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Radiation Damage Calculation Methodology

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ABSTRACT

This report outlines the consensus radiation damage calculation methodology used by the Neutronics Analysis Group at Idaho National Laboratory. This methodology includes calculation of radiation damage rates via the Monte Carlo N-Particle (MCNP) code, and calculation of radiation damage in mixed materials, such as alloys. A consistent method for calculating the average displacement threshold energy of a mixed material has been established by the Neutronics Analysis Group. No standard method currently exists to account for radiation damage caused by neutron-induced transmutations. Only one documented case addresses transmutation radiation damage in materials containing nickel, and that method was incorporated by the Neutronics Analysis Group into their radiation damage calculation methodology.

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Radiation Damage Calculation Methodology

1. Radiation Damage [DPA]

The damage rate in a material is calculated by applying a tally multiplier to a standard flux tally in the Monte Carlo N-Particle (MCNP) code. Applying the tally multiplier card to a flux tally enables calculation of a user-specified reaction rate, defined as:

$$R_x(t) = C \int_0^\infty \sigma_x(E)\Phi(E)dE \quad (1)$$

where:

C = user-specified multiplicative constant,

σ_x (E) = the cross-section for the reaction of interest,

Φ(E) = energy-dependent flux in neutrons/cm^2.

In MCNP, the tally multiplier (FMn) takes the form of "FMn ([C] [mat] [mt])" and performs the integral in Equation 1. For calculating the radiation damage rate, the damage-energy cross section [MeV-b] is needed. This is stored by MCNP and NJOY in MT = 444 (MF = 3).

Next, based on the model created by Norgett, Robinson, and Torrens (i.e., the NRT model) [7], radiation damage can be calculated via Equation 2, assuming constant irradiation conditions:

$$D_x = \frac{\eta}{2E_d} \int_0^T R_d(t)dt \quad (2)$$

where:

η = efficiency correction factor of 0.8,

E_d = average displacement threshold energy,

T = total irradiation time.

It must be noted that η = 0.8 is a historical value (i.e., not experimentally determined). It was arbitrarily set and can be treated as having infinite precision.

MCNP reports tally results normalized per source particle. The FMn tallies are given in units of MeV-barns/cm^2 per source neutron. The normalization factor used to calculate radiation damage is given by Equation 3:

$$F_D = \frac{\bar{\nu}}{Q_f} \quad (3)$$

where:

$\bar{\nu}$ = the average neutrons produced per fission

Q_f = The energy released per fission

For example, if we assume $\bar{\nu}$ = 5.86 and Q_f = 500 MeV, the normalization factor can be determined as follows:

$$F_D = \left(\frac{5.86 \text{ neutrons}}{\text{fission}}\right)\left(\frac{\text{fission}}{500 \text{ MeV}}\right)\left(\frac{\text{MeV}}{1.60218\times10^{-19}\text{MJ}}\right)\left(\frac{1\text{MJ}}{1\text{MW·s}}\right)\left(\frac{3600\text{s}}{\text{hour}}\right)\left(\frac{24 \text{ hrs}}{\text{day}}\right)\left(\frac{1\times10^{-24}\text{cm}^2}{\text{b}}\right) \quad (4)$$

Remember that these values are random variables and *must* be based on experimental data as well as cited. Therefore, the radiation damage in a material is calculated using the flux multiplier results, the normalization factor, the Advanced Test Reactor core power, and the total irradiation time, as per Equation 5:

$$D_x = \frac{F_d \eta}{2E_d} \int_0^T R_d(t) P(t) dt \quad (5)$$

If we assume the reactor power can be approximated as a piecewise function of constant-power irradiation steps, ***and*** that transmutation does not affect the radiation damage, we can simplify Equation 5 to a discrete problem:

$$D_x = \frac{F_d R_d \eta}{2E_d} \sum_{i=1}^{n} P_i t_i \quad (6)$$

1.1 DPA for Mixed Materials

To calculate the total radiation damage in a mixed material (e.g., an alloy), the radiation damage must be calculated for each component (usually each element) comprising the mixed material. This avoids information loss by having MCNP calculate the total directly. The total radiation damage for the mixed material can then be approximated by the sum of the normalized radiation damage for each component times the **atom percent** for that component in the mixed material. For example, given the material FeCrAl, the following separate material definitions would be needed for each material component in the MCNP input file:

```
c   FeCrAl material separated by element for DPA calculations
m101    26054.70c    5.845
        26056.70c    91.754
        26057.70c    2.119
        26058.70c    0.282
m102    24050.70c    4.345
        24052.70c    83.789
        24053.70c    9.501
        24054.70c    2.365
m103    13027.70c    100
```

In the MCNP input, be sure to double check the isotopic abundances. The FMn tally would then take the following form:

```
fc4   cell 9200
f4:n  9200
fq4   s e
fm4 (1.0 101 444) (1.0 102 444) (1.0 103 444)
```

Equation 3 is then used to normalize the FMn tallies for each element of FeCrAl. The total radiation damage is calculated using Equation 7:

$$D_{T,x} = \sum_{j=1}^{J} D_{x,j} n_j \quad (7)$$

where n is the material atom fraction, D_T is the total damage in the material, index x is used for each cell, and index j is used for each element or material component.

Using the example values in Table 1, the total radiation damage would be calculated as:

$$\mathrm{D}_T = (1 * 0.75) + (5 * 0.13) + (3 * 0.12)$$

$$\mathrm{D}_T = 1.76 \quad (8)$$

Table 1. Example values for calculating total radiation damage in a mixed material.

Element	Normalized FMn tally	Atom Percent
Fe (m101)	1	75
Cr (m102)	5	13
Al (m103)	3	12

1.2 Average E_d Value

One issue with DPA calculations relates to the average displacement threshold energy, E_d, values. There is no standard set of E_d values used to calculate DPA, since various literary sources give different values for the same element. For example, updated values from Konobeyev et al. use 37 eV for Si [2], Greenwood and Smither use 25 eV [1], and Heinisch et al. use 35 eV [3]. Such variation in E_d values makes radiation damage a challenging parameter to compare. Radiation damage models are only valid for pure elements. Consequently, the problem with using an E_d-weighted average for mixed materials is that it does not consider either the secondary displaced atoms that are dependent on all possible combinations of recoiling and crystal matrix atoms or the differences in binding energies between compounds and pure elements [4]. In addition, when using a weighted average, it must be assumed that the components in the mixed material are all approximately equal in terms of atomic weight [1]. This assumption works well for bland alloys (e.g. stainless steel), in which most of the alloying components have very similar atomic masses. But this assumption breaks down for alloys with very different atomic masses involved (e.g., FeCrAl). In addition, these models assume the new material to be of a crystalline nature very similar to the crystals of the components — an assumption that breaks down when new chemicals are made, such as silicon carbide ceramic. It also breaks down for high-entropy alloys intended to create an entirely novel crystal. Finding the average E_d value for mixed materials with consideration of secondary displaced atoms is a project in and of itself, and can only be completed through molecular dynamics simulations and extensive computational analyses. Efforts to create more accurate methods have not been developed in a standardized way that enables the performance of such analyses.

Therefore, to create consistency, a methodology for calculating the average E_d value of mixed materials was established by the Neutronics Analysis Group at Idaho National Laboratory. Thus, it is suggested that analysts first reach out to the principal investigator (PI) of that research in order to see if they have an average E_d value they wish to provide. This value will likely be that used in other datasets against which they wish to compare. The PI may want to conduct a literature review, as certain studies have calculated E_d for some of the more novel materials. Otherwise, an average E_d value for mixed materials (e.g., alloys, composites, and compounds) should be used, weighted by the **atomic percent** of the elements in the material. For example, using the example values in

Table 2, the average E_d value would be calculated as:

$$\bar{E}_d = (40 * 0.75) + (40 * 0.13) + (27 * 0.12)$$

$$\bar{E}_d = 3.8 \times 10^{-5}\ MeV \quad (9)$$

Table 2. Example values for calculating the average displacement threshold energy.

Element	Atom Percent	* E_d (eV)
Fe	75	40
Cr	13	40
Al	12	27
*The journal articles for the E_d values used should be cited.		

1.3 Transmutation Radiation Damage for Materials Containing Nickel

Currently, no method accounts for damage caused by neutron-irradiation-induced transmutations (i.e., when the reaction rate in Equation 2 is time dependent). The only documented case in which the reaction rate was found to be time dependent involves nickel [5][6]. Under irradiation, nickel produces the synthetic isotope Ni-59 via the ^{58}Ni(n,γ) reaction. Ni-59 can then undergo an (n,α) or (n,p) reaction, both of which are extremely exothermic. These reactions produce radiation damage currently not being accounted for in neutronics analyses. Moving forward, to account for the radiation damage caused by Ni-59 reactions, the following method established by Greenwood should be used:

1. Perform the radiation damage calculation as normal and assume a constant radiation damage rate for the entire irradiation.
2. Next, perform an activation of only the nickel, and calculate the helium and hydrogen gasses produced. ORIGEN can perform the appm calculations, and these are normalized to the starting quantity; therefore, either 1 or 100 grams can be used as the starting mass.
3. Correct the radiation damage by using Greenwood's correction factors. Initially, these were calculated as 1 DPA for every 567 appm He [5] and every 90 appm H [6] produced in nickel. However, note that, when using ORIGEN, only the helium concentrations should be depended upon, since hydrogen is also produced from sources other than Ni-59. For thermal reactors, Greenwood published an updated helium correction factor that includes (n,α), (n,p), and (n,γ) reactions based on an assumed thermal flux spectrum. This correction factor is 1 DPA for every 548 appm He [6].

Note that a team at Idaho National Laboratory is currently researching this problem, and better methods may eventually be deployed once ORIGEN implements this research.

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