**CALCULATIONS
OF VACANCY CLUSTER PRODUCTION
IN IRON
FOR EBR-II, DFR, FTR, AND ETR**

G. E. RUSSCHER

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By

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INTRODUCTION

Fuel clad swelling and void generation observed in the Dounreay Fast Reactor (DFR)⁽¹⁾ prompted this study which should help to relate those observations to conditions anticipated in the Fast Test Reactor (FTR). Fuel element distension and density changes can shorten the useful in-core fuel life through such mechanisms as coolant channel flow restriction, fuel clad stress concentration near voids, and circumferential fuel clad strain that may exceed the brittle fracture limit. For example, fuel swelling may induce radial fuel cracks that cause stress concentration areas, thinning and subsequently, bursting of the fuel clad.

How can experimental data gleaned from DFR and other nuclear irradiation facilities be most meaningfully applied to the FTR design? While this report does not fully resolve that question, it describes the current correlation technique based on a damage mechanism assuming that void growth around stable vacancy clusters results in isotropic material expansion. Extended vacancy defect size and concentration are calculated. Results of the calculation provide an indication of the relative damage production in different reactor spectra.

The primary objective of this study is to find the best immediate estimate of fuel clad distension and void production in the FTR. A secondary objective is to suggest computer calculations and experimental data needed for the justification and refinement of the correlation technique and its preliminary conclusions.

Due to time and budget limitations, this preliminary study has been confined to the comparative estimation of FTR fuel clad swelling and void production in several reactors using a preliminary correlation technique. The report discusses the damage model used to relate radiation interaction calculations to the material damage. Necessary assumptions used in the calculations are presented in the discussion section.

CALCULATIONAL METHODS

Computer programs have been used to calculate the interactions between neutrons and the irradiated lattice structure in a statistically meaningful way. A spectrum of neutrons produces a spectrum of primary knock-on atoms (PKA) which, in turn, produces defects of various sizes. The PKA energy spectrum is computed with Monte Carlo techniques, using 100,000 neutron histories. A series of discrete two-body atomic interactions are used to calculate the displacement cascade and defect size distribution formed by each PKA.

DAMAGE MODEL

The kinds of calculations performed and the study results published in this report represent, in our current judgement, the most effective way to estimate the number of void nuclei produced in a material by an irradiation environment. These data are sensitive to irradiation neutron spectral differences because PKA production is calculated on the basis of statistical nuclear scattering, and the number of defects/PKA is calculated on the basis of statistical interatomic scattering. Defect production calculations take into account the lattice structure of the specimen. Both elastic and inelastic atomic collisions are accounted for by the Thompson and Wright method. Accurate descriptions of high energy atomic interactions are important to the calculation of energy available for displacing atoms in a fast neutron irradiation. These are refinements over the Kinchin-Pease model, often used to estimate defect production.

Voids are assumed to form, to begin with, by fast neutrons colliding with one or more atoms of the specimen. Some PKA can, in turn, cause additional displacements in a cascade of collisions. The PKA creates lattice defects within a volume called the damage spike or collided atom volume, even though only a small fraction of the atoms in the spike are displaced. Within a very short time, the majority of the displaced atoms return to regular lattice sites. However, some remain as an array of point defects, single interstitial atoms and vacant lattice sites, and extended defects (clusters of interstitials or vacancies).

Two parameters influencing the production of extended defects are the PKA energy or resultant damage spike size, and the density or proximity of spikes. Some defects annihilate each other by recombination, or migrate to sinks or traps. Interstitial clusters may grow to form dislocation loops, or they may be annihilated at other interstitial sinks. Vacancy clusters having 7 or more members are assumed in this study to be stable, but they may continue to grow with time at the expense of mobile vacancies and small clusters.⁽³⁾

Large vacancy clusters may act as nuclei for the development of micrographically visible voids. However, these clusters may also collapse to form dislocation loops, or they may segment, dissociate, and migrate to surfaces, sinks, or traps. The vacancy cluster behavior depends on temperature, controlling the mobility of the void constituents, and on the concentration of competing traps. Other parameters potentially influencing the development of vacancy clusters into voids are the crystal lattice structure, impurities, stress state, and the presence of gases from transmutation reactions.

The stable or large vacancy clusters are primarily assumed in this study to contribute to the volume increase of

the specimen simply by a cumulative summation of their vacant lattice site volumes accompanied by an almost negligible amount of lattice relaxation. However, some intermediate size vacancy clusters may also remain, contributing to the volume increase.

CALCULATION STEP OVERVIEW

The SPECTRA⁽⁴⁾ computer program and published results of studies using the CASCADE-CLUSTER^(5,6) computer programs have been used to calculate the concentration and size distribution of vacancy clusters.

The energy transferred to the PKA was the first step of the calculations for stainless steel in the irradiation environments of DFR, EBR-II, FTR, and the Engineering Test Reactor (ETR). A PKA distribution was calculated for each of the major elements of steel--Fe, Ni, and Cr. Based on the same number of neutron histories from each irradiation spectrum, an effective PKA density for stainless steel was calculated from the PKA density for each of its elements.

The total defect density was then calculated from the effective PKA density using the Thompson and Wright relation of PKA-generated defects to its energy.⁽²⁾ The total defect density was subsequently used to calculate the neutron fluence for each of the irradiation environments capable of creating a statistically complete displacement spike damage state. This state, a lattice filled with displacement cascades, has a gross defect density equal to the defect density within the collided atom volume of a displacement cascade.⁽⁷⁾

The stable vacancy cluster density for stainless steel in each environment was calculated from the PKA energy spectrum and density using Beeler's published α iron vacancy cluster size distributions.^(8,9) Iron PKA spectra, because they are most representative of stainless steel, were used to calculate the vacancy cluster density for each irradiation environment.

Two important parameters, displacement damage superposition and temperature, can influence these results. Displacement damage superposition is defined as the formation of a displacement cascade in a crystal lattice region that has already produced a displacement cascade and still contains an array of stable defects. In effect, the collided atom volumes are superimposed, or at least intersect. A qualitative change in vacancy cluster density due to the probability of damage superposition is projected for each environment.

Subsequently, an effective volume increase was calculated from the volume and density of stable vacancy clusters produced in each environment.

INPUT INFORMATION REQUIRED

The neutron spectra describing each irradiation environment should have at least 15 energy groups above 362 eV. The accuracy of that information is quite important, as evidenced by the sensitivity of the results to relatively small spectral differences among the fast reactors.

Nuclear interaction parameter data are also required in considerable detail. Anisotropic scattering cross sections are needed in the form of cumulative distribution functions of neutron energy and scattering angle. These data are used in the calculation of neutron interaction probability and energy transfer. Interatomic scattering potential data are needed for the calculation of energy transfer from atom to atom.

RESULTS OF CALCULATIONS

PRIMARY KNOCK-ON ATOM ENERGY DISTRIBUTION

The energy distribution of PKA's formed in iron as calculated with SPECTRA and its input computer programs are found in Figure 1 for the DFR, FTR, EBR-II, and ETR. The PKA probability per unit logarithmic energy is presented as a function of logarithmic energy so that each unit area represents the same number of PKA. All curves are normalized to the same area.

The energy distributions of PKA formed in the iron, nickel, and chromium of SS-304 have been calculated. However, iron data, shown in Figure 1, are representative of stainless steel within a few percent, and are used in subsequent calculations.

The neutron spectra producing these PKA are shown in Figure 2. Only those neutrons of energy greater than 362 eV are considered. A minimum neutron energy of 362 eV is required to create a displacement, that is, to transfer the threshold displacement energy, 25 eV, to an iron atom.

PRIMARY KNOCK-ON ATOM DENSITY

SPECTRA calculations for the four environments show the PKA density per unit fluence in irradiated specimens to be the highest for the FTR. Within the accuracy of the calculations, these results (Table 1) are applicable to both 304 and 316 stainless steel. The neutron fluence above the displacement threshold energy, 362 eV, is the basis of these calculations. The PKA density per unit fluence is also shown for the common fluence ($E > 1$ MeV). Table I shows average values of the PKA energy corresponding to the spectra of Figure 1. It is interesting to note that, of the four irradiation environments, the FTR produces the highest PKA density but the lowest average PKA energy. These data are the basis of subsequent calculations found in Tables II and III.

FIGURE 1. Probable Energy of Primary Knock-On Atoms (PKA) Produced in Iron

FIGURE 2. Comparison of Reactor Spectra

TABLE I. Primary Knock-On Atom Density in Steel
(Clad Specimen of 304 or 316 Stainless Steel)

	$\frac{\text{PKA}}{\text{cm}^3}$	$\frac{\text{PKA}}{\text{cm}^3-\phi t}$ $E > 362 \text{ eV}$	$\frac{\text{PKA}}{\text{cm}^3-\phi t}$ $E > 1 \text{ MeV}$	$\bar{E}_{\text{PKA}},$ keV
ETR	5,600	0.153	0.47*	15.3
FTR	16,800	0.460	3.38	10.1
EBR-II	13,000	0.356	1.28	21.7
DFR	13,500	0.370	1.61	18.3

* This value reflects the commonly quoted high energy fluence based on iron activation dosimetry and a fission spectrum averaged cross section of 68 mb. There is a 10% difference between the ETR value and an exact ETR spectrum activation integral.

TABLE II. Vacancy Distribution in Iron

	Gross Defect Density, $\#/\text{cm}^3-\phi t,$ $E > 362 \text{ eV}$	Total Number of Vacancies, $\#/\text{PKA}$	Number of Vacancies in Large (≥ 7) Clusters, $\#/\text{PKA}$	Large Vacancy Cluster Density, $\#/\text{cm}^3-\phi t,$ $E > 362 \text{ eV}$
ETR	17	109	12	0.21
FTR	41	89	10	0.53
EBR-II	62	174	19	0.78
DFR	56	152	17	0.74

TABLE III. Superposition of Radiation Damage in Iron

	Based Upon:		Based Upon:	
	Gross Defect Density, 3 at. %		Clustered Vacancy Density, 1 at. %	
	Defect Density, $\#/\text{cm}^3-\phi t$	Superposition Fluence, cm^{-2} ($E > 362 \text{ eV}$)	Vacancy Density, $\#/\text{cm}^3-\phi t$	Superposition Fluence, cm^{-2} ($E > 362 \text{ eV}$)
ETR	17	1.5×10^{20}	1.8	4.6×10^{20}
FTR	41	6.2×10^{19}	4.6	1.8×10^{20}
EBR-II	62	4.1×10^{19}	6.8	1.3×10^{20}
DFR	56	4.6×10^{19}	6.3	1.4×10^{20}

GROSS DEFECT DENSITY

The total number of defects caused by PKA are calculated by the Thompson and Wright method.⁽²⁾ The energy range of this relation extends beyond the ionization threshold to include both elastic and inelastic atomic collisions. This relation, when applied to the PKA spectra, gives a gross defect density per unit fluence corresponding to the PKA density per unit fluence for each reactor. Table II shows EBR-II to have the highest gross defect density per unit fluence followed by DFR, FTR, and ETR, respectively.

The influence of specimen size upon calculated gross defect density was investigated for typical fuel clad and tensile specimens. These results varied by less than 10% between tensile specimens and 15 mil-fuel clad irradiation specimens. Tensile test and fuel clad specimens were dimensioned $0.36 \times 0.36 \times 7.6 \text{ cm}^3$, and $0.038 \times 0.36 \times 7.6 \text{ cm}^3$, respectively.

LARGE VACANCY CLUSTER PRODUCTION

The large vacancy cluster density per unit fluence is lower in the FTR than in either the DFR or EBR-II. Large cluster data are presented in Table II for stainless steel. They were developed from the calculated PKA density and published vacancy cluster size distributions as a function of PKA energy in iron.⁽⁹⁾ A typical displacement cascade contains about 10% of its vacancies in clusters of 7 or larger. The stability of a vacancy cluster has been approximately related to its size, an assumption which ANNEAL program computations will check. For this study, clusters of 7 or more vacancies are assumed to be stable even at high temperatures. The stable vacancy cluster densities per unit fluence for stainless steel are given in Table II.

RADIATION DAMAGE SUPERPOSITION

When three successive displacement cascades superimpose, large vacancy cluster production (/PKA) nominally increases by a factor of two.⁽¹⁰⁾ The fluence at which displacement cascade superposition could occur is displayed in Table III for each of the environments.

Two criteria have been used to define the threshold of damage state superposition. One is an average vacancy concentration of 3 at.% in a specimen. CASCADE computer calculations show that, within a displacement cascade, an average of 3.2 at.% have been displaced from their stable lattice sites.⁽⁸⁾ Data are shown in Table III predicting the fluence that could produce an average gross defect density (vacancies) of 3 at.% in the specimen.

The other criterion for damage state superposition is a stable vacancy cluster density of 1 at.%. This criterion is qualitatively based on the observation that, within a displacement cascade, about 10% of the defects (0.3 at.%) are found in large stable clusters. When two succeeding cascades are superimposed on the original, 1 at.% stable vacancy clusters can result. This concentration could be experimentally observable as material density change or swelling. Table III also shows the fluence that could produce a 1 at.% stable vacancy cluster density for each of the irradiation environments

A graphic portrayal of the stable vacancy cluster production is shown in Figure 3 where the stable vacancy cluster fraction increases from 10% at fluences below the conservative estimate (gross defect) of damage superposition to 20% beyond the fluence which superimposes three displacement cascades. A stable cluster fraction of 15% is used in the fluence range between the values determined by the two superposition criteria.

FIGURE 3. Stable Vacancy Cluster Production in the Range of Displacement Cascade Superposition

DISCUSSION

RELATIVE IRRADIATION DAMAGE RATING

The calculations indicate the rate of stainless steel irradiation damage per neutron above the energy threshold for PKA production in the reactors studied, to be in the order of $EBR-II \geq DFR > FTR > ETR$. This irradiation environment ordering follows from both gross defect density and vacancy cluster density data of Table II.

Parameters or assumptions used in these calculations can be modified to account for observed differences between the calculated gross defect density and experimental data. The revision of parameters and assumptions used in vacancy cluster calculations may be necessary to account for differences between calculated vacancy cluster density and such experimental data as density change and void population count. However, these alterations should change only the absolute magnitudes but not the relative rating of irradiation damage among the results for different reactors.

MATERIAL SWELLING

The same density change or swelling, calculated to be 1%, occurs in the reactors at the fluences ($E > 362\text{eV}$) as follows: EBR-II, $1.3 \times 10^{20} \text{ cm}^{-2}$; DFR, $1.4 \times 10^{20} \text{ cm}^{-2}$; FTR, $1.8 \times 10^{20} \text{ cm}^{-2}$; and ETR, $4.6 \times 10^{20} \text{ cm}^{-2}$.

This estimate is based on the calculated number of vacancies in a displacement cascade occurring in large, presumably stable clusters ($n \geq 7$). Table II shows the average number of these vacancies produced in the specimen by one neutron distributed over the irradiation spectrum ($E > 362 \text{ eV}$). These vacancy clusters are presumed stable and, thus, their atomic volumes accumulate with fluence.

A calculated vacancy cluster volume of 1 at.% was chosen as a criterion for reactor fluence comparison. Present experimental data indicate up to 1.5% swelling occurs at fluences of about 10^{22} cm^{-2} .⁽¹⁾ The difference between the calculated and experimental fluences suggests that not all large vacancy clusters contribute to swelling. It may well indicate the fraction (call it unstable fraction) of large vacancy clusters collapsing into dislocation loops or other sinks rather than contributing to density change. The temperature history of the specimen may also influence the change in density.

VOID PRODUCTION

The calculations provide the following density estimates of large vacancy clusters per unit fluence ($E > 362 \text{ eV}$) for reactors: EBR-II, $0.78/\text{cm}^3 \phi t$; DFR, $0.74/\text{cm}^3 \phi t$, FTR, $0.53/\text{cm}^3 \phi t$; and ETR, $0.21/\text{cm}^3 \phi t$. These estimates are based for each reactor on the average number of PKA's and the average number of vacancies found in stable clusters in the displacement cascades. These data are shown in Tables I and II, respectively. A comparison of the results shows the same order among reactors in relative damage production per unit fluence as before.

The data can serve as the basis of a preliminary correlation on void generation. If the irradiation environment provides the conditions needed to expand vacancy clusters into voids resolvable by electron microscopy, the void count in an irradiated specimen may be related to the predicted vacancy cluster density. A similar unstable fraction of the large vacancy clusters which do not remain as void nuclei may be needed to establish agreement between the calculated cluster concentration and the number experimentally observed. The size of a void and, consequently, the observable void concentration can be determined, it is believed, by the production and diffusion rates of both gas and vacancies.

UNANSWERED QUESTIONS

Several assumptions were necessary to provide immediate data relevant to the problems of material swelling and void generation. The least serious of these assumptions appears to be the application of BCC alpha-iron data on the probable vacancy cluster size distribution in a displacement cascade to FCC stainless steel. This assumption has been accepted because the differences between cluster distributions in BCC - α -iron and FCC - copper were shown to be small.⁽⁹⁾ However, the influences of crystal grain size, temperature, and impurities have not been considered.

Calculations with superimposed, moderate energy α -iron PKA's have led to the conclusion that two subsequent displacement cascades intersecting an original will approximately double the fraction of defects in the large clusters.⁽¹⁰⁾ The application of this qualitative relation to FCC lattice structure, as well as the linear extrapolation from that relation, are also assumed.

The effect of temperature is not included in these calculations. Present calculations and data, in effect, assume a temperature of absolute zero. Thus, defect cluster data probably over-estimate vacancy production and stability, since those migrating to sinks or becoming trapped are not taken into account. At elevated temperatures, the quantitative predictions for void concentration will probably be different. However, the comparison among void nucleus concentration ($/\text{cm}^3$) for the four irradiation facilities at equal temperature should remain valid, since elevated temperature would produce similar changes in all cases.

SUMMARY

CONCLUSIONS

- Relative irradiation damage per neutron above the displacement threshold yields the following order:
EBR-II $\geq$ DFR $>$ FTR $>$ ETR.
- Equal swelling at the same irradiation temperature is estimated at the fluences ($E > 362$ eV) as follows: EBR-II, $1.3 \times 10^{20} \text{ cm}^{-2}$; DFR, $1.4 \times 10^{20} \text{ cm}^{-2}$; FTR, $1.8 \times 10^{20} \text{ cm}^{-2}$; and ETR, $4.6 \times 10^{20} \text{ cm}^{-2}$. The calculated irradiated condition produced by these fluences is a vacancy production equivalent to a density change of 1%.
- Large vacancy cluster density per unit fluence above 362 eV are:
EBR-II, $0.78/\text{cm}^3 \phi t$; DFR, $0.74/\text{cm}^3 \phi t$; FTR, $0.53/\text{cm}^3 \phi t$; and ETR, $0.21/\text{cm}^3 \phi t$.

RECOMMENDATIONS

An experimental check of the results is necessary. Irradiation and computer experiments providing data on both point defects and extended defects would give a more direct comparison with calculated quantities. Determining the validity of results for these data would increase confidence in the application of the results to more complex engineering properties.

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