**CORRELATION OF RADIATION DAMAGE
TO MATERIALS FOR FFTF**

G. E. RUSSCHER AND R. E. DAHL

MARCH, 1968

**AEC RESEARCH &
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UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

PRINTED BY/ FOR THE U. S. ATOMIC ENERGY COMMISSION

3 3679 00061 0867

BNWL-667
UC-80,
Reactor Technology

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By

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Materials Research Section
Materials Department

March 1968

FIRST UNRESTRICTED
DISTRIBUTION MADE APR 1 '68

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

Printed in the United States of America
Available from
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Springfield, Virginia 22151
Price: Printed Copy \$3.00, Microfiche \$0.65

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ABSTRACT

Radiation damage processes which may be critical to FFTF structural materials are the production of high temperature crystal lattice defects such as vacancy clusters or micro voids, and extended defects, helium production by fast neutron (n,α) reactions with matrix elements, and helium generation in steel by slower neutron (n,α) interactions with boron.

Though high temperature radiation damage appears related to production of voids or extended defects, current correlation techniques are based on equating gross displacement defect production in various neutron spectra. Computer simulation of lattice behavior during irradiation and annealing is being used to develop improved correlation techniques.

Helium generation in nickel alloys and stainless steel leads to embrittlement and loss of ductility. Current correlation is based on gross helium production. However, agglomerated helium atoms probably affect mechanical properties more than the same number of atoms distributed uniformly through the material. Computer simulation of helium migration and agglomeration is aimed at providing an improved correlation basis for the effect of helium production.

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INTRODUCTION

Environmental conditions such as the neutron energy spectrum, neutron flux, and temperature in the Fast Test Reactor (FTR) differ from those in such reactors as Dounreay Fast Reactor (DFR), EBR-II, and ETR. However, data from these and possibly other reactors must be used as a basis of FTR design. Therefore, it is vital that radiation damage correlation techniques provide a sound means for adapting experimental data to FTR conditions. Present damage correlation techniques were developed for tests conducted in thermal reactors; therefore, the applicability of those techniques to the fast reactor environment must be determined.

Present correlation techniques assume gross displacements as a basis. Two irradiations causing the same number of gross displacements are assumed to cause the same property change. Mechanical property data from irradiation in light water reactors and graphite reactors have been correlated satisfactorily on this basis. The general application of this correlation technique to the damage processes anticipated in the FFTF environment may be inaccurate due to the dissimilarity of the damage mechanisms.

Ideal correlation techniques may differ for each damage process. Three potentially significant damage processes are:

1. the production of crystal lattice interstitial and vacancy clusters and their subsequent annealing behavior at high temperature,
2. helium production by fast neutron (n, α) reactions with steel matrix elements, and
3. helium generation by slower neutron interactions with boron impurities.

Current knowledge of the mechanism of each process, the relationship between the mechanisms and the basis of methods for correlating data, and plans for developing or improving those methods are discussed in the following sections of this report.

DISCUSSION

1. CRYSTAL LATTICE DEFECT CLUSTER DAMAGE AT HIGH TEMPERATURES

Defect cluster damage from fast neutron collisions may be a serious problem in FTR materials. Void production, observed in some experimental fuel element cladding specimens, results in significant swelling of the sample.⁽¹⁾ The mechanism of void formation and the size distribution of vacancy clusters, which contribute to the voids through agglomeration, are being investigated. Experimental data have shown that the temperature history of the sample influences the growth of voids. This emphasizes the need of a damage model which includes temperature in the description of the damage process. Current attempts to increase the relevance of damage correlation techniques to void production are based on Beeler's and Johnson's work on vacancy cluster behavior.^(2,3,4)

Preliminary results⁽⁵⁾ show that the ductility and rupture life of stainless steel are drastically reduced, and that yield-strength is significantly increased by exposure to 10^{21} - 10^{22} neutrons/cm² in EBR-II, if the irradiation temperature is higher than half the absolute melting temperature, T_m . Apparently the fast neutrons produce extended or complex defects, such as clusters of vacancies or interstitials, which are stable at temperatures above $T_m/2$. A complex defect structure model is invoked for this high temperature embrittlement because isolated point defects anneal out at temperatures less than $T_m/2$.

Displacement spikes or collision cascades, which result in large, complex defects, are produced by high-energy neutrons.

For example, under ideal conditions, a minimum neutron energy of 50 keV is required to produce a cluster of 10 vacancies in iron. A very intense flux environment may produce extended defects through agglomeration in a defect saturated crystal lattice. The needed damage correlation for complex or extended defects may include the effects of both the fast neutron spectrum and the flux intensity or defect production rate.

Present techniques assume that displacement spike damage is proportional to gross displacement defect production. Several techniques based on this assumption have been successful in correlating low temperature damage in thermal reactors.⁽⁶⁻¹¹⁾ One of the techniques is described in greater detail.

The exposure to neutrons of energy greater than E_L , described by Equation (1), is suggested by Dahl and Yoshikawa.⁽⁶⁾

$$\bar{\Sigma}_D = \int_0^{\infty} \phi(E_n) \Sigma_s(E_n) N(E_n) dE \bigg/ \int_{E_L}^{\infty} \phi(E) dE \quad (1)$$

where

$\bar{\Sigma}_D$ = the effective cross section for the production of a defect in the particular material with a neutron whose energy $> E_L$

$\phi(E_n)$ = the neutron spectrum

$\Sigma_s(E_n)$ = the macroscopic elastic scattering cross section

$N(E_n)$ = the number of displacement defects produced as a result of the collision of a neutron with an atom.

The dependence of $\bar{\Sigma}_D$ on the lower energy limit E_L is calculated for different neutron spectra being correlated. It is found that for a particular value of E_L , there is near-equality among the $\bar{\Sigma}_D$ for a wide range of spectra. This near equality implies that the displacement defect production per unit fluence above

E_L is nearly equal for the spectra. These displacement defects may be produced by the collision of a neutron, a primary knock-on atom (PKA), or atoms of a cascade; however, it is assumed that the radiation damage being correlated is simply based on the total number of defects produced.

The technique accounts for the variation in gross defect production by different neutron spectra, and therefore is a considerable improvement over previous methods which assumed the same fission spectrum in all irradiation facilities. However, all current techniques based on gross displacement production are probably inadequate for precise damage correlation in fast flux reactors. High temperature embrittlement depends heavily on the annealing characteristics of complex defects and only indirectly on the number of gross displacement defects. These circumstances necessitate the reassessment of a preliminary damage analysis of fast reactor data based on the current correlation techniques.

An improved correlation may depend on the annealing characteristics of the large complex defect structures. There are several computer programs which were designed to simulate the behavior of defects in a crystal lattice structure, including the thermal stability or dynamics of the structure. A series of computer programs are in use to study the production and stability of small defect clusters and their interaction with simple point defects. The first program, SPECTRA,⁽¹²⁾ calculates the energy and location of PKA's produced in a material as a function of the incident neutron energy spectrum. Subsequently, the size and physical make-up of the collision cascade produced by each PKA at 0°K can be investigated with the CASCADE and CLUSTER computer programs.⁽¹³⁾ The CASCADE program calculates the numbers and locations of vacancies, interstitials, and replacement interactions. The CLUSTER program, operating in conjunction with CASCADE, records the resultant distribution of

interstitials, vacancies, and defect clusters as a function of their sizes and positions. CLUSTER also recombines Frenkel defect pairs that are unstable at 0°K. The ANNEAL⁽²⁾ program includes temperature by describing thermally induced interactions among the defects, and permits the identification of defects stable at high temperature.

As presently written, this computer code does not permit migration of vacancies, and hence is not applicable to temperatures at which serious void formation is observed. The code will be modified to include vacancy formation. Some of the necessary interaction energies between vacancy complexes have been published recently by Beeler and Johnson.⁽³⁾ The modified code should enable the simulation of spikes in high temperature irradiations of pure metals. In particular the modified codes will permit the study of the agglomeration of vacancies as a function of temperature, flux, and PKA spectrum.

After stable high temperature defects are identified, experiments may be suggested to investigate the correlation of a physical or mechanical property change with the defect density. The computer program calculations will provide a theoretical basis for the correlation of radiation damage data to FTR. The new correlations under investigation will aim at the prediction of high temperature material radiation damage in the FTR environment, based on experimental data.

The DEFECT computer program is being used to calculate the configuration energy for vacancy, interstitial, and impurity atom complexes in crystals. DEFECT calculates defect formation energies, binding energies, and migration energies, as well as geometrical properties of defect complexes. This information will assist the interpretation of experimental data on radiation damage, as well as the study and development of improved helium related damage correlation.

2. HELIUM PRODUCTION BY FAST NEUTRON (n, α) REACTIONS

High energy neutron (n, α) interactions with the matrix elements of stainless steels and nickel alloys produce a significant quantity of helium. The most reactive elements are Fe, N, Ni, and Cr. DePino⁽¹⁴⁾ has calculated the helium concentration to be 136 ppm in 304 stainless steel for a hypothetical fast fluence ($E > 0.18$ MeV) of 6.6×10^{23} neutrons/cm² in the FTR.

These irradiated materials may embrittle, lose ductility, and become more susceptible to fracture. Structural metal failure caused by the production of helium in the matrix elements of the steel is characterized by intragranular or transgranular fracture, and is not necessarily associated with bubble formation. The damage mechanism has not been determined, and its dependence on the movement of helium bubbles, as well as the effect of an interstitial helium atom interaction with either a void or complex defect are also unknown. Inconsistencies have been observed in the comparison of experiments which are based on either helium bubble concentration or atomic helium production.

Present correlation must be based on calculations of the atomic helium concentration produced by fast neutrons in the matrix elements of the materials. These calculations are simply an integration of the neutron spectrum $\phi(E)$ and the macroscopic absorption cross section $\Sigma_a(E)$ for the nuclear interaction.

$$N = \int_0^{\infty} \phi(E) \Sigma_a(E) dE \quad (2)$$

Correlation of radiation damage attributable to helium production could be based on the measurement of the effect, calculation of the helium concentration, and its measurement by spectrographic analysis. Thus, a preliminary prediction of FFTF helium damage would be based on a calculation of predicted helium production in the FFTF and an experimentally based correlation.

Calculations in progress will be used to study the interactions of interstitial atoms with vacancy clusters. They are intended to assess relative importance of helium distributed in the lattice and agglomerated into bubbles. These calculations may suggest experiments to determine the role helium plays in the enhancement or restriction of other defects.

Several other significant problems preclude the complete understanding of the embrittlement caused by matrix element (n, α) reactions. As an example, temperature plays an important role in the diffusion and agglomeration of the atomic helium into a configuration which is critical to the reduction of ductility.⁽¹⁵⁾

3. HELIUM PRODUCTION BY BORON IMPURITIES

Helium production by boron transmutation also may cause materials to lose ductility or become more susceptible to embrittlement and fracture. Boron impurity atoms apparently are concentrated in the grain boundaries of steel during solidification and cooling. Thermal and epithermal neutron absorption causes the boron to emit alpha particles which become helium. This helium is generated in the immediate neighborhood of the grain boundaries and may later agglomerate into bubbles. Thus, it is reasonable that characteristic fracture of metals embrittled by helium from $^{10}\text{B}(\text{n},\alpha)$ reaction is intergranular. Cracks usually originate at triple grain boundaries, then propagate along boundaries exclusively.

Experimental data on the changes in ductility and embrittlement have been related to helium concentration.⁽¹⁵⁾ Such radiation damage data are correlatable to the FTR problems through helium concentration calculations.⁽¹⁴⁾ Calculations predict that the atomic helium produced in the FTR from the $^{10}\text{B}(\text{n},\alpha)^7\text{Li}$ reaction is 13 ppm, based on a thermal fluence of 1.7×10^{19} neutrons/cm² (fast fluence of 6.6×10^{23} neutrons/cm²,

$E > 0.18$ MeV). This accounts for less than 10% of the total helium production in stainless steel of the FTR (149 ppm); the balance is produced by fast neutron (n, α) reactions with matrix atoms. Therefore, helium production by slow neutron absorption is not too significant, but if helium diffuses from grain interiors, its concentration at grain boundaries may enhance the effect of helium from boron.

CONCLUSIONS

Since FTR materials have not been finally selected, all pertinent radiation damage problems cannot be identified. However, the majority of these problems probably fall into either of two damage mechanism categories: (1) Extended lattice defects are created in the crystal structure due to collision cascades, and (2) impurity atoms from nuclear reactions are injected into the lattice, or they agglomerate into precipitates or bubbles.

Correlation of mechanical property radiation damage currently is based on gross defect concentration. Better damage correlation techniques based on this concept, but which also consider the effects of defect cluster behavior in collision cascades, or other defect related damage, are being developed.

Transmutation damage such as helium production, can be correlated on the basis of the number of transmutation nuclear reactions. Improvements in these damage correlations are being studied by computer simulation of damage processes.

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