Nuclear Data for Analysis of Radiation Damage Processes

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Parameters needed to analyze radiation damages for neutron irradiations are presented, taking iron samples irradiated with JMTR neutrons for an example. Special interests have been put on a comparison between results obtained by irradiations for one case with a full neutron spectrum and the other with a Cd-shielded neutron spectrum. A possibility is described that although atomic displacement rates for the two case differ only less than 2 %, production rates of freely migrating defects can differ appreciably, due to recoiled atoms by (n, γ) reactions. More over, it is also suggested that although the median energy of PKA, defined as a PKA energy above (or below) which one half of the total atomic displacements are to be produced, may differ only slightly between the two cases, final radiation effects can be significantly different.

The effects of charged particles emitted with high energies due to nucleon irradiations are stressed in relation to the significance of defects produced by PKAs with lower energies than several keV, especially for the case of irradiations with highly energetic nucleons as anticipated in GeV proton irradiations.

1. Introduction

Nuclear data has been playing important roles in predicting radiation damages of materials to be used in nuclear energy facilities, through giving the atomic density of atoms displaced from normal lattice sites or displacements per atom (dpa) and concentrations of foreign atoms produced by nuclear reactions, mainly gas atoms like helium and hydrogen atoms. The method for calculating the dpa due to collisions in cascades originating from a single knock-on atom produced by reactions with irradiation neutrons have been established and have been written in ASTM standard[1]. That is, once energy spectra of PKA are given, the dpa is calculated based on LSS theory[2] on the partition of PKA energies between electronic excitation energies and kinetic energies of nuclear motion of atoms. In the calculations, an energy partition function called a damage efficiency expressed by Robinson[3] in a fitted equation of reduced PKA energy has been used widely in common. So, the method for calculating dpa is called the NRT model[4].

Present study has been motivated by the possibility that under a certain condition neutron irradiations could bring about quite different consequences upon irradiation effects, even if displacement rates did differ less than a few per cent. The possibility is originating from the fact informed[5] concerning in-reactor irradiation creep experiments performed in Japan Material Testing Reactor (JMTR), in which the irradiation creep rate for the specimen irradiated with the full neutron spectrum has been considerably enhanced by over one order, as compared with that observed for the specimen irradiated with the spectrum, the thermal region of which was cut by appropriate cadmium shieldings with the displacement rates, dpa/s, for the both cases being almost comparable and other conditions pertinent to this kind of experiments being the same, such as the irradiation temperatures and the levels of the applied stress. Although details of the experimental procedures and the results are to be soon published, the significances of the PKA spectra which have recently been recognized and demonstrated experimentally in the context of production rates of freely migrating point defects surviving displacement cascades, therefore closely related with nuclear data, should be examined beforehand, in the present study.

2. Damage parameters and PKA energy spectra

Neutron spectra used to obtain damage parameters for the both cases are given in Ref.[6] and the respective damage parameters are given in Table 1. Although the sample material used actually in the in-reactor irradiation creep experiment is Type 304 stainless steel with the standard composition of chromium(18.5 in mass %), nickel(9.1) and iron(the rest balanced approximately) as major elements, an elemental iron (Fe) is used as an irradiated sample for the present calculations. This simplification will not introduce significant errors on the final consequences, so long as the irradiation time is as short as a few hundreds of hours like the present case.

Note that an apparent large difference in displacement(dpa) cross sections between the two spectra is entirely due to the difference in the total neutron fluxes used in averaging the cross sections, the values of dpa/s differ merely 2 %, where dpa/s for the full spectrum case is larger by the contribution of (n,γ) reactions with the thermal neutrons below 1 eV.

Also given in Table 1 are indexes for PKA spectra, $T_{1/2}$(dpa) and average T in keV. The former is called a median PKA energy, defined as a PKA energy above (or below) which one half of the total atomic displacements are to be produced, while the latter is an energy averaged over the distribution of PKA populations. The median PKA energies $T_{1/2}$(dpa) differing by 10 % between the cases are depicted in Fig. 1 (a) and (b) for the full spectrum case and the thermal neutron shielded case, respectively, in each curve representing cumulative fractions of displacement below a PKA energy of T keV. In the figures, the values of median of PKA distributions are also shown to be 480 and 580 eV for the respective cases, which are far smaller than both values of $T_{1/2}$(dpa) and average T. These cumulative fractions are obtained using PKA energy dependencies of PKA populations and displacements per atom, which are given in Fig. 2(a) and (b) for the respective cases. A remarkable difference in the PKA energy dependencies of dpa between the two cases is the contribution of displacements due to PKAs below 1 keV for the full spectrum case, which are all resulting from (n,γ) reactions with the thermal neutrons below 1 eV. This is demonstrated in Fig. 3(a) and (b) which show contributions of each reaction to the PKA energy spectra as a function of PKA energy, for the respective cases. The results given above have been obtained using an evaluated nuclear data file of JENDL-3.2[7] and the displacement threshold energy Ed of 40 eV according to calculational procedures already described in the previous proceeding[8].

3. What is meant by damage parameters and PKA energy spectra

It has been recognized that defects survival rates are enhanced with an decrease in the median PKA energy. This understanding has been obtained by experimental observations mainly through electrical resistivity measurements for samples irradiated at liquid helium temperatures near 4K, where the nascent defects are maintained as surviving the displacement cascades, and also through large scaled computer simulations of displacement cascades by molecular dynamics calculations starting with a given PKA energy. The result shown in Fig. 4[9] from the literature will best describe the significances of PKA spectra which have on the displacement damage processes: in the figure, fractions of defects that survive cascade quench are given as a function of the median recoil(PKA) energy, as normalized to the dpa calculated by the NRT model. This result indicates that with an increase in PKA energy, although cascade volumes and a number of atoms displaced will increase, fewer defects can be transported beyond a highly agitated cascade core to survive the cascade region, after the kinetic energy is imparted to the lattice atoms and the lattice is cooled (or quench in 10^{-11} s after the initiation of the cascade), and most of the defects, i.e. interstitials and vacancies annihilate by recombinations during this thermal spike. Note that it is the point defects, which survive the cascades and become mobile under temperatures high enough, that bring about radiation effects such as microstructural developments and mechanical properties degradations of the irradiated materials.

The importance of the lower energy PKAs in radiation damages is further demonstrated in Fig. 5[10], in which relative efficiencies of various ions for producing long-range, freely migrating defects are plotted as a function of the median PKA energy. The result has been obtained by measuring quantitatively radiation induced segregation in two concentrated alloys under irradiations with various ions of various energies to alter the median PKA energy but at approximately the same dpa/s, at temperatures ranging 600-900 K. The radiation induced segregation of particular atoms is possible only under the existence of long-range, freely migrating defects. The relative efficiencies here are normalized to that of 1MeV protons. It is noted that the general shape of the curve in Fig. 6 is qualitatively very similar, indicating the lower the PKA energies, the higher the efficiencies of production of freely migrating defects.

It is not so simple for the creep rate enhancement by over one order mentioned above to be understood merely on the analogy of the significance of the lower median PKA energy. In fact, the difference of 10 % between the two $T_{1/2}$ values i.e. 55 keV and 60 keV (Table 1) is insignificant, referring to the $T_{1/2}$ dependencies of the defect survival rates and production efficiencies of freely migrating defects, in Fig. 4 and Fig. 5. However, the occurrence of a large number of population of PKAs peaked below 1 keV, as shown in Fig. 2 for the full spectrum neutron irradiation due to (n,γ) reactions, is considered to play a substantial role in enhancing the creep rate through by itself a relatively high rate of providing freely migrating defects to lattices under the stress.

Furthermore, another result obtained by the sophisticated experiment of measuring the radiation induced segregation in Cu-Au alloy, given in Fig. 6[11], indicates that the effect of lower energy PKAs on enhancing radiation damages is strongly affected by a coexistence of displacement cascades by high energy PKAs. Therefore, the observed creep rate enhancement could be possible under a detailed balance among factors like the existence of peaked PKA distribution below 1keV, a favorable extent of both spatial and temporal distributions of displacement cascades due to high energy PKAs mainly due to (n, n), (n, n’) and (n,α) reactions; note that those distributions due to high energy PKAs being common to the both neutron spectra and only difference being the peaked PKA distribution below 1 keV due to (n,γ) reactions in which are emitted high energy photons peaked around 7.5 MeV from Fe. Any way, the accumulation of experimental data is further required to establish the profound understanding concerning these phenomena.

It is known that light ion irradiations produce lower energy PKAs because of long-range Coulomb interactions, which results in higher survival rates of freely migrating defects, as shown in Fig. 5, for the case of H and He irradiation. Taking niobium for example, cumulative fractions of damage energy below T keV of PKA are given as a function of PKA energy T, in Fig. 7[12], for irradiations with light ions of proton, deuteron and alpha, comparing with fission neutron irradiation, where damage energy multiplied with 0.8/(2Ed) gives number of displacements in the NRT model. Note that these light ion irradiation results in the median PKA energies lower than a few keV.

In view of the importance of the light ion irradiation, light mass charged particles, such as proton, alpha as emitted from nuclear reactions are also considered to produce PKA spectra distributed largely in a lower energy region than a few keV and are considered to contribute more to a formation of freely migrating defects than PKAs produced in fission neutron irradiations. Therefore, importance of nuclear data about charged particles production reaction cross sections and energy spectra of emitted charged particles should be stressed, especially for the case where copious reaction channels become open or nuclear spallations take place and so called internal light ion irradiation can not be neglected.

According to the theoretical prediction, energy distributions of emitted light particles at nuclear spallations are shown to be distributed to be peaked around a few MeV and to be distributed up to several hundreds of MeV[13]. Such a broad energy distribution of light particles is considered to produce both lower energy PKAs and higher energy ones. Moreover, if the cross sections of these light particle emission reactions increase with increasing incident energies of nucleon like neutrons, as is shown in Fig. 8[14], the effect of the internal bombarding by these light particles can be considered to be more important to the radiation damages than currently assumed. So far a number of studies have been done to estimate radiation damages for materials to be used as components of spallation neutron facilities, however, damage parameters like dpa cross sections and gas atom production rates have only been described. However, it seems impractical to evaluate data of both energy spectra and cross sections for a dreadfully large number of reaction channels: instead, realistic and effective representations for these quantities based on sound systematics seem highly desirable for radiation damage analysis.

In summary, the damage parameters for analyzing radiation damage processes were mentioned, referring to significance of lower energy PKAs. Following points are stressed:

1. Taking iron irradiated with JMTR neutrons as an example, a possibility is described that displacement damages produced by PKAs with energies lower than a keV order due to recoiled atoms from (n, gamma) reactions can contribute significantly to radiation damages.
2. From the point of view that highly energetic charged light particles also produce PKAs distributed dominantly in energy regions lower than a keV order, evaluations of nuclear data such as cross sections, and energy spectra of charged particles emitted from the nuclear reactions will be urgently required, for damage analyses of materials to be irradiated with GeV orderÅ nucleons .

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References

[1] Annual Book of ASTM standards, Volume 12.02, E521-96, p.156 and E693-94, p. 254 (1996).

[2] Lindhard J.,. Scharff M. and Schiott H. E., Kgl. Dan. Vidensk. Scjsk. ,Mat.-fys. Medd. 33(14) (1963).
[3] Robinson M. T., Proc. B. N. E. S. Nucl. Fusion Reactor Conf., p.364 (1969).
[4] Norgett M. J., Robinson M. T. andTorrens I. M., J. Nucl. Eng. and Des. 33, 50. (1975).
[5] Y. Kurata, private communication.
[6] Shimakawa S., Komori Y., Nagao Y. and Sakurai F., JAERI-Tech 95-023 (1995).
[7] Nakagawa T. et al., J. Nucl. Sci. Technol., 32 , 1259 (1995).
[8] Aruga T. and Shiraishi K., JAERI-M 89-026, p. 342 (1989).
[9] Jinkle S. J. and Singh B. N., J. Nucl. Mater., 199, 173 (1993).
[10] Hashimoto T., Rehn L. E. and Okamoto P. R., Phys. Rev., B38, 12868 (1988).
[11] Iwase A., Rehn L. E., Baldo P. M. and Funk L., J. Nucl. Mater., 238,224 (1996).
[12] Kirk M. A. and Greenwood L. R., J. Nucl. Mater., 80, 159 (1979).
[13] Daemen L. L. et al., LA-UR 95-1792, p. 487 (1994).
[14] Wechsler M. S., et al., ASTM STP 870, p. 1189 (1985).

Table 1. Comparison of Damage Parameters

	Full Neutron Spectrum	Thermal Neutron Shielded Spectrum
Total Flux (/cm^2/s)	3.5x10^{14}	2.2x10^{14}
Displacement		
Cross section σ_d(b)	150	230
rate (dpa/s)	5.2x10^{-8}	5.1x10^{-8}
$T_{1/2}$(dpa) (keV)	55	60
Average T (keV)	8.5	11.7

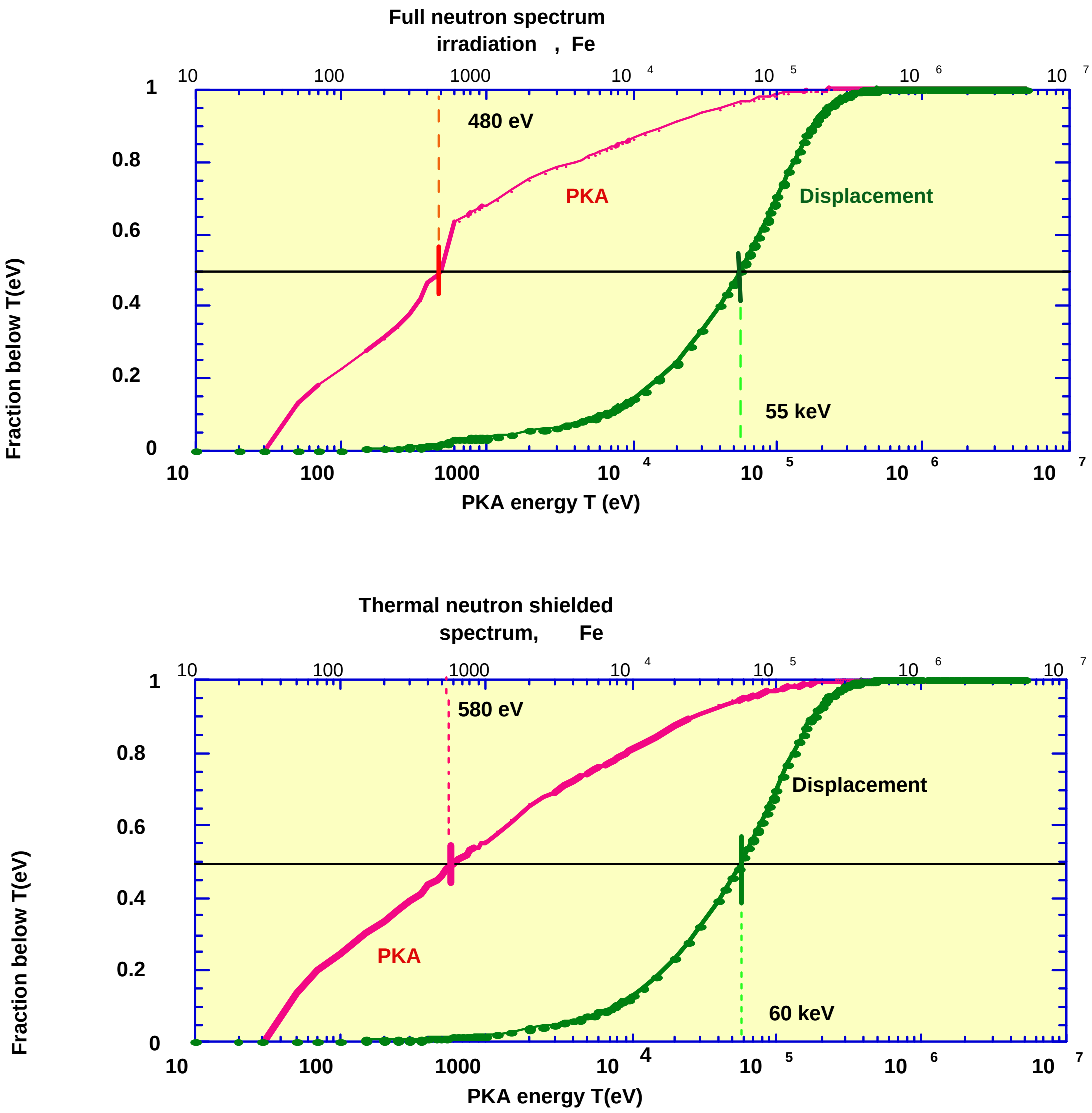

Fig.1. Cumulative fractions of PKAs and displacements below PKA energy T (eV) for Fe sample irradiated with (a:top) full spectrum neutrons and (b:bottom) neutrons of thermal neutron shielded spectrum

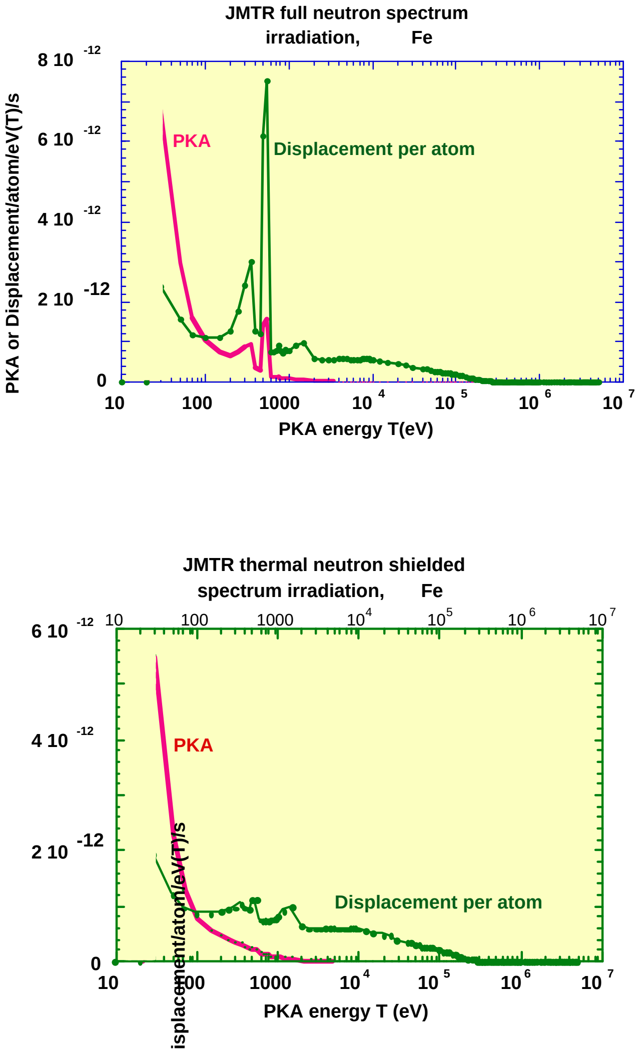

Fig. 2. PKA energy dependencies of PKAs and displacements per atom for Fe sample irradiated with (a) full spectrum neutrons and (b) neutrons of thermal neutron shielded spectrum.

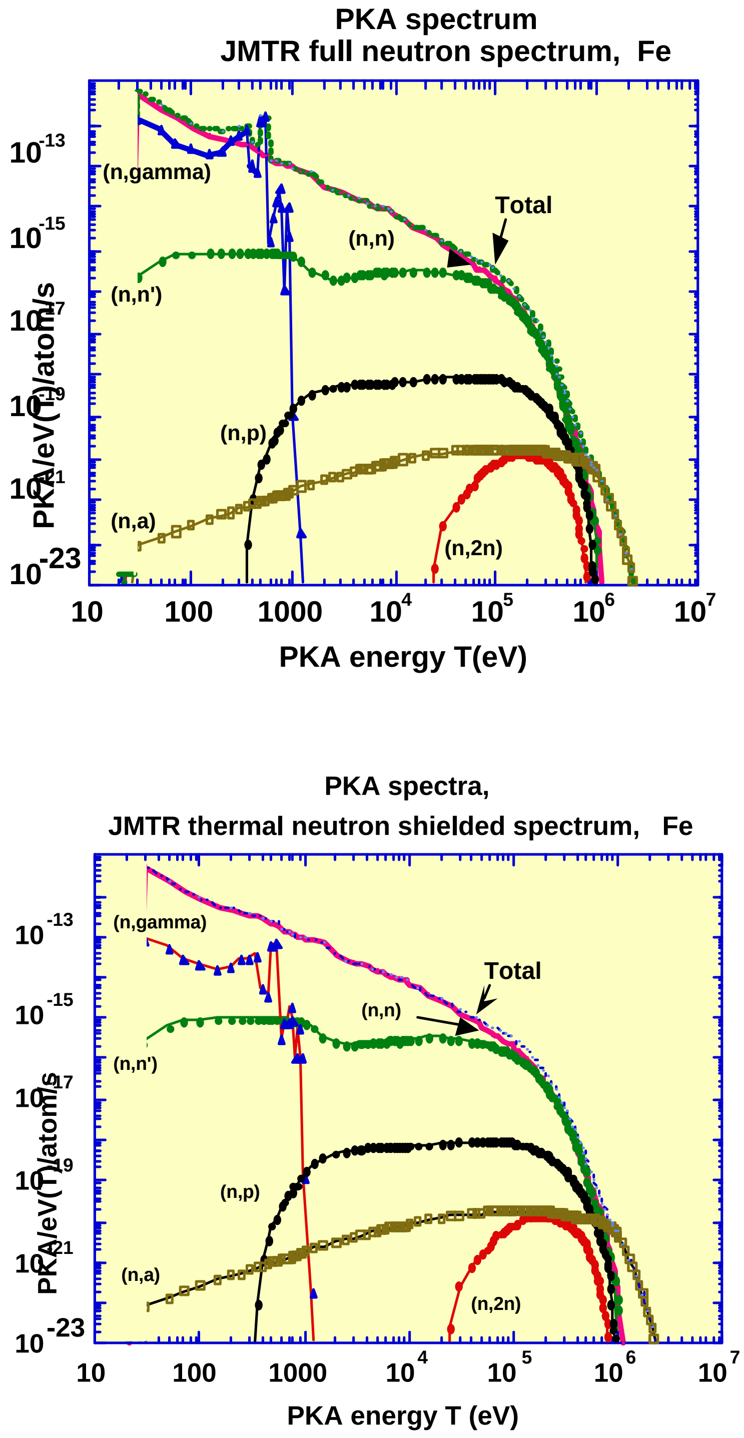

Fig. 3. Reaction wise PKA energy spectra for Fe sample irradiated with (a) full spectrum neutrons and (b) neutrons of thermal neutron shielded spectrum.

Those figures, only captions of which are given below, are kindly requested to be referred to original papers cited.

Fig. 4 . Dependence of the survived (quenched) cascade defect fraction on median PKA energy in copper. The open symbols refer to molecular dynamics calculations[9].

Fig. 5. Relative efficiencies for producing freely migratingdefects as a function of median PKA energy[10].

Fig. 6. Amount of near-surface Au depletion (inverse segregation) as a function of total dpa for 2h dual(1.5MeV He + 800keV Cu) beam irradiation and subsequent He onlyirradiation (solid circles). open diamonds: He only, open circles: 4h dual beams[11].

Fig. 7. Cumulative fractions of damage energy distributions below PKA energy T for different types of irradiating particles[12].

Fig. 8. Helium production cross section versus neutron energy[13].