These assumptions are generally justified for materials used in fission reactors. Irradiation is or will be not continuous, however, in projected fusion reactors, spallation neutron sources and other irradiation facilities. The temporal structure of irradiation is vastly different for different types of devices. It is relatively soft in Tokamak-type fusion reactors, with on- and off-times of the order of several hundreds of seconds; it is moderate to moderately rough in accelerator devices, with on-times varying from ks down to μs (as in the case of spallation neutron sources); and it would be extremely rough in inertial confinement fusion reactors, with extremely short pulse lengths even down to the ns range.

As are radiation damage processes on the whole, possible effects of the temporal structure of the irradiation are controlled by the diffusion and reaction of the mobile irradiation-induced defects, which therefore form the primary subject in the theoretical studies of such effects. Illustrative examples are the temporal evolution of point defect concentrations and fluxes in the presence of given sinks during the transient to steady state after the onset of continuous irradiation [1,2] and during the annealing stage after an instantaneous defect production pulse [2]. Effects of time-dependent defect concentrations and fluxes on microstructural processes and on related macroscopic radiation damage phenomena represent more complicated problems. The first example for such an effect discussed in the literature was irradiation creep [3–5].

In discussing possible pulsing effects, it is useful to distinguish: (1) cumulative microstructural processes which depend mainly on the total dose but not significantly on dose rate and dose rate variations and are thus insensitive to irradiation pulsing, and (2) fluctuation sensitive processes which do depend on dose rate and

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dose rate variations and thus can be sensitive to pulsing under certain conditions. The accumulation of stable defect clusters produced in displacement cascades, the growth of such and other stable structures (cavities, precipitates) and the segregation of impurity atoms to interfaces and grain-boundaries belong to the first class; the surmounting of physical or energetic barriers such as in creep by climb controlled dislocation glide [3–5] or in nucleation processes, respectively, belongs to the second class. A crucial aspect in discussing the sensitivity to pulsing is the relation of the time scales of the externally imposed irradiation (pulse length and distance) to the internal time scales of the production, diffusion and reaction of the irradiation-induced defects.

The present contribution focuses on possible effects of irradiation pulsing on damage accumulation in metallic materials to be used in spallation neutron sources, particularly in the planned European version (European Spallation Source (ESS), see Table 1). Where appropriate, other pulsed accelerator driven systems (ADS) are included into the considerations. The temperatures to be considered in ESS and other ADS applications range from room temperature to about 250°C and 650°C, respectively, but we include even higher temperatures into our discussion. Concerning the material for ESS applications, we have primarily austenitic stainless steels in mind (for which we take Ni to be representative) but the treatment is kept as general as possible (by using, for instance, homologous temperature scales) to allow extensions of the conclusions to other metallic materials, for instance to martensitic/ferritic steels considered for ADS applications (see Tables 2 and 3).

Table 1
Irradiation characteristics

Temporal structure	
Pulse length, τ_p	1 μs
Repetition frequency, ν	50 Hz
Damage rates	
Peak rates	4×10^{-2} dpa/s
	6×10^{-6} Hepa[a]/s
Average rates	2×10^{-6} dpa/s
	3×10^{-10} Hepa/s
Damage yield of one pulse[b]	
Displacements	4×10^{-8} dpa
Surviving Frenkel pairs	10^{-8} pa
Single SIAs/vacancies, $\Delta c_{i,v}$	7×10^{-9} pa
SIAs/vacancies in clusters	3×10^{-9} pa
He atoms, Δc_{He}	6×10^{-12} pa
Cascade density	10^{19} m^{-3}
Cluster density	3×10^{19} m^{-3}
Sink strength increment	3×10^{11} m^{-2}

[a] pa: per matrix atom.
[b] Crude estimates of cascade characteristics, sufficient for the present purpose.

Table 2
Material parameters used in estimates

Atomic volume, Ω	11×10^{-30} m^3
Recombination coefficient, a	5×10^{20} m^{-2}
SIA diffusion coefficient, D_i	
Pre-exponential	10^{-5} m^2/s
Migration energy	1 (to 3)[a] kT_m
Vacancy diffusion coefficient, D_v	
Pre-exponential	5×10^{-5} m^2/s
Migration energy	7 (to 9)[a] kT_m

[a] Defect-impurity binding energy of 2 kT_m.

The paper is structured as follows. In Section 2, we briefly review the basic features of the production, diffusion and reaction of defects as expected under GeV proton irradiation. In Section 3, we compare the mean lifetimes of the mobile defects, particularly of self-interstitial atoms (SIAs) and vacancies, in the microstructure expected to evolve under such irradiation, with the time scales of the irradiation (pulse length and distance) and discuss the related necessary condition for significant pulsing effects ('relative dynamic bias'). In Section 4, we examine whether the pulsing-induced amplitudes of defect fluxes ('absolute dynamic bias') are sufficiently large to influence effects such as cavity nucleation and creep by climb controlled dislocation glide. Finally, in Section 5, we summarise and discuss our conclusions.

2. Basic considerations

2.1. Primary damage

The energy transfer from GeV protons colliding with atoms of the target material is characterised by a broad spectrum of recoil energies of primary knock-on atoms, with medium energies of tens of keV (about 50 keV for stainless steels). Atoms with such high energies have been shown by molecular dynamics (MD) simulations to induce pronounced displacement cascades which extend over several nm and relax to characteristic defect structures within times smaller than ns. The main results of MD simulations of cascades in pure metals are [6,7]: (1) SIAs and vacancies are produced in a localised and segregated fashion with vacancies concentrated in the core of a cascade and SIAs at its periphery, (2) efficient intra-cascade recombination results in a low damage efficiency, (3) substantial intra-cascade clustering of both vacancies and SIAs takes place, (4) SIA clusters in the form of small dislocation loops ('coupled crowdions') are able to perform a fast one-dimensional (1-D) diffusional glide motion even at low temperatures. There are characteristic differences between bcc and fcc metals (for instance between austenitic and martensitic/ferritic

Table 3
Microstructural data used in estimates

	0.2 T_m	0.5 T_m
Maximum bubble density	2×10^{25} m^{-3}	3×10^{20} m^{-3}
Bubble radius at 0.2% He	0.25 nm	12 nm
Bubble sink strength at 0.2% He	6×10^{16} m^{-2}	0.5×10^{14} m^{-2}
Initial dislocation density	3×10^{12} m^{-2}	3×10^{12} m^{-2}
Maximum dislocation density	4×10^{16} m^{-2}	2.5×10^{14} m^{-2}
Maximum total sink strength at 0.2% He	10^{17} m^{-2}	3×10^{14} m^{-2}

steels): both recombination and clustering efficiencies have been found to be lower in bcc than in fcc metals.

Owing to their stochastic nature, displacement cascades represent internal sources of strong defect fluctuations which can crucially affect fluctuation sensitive processes such as cavity nucleation or climb controlled dislocation glide [8,9].

2.2. Temporal and spatial scales in defect reaction kinetics

Crucial internal parameters controlling the possibility of pulsing effects are the temporal and spatial scales related with the diffusion and reaction of the mobile radiation-induced defects. In order to illustrate the meaning and magnitude of these parameters, we consider the evolution of the concentration, c, of a certain type of mobile defects (SIAs, vacancies, clusters) produced randomly in space and time at a production rate P, and diffusing with diffusivity D until getting absorbed by randomly distributed immobile sinks of strength k^2 (sink dominated defect annihilation). In the mean field approximation, this problem is described by the linear diffusion reaction equation [10,11]

$$\partial c/\partial t = P + D\Delta c - Dck^2, \tag{1}$$

where the Laplacian Δ reduces to $\partial^2 c/\partial x^2$ for defects diffusing 1-D in x-direction.

The characteristic spatial and temporal scales defined by (exponential solutions of) Eq. (1) are the mean diffusion range λ and the mean lifetime τ of the defects, respectively,

$$\lambda = k^{-1}, \tag{2a}$$

$$\tau = 1/Dk^2. \tag{2b}$$

According to Eqs. (2a) and (2b), the sink strength k^2 is a key quantity in both the spatial and temporal scales of defect diffusion. In the case of 3-D diffusing defects, the sink strength, k_3^2 is proportional to the density of sinks. For a random distribution of spherical sinks (cavities) of radius r and number density N and dislocations of absorption efficiency Z and density ρ, for instance, k_3^2 is given by [10]

$$k_3^2 = 4\pi rN + Z\rho, \text{ i.e. } \lambda_3 \propto N^{-1/2}\rho^{-1/2}. \tag{3}$$

Differently from the 3-D case, the mean diffusion range of 1-D diffusing defects is, similar as in collision theory, inversely proportional to the absorption cross-section, $\sigma = \pi r^2$ and diameter, d, and the densities N and ρ of spherical sinks and dislocations, respectively, [11]

$$\lambda_1^{-1} = \sigma N + d\rho, \text{ i.e. } \lambda_1 \propto N^{-1}\rho^{-1}. \tag{4}$$

The corresponding sink strength, k_1^2 does not depend linearly, as in the 3-D case, but quadratically on the sink density. Because of this difference, the mean diffusion ranges and, at given D, lifetimes of defects are substantially larger for 1-D than for 3-D diffusion at low to medium sink densities. Disturbances of the 1-D diffusion of SIA clusters by impurities and precipitates in technical alloys such as steels to be considered here are, however, expected to reduce this difference [11].

For SIAs (i) and vacancies (v), we will consider in the following, in addition to the sink term in Eq. (1), a recombination term $aD_ic_ic_v$, where a is the recombination coefficient. For recombination dominated defect annihilation, the spatial and temporal scales given by Eqs. (2a) and (2b) must be modified accordingly (see Section 3.2).

2.3. Role of asymmetries in the production and properties of SIA and vacancy-type defects

Radiation damage accumulation in metallic materials is crucially determined by different types of asymmetries in the production and properties of SIA and vacancy-type defects.

(1) Single SIAs diffuse substantially faster and arrive earlier at sinks than single vacancies. This difference defines a ‘dynamic bias’ which is relevant in pulsing effects and has therefore to be considered in the following.

(2) Because of the larger magnitude of their relaxation volume, SIAs interact stronger with dislocations than vacancies resulting in an excess vacancy concentration. This difference, termed ‘dislocation bias’, has been considered previously to be the main cause of vacancy accumulation in the form of voids [9,10].

(3) The production, stability and mobility of clusters of SIAs and vacancies are different. Thus, in the temperature range of void swelling (annealing stage V

between 0.4 and 0.5 T_m), SIA clusters are thermally stable while small vacancy clusters decay rapidly such that, in this temperature range, effectively only SIA clusters are produced in cascades. This so-called 'production bias' [12], in conjunction with the escape of 1-D diffusing SIA clusters to sinks, has been shown to result in a high excess vacancy concentration representing a strong driving force for void swelling under cascade damage conditions [11].

On proper relative scales, the 'dislocation bias' amounts to a few percent whereas the 'production bias' is generally higher under cascade damage conditions and can reach even tens of percent. Under pulsed cascade damage conditions, the 'dynamic bias' would be masked by these types of biases unless it reaches comparable orders of magnitude.

3. Lifetimes of defects – dynamic bias

3.1. Sink strengths and diffusion ranges

As shown in Section 2.2, the sink strength k^2 is the key quantity in both the spatial and temporal scales of defect diffusion and reaction which control the occurrence of pulsing effects. Since no reliable information on the microstructure evolving under pulsed irradiation is presently available we base our discussion of pulsing effects on knowledge about the microstructure evolving under an equivalent continuous irradiation, and check the validity of this procedure after having reached preliminary conclusions.

Since we are primarily interested in stainless steels (for which defect diffusion parameters of Ni are taken to be representative) we consider microstructures expected to occur in fcc metals under irradiation conditions corresponding to GeV proton irradiation as in ESS and use homologous temperature scales to keep the discussion as general as possible (see Table 2; note as a guideline that room temperature corresponds to about 1/6 T_m for stainless steels). We may assume that helium bubbles and dislocations (including defect clusters and dislocation loops) form the main microstructural components. For both, some knowledge is available for low as well as high temperatures (see Table 3).

(1) *Helium bubbles*: At low temperatures, $T \leqslant 0.2\ T_m$, electron diffraction from bubble lattices in Ni and stainless steel [13] indicates maximum bubble densities of about $2 \times 10^{25}\ m^{-3}$ reached by He implantation at room temperature. At higher temperatures, the evolution of He bubbles has been systematically investigated as a function of temperature [14,15]. The nucleation of He bubbles ceases at low He concentrations depending on temperature. The bubble nucleation process has been identified to be 'He diffusion controlled' and 'He dissociation controlled' characterised by low and high activation energies at medium and high temperatures, respectively. The transition between the two regimes occurs around 0.5 T_m, where bubble densities between 10^{20} and $10^{21}\ m^{-3}$ are expected for the mean He generation rates considered here. Below 0.5 T_m, He bubbles are most likely overpressurised and contain a high density of He atoms, whereas they tend to establish thermal equilibrium above 0.5 T_m. In estimating bubble sizes, we assume that the He density in bubbles is of the order of the matrix atom density even up to 0.5 T_m. A schematic plot of bubble densities and radii vs. homologous temperature is presented in Fig. 1. Note that similar features have been found for martensitic/ferritic steels and other bcc metals [15].

(2) *Dislocations*: At low temperatures, $T \leqslant 0.2\ T_m$, the dislocation density in well-annealed metals starts from values below $10^{13}\ m^{-2}$. Under irradiation, the dislocation density increases by loop formation, growth and coalescence and approaches at a few dpa maximum values of the total density, including loops, between 10^{16} and $10^{17}\ m^{-2}$ (probably controlled by the cascade size). At $T \sim 0.5\ T_m$, dislocation densities have been found to saturate between 10^{14} and $10^{15}\ m^{-2}$ [16]. Between 0.2 and 0.5 T_m, we assume Arrhenius behaviour as for bubble densities. Note that the densities of defect clusters and dislocation loops at given dose and their increase with dose have been found to be significantly

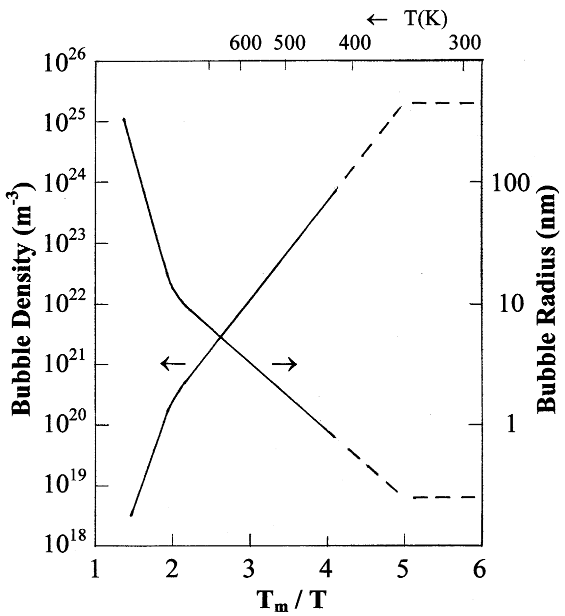

Fig. 1. Schematic plots of bubble density and radius for 0.2% He vs. homologous temperature. Bubble data given in Table 3 are used. For Ni (stainless steel) the corresponding absolute temperature scale is shown on the upper axis. The lines are broken at low temperatures where bubbles are not resolvable for 0.2% He.

lower and slower, respectively, in bcc than in fcc metals, most likely due to lower intra-cascade clustering efficiencies [17].

To translate these microstructural parameters to sink strengths for the annihilation of 3-D diffusing defects we use Eq. (3) assuming for the absorption efficiency $Z = 1$. The resulting partial and total sink strengths are plotted vs. homologous temperature in Fig. 2 for 0.2% He corresponding to about 13 dpa. In martensitic/ferritic steels and other bcc metals, the total sink strength is expected to be lower because of the smaller contribution of the dislocation structure.

The mean diffusion range corresponding to the total sink strength is found to be comparable with the mean distance between cascades of one irradiation pulse in ESS (and other ADS) only at the very beginning of irradiation but to decrease to substantially lower values during damage accumulation. This means that cascades of one pulse act, independent of their lifetime, separately and, consequently, in the same way as if they were produced continuously over the whole pulsing period. From this we can draw the conclusion that possible effects of internal point defect fluctuations associated with cascades would be independent of anticipated pulsing-related effects under ESS irradiation conditions, and even more so for longer pulse lengths anticipated in some other ADS such as transmutation or irradiation facilities.

For the microstructural parameters of the 'developed' structure discussed above, the mean diffusion ranges of 1-D diffusing defects are, according to Eq. (4), only somewhat larger than those for 3-D diffusion at

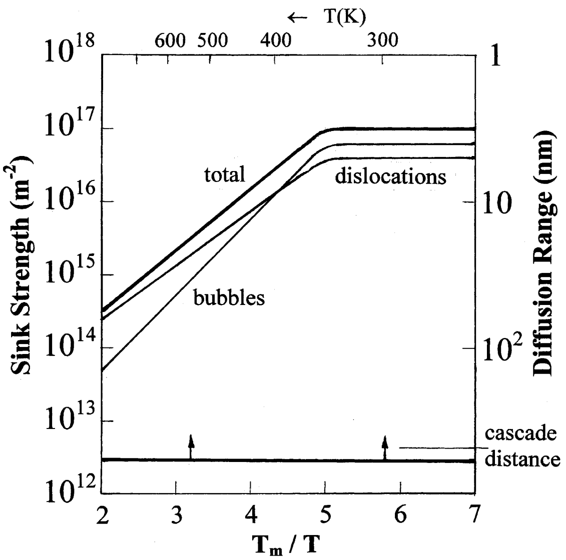

Fig. 2. Schematic plots of partial and total sink strength and corresponding 3-D diffusion range for initial and 'developed' structures, respectively, vs. temperature. Arrows indicate evolution of total sink strength. Data given in Table 3 are used.

temperatures below and around 0.2 T_m but substantially larger at 0.5 T_m, where they come close to the mean distance between cascades. Our foregoing conclusion concerning the independence of individual cascade remains valid, however, even at these high temperatures.

3.2. Lifetimes of point defects

The relations between the time scales in the diffusion-reaction kinetics of the relevant mobile defects to the time scales of pulsing (pulse length, τ_p and frequency ν) are suited to provide criteria for the occurrence of pulsing effects. We focus here on the role of single SIAs and vacancies in possible pulsing effects. In estimating their lifetimes we use the (3-D) diffusion properties given in Table 2 ($D_i/D_v \gg 1$) and the total sink strength plotted in Fig. 2. In the case of sink dominated defect annihilation, the defect lifetimes are given by Eq. (2b) – independent of the temporal structure of irradiation. At the beginning of (continuous or pulsed) irradiation, the lifetime of the fast SIAs is indeed controlled by annihilation at existing sinks for the whole temperature range considered here, but the lifetime of the slow vacancies is initially limited by recombination. Subsequently, the SIA lifetime becomes also recombination limited due to the accumulation of vacancies during the time required for them, on the average, to reach sinks. At the high temperature side, the slowly accumulating sinks become, however, again dominant in defect annihilation.

In the schematic plots shown in Fig. 3, we have used a number of simplifying approximations which suffice for the order of magnitude estimates required here. Since recombination depends in a complicated way on the temporal structure of irradiation due to its bi-linear dependence on the concentrations of SIAs and vacancies, we have taken for the lifetimes limited by recombination values for quasi-steady state under a fictitious continuous irradiation corresponding to the time-averaged real irradiation with defect production rates $\langle P_i \rangle = \langle P_v \rangle$. For the vacancy concentration needed in this case, we have changed schematically to an upper bound value, $c_v^{max} = 0.01$ (corresponding to a recombination volume of 100 Ω) when this would be exceeded according to the kinetic equations. For the 'developed structure' (reached at a few dpa) we have taken the values for that defect annihilation mechanism which is dominant at the temperature considered (for typical limits see also [18]). In summary we have assumed

$$\tau_i^{-1} = D_i k_o^2, \quad \tau_v^{-1} = a\langle P_{i,v} \rangle / k_o^2 \quad \text{for } 0 < t < \tau_v, \tag{5a}$$

$$\tau_{i,v}^{-1} = D_{i,v} a c_v^{rec} \quad \text{for } t > \tau_v, \quad k^2(t) < a c_v^{rec}, \tag{5b}$$

$$\tau_{i,v}^{-1} = D_{i,v} k^2 \quad \text{for } t > \tau_v, \quad k^2(t) > a c_v^{rec}, \tag{5c}$$

where

$$c_v^{rec} = \mathrm{Min}\{(\langle P_{i,v} \rangle / a D_v)^{1/2}, c_v^{max}\}. \tag{5d}$$

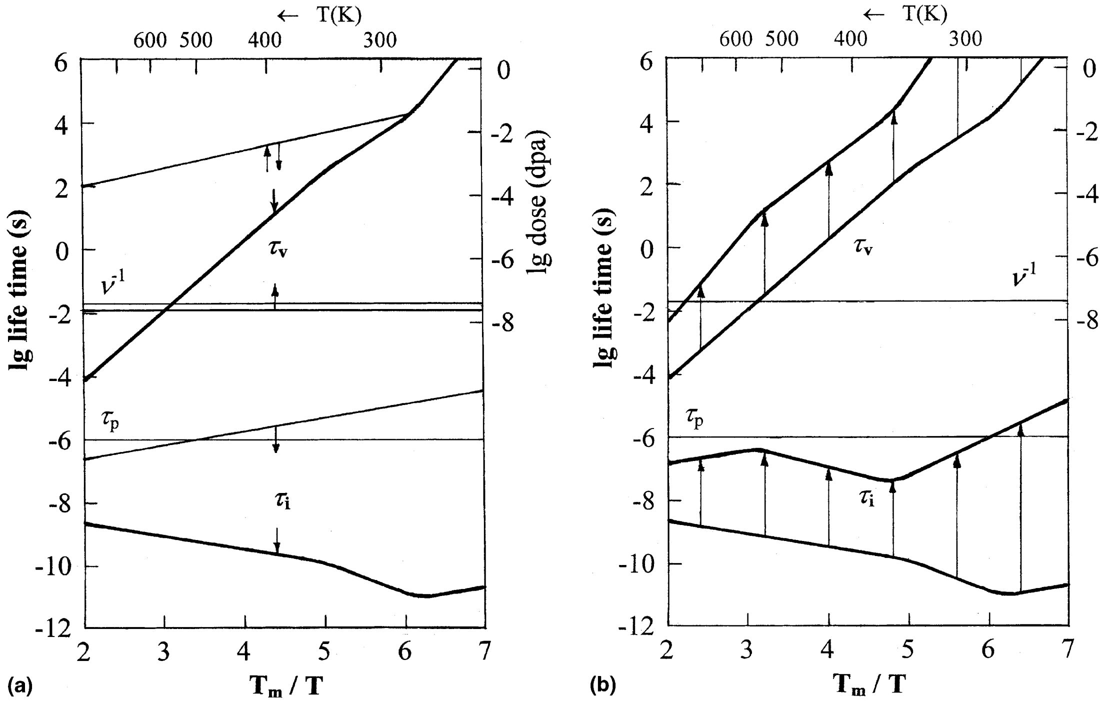

Fig. 3. Schematic plots of lifetimes of SIAs and vacancies for initial and 'developed' structures, respectively, vs. temperature, (a) for pure metal (arrows indicate evolution), (b) possible effects of impurities in technical alloys for 'developed' structures (arrows indicate impurity-induced changes). The total sink strength shown in Fig. 2 and the defect diffusion data given in Table 2 are used.

Note that, except at the very beginning of irradiation, i.e. at all $t > \tau_v$ the ratio of the lifetimes of the two defects is given by $D_v/D_i \ll 1$ in both limiting cases of defect annihilation.

Fig. 3 shows that the lifetimes of SIAs and vacancies differ not only by orders of magnitude, as expected, but are in opposite relation to the time scales of irradiation in ESS (ADS) in most parts of the parameter range covered. The lifetime of SIAs is shorter than the pulsing period and, except at the very beginning of irradiation, even shorter than the pulse length in the whole temperature range considered. In contrast to this, the vacancy lifetime is larger than the pulse length in the whole temperature range and, except at the very beginning of irradiation, even larger than the pulsing period up to about 1/3 of T_m, i.e. in the whole temperature range for ESS applications. Accordingly, under such conditions, the appearance of SIAs is closely bound to the pulses whereas the vacancies are continuously present as under continuous irradiation. Even a significant reduction of the effective diffusivities of the defects by binding to impurities would not change these conclusions much as illustrated in Fig. 3(b). Similar conclusions hold for other ADS where the pulsing frequency is similar but the pulse length is longer by three orders of magnitude than in ESS. The striking difference in the behaviour of SIAs and vacancies under such irradiation conditions relative to the time scales of pulsing indicates the possibility of pulsing effects which must be therefore examined further.

The two other types of defects, which are mobile in the considered temperature range, are He interstitial atoms and SIA clusters in the form of small glissile dislocation loops. Since both are, as single SIAs, fast diffusing defects, their appearance is closely bound to the pulses of ESS (or ADS).

3.3. Dynamic bias

The quantity characterising fluctuations in the arrival rates of SIAs and vacancies at sinks is the 'excess defect flux' $D_i c_i - D_v c_v$. According to the preceding section, under ESS irradiation conditions, the SIA concentration follows closely the course of irradiation while the vacancy concentration is only weakly influenced by pulsing and keeps closely to the value for an equivalent continuous irradiation. After a transient, i.e. for $t > \tau_v$, the defect fluxes become periodic and for vanishing dislocation and production bias, $k_i^2 = k_v^2$ $(Z_i = Z_v)$, $P_i = P_v$, the 'excess defect flux' fluctuates around 0. This is illustrated in Figs. 4(a,b). The quantity characterising fluctuations of the sizes of sinks (clusters, cavities) and the climb position of dislocations is the excess number of SIAs or vacancies absorbed by such sinks, determined by the integral of the 'excess defect flux', $D_i c_i - D_v c_v$,

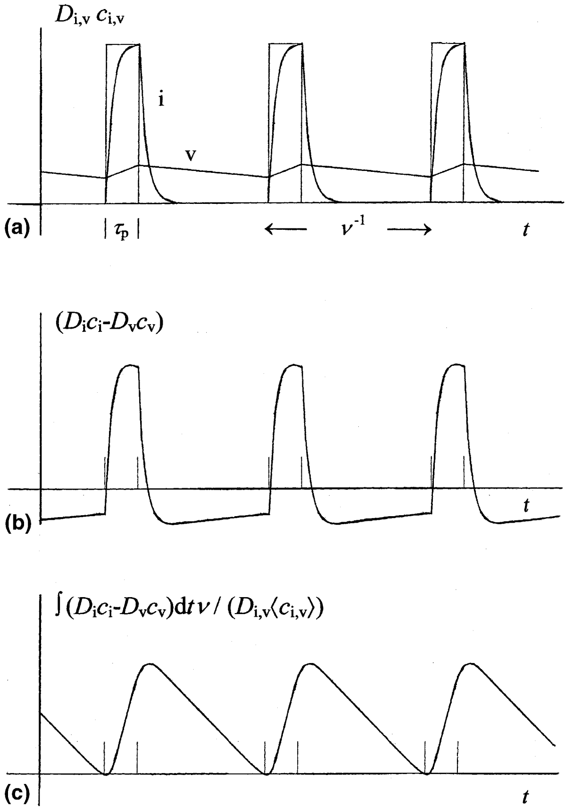

Fig. 4. Sketch of time dependence of (1) SIA and vacancy fluxes (a), (2) difference of both fluxes (b), (3) time integral of flux difference (c) induced by pulsed irradiation for $P_i = P_v$, $t \gg \tau_v$.

over time as illustrated in Fig. 4(c). We define the amplitude of this quantity as the 'absolute dynamic bias'

$$\Delta = \mathrm{Ampl}\left\{ \int (D_i c_i - D_v c_v)\, dt \right\}. \quad (6)$$

For non-vanishing dislocation and production bias, the time average of $D_i c_i - D_v c_v$ would have to be subtracted in the integrand of Eq. (6). The pulse-induced fluctuation amplitudes of the excess number of SIAs/vacancies, $n_{i,v}$, contained in a spherical sink of radius r or attached to a dislocation segment of length l are obtained by simply multiplying Δ with $4\pi r/\Omega$ and l/Ω, respectively, where Ω is the volume per matrix atom. The values of Δ depend, of course, via $D_{i,v} c_{i,v}$, on defect production rate and total sink strength.

It is useful for the further discussion to relate Δ to the integral of the flux of one of both types of defects (or of the average flux of both for non-vanishing dislocation and production bias) over one pulsing period and to define this quantity as the 'relative dynamic bias'

$$\delta = \Delta \nu / (D_{i,v} \langle c_{i,v} \rangle). \quad (7)$$

For the case of sink dominance (which is more important for pulsing effects than the case of recombination dominance), this quantity has the advantage to depend explicitly only on the external and internal time scales of pulsing and defect diffusion, τ_p, ν^{-1} and $\tau_{i,v}$, respectively, but not, as Δ, explicitly on defect production rate and total sink strength. In this case, the 'relative dynamic bias' δ defined by Eq. (7) has limiting values of 0 and 1 for continuous irradiation, $\nu\tau_p = 1$, and extreme pulsing conditions, $\nu\tau_p \ll 1$, $\tau_i \ll \tau_v$, $\nu\tau_i \ll 1$, respectively. For a rectangular temporal structure of irradiation with constant and vanishing damage rates during and between the pulses, respectively (as assumed here for ESS), where the piecewise solutions of Eq. (1) for the concentrations of SIAs and vacancies depend on time exponentially, Eq. (7) can be expressed as a function of exponential functions of ratios of the external and the internal time scales, i.e. of τ_p, ν^{-1}, τ_i and τ_v. The corresponding complicated expression is not quoted here explicitly [19], but δ is plotted in Fig. 5 vs. τ_v/τ_p and $\nu\tau_i$ for $\tau_i \ll \tau_v$ and for the moderate and extreme pulsing, $\nu\tau_p = 0.5$, and $\nu\tau_p \ll 1$, respectively. In these cases, $\delta \approx \tau_v/\tau_p$ for $\tau_v/\tau_p \ll 1$ ($\tau_v/\tau_p < 0.5$), $\delta \approx 0.5$ and $\delta \approx 1$, respectively, for $\tau_v/\tau_p \gg 1$ ($\tau_v/\tau_p > 2$) and $\nu\tau_i \ll 1$ ($\nu\tau_i < 0.05$), and $\delta \approx \tau_p/16\tau_i$ and $\tau_p/8\tau_i$, respectively, for $\nu\tau_i \gg 1$ ($\nu\tau_i > 0.2$).

Accordingly, we find $\delta \approx 1$ for ESS and other ADS irradiation conditions for the whole temperature range considered here, provided the sink strength assumed is representative for such conditions. This represents a quantification of the indication for a possibility of pulsing effects in ESS and other ADS obtained by discussing the defect lifetimes in their relation to the time scales of pulsing. We emphasise here that the main conditions for this conclusion, $\tau_i \ll \tau_v, \nu\tau_i \ll 1$, are certainly fulfilled even though the values for the defect lifetimes, $\tau_{i,v}$, may be uncertain by one to two orders of magnitude due to uncertainties in defect diffusivities and sink strengths.

A value of the relative dynamic bias, δ, sufficiently close to one represents, however, only a necessary but not a sufficient condition for pulsing effects. Significant fluctuations in sizes of clusters and climb position of dislocations require, in addition, a sufficiently large value of the absolute dynamic bias Δ. For the case of sink dominated defect annihilation, Δ can be expressed by δ as (see Eq. (7))

$$\Delta = \delta D_{i,v} \langle c_{i,v} \rangle / \nu = \delta \langle P_{i,v} \rangle / \nu k^2. \quad (8)$$

In this case, Δ depends linearly on the concentration of SIAs/vacancies produced in one pulse, $\Delta c_{i,v} = \langle P_{i,v} \rangle / \nu$.

According to Eq. (8), the requirement of a minimum value of Δ for a certain pulsing effect would impose conditions on the quantities defining Δ. For given average defect production rate and temperature (defining δ and k^2), for instance, pulsing effects would be limited to

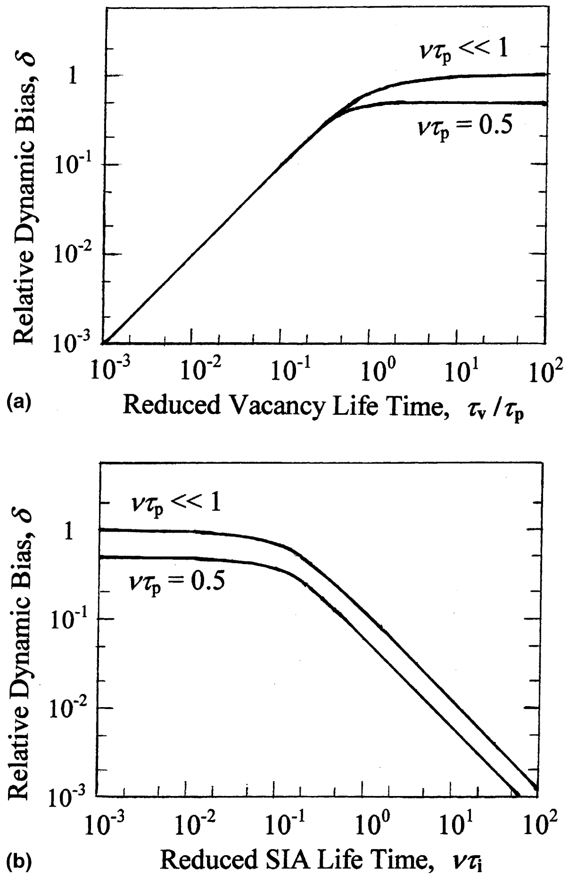

Fig. 5. 'Relative dynamic bias', δ, vs. normalised lifetimes of vacancies and SIAs, τ_v/τ_p and $\nu\tau_i$ for moderate and extreme pulsing, $\nu\tau_p = 0.5$, and $\nu\tau_p \ll 1$, respectively.

pulsing frequencies below a certain maximum value. Above the minimum value of Δ, the average rate of a continuing process such as creep by climb controlled dislocation glide may be expected to be proportional to $\nu\Delta$ which would not depend on ν, according to Eq. (8), for constant δ. The minimum value of Δ is, however, the crucial quantity in discussing the possibility of pulsing effects under ESS (ADS) irradiation conditions for which we may assume $\delta \approx 1$.

We mention here that a quantity analogous to Δ to characterise the effect of pulsing on diffusion fluxes of He atoms, can be defined by substituting in Eq. (6) of $D_i c_i - D_v c_v$ by of $D_{He}c_{He} - \langle D_{He}c_{He}\rangle$.

4. Effects of dynamic bias

4.1. Damage yield of one pulse

Eq. (8) underlines the central role of the 'damage yield of one pulse' in pulsing-induced fluctuations. The striking common feature of the values given in Table 1 for ESS irradiation conditions (values for other ADS are similar) is their extremely low level (see below for comparison with relevant reference values). This will be a crucial point in the following discussion.

From the low concentrations of SIAs and vacancies produced in one pulse we can draw an immediate conclusion on the role of pulsing in the contribution of mutual recombination to defect annihilation. Principally, this contribution is expected to be increased by concentrating irradiation, for a given average damage rate, in short pulses which would clearly be a positive effect of pulsing. The sink strength of the vacancies for recombining with SIAs produced in one and in the very first pulse, $a\Delta c_v \approx 3 \times 10^{12}$ m^{-2}, is indeed comparable with the sink strength expected at the beginning of irradiation for well-annealed metals. But the contribution of one pulse to defect annihilation becomes negligibly small compared to that of the vacancies accumulating during their lifetime and to that of fixed sinks (cavities and dislocations) developing in the course of irradiation (see Fig. 2 for comparison). From this, we may conclude that the contribution of recombination to defect annihilation is not significantly enhanced by the concentration of irradiation in short pulses.

In this context, a comment on radiation enhanced diffusion is worth adding. The contribution of vacancies produced in one pulse to the mean square atomic displacement, for instance, $\langle r^2\rangle \approx 6D_v\Delta c_v\tau_v \approx 6\Delta c_v/k^2 < 2 \times 10^{-20}$ m^2, is clearly below atomic scales in the whole parameter range considered here. Thus, many pulses are needed to get significant diffusion. We emphasise here that radiation enhanced diffusion and a possibly associated impurity segregation are accumulative processes which are anyway insensitive to pulsing. In the following, we discuss two typical fluctuation sensitive processes which can be sensitive to pulsing if the fluctuation amplitudes defined by Δ are sufficiently large, namely, cavity nucleation and irradiation creep by climb controlled dislocation glide.

4.2. Does pulsing affect cavity formation?

Under the concurrent production of He atoms and displacement defects, cavity formation starts with the clustering of He atoms and vacancies [14]. The formation of stable He bubbles by this clustering process is limited due to the reduction of the concentration of mobile He atoms upon their absorption by the increasing density of bubbles. At typical void swelling temperatures (between 0.4 and 0.5 T_m), the slow growth of such bubbles by the absorption of subsequently produced He atoms changes under certain conditions to faster growth driven by a supersaturation in the vacancy concentration by which the bubbles transform effectively to voids [9,14]. The formation of bubble nuclei from small He-vacancy complexes is expected to be the process most sensitive to pulsing in this sequence of processes.

Small He-vacancy complexes are stabilised by the absorption of He atoms and vacancies but destabilised by the absorption of SIAs and the emission of vacancies. The kick-out of one or more He atoms from a vacancy by an SIA represents the most extreme example for such a destabilisation process. Accordingly, bubble formation from small He-vacancy complexes may be considered to be the result of a competition between constructive and destructive atomic reaction processes. This underlines our suspicion that pulsing-induced temporal fluctuations in the defect fluxes could indeed be crucial in bubble formation – provided their amplitudes are significant.

The significance of fluctuations in the defect fluxes for bubble formation can be examined on the basis of the 'absolute dynamic bias' Δ discussed above and its analogue for He interstitial diffusion. A simple upper bound estimate for the mean fluctuations in the number of He atoms and vacancies, $\Delta n_{\mathrm{He,v}}$, contained in a bubble nucleus is obtained by assuming, in addition to $\delta \approx 1$, that all of the fast diffusing He interstitials and SIAs produced in one pulse are instantaneously and completely distributed over all existing bubble nuclei. Assuming further that the density of the resulting stable bubbles, N_{b}, represents a lower limit for bubble nuclei in the bubble nucleation phase ($k^2 \to 4\pi r N_{\mathrm{b}}$ in Eq. (8)), we may write for the fluctuations in the number of He atoms and vacancies per pulse and per bubble

$$\Delta n_{\mathrm{He,v}} < \Delta c_{\mathrm{He,v}} N_{\mathrm{b}} \Omega. \tag{9}$$

According to Eq. (9), a simple rescaling of Fig. 1 using Table 1 for $\Delta c_{\mathrm{He,v}}$, as is done in Fig. 6, indicates extremely small values, $\Delta n_{\mathrm{He,v}} \ll 1$ below 0.5 T_{m}, particularly for He atoms. This means that, under ESS (ADS) irradiation conditions, pulsing-induced fluctuations in the number of defects per bubble nucleus may be considered to be negligible and even meaningless since they would be completely masked by intrinsic fluctuations in the arrival of defects – provided bubble densities typical for continuous irradiation (of austenitic and martensitic/ferritic steels) are representative for pulsed irradiation conditions. Only above 0.5 T_{m}, i.e. beyond the applicability ranges of ESS and other ADS, SIA fluxes, pulsing with irradiation, could play a role in bubble nucleation. The upper bound estimate for $\Delta n_{\mathrm{He,v}}$ provides also an idea of how much the pulsing frequency would have to be reduced, at given average defect production rate, to get pulsing effects. For a more quantitative determination of such a limit, more accurate representations of the dynamic bias, Δ and δ, as given above would have to be evaluated.

An alternative aspect for judging the significance of pulsing for bubble formation is the number of pulses

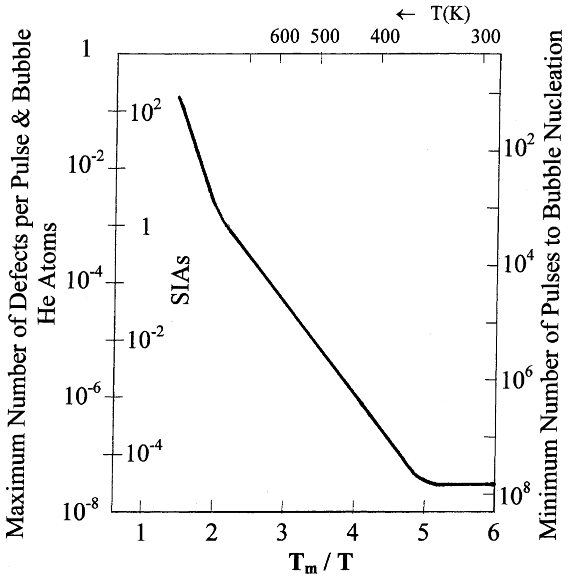

Fig. 6. Number of defects (He atoms and SIAs) per pulse and bubble (nucleus) (right abscissa) and minimum number of pulses to bubble nucleation (left abscissa) vs. temperature, obtained by rescaling bubble densities of Fig. 1.

required to reach typical bubble densities, n_{p} which is, of course, related to the number of He atoms per pulse and per bubble, Δn_{He}. Assuming that as few as two He atoms in a vacancy (or vacancy complex) form a stable bubble nucleus we find a lower bound estimate

$$n_{\mathrm{p}} > 2 N_{\mathrm{b}} \Omega \Delta c_{\mathrm{He}} = 2/\Delta n_{\mathrm{He}}. \tag{10}$$

Using this relation in conjunction with the values plotted in Figs. 1 and 6, we may conclude that, at least below 0.5 T_{m}, i.e. at temperatures applied in ESS and ADS, a very large number of pulses ($>10^3$) is required to reach significant bubble densities. This indicates that, in this temperature range, bubble formation is, effectively, a cumulative process which is not expected to be sensitive to pulsing.

The latter conclusion is supported by the following arguments. The formation of stable bubbles appears to be the result of placing and redistributing He atoms into and between vacancies and vacancy complexes, respectively. This is obviously a stochastic process equivalent to a Monte Carlo game which may be arbitrarily interrupted and continued without any effect on the result. Consequently, bubble formation is essentially a pulsing insensitive cumulative process.

The arguments against pulsing effects on bubble formation based on the number of defects provided per pulse and bubble can also be applied to the transformation of bubbles to voids under a vacancy supersaturation and are even stronger in this case since the fluctuation in the number of defects relative to the defects contained in a bubble is, of course, much smaller in bubbles close to that transformation, and even more so in the resulting faster growing voids, than in atomistically small bubble nuclei. We may conclude that irradiation pulsing in ESS and other ADS does not affect the evolution of cavities in any of its phases and, consequently also not the associated overall swelling and irradiation hardening by cavities.

For He production rates substantially lower than under ESS irradiation conditions, cavity or void nucleation at typical void swelling temperatures (between 0.4 and 0.5 T_m), could be essentially controlled by vacancy clustering driven by vacancy supersaturation which would have to be considered sensitive to pulsing even for the pulsing time scales of ESS and other ADS. A detailed treatment of cavity nucleation under such conditions, for instance by the method of meta-stable and unstable limiting cycles [20] is much more complicated than our above procedure – but, fortunately, not needed here.

4.3. Does pulsing affect irradiation creep?

Irradiation creep is basically due to climb of dislocations by some kind of bias in the absorption of SIAs and vacancies. In detail, several mechanisms of irradiation creep may be distinguished (see [9] for review). Amongst these mechanisms, the climb-induced release of a dislocation from an obstacle such as a precipitate, defect cluster or cavity, allowing it to glide to another obstacle, is the most pulsing sensitive mechanism which we consider therefore in the following.

We may use the same strategy as for cavity formation to judge the sensitivity of this 'climb controlled glide' to pulsing-induced fluctuations in the defect fluxes. An upper bound estimate for the mean-climb amplitude of an edge dislocation, d_{cl} is obtained by assuming that all SIAs produced in one pulse are instantaneously, homogeneously and completely distributed over all existing dislocations, meaning $\delta \approx 1$; $k^2 \to \rho$ in Eq. (8):

$$d_{cl} < \Delta c_i / b\rho, \tag{11}$$

where b is the Burgers vector. Using for Δc_i the value given in Table 1, we find that $d_{cl} \ll b$ for $\rho > 10^{12}$ m^{-2}, i.e. for all dislocation densities given in Fig. 2. Thus, the pulsing-induced mean-climb amplitude is atomistically small and therefore insignificant under ESS and similar ADS irradiation conditions. For a given average defect production rate, the mean-climb amplitude would become significant only at pulsing frequencies orders of magnitude lower than in ESS (ADS).

In order to rule out pulsing effects on creep by climb controlled glide we have also to examine the effect of fluctuations in the climb amplitude of dislocation. For a spatially random defect production, such fluctuations are negligibly small as long as the mean-climb amplitudes are atomistically small. Large intrinsic point defect fluctuations as occurring in cascades are required to get significant fluctuations in climb amplitudes. As has been shown already in Section 3.1, the mean distance between cascades of one pulse in ESS (ADS) is so large, however, that such cascades act independently and, consequently, in the same way as if they were produced continuously over the whole pulsing period. This conclusion is even strengthened in the case of creep by climb controlled glide since the mean distance between cascades of one pulse acting on a certain dislocation (estimated to be several hundred μm under ESS and similar conditions) is even significantly larger than the mean distance between cascades in the whole volume. Thus, we may conclude that pulsing in ESS (and similar ADS facilities) does not affect creep by climb controlled glide. The same conclusion holds, of course, for less pulsing sensitive radiation creep mechanisms.

5. Summary and conclusions

In the course of planning high power spallation sources for various purposes, the question arose whether the pulsed nature of the bombarding protons and neutrons would lead to radiation damage different from that known from continuous sources. If this were so,

1. the enormous database accumulated in fission and fusion materials research could hardly be transferred to the spallation case, and
2. irradiation facilities supplying pulsed neutrons would be of little value for fission and fusion applications.

In the present paper, we have examined this question for structural metallic materials in spallation targets. In order to quantify the discussion, we have used, as an example, parameters typical for stainless steels considered for the planned ESS. However, the conclusions should also hold for other ADS facilities in the MW power range using martensitic/ferritic steels instead of stainless steels – except perhaps close to the highest temperature considered for such facilities.

Since a full-scale ab initio modelling of radiation damage phenomena is not possible, we have based our considerations of possible pulsing effects on typical microstructural data for comparable continuous irradiation.

On this basis, we have shown:

1. The relations between the lifetimes of mobile SIAs and vacancies in such structures and the time scales of pulsing, quantified by a 'relative dynamic bias', are in favour of pulsing effects.
2. The damage yield of one pulse, controlling the 'absolute dynamic bias', is much too small, however, to allow any significant pulsing effect, even on processes considered to be pulsing sensitive such as cavity

nucleation and creep by climb controlled dislocation glide.

Doubts concerning the use of microstructural data typical for continuous irradiation can be ruled out safely: important processes in the microstructural evolution such as the generation of stable dislocations loops of SIA type in cascades, their coalescence and growth into the dislocation network are essentially cumulative and therefore insensitive to the temporal structure of irradiation. We have argued that, at the temperatures of interest, even cavity formation is essentially a cumulative process. On the other hand, for the structures assumed, the damage yield of one pulse in ESS and similar facilities is so far away from being sufficient for significant pulsing effects in the temperature range considered for ESS applications, that changes of the microstructural data by even one or two orders of magnitude would not change the conclusion.

It is therefore justified to answer the question formulated in the title of this paper with a clear 'no'.

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