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EFFECT OF NEUTRON SPECTRA ON THE SWELLING OF CERAMIC
INSULATORS AND IMPLICATIONS FOR THERMIONIC REACTOR DESIGN *

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ABSTRACT

Data on swelling and loss of vacuum integrity of alumina due to neutron irradiation are now available for several neutron spectra. A neutron-energy-dependent model for predicting relative numbers of neutron-induced atomic displacements in alumina in different neutron spectra is presented which correlates the available swelling data reasonably well. The model has been extended to yttria and magnesia. It is shown that neutron fluence, $E_n > 0.1$ MeV, is an adequate parameter for use in practical applications for predicting relative numbers of neutron-induced atomic displacements in each of the three materials.

The implications of neutron-induced damage to ceramic insulators for thermionic reactor design are discussed, and core designs optimized for minimum neutron damage to in-core ceramic insulators at constant core electric power density are described.

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Introduction

Sufficient data has been accumulated on the damaging effects of fast neutrons to ceramic insulators^{1,2,3} to make it clear that these effects must be considered in the design of long-lifetime thermionic reactors.

Fast neutron damage has been shown to result in a decrease in the thermal conductivity of Al_2O_3 and Y_2O_3 and to have a minor adverse effect on the electrical resistivities of these materials; however, the major effect produced in Al_2O_3 is that of anisotropic distortion of the Al_2O_3 grains which results not only in dimensional increases, but also of micro-cracking of the ceramic structures with concomitant losses of vacuum integrity, strength, and eventually of structural integrity.

Because the damage resulting from a neutron collision with a lattice atom is dependent on the energy of the neutron, the amount of damage done per unit time to a ceramic material in a radiation field will depend on the neutron-energy spectrum as well as on the magnitude of the neutron flux. Test-reactor environments with correct neutron-flux magnitudes and spectral shapes to match the conditions of intermediate- and fast-neutron spectrum thermionic reactors do not exist. Thus, it has been necessary to conduct irradiation damage experiments on ceramic and ceramic-metal components in test reactors with neutron spectra which differ significantly from those of reference thermionic reactor designs. It is therefore imperative that adequate methods be developed for treating the effects of spectral shape on damage rates in order that experimental data can be applied to thermionic reactor design.

One method of comparing neutron-damage data obtained in different neutron spectra is to compare radiation exposure in terms of neutron fluences for neutrons with energies greater than 1.0 MeV, independent of the detailed spectra of such neutrons. This practice was widely followed until recent years and lingers today in some quarters. It is a very poor way of measuring the actual neutron-damage fluence experienced by an irradiated specimen. This is shown by the data plotted in Fig. 1 where volume swelling of small-grain, General Electric Co. Lucalox Al_2O_3 is used as the measure of actual damage and the fluence of neutrons, $E_n > 1.0$ MeV, is used as a measure of irradiation exposure. The data obtained in water-moderated reactors with MTR-type fuel elements, i.e., the Omega West Reactor (OWR) and the Engineering Test Reactor (ETR), do not correlate well with the data obtained in the fast-neutron-spectrum Experimental Breeder Reactor (EBR-II). This type of discrepancy has led to the formulation of a simple neutron-damage model which better approximates the effect of neutron energy on the relative damage potentials of different neutron spectra. The model has been used to predict the relative numbers of atomic displacements occurring per unit of neutron fluence in Al_2O_3 , Y_2O_3 , and MgO for different neutron spectra.

Theoretical Model

The model for ceramic materials has been developed following the general methods of Kinchin and Pease.⁴ It provides a means of estimating the dependence on neutron energy of the average number of atomic displacements resulting from a neutron interaction. The total number of displacements produced per unit of total neutron flux can then be obtained by a convolution integral of the displacement function and the neutron energy spectrum.

Damage is assumed to be proportional to the number of atoms displaced in the irradiated material. The damage model can thus be used to compare neutron-damage data from reactors with different neutron spectra; it is unnecessary to determine the constants of proportionality.

A "damage parameter" is defined as:

$$\text{Damage Parameter} = K_1 \int_0^{\infty} [\text{RD}(E)] \phi(E) dE$$

where:

K_1 = proportionality constant, arbitrarily set equal to 0.001,

$\phi(E)$ = neutron flux, $n/\text{cm}^2\text{-sec-MeV}$, normalized to $\int_{1.0 \text{ MeV}}^{\infty} \phi(E) dE = 1$, and

$\text{RD}(E)$ = relative damage = number of atomic displacements resulting from the interaction of a neutron with energy E and the insulating material.

The neutron-energy-dependent coefficient of the neutron flux, RD , is defined for each of the materials of interest by:

$$[\text{RD}(E)]_{\text{Al}_2\text{O}_3} = [2\sigma_s^{\text{Al}}(E) \nu_{\text{Al}}(E) + 3\sigma_s^{\text{O}}(E) \nu_{\text{O}}(E)]$$

$$[\text{RD}(E)]_{\text{Y}_2\text{O}_3} = [2\sigma_s^{\text{Y}}(E) \nu_{\text{Y}}(E) + 3\sigma_s^{\text{O}}(E) \nu_{\text{O}}(E)]$$

and

$$[\text{RD}(E)]_{\text{MgO}} = [\sigma_s^{\text{Mg}}(E) \nu_{\text{Mg}}(E) + \sigma_s^{\text{O}}(E) \nu_{\text{O}}(E)]$$

where:

$\sigma_s(E)$ = microscopic neutron-scattering cross section,

and

$\nu(E)$ = number of atomic displacements created per neutron-scattering event.

Atomic displacements result both from scattering events between neutrons and atoms and scattering events between displaced atoms and other atoms in the crystal structure. An atom which is displaced by a neutron is called a primary knock-on atom, and the energy transmitted by the neutron to the primary knock-on atom is denoted by E_p . The energy, E_d , required to displace an atom from its crystal location is assumed to be 25 eV. If E_p is less than 25 eV, it is assumed that the energy is dissipated as heat and no displacement occurs. If E_p is between 25 and 50 eV, one displacement takes place, and above 50 eV, the number of displacements is linearly dependent on E_p up to an ionization energy limit, E_I . Above this energy the primary knock-on atoms lose an increasing proportion of their energy to lattice atoms through inelastic ionizing interactions which do not result in displacements. For simplicity, it is assumed that this transition is abrupt and that the number of displacements produced per primary knock-on atom is constant for $E_p > E_I$.

The target atoms can be ionized only if the kinetic energy of the electrons of the moving atom is of the order of magnitude of the excitation energy of the target electrons. Seitz⁵ suggested that the threshold kinetic energy of the moving electrons required to cause ionization is $\sim I/8$, where I is the value of the forbidden-energy gap for an insulator. However, present correlations for alumina indicate that $I/6$ may be a better assumption for this threshold. Applying this relationship to the energy of the primary knock-on atom, the threshold for ionization is:

$$E_I = \frac{M}{m_e} \left(\frac{I}{6} \right)$$

where:

- M = average mass of target atoms,
- m_e = mass of an electron, and
- I = forbidden-energy gap for the target insulator, assumed to be 10 eV for the materials considered.

Assuming isotropic elastic scattering, the average energy transmitted to a primary knock-on atom per collision with a neutron of energy E is given by:

$$E_p = \frac{2M}{(1+M)^2} E$$

where M is the mass of the primary knock-on atom.

Perturbations due to the anisotropy of elastic scattering have not been included because the magnitudes of the neutron fluxes, for neutron energies at which such effects are important, are small for neutron spectra of interest. Inelastic scattering, $(n, 2n)$, and (n,p) reactions have been omitted for the same reason.

The average number of displacements per neutron interaction, $\nu(E)$, is a function of E_p and is given by:

$$\nu(E_p) = \begin{cases} 0, & E_p < 25 \text{ eV} \\ 1, & 25 \text{ eV} \leq E_p < 50 \text{ eV} \\ E_p/50, & 50 \leq E_p \leq E_I \\ E_I/50, & E_I < E_p \end{cases}$$

By applying the equations above to the materials of interest, the relative numbers of displacements per unit volume and unit of neutron flux can be calculated as functions of neutron energy. These functions are shown for Al_2O_3 , Y_2O_3 , and MgO in Fig. 2. These relative displacement functions are multiplied by the normalized neutron fluxes and integrated over all neutron energies to find the damage parameters. Molecular densities are included in the functions in Fig. 2 in order that different materials may be compared; densities are not included in correlations of experimental results presented in subsequent figures.

A significant aspect of the curves in Fig. 2 is the importance of neutrons with energies in the range of 0.1 to 1.0 MeV insofar as displacement production is concerned. It is because of this and the fact that

neutron energy distributions vary markedly in this energy region for different reactor types that neutron fluence for $E_n > 1.0$ MeV is such a poor method of correlating experimental ceramic irradiation data from different reactors. The variation of neutron spectrum shape is shown in Fig. 3 for three locations in the EBR-II, a typical fast-neutron-spectrum thermionic reactor, and an MTR-type reactor; these plots are normalized such that

$\int_0^\infty \phi(E) dE = 1$. The spectra for the EBR-II were calculated from published data.⁶ The thermal-test-reactor spectrum is typical of water-moderated reactors with MTR type fuel elements.

The importance from a damage standpoint of neutrons with energies in the range $0.1 < E_n < 1.0$ MeV is illustrated more directly in Fig. 4 where the damage (as predicted from the atomic displacement rate) caused by neutrons with energy greater than a given value, E_n , is plotted as a function of E_n for the reactor spectra given in Fig. 3. For the above-the-core position in Row 7 of the EBR-II, 58% of the displacements calculated by this model are caused by neutrons with energies less than 0.4 MeV and only 8% are caused by neutrons with energy greater than 1.0 MeV.

Application of Model to Various Neutron Spectra

In order to compare the damage potential of various reactor spectra it is convenient to have a standard reference spectrum. A logical choice for such a reference is the Watt fission spectrum, $\phi_w(E)$.⁷ This spectrum has been used to generate a quantity called the spectral damage factor (SDF) which is defined as follows:

$$SDF = \frac{\int_0^\infty (RD) \phi_a(E) dE}{\int_0^\infty (RD) \phi_w(E) dE}$$

where the various spectra are normalized by the relationship

$$\int_{1.0 \text{ MeV}}^\infty \phi(E) dE = 1 .$$

The SDF is the ratio of the number of atomic displacements produced in a given material per unit time by an arbitrary neutron spectrum, $\phi_a(E)$, to that which would be produced by the Watt fission spectrum when the magnitude of each spectrum has been adjusted to give the same integrated value for the neutron flux, $E_n > 1.0$ MeV.

Typical values of the SDF for various positions in the EBR-II, for the water-moderated OWR, and for a typical fast-neutron-spectrum thermionic reactor are given in Table I. From this table one finds, for instance, that a fluence of 10^{21} n/cm², $E_n > 1.0$ MeV, at the axial centerline of Row 7 in the EBR-II produces as many atomic displacements in Al₂O₃ as a fluence of $5.28/1.34 \Rightarrow 3.94 \times 10^{21}$ n/cm², $E_n > 1$ MeV, in the OWR (or ETR).

Irradiation data are usually reported for fast neutron fluences ($E_n > 1.0$ MeV), a practice which is related to the fact that thresholds for nuclear reactions used to measure fast neutron fluence are generally 1.0 MeV or greater. If the fast neutron fluence is multiplied by the SDF then

the product is proportional to the total damage that would be predicted by the damage model.

Correlation of Theory and Experimental Data

The product SDF times the neutron fluence, $E_n > 1.0$ MeV, has been used to correlate the experimental data. The data given in Fig. 1 have been correlated in this manner and are shown in Fig. 5. The standard deviation of all the points around the least-squares linear fit is 23%.

Neutron induced swelling data obtained in water-moderated, MTR type reactors for Lucalox Al_2O_3 is plotted vs neutron fluence, $E_n > 1.0$ MeV, in Fig. 6. There are essentially no differences between the neutron spectra in such reactors; thus, the data should correlate reasonably well as a simple function of neutron fluence. The standard deviation of the points around the least squares fit is 27%. This compares with the standard deviation of 23% for the fit to all of the Lucalox Al_2O_3 data given in Fig. 5 and suggests that the deviations may be due primarily to variations in such properties as internal stress states or possibly specimen impurity level and grain size rather than to an improper evaluation of the effect of spectral differences.

It is probable that radiation damage to Al_2O_3 , as evidenced by volume swelling, is not a linear function of neutron fluence; however, because of the spread in the data, there appears to be no reason to attempt a more complex fit than a straight line at this time.

The correlation of the data by the model is reasonably good. However, the model is more complicated to use than a correlation with neutron fluence only. Therefore, an approach similar to that suggested by Dahl and Yoshikawa⁸ was investigated in an attempt to find a threshold energy, E_T , by which a damage neutron fluence ($E_n > E_T$) can be defined. Values of the quantity C, given by

$$C = \frac{\int_0^{\infty} (RD) \phi(E) dE}{\int_{E_T}^{\infty} \phi(E) dE}$$

where $\int_{1.0 \text{ MeV}}^{\infty} \phi(E) dE = 1$ and E_T was varied from 0.05 to 1.0 MeV, were calculated for each material of interest for several different reactor neutron spectra.

If a value of E_T can be found that results in the same value of C for various neutron spectra, then it should be possible to correlate neutron induced damage by neutron fluence, $E_n > E_T$.

The results of these calculations are shown in Figs. 7, 8, and 9 for Al_2O_3 , Y_2O_3 , and MgO , respectively. The data for Al_2O_3 and Y_2O_3 converge at a value of $E_T \sim 0.15$ MeV, and the MgO data converge below 0.1 MeV. The variation in the value of C at 0.1 MeV is less than 15% for all three materials. It is doubtful that the accuracy of the model or of calculated neutron spectra is much better than this. Therefore, it is reasonable to use the fluence of neutrons with $E_n > 0.1$ MeV to compare the damage potential of various neutron spectra for all three materials.

The data given in Fig. 1 is plotted as a function of neutron fluence ($E_n > 0.1$ MeV) in Fig. 10. The standard deviation for this correlation is 27%; although not quite as good as the correlation given in Fig. 5, it may be more useful in practical application.

Implications for Thermionic Reactor Design

The neutron-damage model described above has been used to compare alumina insulator lifetimes for several different thermionic reactor designs.

Radiation damage to the alumina insulators was calculated for several different thermionic-reactor core designs, including fast-neutron-spectrum as well as highly moderated cores. The study was restricted to reactor designs containing conventional, internally-fueled thermionic emitters.

Neutron spectrum calculations were made for thermionic reactor cores based on two thermionic fuel element (TFE) designs, one containing tungsten-clad UO_2 -fueled emitters and the other tungsten-clad Mo- UO_2 -cermet fueled emitters. The composition of the TFEs is given in Table II. Although the specific core designs for which calculations were made are typical of those containing small-diameter emitters (i.e., ~ 13 mm diam), the results of the study also apply, with limitations, to a much larger class of reactor designs.

The moderator was included in the reactor cores as Hastelloy-N clad ZrH_x rods. Cases for which the zirconium was alloyed with 5 w/o uranium as well as cases with unfueled moderator rods were considered. The reactor coolant, assumed to be sodium-potassium alloy, comprised between 11 and 19% of the core volumes. The variation in coolant volume fraction is due to the different coolant requirements of moderated, unmoderated, and driver-fuel designs.

The diameters of TFEs, fueled moderator rods, and unfueled moderator rods included in specific core designs were 17.3, 17.8, and 18.3 mm., respectively. The reactor cores consisted of hexagonal arrays of TFEs and moderator rods, with coolant flow in the interstices between the rods.

In order to make meaningful comparisons of insulator damage between different reactor designs it is necessary to establish a basis for comparison. The ground rule used in this study was to keep the electric-power output per unit of core volume constant for the several reactor designs considered. It was also assumed that the thermionic efficiency was the same for all cases.

Relative damage to insulators for constant electric power output per unit of core volume as a function of the number of moderator rods per TFE is plotted in Fig. 11. The results are normalized so that the insulator damage for an unmoderated core of UO_2 -fueled TFEs is unity. The hydrogen-to-uranium ratios in the designs considered in Fig. 11 ranged from 0 to 36.

An optimum amount of moderating material for minimum insulator damage at constant core electric power density is indicated in Fig. 11 for each of the TFE-moderator rod combinations. Reductions in insulator damage, compared to an unmoderated UO_2 -fueled TFE core, by factors of two and three are obtainable for cores containing fueled and unfueled moderator rods, respectively. The inclusion of only one unfueled moderator rod for every three TFEs would reduce insulator damage by almost a factor of two.

The conclusions from Fig. 11, stated differently are: the fractional reduction in insulator damage resulting from the inclusion of hydrogenous moderator is larger than the fraction of core volume occupied by the moderating material. This statement is true for core-volume fractions of moderator that are less than, or equal to, the optimum amounts indicated for each of the TFE-moderator rod combinations.

Reductions in insulator damage due to the inclusion of moderating materials can be equated directly by equal factors to either increased insulator lifetimes, increased core power densities, or increased factors of safety.

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TABLE I. EFFECT OF NEUTRON SPECTRA ON SDF

<u>Reactor</u>	<u>SDF</u>
EBR-II axial centerline, Row 7	5.28
EBR-II top of core, Row 7	6.34
EBR-II top of core, Row 4	2.98
OWR, water moderated thermal reactor	1.34
Fast-neutron-spectrum thermionic reactor	2.08

TABLE II. COMPOSITION OF THERMIONIC FUEL ELEMENTS

<u>Component</u>	<u>Volume Fraction of TFE</u>
Fueled emitter	0.485
Cesium gap	0.036
Collector	0.118
Sheath insulator	0.111
Outer sheath	0.058
Intercell space	0.192

- Fig. 1. Volume swelling of Lucalox Al_2O_3 vs fast neutron fluence.
- Fig. 2. Relative atomic displacements in insulating materials vs incident neutron energy.
- Fig. 3. Typical neutron spectra of interest to thermionics materials-irradiation program.
- Fig. 4. Integrated fractional neutron damage as functions of neutron energy for alumina.
- Fig. 5. Volume swelling data of Fig. 1 correlated by product of spectral damage factor and neutron fluence.
- Fig. 6. Volume swelling of Lucalox Al_2O_3 in moderated test reactors vs neutron fluence.
- Fig. 7. Ratio of damage parameter for Al_2O_3 to integrated neutron flux vs lower limit of integration.
- Fig. 8. Ratio of damage parameter for Y_2O_3 to integrated neutron flux vs lower limit of integration.
- Fig. 9. Ratio of damage parameter for MgO to integrated neutron flux vs lower limit of integration.
- Fig. 10. Volume swelling of Lucalox Al_2O_3 vs neutron fluence, $E_n > 0.1$ MeV.
- Fig. 11. Relative damage to insulators for constant electric power per unit core volume vs number of moderator rods per TFE.

Fig. 1. VOLUME SWELLING OF LUCALOX Al_2O_3 vs FAST NEUTRON FLUENCE.

Fig. 2. RELATIVE ATOMIC DISPLACEMENTS IN INSULATING MATERIALS vs INCIDENT NEUTRON ENERGY.

Fig. 3. TYPICAL NEUTRON SPECTRA OF INTEREST TO THERMIONICS MATERIALS-IRRADIATION PROGRAM

Fig. 4. INTEGRATED FRACTIONAL NEUTRON DAMAGE AS FUNCTIONS OF NEUTRON ENERGY FOR ALUMINA

Fig. 5 VOLUME SWELLING DATA OF Fig.1 CORRELATED BY PRODUCT OF SPECTRAL DAMAGE FACTOR AND NEUTRON FLUENCE.

Fig. 6. VOLUME SWELLING OF LUCALOX Al_2O_3 IN MODERATED TEST REACTORS vs NEUTRON FLUENCE

Fig. 7. RATIO OF DAMAGE PARAMETER FOR Al_2O_3 TO INTEGRATED NEUTRON FLUX vs LOWER LIMIT OF INTEGRATION.

Fig. 8. RATIO OF DAMAGE PARAMETER FOR Y_2O_3 TO INTEGRATED NEUTRON FLUX vs LOWER LIMIT OF INTEGRATION.

Fig. 9. RATIO OF DAMAGE PARAMETER FOR MgO TO INTEGRATED NEUTRON FLUX vs LOWER LIMIT OF INTEGRATION.

Fig.II. RELATIVE DAMAGE TO INSULATORS FOR CONSTANT ELECTRIC POWER PER UNIT CORE VOLUME vs NUMBER OF MODERATOR RODS PER TFE.