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Conditions for effects of radiation pulsing

H. Trinkaus *, H. Ullmaier

Forschungszentrum Jülich, Institut für Festkörperforschung, ESS-Projekt, 52425 Jülich, Germany

Abstract

The possibility of pulsing effects on radiation damage is due to differences in the delay times of relevant defect reactions and/or to the non-linear dependence of such reactions on defect production rates. Thus, significant pulsing effects require (1) proper relationships of the internal time scales of defect production and reaction to the time scales of pulsing and (2) sufficiently large pulsing induced fluctuations in relevant microstructural variables. We show that the first condition, which we quantify by a 'relative dynamic bias', is indeed fulfilled in wide ranges of the main irradiation parameters. The second condition, quantified by an 'absolute dynamic bias', is, however, found to restrict the parameter ranges of possible pulsing effects substantially. For planned spallation neutron sources and similar accelerator driven systems facilities we find, for instance, that, in the temperature range of interest, the defect yield of one pulse (controlling the absolute dynamic bias) is much too small to allow any significant pulsing effect. We introduce and discuss maps for the occurrence of significant pulsing effects in the space of the main irradiation parameters.

1. Introduction

Most experimental and theoretical studies of radiation damage in metals and alloys consider the damage-generating irradiation to be homogeneous and continuous. Irradiation is or will be not continuous, however, in projected magnetic or inertial confinement fusion reactors (MCFR/ICFR), spallation neutron sources (SNS) and other accelerator driven systems (ADS). In spite of this, only a few theoretical studies on possible pulsing effects have been published [1,2].

As are radiation damage processes on the whole, possible effects of the temporal structure of the irradiation are controlled by the production, diffusion and reaction of the mobile defects. Defect reactions occur delayed with respect to their production and the corresponding delay times are different for different defect reactions. Most defect reactions depend non-linearly on defect concentrations (second and higher order reactions) and, because of this, on defect production rates. In fact, differences in the delay times and/or non-linearities in the rate dependence of defect reactions represent the two reasons for possible pulsing effects on radiation damage.

Accordingly, crucial aspects in discussing possible pulsing (cycling) effects are (1) the relation of the time scales of the externally imposed irradiation (on- and off-times) to the internal time scales of the production, diffusion and reaction of the irradiation induced defects, and (2) the sensitivity of certain processes to fluctuations in the concentrations and absorption rates of mobile defects. Thus, some microstructural processes are insensitive, while other ones are sensitive to such fluctuations. The accumulation and growth of stable clusters produced in displacement cascades belongs to the first class, whereas cavity nucleation and creep by climb controlled dislocation glide may be considered to belong to the second.

Recently, we have shown that even (non-linear) defect fluctuation sensitive processes are not significantly affected under the pulsing conditions of planned SNS, such as the European version ESS, and similar ADS facilities where, in the temperature range of interest, the damage yield of one pulse or period (controlling the 'absolute dynamic bias') is much too small to allow any

* Corresponding author. Tel.: +49-2461 616460; fax: +49-2461 612620.

E-mail address: h.trinkaus@fz-juelich.de (H. Trinkaus).

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significant pulsing effect [3]. In the present contribution, we examine necessary conditions for the occurrence of pulsing effects on radiation damage in structural metallic materials in dependence upon the main irradiation parameters (pulse length and frequency, peak or average damage rates, temperature).

Concerning the material, we have primarily austenitic stainless steels in mind (for which we take Ni to be representative concerning defect properties) 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.

2. Conditions for pulsing effects in terms of dynamic bias

2.1. Temporal scales in defect reaction kinetics

For the irradiation conditions of interest here, the lifetimes of the mobile radiation induced defects are diffusion controlled. For sink dominated defect annihilation, the mean lifetimes, τ, of a certain type of a mobile defect is given by [3,4]

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

where D is the diffusivity of the defect considered and k^2 is the sink strength for its annihilation. According to Eq. (1), the sink strength k^2 is a key quantity in the 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 (with a dose rate comparable with the average dose rate of the pulsed irradiation). This is justified since a substantial part of the defect evolution is essentially cumulative. Concerning the evolving bubble structure, we take into account here, differently from our previous analysis for ESS [3], the effect of the (average) helium production rate, P_{He} on the bubble density, N_{b}, assuming that N_{b} scales as $P_{\mathrm{He}}^{1/2}$ [5,6], since P_{He} is different for the different devices which we want to compare (ESS and ADS: $\langle P_{\mathrm{He}}\rangle \approx 3\times10^{-10}$ s^{-1}; MCFR and ICFR: $\langle P_{\mathrm{He}}\rangle \approx 10^{-11}$ s^{-1}).

In the temperature range of interest here (see below), possible pulsing effects are due to the fact that self-interstitial atoms (SIAs) diffuse substantially faster and arrive earlier at sinks than vacancies. In Fig. 1, the lifetimes of SIAs and vacancies are plotted vs. reciprocal homologous temperature, T_{m}/T (T_{m}: melting temperature), on the basis of the sink strengths used in Ref. [3] (where recombination is taken into account). The large difference between the lifetimes of SIAs and vacancies is evident. It should be noticed, that in this respect, small SIA and vacancy clusters, the lifetimes of which are also diffusion controlled between 0.2 and 0.4 T_{m} behave similarly as single SIAs and vacancies.

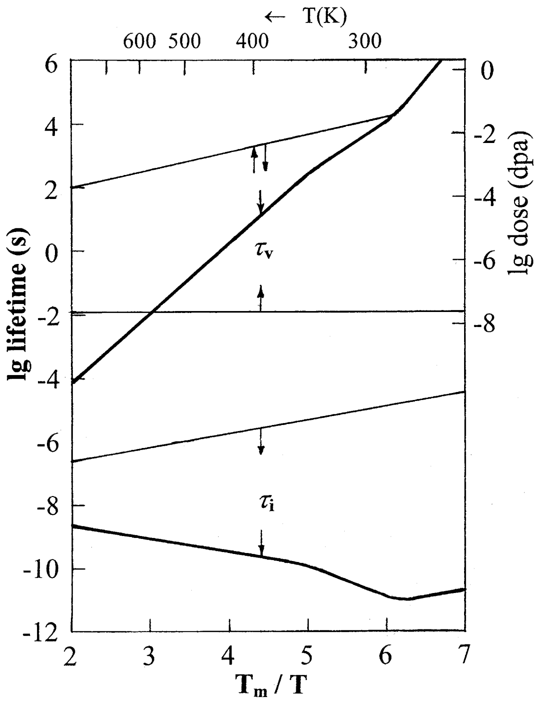

Fig. 1. Schematic plot of logarithms of lifetimes of SIAs and vacancies for initial and 'developed' structures, respectively, vs. reciprocal homologous temperature, used in Ref. [3] for discussing possible pulsing effects in ESS. For Ni (stainless steels) the corresponding absolute temperature scale is shown on the upper axis. Arrows indicate evolution of total sink strength [3].

2.2. Dynamic bias

For pulsing effects, the time dependencies of the 'defect fluxes' $D_{\mathrm{i}}c_{\mathrm{i}}$, and $D_{\mathrm{v}}c_{\mathrm{v}}$ and their difference, the 'excess defect flux' $D_{\mathrm{i}}c_{\mathrm{i}}$–$D_{\mathrm{v}}c_{\mathrm{v}}$ are crucial quantities. In Fig. 2(a) and (b), these dependencies are sketched for pulsing conditions for which the difference between the diffusivities of the two defects is relevant.

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 $D_{\mathrm{i}}c_{\mathrm{i}}$–$D_{\mathrm{v}}c_{\mathrm{v}}$ over time as illustrated in Fig. 2(c). We define the amplitude of this quantity as the absolute dynamic bias

$$\Delta = \mathrm{Ampl}\left\{ \int (D_{\mathrm{i}}c_{\mathrm{i}} - D_{\mathrm{v}}c_{\mathrm{v}})\,\mathrm{d}t \right\}. \tag{2}$$

The value of Δ depends, via $D_{\mathrm{i,v}}c_{\mathrm{i,v}}$ on defect production rate and total sink strength. The pulsing induced fluctuation amplitudes of the excess number of SIAs/

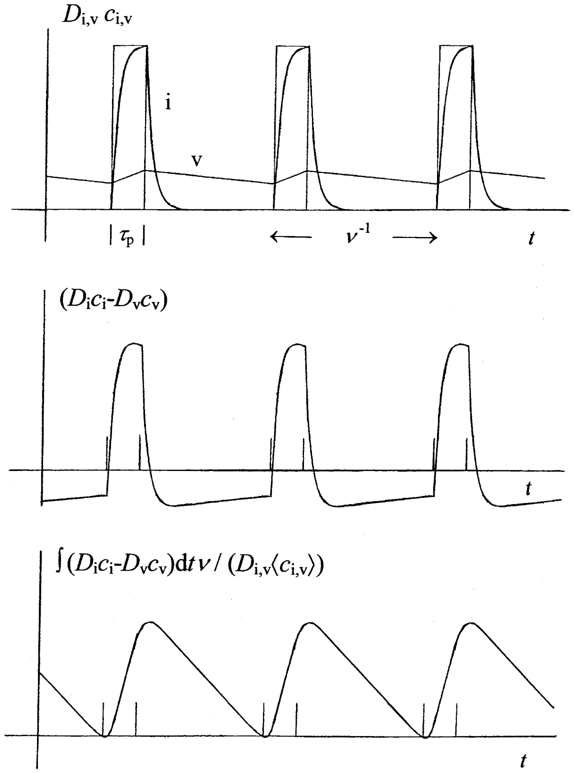

Fig. 2. Sketch of time dependence of (1) SIA and vacancy fluxes (top), (2) difference of both fluxes (middle), and (3) time integral of flux difference (bottom) induced by cyclic pulsed irradiation for $P_i = P_v$, $t \gg \tau_v$.

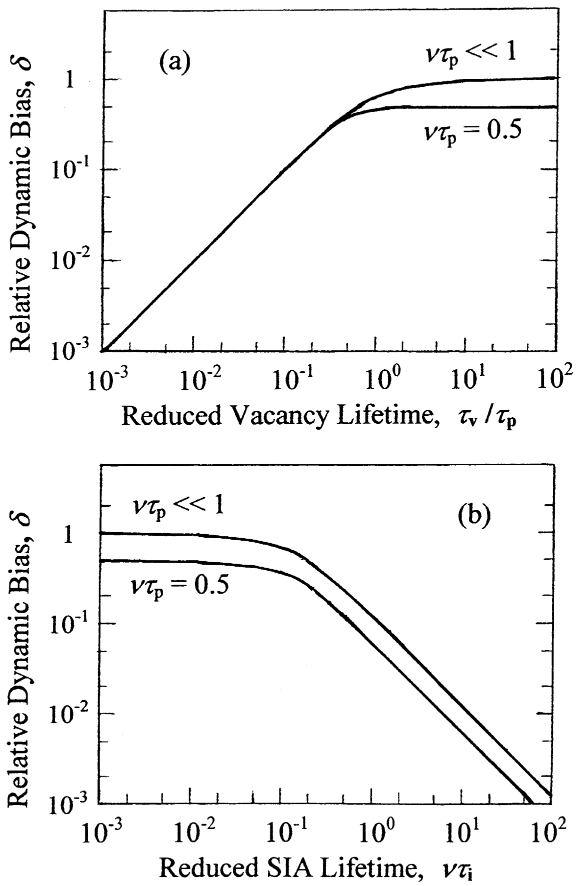

Fig. 3. Relative dynamic bias, δ, vs. normalised lifetimes of vacancies and SIAs, τ_v/τ_p and $\nu\tau_i$, for moderate and extreme cyclic pulsing, $\nu\tau_p = 0.5$ and $\nu\tau_p \ll 1$, respectively.

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.

It is useful 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'

$$\delta = \Delta\nu/(D_{i,v}\langle c_{i,v}\rangle). \tag{3}$$

For the case of sink dominance, this quantity has the advantage to depend explicitly only on the external and internal time scales of pulsing/cycling and defect diffusion (pulse length, τ_p, pulsing period ν^{-1} and defect lifetimes $\tau_{i,v}$), but not explicitly, as Δ, on defect production rate and total sink strength. In Fig. 3, δ is plotted vs. τ_v/τ_p and $\nu\tau_i$ for $\tau_i \ll \tau_v$ and for moderate and extreme pulsing, $\nu\tau_p = 0.5$, and $\nu\tau_p \ll 1$, respectively. In these cases, significant values of $\delta \approx 0.5$ and 1, respectively, are reached for $\tau_v/\tau_p > 2$ and $\nu\tau_i < 0.05$. Note that $\nu\tau_i < 0.05$ is fulfilled for all irradiation conditions considered here.

A sufficiently large value of the relative dynamic bias, say $\delta > 0.5$, represents, however, only a necessary but not a sufficient condition for noticeable pulsing effects. Significant fluctuations in sizes of clusters and climb position of dislocations or other microstructural quantities 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 and thus may be written as

$$\Delta = \delta D_{i,v}\langle c_{i,v}\rangle/\nu < \delta\langle P_{i,v}\rangle k^2\nu, \tag{4}$$

where $\langle P_i\rangle \approx \langle P_v\rangle$ ($\approx 10^{-7}$ s^{-1}) are the production rates of SIAs and vacancies, respectively. The expression on the right hand side of Eq. (4) represents an upper bound estimate for Δ since recombination is neglected there. This estimate depends linearly on the concentration of SIAs/vacancies produced in one pulse or period (the 'defect yield per pulse'), $\Delta c_{i,v} = \langle P_{i,v}\rangle/\nu$. According to Eq. (4), 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 rates and temperature (defining δ and k^2), for instance, pulsing effects would be limited to pulsing frequencies ν below a certain maximum value.

3. Applications

Processes to be considered here are those which are sensitive to fluctuations in the excess fluxes of SIAs or vacancies. Prominent examples are cavity nucleation and creep by climb controlled dislocation glide.

3.1. Conditions for pulsing effects on cavity nucleation

Under the concurrent production of He atoms and displacement defects, cavity formation is initiated by bubble nucleation from small He-vacancy complexes [5,6] which can be sensitive to pulsing under certain conditions to be examined here. 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. In the temperature range of interest, bubble nuclei are most sensitive to the absorption of SIAs. Significant pulsing effects require, in addition to a substantial value of δ, that the pulsing induced fluctuation in the number of vacancies per bubble nucleus is at least of the order of 1 since, for smaller fluctuations, the temporal correlation with the cyclic pulsing is lost. Using Eq. (4) we may express these conditions (somewhat more general than in Ref. [3]) as

$$\delta > 0.5, \tag{5a}$$

$$(4\pi r_0/\Omega)\delta\langle P_{i,v}\rangle/k^2\nu > \Delta n_{i,v} > 1, \tag{5b}$$

where r_0 is the absorption radius of a bubble nucleus. For given average defect production rates, dose, He concentration, c_{He}, and temperature (defining δ and k^2), Eqs. (5a) and (5b) define the frequency range where pulsing effects can be significant, Eq. (5a) its lower, Eq. (5b) its upper boundaries. In Fig. 4, such ranges are plotted vs. reciprocal homologous temperature for $\langle P_{i,v}\rangle = 10^{-7}$ s^{-1}, $c_{He} > 10^{-4}$, and otherwise for duty factors $\nu\tau_p$ and average He production rates, $\langle P_{He}\rangle$, characteristic for fusion reactors and spallation sources ($\nu\tau_p = 0.5$ for MCFR; $\nu\tau_p = 5\times10^{-5}$ for ESS/short pulse; $\langle P_{He}\rangle = 10^{-11}$ s^{-1} for MCFR/ICFR; $\langle P_{He}\rangle = 3\times10^{-10}$ s^{-1} for ADS/ESS).

This significance map confirms our earlier conclusion that the high frequency of 50 Hz in ESS and other ADS facilities does not allow any pulsing/cycling effect on bubble nucleation, simply because of the extremely small defect yield per pulse at such high frequencies [3]. Note that this argument is rather general since it applies to any other irradiation induced nucleation process, and even to mutual recombination of SIAs with vacancies for which k^2 in Eq. (5b) may be interpreted as the sink strength of the slow vacancies for the annihilation of the fast SIAs [3].

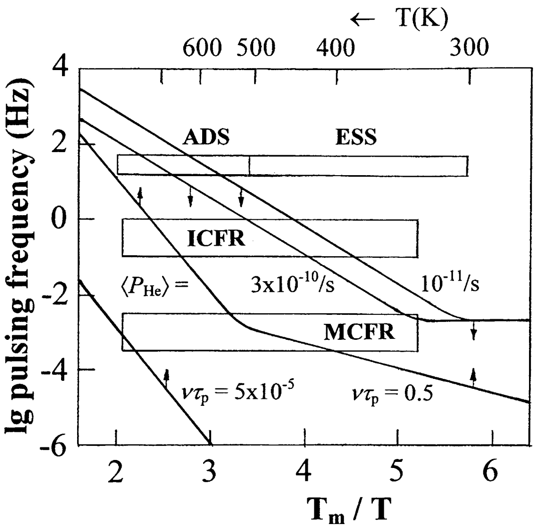

Fig. 4. Map in the frequency/temperature space for the occurrence of significant pulsing effects on bubble nucleation assuming duty factors, $\nu\tau_p$, and average He production rates, $\langle P_{He}\rangle$ characteristic for fusion reactors and spallation sources ($\nu\tau_p = 0.5$ for MCFR; $\nu\tau_p = 5\times10^{-5}$ for ESS/short pulse; $\langle P_{He}\rangle = 10^{-11}$ s^{-1} for MCFR/ICFR; $\langle P_{He}\rangle = 3\times10^{-10}$ s^{-1} for ADS/ESS). The lower and upper bounds are defined by the conditions of significant relative and absolute 'dynamic bias', respectively. Arrows point into the range of pulsing effects. The map shows that the high frequency (50 Hz) in ESS and other ADS facilities does not allow significant pulsing effect on bubble nucleation. Cyclic irradiation pulsing in fusion reactors could have some effect on bubble nucleation, which is, however, negligibly small for the aspired small off-times compared to the on-times.

Irradiation pulsing/cycling in fusion reactors could have some effect on bubble nucleation. For the aspired condition of on-times being large compared to off-times, the expected effect becomes, however, negligibly small.

It is noticed here that, at elevated temperatures, direct void nucleation under an irradiation induced vacancy supersaturation without major assistance of gas would be more sensitive to pulsing than bubble nucleation. In this case (not interesting in the present context), the lifetime of void nuclei would be dissociation rather than diffusion controlled.

3.2. Conditions for pulsing effects on irradiation creep

The most pulsing sensitive creep mechanism is the release of dislocations from obstacles such as precipitates, defect clusters or cavities, by irradiation induced climb allowing them to glide to other obstacles [7].

Significant pulsing effects on this type of creep require, in addition to a substantial value of δ, that the pulsing induced climb amplitude of the dislocations, d_{cl} must be sufficiently large, $d_{cl} > d^*_{cl}$, depending on the size of the obstacles (>0.5 nm). Using Eq. (4) we may express these conditions (somewhat more general than in Ref. [3]) as

$$\delta > 0.5, \tag{6a}$$

$$d_{cl} = \delta \langle P_{i,v} \rangle / bk^2 v > d^*_{cl}. \tag{6b}$$

The analysis of these conditions shows that irradiation pulsing in fusion reactors, SNS or other ADS irradiation facilities does not significantly affect creep by climb controlled dislocation glide. The same conclusion holds, of course, for less fluctuation sensitive radiation creep mechanisms.

It can also be shown that the mean distances between displacement cascades produced during one pulse are so large that they act independently of each other and, consequently, in the same way as if they were produced continuously over the whole pulsing/cycling period. Thus, we may conclude that cyclic pulsing in fusion reactors, SNS or other ADS irradiation facilities does not significantly affect radiation creep.

4. Conclusions

In the present contribution, two important necessary conditions for significant effects of cyclic irradiation pulsing on radiation damage in metallic materials are discussed in terms of a relative and absolute dynamic bias, introduced to quantify the temporal separation of the reactions of SIAs and vacancies with relevant components of the microstructure (relative dynamic bias) and the pulsing induced magnitude of fluctuations in sizes or positions of such components (absolute dynamic bias), respectively. For otherwise fixed parameters (duty factors, average defect production rates, temperature) the relative and absolute dynamic bias define lower and upper limits of the frequency range where pulsing effects can be significant. Outside this range, the radiation damage resulting from pulsed irradiation is virtually the same as that occurring under continuous irradiation with defect production rates equal to the average rates of the pulsed irradiation.

Our criteria for significant pulsing effects are applied to two typical pulsing sensitive processes occurring under the irradiation conditions considered: cavity nucleation associated with He production and creep by climb controlled dislocation glide. The criteria are, however, more general and apply to other irradiation induced processes as well. The main results of our analysis are:

(1) In the temperature range of interest, the high pulsing frequencies in SNS and similar irradiation facilities do not allow any significant (non-linear) pulsing effect because of the extremely small defect yield per pulse or period at such high frequencies.
(2) Irradiation cycling in MCFR/ICFR could affect cavity nucleation at elevated temperatures. But the effects are most likely weak.

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