http://dx.doi.org/10.5516/NET.01.2013.051

A SIMPLE METHOD TO CALCULATE THE DISPLACEMENT DAMAGE CROSS SECTION OF SILICON CARBIDE

JONGHWA CHANG*, JIN-YOUNG CHO, CHOONG-SUP GIL, and WON-JAE LEE
Korea Atomic Energy Research Institute
*Corresponding author. E-mail : jhchang@kaeri.re.kr

Received June 25, 2013
Accepted for Publication February 19, 2014

We developed a simple method to prepare the displacement damage cross section of SiC using NJOY and SRIM/TRIM. The number of displacements per atom (DPA) dependent on primary knock-on atom (PKA) energy was computed using SRIM/TRIM and it is directly used by NJOY/HEATR to compute the neutron energy dependent DPA cross sections which are required to estimate the accumulated DPA of nuclear material. SiC DPA cross section is published as a table in DeCART 47 energy group structure. Proposed methodology can be easily extended to other materials.

KEYWORDS : Displacement Per Atom (DPA), Cross Section, Silicon Carbide, Compound Material

1. INTRODUCTION

Silicon carbide (SiC) is an important material in advanced nuclear systems such as high temperature gas reactor (HTGR), and advanced nuclear fuel and fusion reactors. In a TRISO fuel particle, β-SiC is used as a coating barrier over a fuel kernel to protect fission product release. Irradiated material property data are important in estimating the safety life time of components in a nuclear reactor and a nuclear fuel. Traditionally, accumulated fast neutron fluence was used as a measure of irradiation. However, it becomes more important to use accumulated DPA as a measure, since an electron or an ion beam is often used in irradiation experiments to reproduce real environmental conditions of the material, to shorten experiment time, and to reduce the experiment cost. Moreover, it is not reasonable to use a fluence measure when the neutron spectrum is significantly different from that of the experiment since the cascade of damage is dependent on incident neutron energy and target material. It is necessary to use the DPA measure when designing a new type of reactor such as HTGR, SFR, and fusion reactor, and when introducing a new material such as SiC.

NJOY[1] is a neutron cross section processing code widely used in generating a multi-group library for neutron transport calculation. NJOY has a module HEATR which can compute the damage energy using Norgett-Robinson-Torrens (NRT) model[2] by tracking the recoil energy in all possible reaction channels such as scattering, capture, inelastic, etc. However, its capability is limited to handling single isotope material which is the generic limitation of NRT model.

For compound materials, a simple method can be applied by taking a weighted average of isotope-wise DPA cross sections. However, this approach is not accurate since the primary recoiled atom can collide with other kinds of atoms having different charge number and mass.

To handle this situation accurately, Parkin and Coulter [3] proposed a set of differential equations on the displacement functions. To solve the set of equations, differential cross sections and displacement trapping probability must be known a priori. A computer code, SPECOMP, was developed by Greenwood [4] using the Parkin and Coulter theory. However, SPECOMP has a limitation on the reaction type, such as (n,t) or (n,α), and the cross section library. Recently Heinisch et al.[5] calculated the DPA cross section of SiC using SPECOMP and SRIM-2000[6,7]. They take weighted average of the four possible cases of the primary knock-on atom (PKA) and the secondary knock-on atom. The results were published in a table of numerical values in 100 group structure.

Parkin and Coulter's method incorporates a complicated process in order to obtain the number of damages for a given PKA energy. For each combination of recoiled atom, target atom, and displaced atom, a set of integro-differential equations on the neutron energy dependent net displacement function is solved with a given differential cross section and electron stopping power. The electron stopping power should be prepared in advance using a code such as TRIM. [7] The equation requires a recoil energy dependent displacement probability and a trapping probability. These are simply approximated by identical particle collision case, which is not true for compound material. Then, the spectral averaged damage is obtained.

TRIM can compute the damage per PKA for a compound material by calculating the electron stopping power using more rigorous treatment on displacement and trapping using isotope-wise threshold displacement energy per each collision through the Monte-Carlo simulation. We can get more accurate results by incorporating the TRIM results in the process of NJOY calculation. The authors propose a simple hybrid method using TRIM and NJOY which can take care of all reactions combined with an up to date cross section library. In this study, the NJOY/HEATR module has been modified to incorporate the TRIM results. For comparison with Heinisch model, the generated DPA cross section for 47 energy group is applied to a neutron transport code, DeCART.[14]

2. LATTICE PARAMETERS

There are more than 250 polytypes of silicon carbide. Among them 3C-SiC, so-called β-SiC, which is formed under 1700oC, is a candidate material for nuclear applications. SiC is a ceramics material and can be modeled as a zinc-blend two-component covalent material.

The collision cascade of recoiled nuclides is characterized by threshold displacement energy, lattice binding energy and surface binding energy. Among them, threshold displacement energy that causes Frenkel pairs is the most important parameter in estimating the displacement damage. This parameter is dependent on the lattice structure and can be obtained by an irradiation experiment, or predicted by molecular dynamics simulations. Many experimental results were published on SiC, however, reported values vary widely, 20-50 eV for C defect and 35-110 eV for Si defect . Zinkle and Kinoshita [8] recently recommended 20 eV for C and 40 eV for Si.

There are many studies to predict threshold displacement energy using molecular dynamics simulations. Devanathan et al.[9] obtained 20 eV for C displacement and 35 eV for Si displacement. Heinish et al. adopted these values. [5] Recently, Lucas and Pizzagalli [10] pointed out that the different empirical potentials used in the previous studies may result in poor prediction of Frenkel pairs production for covalent materials. They made a more fundamental approach to produce the inter-atomic potential using electron the density functional theory (DFT) [11]. They obtained the lattice averaged threshold displacement energy as 19 eV for C sublattice and 38 eV for Si sublattice. Those values are largely different from that of single element material, 31 eV for graphite and 25 eV for silicon.[1] The difference comes from the difference in lattice structure. The result of Lucas and Pizzagalli was adopted in this study, 19eV and 38eV as the threshold displacement energy for C and Si, respectively.

Lattice binding energy is required for SRIM/TRIM simulation. This value, the vacancy formation energy, can be predicted using another molecular dynamics calculation. This study adopted Huang and Ghoneim's result [12]; 2.63eV and 3.25eV for C and Si, respectively.

Surface binding energy, which is less important in the DPA estimation, can be derived from heat of vaporization for each element. This study adopted 7.4 eV and 4.7eV for C and Si, respectively.

3. NRT MODEL IN NJOY

The NRT model in the NJOY/HEATR module deviates from the original NRT model in the low energy cutoff and also deviates from the modified NRT model which multiplies the efficiency factor (0.8) to the number of vacancies. This factor can be taken into account when deriving the DPA cross section from the NJOY produced damage production cross section ($E\sigma$) (MT=444). NJOY sums up the cross section multiplied by damage energies, which is the damage production cross section representing the effective kinetic energy of recoiled atom for possible reaction types (k) at a given incident neutron energy (E_n).

$$\left(E\sigma\right)_{DPA} = \sum_k E_{d,k}\sigma_k\left(E_n\right)$$

For a specific reaction, the damage function is calculated using the following formulae.

$$\mathrm{df} = 0 \quad \text{when} \quad E_{PKA} < E_d\,,$$

$$\mathrm{df} = \frac{E_{PKA}}{1 + F_L g\left(\varepsilon\right)} \quad \text{when} \quad E_{PKA} \geq E_d\,,$$

where, E_{PKA} is the kinetic energy of the PKA, E_d is the threshold displacement energy,

$$F_L = \frac{0.0793 Z_R^{2/3} Z_L^{1/2} \left(A_R + A_L\right)^{3/2}}{\left(Z_R^{2/3} + Z_L^{2/3}\right)^{3/4} A_R^{3/2} A_L^{1/2}}\,,$$

$$g\left(\varepsilon\right) = 3.4008\varepsilon^{1/6} + 0.40244\varepsilon^{3/4} + \varepsilon\,,$$

$$\text{and} \quad \varepsilon = \frac{E_{PKA}}{30.724 Z_R Z_L \left(Z_R^{2/3} + Z_L^{2/3}\right)^{1/2}} \frac{A_L}{A_R + A_L}\,.$$

Z_L and A_L are the charge number and atomic mass of lattice atom, Z_R and A_R are those of recoiled atom.

The number of displacement per atom, N_d, is calculated as

$$N_d = \frac{1}{2E_d}\mathrm{df}\,.$$

The NRT model can effectively handle the electron energy loss of the recoil atom for single element material. A simple method may be devised for SiC by considering four cases of collision cascade as proposed by Heinish et al.[5] These are C-C (PKA C collides with C), C-Si (PKA C collides with Si), Si-Si (PKA Si collides with Si), and Si-C (PKA Si collides with C). The effective displacement energy of chains can be derived from the maximum energy

sharing between recoiled atoms. Energy sharing is dependent on the collision angle and the maximum energy sharing is given by $4M_1M_2/(M_1+M_2)^2$ (=0.84 between 12C and 28Si). The effective displacement energies were obtained: 19 eV(C-C), 45.2 eV(C-Si), 38 eV(Si-Si), and 22.6 eV (Si-C). Then, the average DPA cross section is obtained by averaging the 4 cases.

$$\sigma_{dpa} = \frac{1}{2}\left[0.25\frac{E\sigma_{CC}}{2E_{d,CC}} + 0.25\frac{E\sigma_{CSi}}{2E_{d,CSi}} + 0.25\frac{E\sigma_{SiSi}}{2E_{d,SiSi}} + 0.25\frac{E\sigma_{SiC}}{2E_{d,SiC}} \right]$$

The damage production cross sections, $E\sigma$ are obtained from the NJOY calculation with the effective displacement energies. This method is similar to Heinish et al.'s method but it lacks consideration on the succeeding collision cascade accurately.

4. PROPOSED METHOD

SRIM/TRIM is known as the most reliable and accurate tool in modeling the ion collision cascade problems. It considers electron energy loss and nuclear collision energy loss as well as atomic collision for ion beam irradiation. SRIM/TRIM has been validated by wide ranges of users for the last 30 years since the code was released to the public. The number of vacancies can be computed by a Monte Carlo simulation for a given lattice parameters of target materials such as threshold displacement energies.

The NJOY/HEATR module computes PKA energy spectra accurately for every reaction using up to date nuclear data library. NJOY is widely accepted in the reactor physics community to provide the multi-group cross sections for transport calculation codes. Even though NJOY can handle only one nuclide, the damage production cross section can be computed for compound material as long as we provide the number of vacancies of the compound material per PKA energy. For the case of SiC, we can provide the number of vacancies for ^{12}C as PKA (or ion in TRIM), then make NJOY run for ^{12}C to produce the number of vacancies depending on incident neutron energy. Similar work for ^{28}Si as PKA calculates the contribution of Si PKA on the neutron energy dependent DPA. By adding these two contributions, we can find the neutron energy dependent damage cross section of SiC.

Series of TRIM simulations on SiC, varying ion energies, generate the number of vacancies per ion of ^{12}C and ^{28}Si for the elastic scattering. Similar TRIM simulations for ^{13}C and ^{29}Si ion can be made to prepare the damage factor table for capture reaction. For less important reactions and energy ranges, we can rely on the NJOY's NRT model. The TRIM calculation accurately simulates the collision cascade in the SiC lattice with given lattice parameters, the threshold displacement energy, the lattice binding energy, and the surface binding energy for carbon and silicon atoms in the solid.

To make use of these values, the damage factor is calculated by df = $2E_dN_d$. The E_d value used in previous equation is somewhat arbitrary since NJOY computes the damage production cross section which needs to be divided by $2E_d$ to obtain the DPA cross section. In an attempt to minimize time-consuming TRIM simulations, the NRT model is relied on when the damage factor table is not provided. E_d is determined so that the NRT model will give the same number of vacancies as the TRIM simulation at elastic scattering up to highest PKA energy of interest. For C PKA, we obtain 31.07 eV at 446 keV PKA energy. For Si PKA, we obtain 33.23 eV at 100 keV PKA energy. These effective displacement energies will give the same number of vacancies in both TRIM and NRT calculations for a given PKA energy. The TRIM simulation is performed down to cutoff energy, which is the minimum value among the threshold displacement energy of a nuclide in the compound material. The TRIM simulation takes into account important reaction types, such as elastic and capture, up to important PKA energy.

We devise an interpolation scheme to compute the damage function from the prepared table based on the energy dependency of the NRT model;

$$\mathrm{df} = \frac{E_{PKA}}{a + bE_{PKA}},$$

where the parameters a and b are determined by the table grid values for which the TRIM calculation is performed.

Two NJOY/HEATR runs are made for ^{12}C and ^{28}Si, each run will produce the damage production cross section, $E\sigma_C$ and $E\sigma_{Si}$. The DPA cross section is calculated by summing up the contributions of nuclides in the compound, then dividing by the total number of atoms in a molecule :

$$\sigma_{dpa} = \frac{1}{2}\left[0.5\frac{E\sigma_C}{2E_{d,C}^{eff}} + 0.5\frac{E\sigma_{Si}}{2E_{d,Si}^{eff}} \right].$$

5. RESULT

Cross sections for NJOY were taken from ENDF/B-VII.1 [13]. Carbon and silicon have small fraction of isotopes. Abundance of ^{12}C is 98.9% and ENDF/B library cross section is for natural carbon. We regarded C in SiC as 100% ^{12}C in this study. ^{28}Si is a dominant contributor to damage due to a larger scattering cross section than other isotopes. Moreover, big resonances at 55.7, 182.6, 565.7, 812.5, 964.1 keV should make a significant contribution to the damage cross section. However the 47 group structure smears those resonances. We have treated Si in SiC as ^{28}Si although its abundance is 92.23%. The resulting DPA cross section may be slightly overestimated due to

a larger cross section of ^{28}Si, however, it will not be a problem for practical usage.

The calculation was done at 300K. Since ^{12}C and ^{28}Si has no significantly narrow resonances, there should be no problem in using the results up to reasonably high temperatures.

Figures 1 and 2 display the number of vacancies (or displacement) per atom produced by ^{12}C and ^{28}Si PKA, respectively. The results are for the elastic scattering reaction. In the NRT model, the number of displacements below the threshold displacement energy is zero. At the threshold energy, the value is taken to be 0.5 as the NRT model used in NJOY. In low PKA energy region, where the electron energy loss is negligible, the number of displacements are proportional to the PKA energy divided by 2 times of the threshold displacement energy. It is shown that at high PKA energy region, roughly above 10 keV, the electron energy loss becomes significant. The TRIM result at low energy region for compound material shows nonlinear behavior lying between both NRT predictions. This is as expected since NRT results represent extreme cases where succeeding cascade occurs with the same nuclides.

Figure 2 shows the number of displacements per atom for ^{28}Si PKA. In the Figure, the TRIM result is out of the NRT model range. It is originated from numerical truncation error since TRIM shows the result with a single digit. In the middle PKA energy range, the number of displacements per atom predicted by TRIM approaches to that of Si-C cascade and it approaches that of Si-Si cascade at high energy.

Figure 3 displays the DPA cross section generated by the proposed method up to 150 MeV which is the new ENDF/B-VII energy range. ^{12}C is the main contributor to damage for neutron energy below 1 MeV. In higher energy range, ^{28}Si becomes the main contributor to damage. Strong resonance at 55.7 keV and 182.6 keV is noticeable.

Figure 4 and 5 compare the DPA cross sections given by Heinish et al.[5] and its condensation to the DeCART 47 group structure[14] up to 20 MeV. Results by the NJOY/NRT model and the NJOY/TRIM model are also compared. It is shown that resonance structure was smeared in the 47 group structure. It is more practical to generate the DPA cross section in a wide group structure considering neutron transport calculations, even if the proposed method can produce a finer group structure of DPA cross section. The simplest 4 chains averaging method using the NJOY results is a much higher value at MeV regions. This is expected

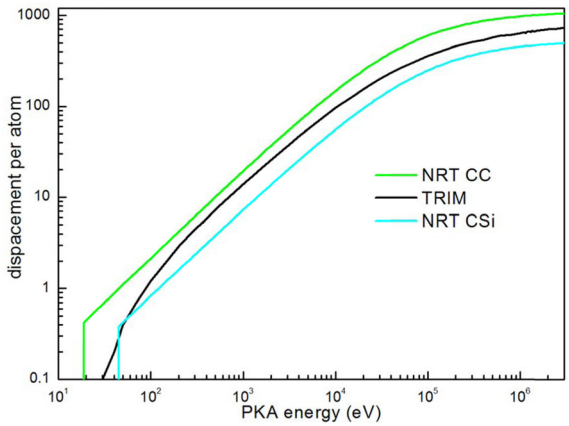

Fig. 1. Displacement Per Atom in SiC by Carbon PKA.

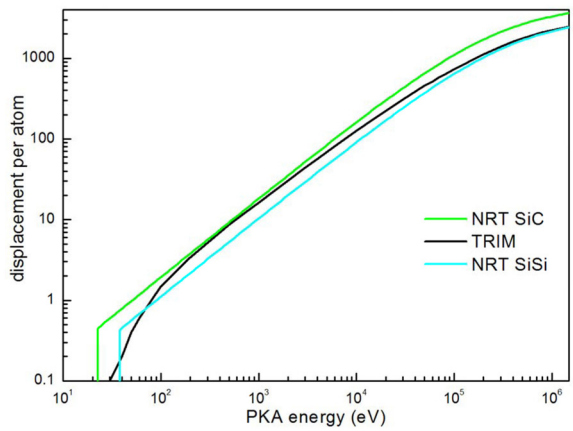

Fig. 2. Displacement Per Atom in SiC by Silicon PKA.

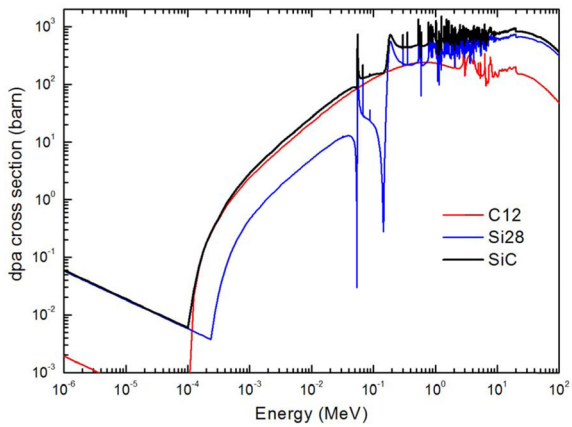

Fig. 3. DPA Cross Section of SiC and Element-wise Contributions.

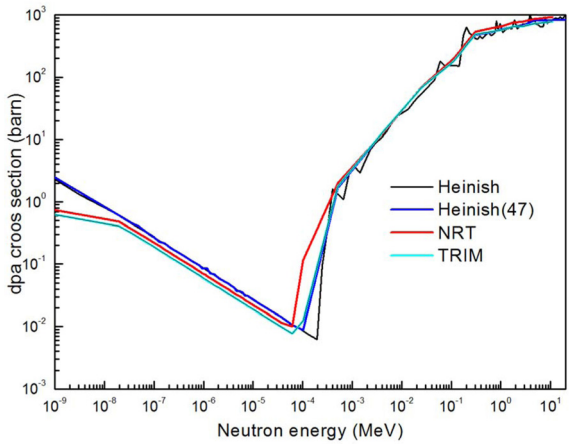

Fig. 4. DPA Cross Section of SiC in Logarithmic Scales.

Table 1. DPA Cross Section of SiC

lower energy (eV)	Heinish (barns)	Present (barns)	lower energy(eV)	Heinish (barns)	Present (barns)
1.00E-05	4.036E+00	7.258E-01	3.93E+00	4.188E-02	2.923E-02
1.24E-02	6.125E-01	4.087E-01	4.45E+00	3.700E-02	2.747E-02
3.06E-02	4.580E-01	3.122E-01	5.04E+00	3.700E-02	2.584E-02
4.28E-02	4.064E-01	2.679E-01	5.72E+00	3.570E-02	2.424E-02
5.69E-02	3.307E-01	2.278E-01	6.48E+00	3.110E-02	2.278E-02
8.20E-02	2.777E-01	1.928E-01	7.34E+00	3.110E-02	2.138E-02
1.12E-01	2.300E-01	1.670E-01	8.32E+00	2.716E-02	1.891E-02
1.46E-01	2.155E-01	1.475E-01	1.21E+01	2.370E-02	1.666E-02
1.84E-01	1.810E-01	1.265E-01	1.37E+01	1.936E-02	1.346E-02
2.71E-01	1.540E-01	1.070E-01	2.90E+01	1.401E-02	9.810E-03
3.58E-01	1.318E-01	9.181E-02	4.79E+01	1.059E-02	7.650E-03
5.03E-01	1.134E-01	7.999E-02	7.89E+01	8.700E-03	1.243E-02
6.25E-01	1.040E-01	7.154E-02	1.30E+02	1.666E+00	1.771E+00
7.82E-01	9.359E-02	6.509E-02	2.03E+03	1.365E+01	1.355E+01
9.10E-01	8.430E-02	6.166E-02	9.12E+03	6.836E+01	6.785E+01
9.71E-01	8.430E-02	6.002E-02	6.74E+04	1.949E+02	1.811E+02
1.01E+00	8.430E-02	5.869E-02	1.83E+05	4.765E+02	4.858E+02
1.07E+00	8.430E-02	5.717E-02	4.98E+05	5.263E+02	5.363E+02
1.13E+00	8.430E-02	5.591E-02	8.21E+05	5.922E+02	5.791E+02
1.17E+00	8.430E-02	5.452E-02	1.35E+06	6.504E+02	6.405E+02
1.24E+00	7.199E-02	5.157E-02	2.23E+06	6.870E+02	6.617E+02
1.46E+00	6.860E-02	4.657E-02	3.68E+06	8.092E+02	7.243E+02
1.86E+00	5.774E-02	4.124E-02	6.07E+06	8.251E+02	7.784E+02
2.38E+00	4.949E-02	3.426E-02	2.00E+07	-	-

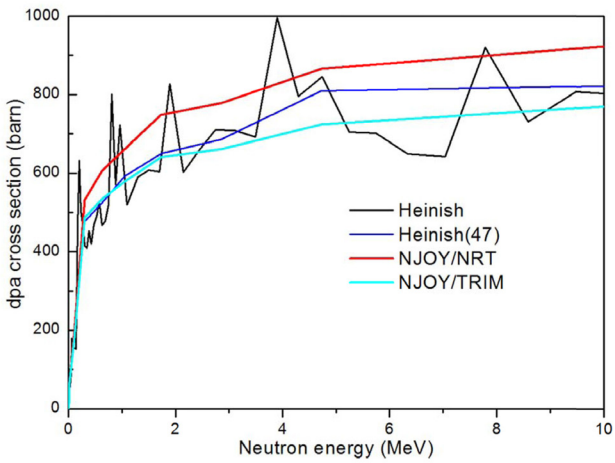

Fig. 5. DPA Cross Section of SiC in Linear Scales.

since C-C chain gives many more vacancies than the TRIM calculation on SiC and C-C chain is the most frequent collision event due to less atomic mass. Comparison of the results by Heinish and the proposed NJOY/TRIM models shows that there are significant differences between two models at high neutron energy above 2 MeV where the electronic energy loss and secondary chain effect become more significant. This may come from the fact that we prepared the TRIM table up to 446 keV for C elastic and 100 keV for Si elastic, which corresponds to roughly 2 MeV neutron energy. This difference may be narrowed by preparing TRIM table to higher ion energy. However, below 2 MeV which is an important energy region for the fission reactor problem, the difference is not significant. Numerical data of SiC for the DeCART 47 group structure is given in Table 1.

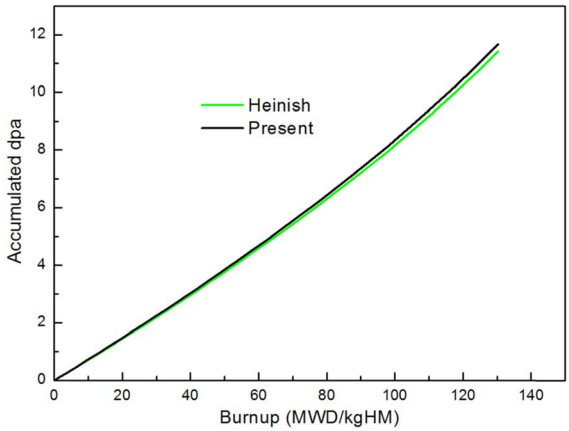

Fig. 6. Comparison of Accumulated DPA on a FCM Fuel.

The accumulated DPA is computed by

$$\text{dpa} = \int_0^T dt \int_0^\infty d\xi \sigma_{dpa}(\xi)\phi(\xi,t)$$

where $\phi(\xi,t)$ is the neutron flux with lethargy ξ at time t. The DPA cross section, σ_{dpa} takes a unit of barn per atom and the unit of flux is per cm^2•s so that the accumulated DPA is in the unit of per atom. Figure 6 compares accumulated DPA for a typical fully ceramic micro-encapsulated (FCM) fuel assembly [15] using the DeCART 47 energy group transport calculation. Present DPA cross section results in about a 2.2% higher value than that by Heinish's DPA cross section.

6. CONCLUSION AND RECOMMENDATIONS

Recently, irradiation properties of new materials are mostly measured using an electron accelerator or an ion accelerator where the irradiation can be performed at an accurately controlled condition in a shorter time than a neutron irradiation in a test reactor. So many research results are published in DPA units as a measure of irradiation. However, DPA cannot be measured accurately due to the recombination nature of vacancies. The best way to use DPA measure is to predict the DPA accumulation using a rigorous and accurate DPA prediction model.

We have devised a simple yet accurate method to produce a DPA cross section using widely used computer codes SRIM and NJOY. NJOY/HEATR is modified to handle the damage function table. The DPA cross section of SiC is generated following the proposed method and its numerical values in 47 energy group are provided for DeCART neutron transport calculation. The proposed method can be extended to other group structure and the numerical data for them will be available.

REFERENCES

[1] R.E. MacFarlane and A.C. Kahler, Methods for Processing ENDF/B-VII with NJOY, Nuclear Data Sheets 111, 2739-2890 (2010).
[2] M.J. Norgett, M.T. Robinson, and I.M. Torrens, A Proposed Method of Calculating Displacement Dos Rates, Nucl. Eng. and Design 33, 50-54 (1975).
[3] D.M. Parkin and C. Alton Coulter, Displacement Cascades in Polyatomic Materials, J. of Nucl. Mat. 117, 340-344 (1983).
[4] L.R.Greenwood, SPECOMP Calculations of Radiation Damage in Compounds, Sixth ASTM-EURATOM Symposium on Reactor Dosimetry, Jackson Hole, Wy. USA, June 1-5, 1987. also L.R. Greenwood, ASTM STP 1001, 598 (1989).
[5] H.L. Heinish et al, Displacement damage in silicon carbide irradiated in fission reactors, J. of Nucl. Mat. 327, 175-181 (2004).
[6] J.F. Zeigler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solids, Pergamon, New York, 1985.
[7] J.F. Zeigler, SRIM-2000, Code and manuals available on http://www.srim.org/ (2001, 2013).
[8] S.J. Zinkle and C. Kinoshita, Defect production in ceramics, J. of Nucl. Mat. 251, 200-217 (1997).
[9] R. Devanathan, W.J. Weber, and F. Gao, Atomic scale simulation of defect production in irradiated 3C-SiC, J. Appl. Phys. 90(5) 2303-2309 (2001).
[10] G. Lucas and L. Pizzagalli, Ab initio molecular dynamics calculations of threshold displacement energies in silicon carbide, Phys. Rev. B 72, 161202 (2005).
[11] W. Kohn and L.J. Sham, Self-Consistent Equations Including Exchange and Correlation Effects, Phys. Rev. 140, A1133-1138 (1965).
[12] H. Huang and N. Ghoniem, Molecular dynamics calculations of defect energetics in b-SiC, J. Nucl. Mat. 212-215, 148-153 (1994).
[13] M.B. Chadwick et al., ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields, and Decay Data, Nucl. Data Sheets 112(12) 2887-2996 (2011).
[14] J.Y. Cho et al., DeCART2D v1.0 User's Manual, KAERI/TR-5116/2013, Korea Atomic Energy Research Institute (2013).
[15] W.J. Lee et al., Fully Ceramic Micro-encapsulated (FCM) Replacement Fuel Assembly for LWRs, ICAPP 2013, Jeju Island, Korea, April 14-18, 2013 (2013).