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Numerical Evaluation of Material's Degradation under Various Irradiation Conditions

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ABSTRACT

Structural materials during a use in a fission reactor are subjected to irradiation by high energy neutrons, and therefore the properties, functions, and performances of materials become gradually degraded. Since keeping material's integrity is a key to success towards a stable operation of reactors, materials degradation due to irradiation should be taken into account when reactor design and maintenance are considered. One of the issues to be solved for realization of a long term operation is how the integrity of materials in use is ensured for a long time beyond an actually-experienced operation period. To do this, the future ageing behavior of materials should be understood *in advance*. Usually, an attempt is made to understand the behavior using the existing irradiation facilities with accelerated irradiation environments. In many cases, however, material's behavior depends much on acceleration coefficients and its dependency is not so simple. To overcome such a difficulty, a numerical simulation study is employed. In the present study, reaction rate analysis is performed to investigate material microstructure changes under various irradiation conditions. Our results have shown that volume swelling due to irradiation is very much different depending on irradiation conditions provided by such irradiation facilities as HFIR, JOYO, KUR, 14MeV fusion neutrons. Based on the results, irradiation correlation methodology is discussed to predict material's degradation under arbitrary irradiation conditions using the actual existing data obtained with accelerated irradiation conditions.

KEYWORDS

Radiation damage, Rate theory, Defect cluster, Neutron spectrum, Irradiation correlation

ARTICLE INFORMATION

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1. Introduction

In fission reactors, component materials are exposed to irradiation by high energy neutrons, where athermal point defects (vacancies and self-interstitial atoms (SIAs)) are produced by atomic displacements. Those defects may diffuse freely in the material depending on temperatures, and form such clusters as voids (vacancy clusters) and dislocation loops (SIA clusters), which will cause microstructural changes and accompanying macroscopic property changes (e.g., volume swelling, ductility loss, and embrittlement). To keep the integrity of reactors, material degradation due to irradiation should be considered when reactor design and maintenance are considered.

In Japan, the operational period of fission reactors is restricted primarily less than 40 years by the safety regulation revised in 2012. The operational period can be extended until 60 years provided that reactor integrity is validated by performing so-called the special inspection. Since the operational experiences of Japanese commercial reactors are around 40 years at the longest, actual data of material degradation do not exist for longer irradiations than 40 years. Material's behavior due to such an unexperienced, longer term irradiation must be obtained by extrapolating the data obtained at other accelerated irradiation facilities.

Dpa (displacement per atom) is one of the most important parameters to characterize irradiation conditions. Dpa indicates an irradiation dose that is obtained using incident energy spectra, incident flux, and cross section of reactions between incident particles and target materials. Dose rate (dpa/s) is usually employed as an acceleration coefficient in the research field of materials irradiation.

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Unfortunately, however, dpa is not linearly correlated with actual materials degradation due to irradiation, which cannot always be a good index for describing materials degradation. Namely, even when dpa values are identical, microstructural and mechanical property changes are different depending on irradiation facilities employed. In the present study, a relationship between dpa (or dpa/s) and material's microstructure in ferritic steel is theoretically evaluated, where defect production and cluster formation processes are simulated using the reaction rate theory equations.

2. Quantification of irradiation condition

2.1. Numerical method

Various irradiation facilities were quantified by the SPECTER code [1], which can calculate the dpa rate, PKA (primary knock-on atom) energy spectra, gas production rate, and total damage energy (Kerma) for 41 material elements, using the input data of incident neutron energy spectra ranging from 10^{-10} to 20 MeV. All nuclear reaction cross-section data was obtained from ENDF/B-V distributed by Brookhaven National Laboratory. The target element for the evaluation is iron, which is a major element of steel.

2.2. dpa rate of various irradiation facilities

The dpa rates were calculated by the SPECTER code for such existing irradiation facilities as HFIR (The High Flux Isotope Reactor at ORNL), JOYO (Experimental fast reactor at JAEA), KUR (Kyoto University Reactor), JMTR (Japan Materials Testing Reactor), and 14 MeV fusion neutrons.

Figure 1 shows the dpa rate as a function of neutron flux for various irradiation facilities. As shown in the figure, neutron flux and the corresponding dpa rate are quite different depending on irradiation facilities. When the data of the core region of reactors is however focused, the dpa rate is in proportion to the neutron flux. By means of the least squares method, a relationship between them is obtained as follows;

$$P = 7.9 \times 10^{-26} \phi ,$$

where P is the dpa rate in dpa/s, ϕ is the neutron flux in n/m^2/s. In the following section, the influence of dpa rate on microstructural changes in steel is discussed based on the rate theory analysis.

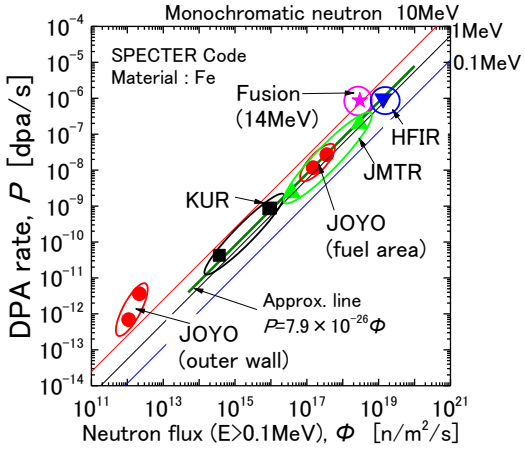

Fig. 1 DPA rate (dpa/s) as a function of neutron flux for various irradiation facilities

3. Simulation of defect cluster formation process

3.1. Reaction rate theory model of defect accumulation processes

We have developed the defect cluster formation model on a basis of the reaction rate theory, where defect energetics obtained by the molecular dynamics study [2] was employed. With the energetics thus employed, the nucleation process of defect clusters is appropriately described and hence, for example, the critical size for nucleation, an important parameter in the nucleation theory, can be defined dynamically during calculations and appropriately incorporated into the model, although the conventional nucleation model [3] was described with only a simple, inflexible assumption. Model assumptions employed here are as follows.

- Only Frenkel pairs are produced by atomic displacements.
- Only single vacancies and self-interstitial atoms (SIAs) are mobile.
- An SIA cluster is immobile.
- A vacancy and an SIA are mutually annihilated by recombination reaction.
- An interstitial-type dislocation loop (an SIA cluster) has disk shape, and a void (vacancy cluster) has spherical shape.
- Dislocation lines play a role as a permanent sink for mobile point defects.
- The dislocation bias effect is considered in which a dislocation line prefers absorbing SIAs to absorbing vacancies. Dislocation loops created during irradiation have also the bias effect.

Using the assumptions above, reactions of point defect diffusion, defect clustering, recombination reaction, and absorption to sinks were considered. Rate equations were simultaneously solved using the Gear method [4] to obtain time evolution of defect clusters. The defect clusters created during irradiation were followed for every cluster size of up to a 500-member cluster. The clusters greater than the 500-member cluster are all classified into one group category, where only the average concentration and average size are defied for the group. Note that, in order to understand the effect of dpa rate clearly, so-called the effect of cascade clusters on microstructural evolution was regarded beyond the scope of the present study. The cascade cluster effect is under investigation and published in the near future. In addition, the effect of 1D-motion of SIA clusters is not also considered here because our target is ferritic steel that contains a lot of impurity atoms which may play a role as an obstacle for the movement [5].

3.2. Time evolution of defect concentrations

Figure 2 shows the time evolution of defect concentrations when the dpa rate is fixed at 1×10^{-8} dpa/s and temperature is 573 K. In the early stage of irradiation (around 10^{-5} s), the concentrations of

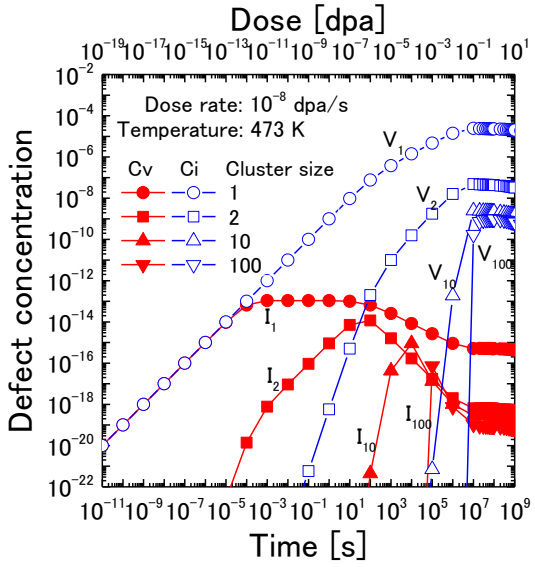

Fig. 2 Time evolution of defect concentration for 1×10^{-8}dpa/s and 573 K

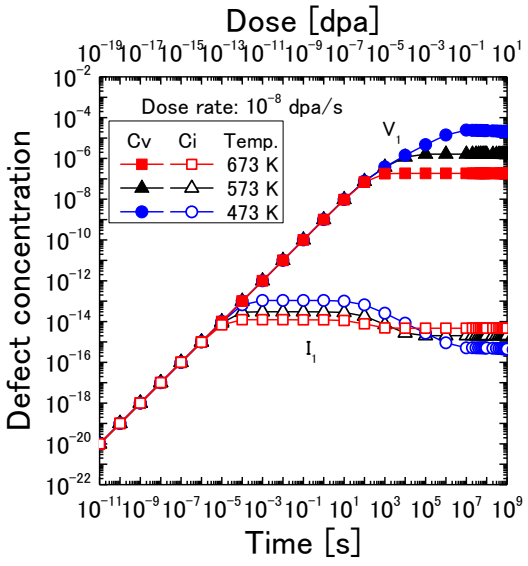

Fig. 3 Time evolution of point defect concentration at the temperature of 473 K, 573 K, and 673 K.

point defects increase by atomic displacements. And then, SIAs are absorbed by dislocation lines and dislocation loops start to form at 10^{-5} - 10^{1} s. Finally, after voids are formed at 10^{-1} - 10^{7} s, defect concentrations are saturated.

Figure 3 shows the time evolution of point defect concentrations at temperatures of 473 K, 573 K, and 673 K. The concentrations vary depending on temperature, even when the dpa rate is fixed at the same value. Vacancy concentration increases with decreasing irradiation temperature at the late stage of irradiation. This is because defect annihilation at a permanent sink takes less frequently due to less mobility of point defects under lower temperatures.

3.3. Vacancy supersaturation and void critical size

In order to understand defect accumulation processes in more detail, defect diffusion flux in the material matrix was defined by the product of diffusion coefficient (D) and defect concentration (C), where the diffusion coefficient is expressed by

$$D \propto \exp\left(-\frac{E_m}{kT}\right).$$

Here, D is the diffusion coefficient, E_m is the migration energy, T is temperature, k is the Boltzmann constant. Using the defect diffusion flux defined here, vacancy supersaturation was defined by a difference between the fluxes of vacancies and SIAs in the material matrix, as follows;

$$S_V = \frac{D_V C_V - D_I C_I}{D_V C_V^{eq}},$$

where C_V^{eq} is the equilibrium vacancy concentration. The subscript V and I denotes a vacancy and an SIA, respectively. When the value of vacancy supersaturation is positive, the vacancy flux is greater than the SIA flux, in which voids grow easily.

Figure 4 shows the time evolution of vacancy supersaturation. In the early stage of irradiation, the vacancy supersaturation is negative, because the mobility of an SIA is higher than that of a vacancy. And then, the vacancy supersaturation turns to positive by the effect of dislocation bias, in which voids start to nucleate and grow up stably. The figure shows that it takes longer time to reach positive values when the mobility of point defects is lower at lower temperatures. Figure 5 shows the temperature dependence of vacancy supersaturation as a function of dpa rate. The dose is fixed at 1 dpa. At low temperatures the vacancy supersaturation takes a negative value. In contrast, the vacancy supersaturation is positive at high temperatures, where the values slightly decrease with increasing temperature.

The vacancy supersaturation is only an index that presents a balance between the influxes of vacancies and SIAs to a void. In order to understand void nucleation process precisely, the thermal stability of voids should also be considered. When those considerations are made, the critical size of void nucleation is evaluated as a function of temperature and dpa rate, as shown in Figure 6. The critical size is an index showing how easy void nucleation takes place. When temperature is larger, the critical size becomes larger, indicating that a void is difficult to nucleate.

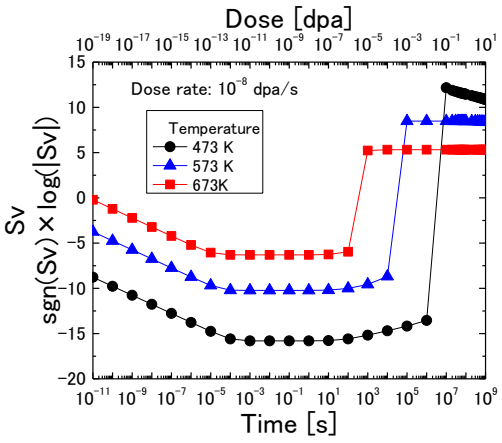

Fig. 4 Time evolution of vacancy supersaturation

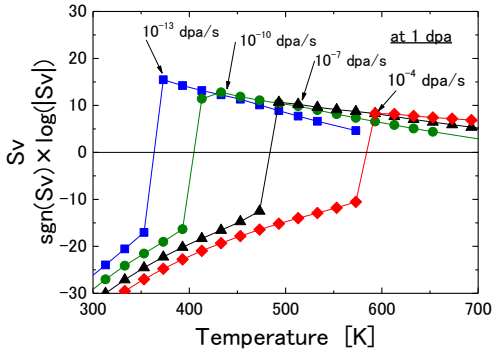

Fig. 5 Temperature dependence of vacancy supersaturation as a function of dpa rate

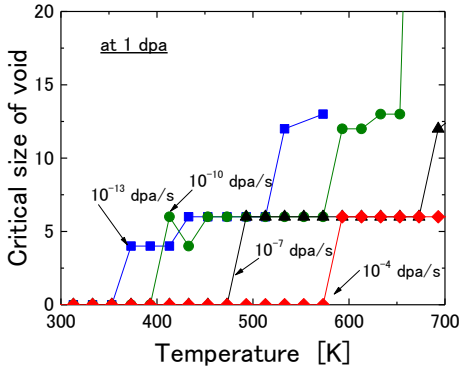

Fig. 6 Temperature dependence of void critical size for nucleation as a function of dpa rate

3.4. Average diameter and number density of voids, and volume swelling

Figure 7 shows temperature dependence of void number density as a function of dpa rate. As shown in the figure, the number density of voids has a peak against temperatures. At the lower temperature side, a void hardly nucleates due to low mobility of vacancies. While, at the higher temperature side, a void is thermally unstable in which the critical size is larger. Figure 8 shows the temperature dependence of void average diameter as a function of dpa rate. The diameter becomes larger when temperature increases. At a given temperature, the diameter is larger when the dpa rate decreases. The results may indicate that a small number of large voids are created under high temperatures, while a large number of small voids are formed to be distributed in the material at low temperatures. This behavior is qualitatively consistent with experiments [6].

The swelling was calculated using the following equation,

$$S = \bar{i} \times \rho_{void},$$

where S is the calculated swelling, $\bar{i}$ is the average size of voids, ρ_{void} is the number density of voids. Notice that, a void in consideration here is restricted for a relatively large void with diameter greater than 1 nm.

Figure 9 shows the calculated void swelling as a function of temperature for various dpa rates. Again, the swelling has a peak against temperature. When temperature is low enough, defect diffusivity is too small, and hence, a void cannot form. With increasing temperatures, the diffusivity gradually increases and a void becomes easy to be created. However, when temperature becomes too high, the thermal stability of voids becomes decreased, which results in less formation of voids. Thus, the peak is formed by a balance between the defect diffusivity and void thermal stability. Interestingly, moreover, the peak temperatures depend on the dpa rate as shown in the figure. The peak shifts to the higher temperature side when the dpa rate increases. This is reflected by the fact that above balance is changed depending on the dpa rate. Since the dpa rate represents an acceleration coefficient that is a characteristic parameter of irradiation facilities, the peak shift shown here should be considered when accelerated irradiation is performed.

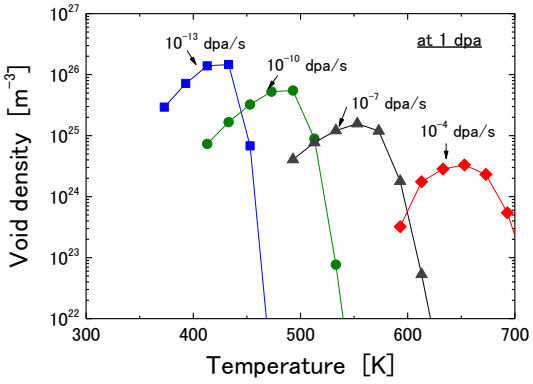

Fig. 7 Temperature and DPA rate dependence of number density of voids

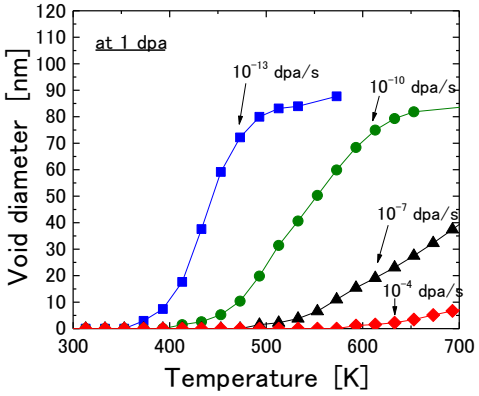

Fig. 8 Temperature and dose rate dependence of the void average diameters

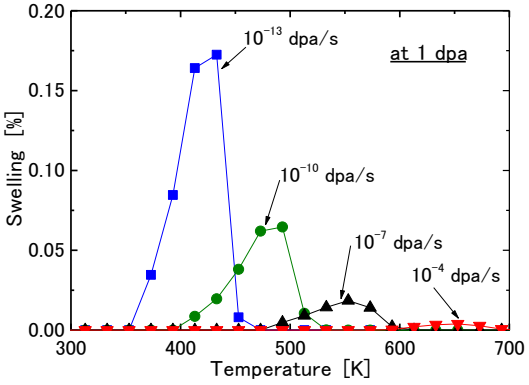

Fig. 9 Void swelling as a function of temperature for various DPA rate ranging from 10^{-13} dpa/s to 10^{-4} dpa/s.

4. Conclusion

Numerical simulations have been performed to clarify a relationship between irradiation conditions and material degradation behavior. The results are as follows;

1. We developed numerical models for microstructural evolution due to irradiation, which contain the detail process of defect cluster nucleation.
2. In the early stage of irradiation, the vacancy supersaturation takes a negative value. And then, the vacancy supersaturation becomes positive due to dislocation bias effect, when voids start to nucleate and grow.
3. The critical size of void nucleation increases with increasing temperature. When temperature is fixed, the critical size increases with decreasing the dpa rate. The critical size is determined by a balance between the vacancy supersaturation and the thermal stability of voids.
4. The void swelling has a peak against irradiation temperature, which shifts to the higher temperature side when the dpa rate increases.

From these understandings obtained here, the following suggestions should be considered when reactor design and maintenance are performed. Namely, when material degradation is evaluated by accelerated irradiation facilities, it is necessary to consider a difference in peak temperatures for void swelling.

Acknowledgement

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