MASTER

CALCULATION OF RADIATION DAMAGE IN INSULATORS FOR FUSION REACTORS*

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The MORSE neutron transport code has been used to characterize the neutron and gamma ray fluxes in a typical irradiation cave from a Li(d,n) neutron source. Recoil-atom damage and gas generation have been evaluated for Al_2O_3 and Si_3N_4 using the damage program DON.

1. INTRODUCTION

The problem of radiation damage to insulators in critical regions of fusion reactors requires examination over the next few years. Insulator needs for fusion reactors have been characterized into four divisions [1]: 1) insulators for neutral beam injectors, 2) insulators for injector power supplies, 3) insulators for the direct converters, and 4) insulation for the magnets. The insulators used for the neutral beam injectors will experience the most severe radiation exposure, because they are nearest the plasma.

It is necessary to identify insulator candidate materials by determining the displacement damage as well as the gas production resulting from interactions with high fluxes of high energy neutrons. Testing of candidate materials for use as insulators in fusion reactors will be performed using irradiation facilities such as the proposed FMIT facility at HEDL. Prior to actual irradiation, promising insulator candidate materials should be identified by simulating damage induced by neutrons and gamma-rays at such a facility.

2. CHARACTERIZATION OF NEUTRON SPECTRA

The fluxes of neutrons and gamma-rays in a typical irradiation cave at a Li(d,n) neutron facility have been determined by using the three-dimensional transport code MORSE.

Neutrons are generated by a Monte-Carlo source routine that simulates neutrons produced by deuteron stripping and compound nuclear reactions in a thick lithium target. Parameters of the routine have been selected to produce an adequate simulation of the neutron spectra measured by Saltmarsh et al. at angles of 0° to 90° [2]. The time-of-flight yields of Saltmarsh et al. have been extrapolated to large angles and have been integrated to give an estimate of the neutron yield emitted into 4π solid angle. The resulting total yield at 40 MeV has been fitted to the yields at lower energies [3], and

a value of 2.2×10^{17} neutrons/C has been obtained as the total neutron yield produced by 30 MeV deuterons. This yield has been used to translate the results from MORSE to conventional flux units.

Neutrons, once generated, are followed by the MORSE transport code until their energy falls below 1.1 eV. The neutron flux consists of two components: the uncollided flux and the collided flux. The uncollided flux is obtained by determining the probability that a neutron on its first flight will reach a particular detector. The collided flux is determined at each collision point by computing the probability that the neutron on its next flight could reach a particular detector.

Flux characterizations have been made for the case of 30 MeV deuterons incident upon a lithium target that is 100 mm wide, 10 mm high, and whose thickness just exceeds the deuteron range. To assay the alteration of the flux by the presence of a test assembly containing test samples, flux characterizations have been made at several points along the center line of a rectangular $300 \times 200 \times 200$ (mm)³ block of iron located ad-

Fig. 1. Geometry used for flux characterizations.

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adjacent to the lithium source. The total thickness of iron is 1.572 g/(mm)^2 . The geometry is shown in fig. 1. Both the collided and the uncollided fluxes have been determined. The material within the test assembly attenuates the uncollided flux, while it produces the collided flux. As the depth within the test assembly increases, the relative importance of these two components changes. This change is shown in fig. 2. The dependence upon depth within the test assembly of the total neutron flux and the average neutron energy is listed in table 1. The neutron flux decreases by two orders of magnitude from the front to the back of the test assembly, and the average neutron energy decreases by a factor of nearly three. For less dense test assemblies the variation of flux and average neutron energy is reduced.

The contribution to the neutron flux arising from the walls of the irradiation cave has been determined by comparing the flux within a test assembly of iron surrounded by a void with the flux obtained when the test assembly is located within an irradiation cave having walls of high density concrete. The flux contribution from the walls of the cave was found to be less than 1% of the total flux. Consequently, most of our flux characterizations have been made for the case of a test assembly surrounded by a void. This geometry greatly reduces the computer time required to obtain acceptable results.

The coupled neutron-gamma transport problem has been analyzed for a test assembly surrounded by a void and for the case of the test assembly located in a cave having walls of high density concrete. At each collision point a test was made to determine whether or not a gamma-ray was generated. If generated, the gamma-ray was stored in a particle bank buffer while the history of the parent neutron was completed. Each gamma-ray was considered as a separate particle; its history was followed from its point of generation using the same tests and the same flux estimators that were used for neutrons. Gamma-rays were followed until their energy fell below 10 keV. As in the case of the uncoupled neutron

Fig. 2. Variation of the uncollided and collided fluxes with depth in the test assembly.

transport problem, the gamma-ray production in the walls of the irradiation cave did not contribute significantly to the gamma spectra within the test assembly. Typical neutron and gamma-ray spectra are shown in fig. 3, and the dependence upon depth in the assembly of the total neutron and gamma-ray fluxes is shown in fig. 4.

The neutron transport calculations were performed using the cross sections of Alsmiller and Barish [4]. These cross sections have 47 neutron groups extending to 50 MeV and 21 gamma-ray groups extending to 14 MeV. As these cross sections are available for only a limited number of elements that does not include many of those elements comprising insulators, the neutron flux was characterized by assuming the entire test assembly to be filled with iron and the walls of the irradiation cave to be high-density concrete. The resulting flux spectra have been used as

Table 1
Spectrum averaged displacement and gas production cross sections for Al_2O_3

Sample depth (mm)	5.0	10.0	20.0	40.0	80.0	140.0	190.0
Flux $\{10^{20} \frac{\text{neutrons}}{\text{m}^2 \cdot \text{A} \cdot \text{s}}\}$	2.38	1.87	1.09	0.587	0.224	0.0722	0.0226
$\bar{E}_n$ (MeV)	10.69	10.18	8.60	7.85	5.83	3.93	3.68
$\bar{\Sigma}_d(\text{Al})$ (kb)	1.067	1.073	1.025	0.996	0.932	0.782	0.869
$\bar{\Sigma}_d(\text{O})$ (kb)	0.411	0.413	0.395	0.385	0.361	0.304	0.337
$\bar{\Sigma}_H$ (mb)	23.7	23.0	21.7	18.9	13.7	6.85	7.13
$\bar{\Sigma}_D$ (mb)	5.31	5.54	5.00	3.85	3.07	1.57	1.78
$\bar{\Sigma}_T$ (mb)	1.29	1.44	0.990	0.902	0.660	0.361	0.358
$\bar{\Sigma}_{\text{He}}$ (mb)	49.0	45.7	46.0	40.5	29.0	14.1	14.6
$\bar{\Sigma}_{\text{C(gas)}}$ (mb)	73.9	75.7	73.7	64.1	46.5	22.9	23.9

Fig. 3. Neutron and gamma-ray spectra at point (0,0.5 mm) within the test assembly.

input to the damage program DON [5]. DON uses ENDF/B-IV cross sections, and hence damage calculations can be performed for many materials of interest for use as insulators. The Alsmiller cross sections are used in MORSE to transport high energy neutrons to lower energies. Once the flux spectrum has been characterized, the ENDF/B-IV cross sections are used with DON to evaluate the damage in materials. For that part of the neutron spectrum having energy greater than 20 MeV, the cross section used in DON is the 20 MeV cross section.

3. DAMAGE

Damage calculations consisting of recoil-atom damage and gas generation have been performed for the monatomic materials C, Al and Fe using a displacement energy of 25 eV for all three

Table 2
Spectrum averaged displacement and gas production cross sections for C, Al and Fe at a sample depth of 10 mm. The flux and the average neutron energy are the same as listed in table 1.

Material		C	Al	Fe
$\bar{\sigma}_d$	(kb)	0.751	2.312	3.464
$\bar{\sigma}_H$	(mb)	---	39.1	109.9
$\bar{\sigma}_D$	(mb)	---	9.90	10.6
$\bar{\sigma}_T$	(mb)	---	3.60	6.39
$\bar{\sigma}_{He}$	(mb)	52.0	39.9	29.3
$\bar{\sigma}_{He(gas)}$	(mb)	52.0	92.5	158.2
He/dpa	(10 ⁻⁶)	69.2	17.26	8.46

Fig. 4. Variation of total neutron and gamma-ray fluxes with depth in the test assembly.

materials. Displacements are related to the damage energy $\nu(T)$ by the modified Kinchin-Pease relation:

$$\begin{aligned}
 N_d &= 0 & 0 \leq T < E_d \\
 N_d &= 1 & E_d < T \leq 2E_d \\
 N_d &= \frac{0.8 \nu(T)}{2E_d} & 2E_d < T
 \end{aligned} \quad (1)$$

The spectrum averaged displacement cross sections, the gas production cross sections, and the ratio of helium production to displacements at a depth of 10 mm within the test assembly are listed in table 2 for C, Al and Fe. The He/dpa ratio for Fe decreases by 34% from the front to the back of the test assembly, while, for Al and C, the ratio decreases by factors of 2.0 and 3.8, respectively. Fe has a positive Q-value (nuclear reaction energy) for the (n, α) reaction and even low energy neutrons can induce this reaction. In contrast, both C and Al have negative Q-values for the (n, α) reaction, and, as the average neutron energy decreases, fewer neutrons are able to induce (n, α) reactions in these materials.

For polyatomic materials the relationship between damage energy and displacements is expected to be more complicated than is indicated by the Kinchin-Pease relation. An initial estimate of the recoil-atom damage in the binary insulators Al₂O₃ and Si₃N₄ has been obtained by replacing the Kinchin-Pease relation by the total displacement functions generated for these materials by Parkin and Coulter [6]. For a binary

Table 3

Spectrum averaged displacement and gas production cross sections for Si_3N_4 .

Sample depth (mm)	5.0	10.0	20.0	40.0	80.0	140.0	190.0
Flux $\{10^{20} \frac{\text{neutrons}}{\text{m}^2 \cdot \text{A} \cdot \text{s}}\}$	2.38	1.87	1.09	0.387	0.224	0.0722	0.0226
E_n (MeV)	10.69	10.18	8.60	7.85	5.83	3.93	3.68
$E_d(\text{Si})$ (kb)	0.365	0.366	0.345	0.329	0.290	0.229	0.246
$E_d(\text{N})$ (kb)	0.423	0.421	0.398	0.380	0.335	0.266	0.285
E_H (mb)	106.1	108.4	94.8	86.9	65.3	39.0	39.1
E_D (mb)	10.4	10.1	9.86	8.15	6.11	2.98	3.20
E_T (mb)	6.50	5.98	5.95	4.88	3.64	1.90	2.02
E_{He} (mb)	127.7	127.8	111.8	101.6	75.5	42.7	43.1
E_{gas} (mb)	250.7	252.3	222.4	201.5	150.8	86.6	87.4

material four displacement functions are required to describe the displacements of atoms of types A and B by primary knock-on atoms of types A and B. The total number of displaced atoms of each type is obtained from a sum, weighted according to the atomic fraction of each atom, of the displacements produced by each type of primary knock-on atom.

Gas generation in the binary materials was determined from a weighted sum of the gas generation from the atoms of each type. Tabulations of the dependence on depth within the test assembly of the spectrum averaged displacement cross sections for gas generation are given in tables 1 and 3.

4. DISCUSSION OF RESULTS

4.1. Neutron-Induced Damage

Evaluation of an insulator candidate material solely on the basis of gas generation depends upon nuclear cross sections and is straightforward. Using this criterion only, Al_2O_3 should be a better insulator candidate material than Si_3N_4 .

Evaluation of an insulator on the basis of displacements is more complicated and depends, among other things, upon the value used for the displacement energy. Comparison of the spectrum averaged displacement cross sections in tables 1 and 3 shows the displacement cross section for Al in Al_2O_3 to be more than twice as large as the spectrum averaged displacement cross section for either Si or N in Si_3N_4 . Much of this large cross section arises from the low value used for the displacement energy when generating the total displacement functions for Al in Al_2O_3 . The displacement energies used in generating the displacement functions for Al_2O_3 and Si_3N_4 were 18 and 72 eV, respectively, for Al and O in Al_2O_3 and 60 eV for both Si and N in Si_3N_4 .

The value of $E_d = 18$ eV for Al in Al_2O_3 was reported by Pellis and Phillips [7]. Compton and Arnold [8] have reported the displacement energy of Al in Al_2O_3 to be 40 eV. As the number of displacements is essentially inversely propor-

tional to E_d , the spectrum averaged displacement cross section for Al in table 1 may be as much as a factor of two too large. This illustrates the difficulty in evaluating materials on the basis of displacement cross sections.

4.2. Damage from Other Mechanisms

In addition to displacements induced by primary knock-on atoms from neutron-atom collisions, displacements arising from other mechanisms need to be included in a full evaluation of candidate materials. Some mechanisms that can produce displacements are: 1) recoils from (n,γ) radiative absorption, 2) recoils from interactions with gamma rays, and 3) stopping of particles from reactions such as (n,p) and (n,α) .

4.2.1. Radiative absorption

For all the elements considered herein, the (n,γ) reaction has a positive Q-value of several MeV, and hence even thermal neutrons can produce displacements through this mechanism. The nuclear recoil energies of C, Al, Si and Fe from (n,γ) reactions with thermal neutrons are 1003, 771, 473 and 389 eV, respectively [9]. The (n,γ) channel has not been included in the damage program DON. In general, the cross section for the (n,γ) reaction is in the mb range. As this is much less than the total neutron cross section, this mechanism is estimated to produce fewer than 10% of the total displacements.

4.2.2. Gamma-ray interactions

4.2.2.1. Electrons

Energetic electrons are produced by pair-production, photoelectric, and Compton interactions. The threshold for pair-production is 1.02 MeV, while the electron energy required to transfer the displacement energy to an atom is about 0.5 MeV. Since energy is shared equally between the electron and positron, the gamma-ray energy for producing displacements is approximately 2 MeV. As can be seen from fig. 3, the gamma-ray spectrum decreases rapidly with increasing energy. Hence few displacements should be produced by the pair-production mechanism. In general, for

gamma-ray energies less than 10 MeV, only photoelectric and Compton processes need be considered [10].

Displacement cross sections for photoelectric and Compton electrons have been calculated by Oen and Holmes [11] using a displacement energy of 25 eV. Displacement cross sections for Compton electrons dominate for gamma-ray energies less than 5 MeV. For elements with $Z < 50$, the Compton displacement cross section is at most three barns. Compared to the spectrum averaged displacement cross sections from recoil atoms, and considering the comparatively small flux of gamma rays, displacements produced by recoils from energetic electrons should be negligible.

4.2.2.2. Nuclear processes

Oen and Holmes estimated the displacement cross section for nuclear Compton and photoelectric processes. The Compton process was considered to be negligible, while the photoelectric process can become significant for gamma energies above 15 MeV. For the spectra generated by MORSE, neither process is important.

4.2.2.3. Low energy photons

Displacements can be produced in certain solids, such as the alkali halides, by photons in the x-ray region [12]. This mechanism is expected to vary from one material to another but is thought to be of little importance for metals and covalent crystals.

4.3. Particle Production

Interactions such as (n,p) and (n, α) produce charged particles that can produce displacements. In many cases the Q-value associated with charged particle production is negative, but, in the case of the (n,p) reaction on ^{14}N , it is positive. Charged particles produced in such reactions will, in general, lose much of their energy through electronic excitation. However, a particle of mass A must have an energy greater than A keV before it can transfer enough energy to produce electronic excitation in either metals or insulators. Below this energy all energy must be dissipated through other processes that may produce displacements. In addition, rearrangements following ionization processes in insulators is expected to produce displacements.

5. CONCLUSIONS

As an initial step in evaluating insulators for use in regions of a fusion reactor having high fluxes of high energy neutrons, damage calculations including recoil-atom damage and gas production by neutrons have been performed for several monatomic materials and for the two binary insulators Al_2O_3 and Si_3N_4 . In the test assembly considered the flux varies by two orders of magnitude and the average neutron energy changes by a factor of nearly three from the front to the back of the test assembly. The

He/dpa ratio for Fe (positive Q-value) decreases by 34% while the He/dpa ratio for C, Al, Al_2O_3 , and Si_3N_4 (negative Q-values) decreases by a factor between 2.0 and 3.8 from the front to the back of the test assembly. When the density of the material in the test assembly is decreased, the variation with depth of the flux and the He/dpa ratio decreases.

In addition, in a mixed environment of neutron and gamma-rays, displacements produced by gammas as well as displacements resulting from ionization processes need also be considered. As a first approximation it is judged that displacements resulting from recoils due to energetic electrons produced from gamma-rays are probably less than a 10% contribution. The problem of ascertaining the importance of electron excitation and ionization in producing displacements in insulators is complicated and is expected to be material dependent.

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