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The Effect of Neutron Energy On Defect Production In Alumina

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ABSTRACT

In a deuterium-tritium fusion reactor, neutrons are produced at an energy of about 14 MeV. When these neutrons and other lower energy neutrons from (n,xn) reactions travel through insulating materials in the reactor, displacement damage will occur as they interact with atoms in the insulating material. Using the Monte Carlo Code TRIM (Transport Ions in Matter), calculations using Al and O ions at energies that simulate expected recoil energy levels in alumina (Al_2O_3) were performed. This study focuses on recoil energy ranging from 100 eV to 10 MeV. In addition, recoil energy spectra for aluminum and oxygen were calculated for 1 MeV and 14 MeV neutrons using the computer code, SPECTER. Combining the TRIM and SPECTER calculations, it was possible to determine the number of aluminum and oxygen vacancies created in alumina resulting from a specific neutron energy.

INTRODUCTION

Ceramic insulators for fusion applications are subject to neutron bombardment which can degrade electrical and mechanical properties. An essential part of understanding this degradation is understanding radiation produced atomic displacements in materials. Such displacements can be calculated with modern computer codes. For example, TRIM can calculate the the number of vacancies, displacements, interstitials, and the percentage of energy lost to ionization, phonons, and vacancies. The simulations for alumina (Al_2O_3) reported here were made using the computer codes TRIM and SPECTER.

TRIM is a monte carlo code that calculates the slowing down and scattering of energetic ions in amorphous targets [1]. SPECTER simulates damage

caused by mono-energetic specific neutron irradiation. Interfacing the results from TRIM and SPECTER, it was possible to calculate the number of aluminum (Al) and oxygen (O) vacancies for 1 MeV and 14 MeV neutrons into (Al₂O₃).

THEORY OF BOMBARDING NEUTRONS

Fig. 1. shows a density of neutrons ρ_n with velocity v (neutron flux ϕ_n) traveling through a cross-section (σ) with density (ρ). In a neutron flux, neutrons will produce some type of interactions with atoms in its path. The number of interactions (n') per second (t) is the interaction rate. Interaction rate per atom (κ_i) per neutron flux is called the neutron cross-section with units of barns (10^{-24} cm^2) denoted by $\sigma_n = \kappa_i / \phi_n$ where i is a type of interaction (i.e., absorption, displacement, fission, etc.).

Manipulating the σ_n equation by substituting for κ_i , σ_n and ϕ_n :

$$\sigma_n = \kappa_i / \phi_n \quad (1)$$

$$\sigma_n = n' / t \text{ atom} \cdot \rho_n v \quad (2)$$

$$\sigma_n = n' A / n \text{ atom} \quad (3)$$

where A is an area in cm^2 .

We see that σ_n represents the number of interactions per atom incident neutron per area.

Rearranging the σ_n equation, the number of interactions per atoms can be calculated for non mono-energetic incident neutrons.

$$\kappa_i = \sigma_n \phi_n \quad (4)$$

where $\kappa_i = \int \sigma_i(E) \phi'_E dE$ (the interaction rate per atom)

$\sigma_i(E)$ = cross-section for neutrons of energy E and interaction type i

$\phi'_E dE$ = the flux of neutrons with energies between E and $E + dE$

In a mono-energetic case:

$$\kappa_i = \sigma_i \phi \quad (5)$$

$$n_i = \kappa_i t = \sigma_i \phi t \quad (6)$$

and $\phi t = \Phi$ = fluence. If ϕ is a function of time, then the number of interactions per atom is [2]:

$$n_i = \sigma_i \int \phi(t) dt \quad (7)$$

The displacement of atoms by incident neutrons is produced by elastic or inelastic collisions. Figure 2. illustrates the type of interactions that can occur. Atoms that are displaced by neutrons are called Primary Knock-on Atoms (PKA's). If substantial energy is transferred ($T \gg E_d$, where T is transferred energy and E_d it the threshold energy or the minimum energy required to displace an atom from its lattice site), then the PKA is capable of displacing additional atoms called Secondary Knock- on Atoms (SKA's). Higher Knock-on Atoms or recoil atoms maybe displaced by SKA's. If $T < E_d$, then the atom cannot escape its site and any energy absorbed is lost as thermal lattice vibrations called phonons. Interstitials are created when energetic ions come to a rest ($T = 0$) occupying space between lattice sites or vacant sites.

Inelastic collisions can cause electron excitation or nuclear reactions. Ionization occurs by an energetic ion transferring energy to orbital electrons of atoms in which the energy will be given off as photons or phonons described in Itoh and Tanimura [3]. In a nuclear reaction, neutrons are absorbed by the atom's nucleus causing transmutation. The cross-sections and the types of nuclear reactions that occurred for a 1 MeV and 14 MeV neutron into Al and O are shown in Table 1. taken from references [4]&[5].

Energy loss in the TRIM calculations can be accounted for in ionization, vacancies, and phonons.

Displacements in the TRIM code is based on Kinchin-Pease Theory [6] and the Lindhard Energy-Partitioning Theory [7]. The SPECTER code incorporated the Lindhard Energy-Partitioning Theory.

SIMULATIONS

PART I TRIM

TRIM computations were made by assigning specific PKA energies, ranging from 100 eV to 10 MeV, to Al and O bombarding ions into Al_2O_3 . To determine the vacancy production for each Al and O recoil atom that were created by the Al and O incident ions, the vacancy text file was analyzed. The vacancy text file is a standard output of the TRIM program which calculates the number of vacancies per depth interval [8]. The data in the vacancy text file was integrated by using the summation technique to calculate the number of Al and O recoil atom vacancies produced at each PKA energy for each Al and O incident ion. Using the results, a graph was constructed shown in figure 3., plotting the Number of Vacancies vs. PKA Energy. The graph consists of six curves: Al PKA's creating Al recoil atoms (Al, Al); Al PKA's creating O recoil atoms (Al, O); O PKA's creating O recoil atoms (O,O); O PKA's creating Al recoil atoms (O,Al); the sum of Al and O PKA's creating Al recoil atoms (Al,Al) + (O,Al); and the sum of Al and O PKA's creating O recoil atoms (Al,O) + (O,O). A 7th order polynomial was fitted to each curve. The equation for each curve describes the number of vacancies created for a given PKA energy for a specific type of PKA and recoil atom.

PART II SPECTER

SPECTER simulations were calculated for 1 MeV and 14 MeV neutrons into Al and O. The output data from SPECTER are the recoil energy spectra for Al and O PKA's. These data was distributed into bins and distribution function graphs, the probability that an interaction might occur per unit PKA energy, were plotted for 1 MeV and 14 MeV neutrons into both Al and O which are shown in figure 4.

Figure 5. illustrates the nomenclature of the TRIM and SPECTER computer codes. Table 2 shows the adjustments of the TRIM data before it was analyzed. The incident ions, PKA's and SKA's were treated as PKA's, SKA's and recoil atoms respectively.

The TRIM results and the SPECTER data were interfaced by substituting the midpoint energy for each bin in SPECTER data into the 7th order polynomial equations from TRIM calculations to determine the number of vacancies (N_v) created for a PKA of that midpoint recoil energy. N_v was normalized by multiplying it by the fraction of the PKA's with each midpoint energy. These normalized N_v values were summed together to calculate the total number of vacancies per neutron. This procedure was done for each type of interaction for both 1 MeV and 14 MeV neutrons.

The SPECTER calculations were for neutron interactions with pure Al and O separately. In the case of Al_2O_3 , the stoichiometry must also be taken in consideration. For a neutron bombarding into Al_2O_3 , the probability of interaction with an Al atom is two-fifths and with an O atom three-fifths. Therefore the number of vacancies for (Al,Al); (Al,O) and (O,Al); (O,O) was multiplied by the stoichiometry 40% and 60% respectively. In addition, the neutron cross-section [4]&[5], the measure of the probability of occurrence of a neutron-Al or neutron-O collision was included in the computations.

RESULTS

Table 3. shows the results of calculations comparing them with the neutron cross-sections.

DISCUSSION

STATISTICAL PRECISION

Before starting the TRIM calculations, several calculations were performed to determine how many ions would give good statistical calculations. Using 100 keV Al bombarding into Al_2O_3 , and keeping the input data parameters constant, calculations were made at 200 ions, 400 ions, 500 ions, 1000 ions, 4500 ions and 6000 ions. Although the statistics were excellent for 6000 ions and 4500 ions showing smooth curves, using these values for the ions would have been computer time-consuming. Comparing the graphs of 200 ions, 400 ions, 500 ions and 1000 ions, we decided to use 1000 ions based on how free the graphs were from irregularities.

VACANCY TEXT FILE

When examining the vacancy text file in TRIM, the data in the vacancy text file was arranged in columns and labeled Depth, PKA, SKA, and SKA. In a calculation with a target containing A and B atoms, the depth is the distance of the bombarding ion when it interacted with an atom in the target. The Primary Knock-on Atom (PKA) column contains all the A and B atom that were displaced by the bombarding ions, the first Secondary Knock-on Atom or SKA column contains all the A recoil atoms that were displaced, and the second SKA column contains all the B recoil atoms that were displaced. Subsequently the PKA's

calculated in TRIM are being used as SKA's, therefore it was important to separate the A and B atoms in the PKA column of the vacancy text file.

The collision text file in the TRIM code gives a summary of the PKA atoms by element. Therefore it was possible to decipher the number of PKA A atoms and PKA B atoms at each recoil energy. To create statistical consistency, the collision text file was activated for a certain number of bombarding ions to interact with at least 100 ions for each energy. Determining the total number of SKA for the A atoms and B atoms, the number of PKA's for each A atoms and B atoms were summed. The values of the PKA A atoms and the PKA B atoms were added to the SKA atoms and the KS B atoms respectively to give the total of the SKA atoms for each element.

Comparing the total number of vacancies calculated using the vacancy text file and the collision text file with the standard output of the total number of vacancies given by TRIM, there was an error ranging from 0% to 3%.

RANDOM SEED GENERATOR

TRIM computer code has a manual random seed generator. Meaning each time the same calculation is performed the results are identical. To change the results of the calculations, the random seed number would have to be changed. Since there was no automatic random seed generator, all calculations were computed with the random seed number zero. The TRIM program suggested zero was the best seed to use. A problem evolved when trying to determine the number of vacancies for an Al ion bombarding into alumina at 2 keV. In the vacancy text file, the summation of the three columns, the PKA column and the SKA columns for Al and O, equal the number of vacancies produced. For the calculation at 2 keV, the summation of all three columns was higher than the total number of vacancies that the program gave. Computing this calculation at

a different random seed number of 2, this error was corrected. We believe there is a bug in the program at an Al ion bombarding Al_2O_3 at 2 keV. This problem did not occur with an O ion bombarding Al_2O_3 at 2 keV.

THRESHOLD ENERGY

Although the threshold energy, E_d , is at issue for non-metals, it is a fundamental parameter in evaluating the defect production in all calculations. For example, in non-metals, E_d depends on bond strength which can predict the spacing availability between atoms in the structure for accomadating interstitial atoms [9]. Displacement energies of 18eV for Al and 72 eV for O were used in the Al_2O_3 simulations which these values were calculated by Pells and referenced from Parkin and Coulter [10].

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Fig. 1. Neutron flux Φ_n approach cross-section σ_n with density ρ_n .

Fig. 2. Types of interactions that occur under neutron bombardment. Other types of transmutations $[(n,p),(n,2n),(n,\gamma)]$ and (n,d) are also possible.

FOR O-16

**@ $E_n = 1.00$ MeV
 $\sigma_{tot} = 8.22$ Barns
100% elastic**

**@ $E_n = 14$ MeV
 $\sigma_{tot} = 1.62$ Barns
59.0% elastic
29.3% inelastic
7.8% n, α
0.9% n,d
2.9% n,p**

FOR AL-27

**@ $E_n = 1.00$ MeV
 $\sigma_{tot} = 2.37$ Barns
99.8% elastic
0.2% inelastic**

**@ $E_n = 14$ MeV
 $\sigma_{tot} = 1.74$ Barns
37.4% elastic
48.1% inelastic
7.2% n, α
1.5% n,d
4.5% n,p
1.4% n,2n**

Table 1. shows neutron cross-sections and the types of interactions that occurred for 1 MeV and 14 MeV neutrons into Al and O.

Fig. 3.

Fig. 4a. **ALUMINIUM 1 MeV**

Fig. 4b. **OXYGEN 1 MeV**

Fig. 4c.

ALUMINIUM 14 MeV

Fig. 4d.

OXYGEN 14 MeV

Fig. 5. The nomenclature of TRIM and SPECTER computer codes.

TRIM COMPUTER CODE

THEORY

Table 2. A Display of how TRIM and SPECTER data was adjusted before computations.

THEORY	Incident Ion	PKA	SKA	Recoil Atom
TRIM		Incident Ion	PKA	SKA
SPECTER	Neutron	PKA		

Number of Vacancies per Atom (DPA)

Table. 3a.

	1 MeV		14 MeV	
Al,Al	.3 DPA	300 barns	.8 DPA	800 barns
O,Al	2.1 DPA	2100 barns	.5 DPA	500 barns
Al,O	.2 DPA	200 barns	.4 DPA	400 barns
O,O	4 DPA	4000 barns	.2 DPA	200 barns

Table. 3b.

	1 MeV		14 MeV	
Al	2.4 DPA	2400 barns	1.3 DPA	1200 barns
O	4.2 DPA	4200 barns	.6 DPA	600 barns

$$\left(\Phi \frac{n}{\text{cm}^2} \right) \left(\sigma_{\text{sect}} \text{ cm}^2 \right) = n$$

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