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Primary displacement damage calculation induced by neutron and ion using binary collision approximation techniques (MARLOWE code)

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1. Introduction

The level of damage expected in future fusion reactors conditions is such that the performance of materials and components under these extreme irradiation conditions is still unknown. Considering this scenario, the study of the effects of energetic neutrons generated in fusion reactors on materials is one of the most important research topics to be carried out during next years.

The effects of neutron irradiation on materials involve, from a fundamental point of view, two physical phenomena: i) the displacement of atoms from their equilibrium positions in the lattice, which creates point defects, and ii) the generation of nuclear transmutation reactions that contribute to the formation of impurities inside the material, with He and H as the most important ones. The ratio between the levels of He and H, and the amount of point defects is one of the main parameters to understand the effect of the radiation on materials

In order to emulate the neutron irradiation that would prevail under fusion conditions, two approaches are contemplated:

a) on one hand different kinds of current neutron sources to emulate the fusion irradiation environment are available, as for example
   - Fission power reactor
   - Spallation sources
   - Striping Sources: The objective of t*he International Fusion Materials Irradiation Facility (IFMIF*) [1] will be to provide an intense neutron source with adequate energy spectrum to test the suitability of candidate materials for future nuclear fusion power reactor (DEMO). IFMIF will constitute an essential tool in the international strategy towards the achievement of future fusion reactors.

b) on the other hand, as these neutron sources have a number of problems and very strict operating conditions, (e.g. the radiological risks), to emulate the effects of fusion neutron on materials, some other facilities can be used. One example is the Spanish initiative TechnoFusión facility [2] which purpose is to serve as technological support for IFMIF and DEMO. The Material Irradiation Experimental Area of TechnoFusion will emulate the extreme irradiation fusion conditions in materials by means of three ion accelerators: one used for self-implanting heavy ions (Fe, Si, C,…) to emulate the displacement damage induced by fusion neutrons and the other two for light ions (H and He) to emulate the transmutation induced by fusion neutrons. This Laboratory will play an essential role in the selection of functional materials for DEMO reactor since it will allow reproducing the effects of neutron radiation on fusion materials. Ion irradiation produces little or no residual radioactivity, allowing handling of samples without the need for special precautions.

Currently, two different methods are used to calculate the primary displacement damage by neutron irradiation or by ion irradiation. On one hand, the displacement damage doses induced by neutrons are calculated considering the NRT model [3] based on the electronic screening theory of Linhard. This methodology is commonly used since 1975. On the other hand, for experimental research community the SRIM code is commonly used to calculate the primary displacement damage dose induced by ion irradiation. Therefore, both methodologies of primary displacement damage calculation have nothing in common. However, if we want to design ion irradiation experiments capable to emulate the neutron fusion effect in materials, it is necessary to develop comparable methodologies of damage calculation for both kinds of radiation. It would allow us to define better the ion irradiation parameters (Ion, current, Ion energy, dose, etc) required to emulate a specific neutron irradiation environment.

Therefore, our main objective was to find the way to calculate the primary displacement damage induced by neutron irradiation and by ion irradiation starting from the same point, that is, the PKA spectrum.

2. Methodology

Neutron irradiation induces elastic and inelastic nuclear reactions. The subsequent displacement of ions (Primary Knock-on Atoms [PKA] spectrum) is generated by both elastic and inelastic nuclear reactions, the elastic nuclear reactions contributing generally in more than 90% to the displacement damage [4]. This varies with the isotope analyzed. However, the PKA spectrum itself (displacements atomic induced by neutron irradiations) mainly induces elastic atomic reactions producing displacement damage. The combination of the neutron irradiation and PKA spectra (induced by neutrons) will produce different types of defects; on one side Interstitials, Vacancies and Clusters thereof can be produced by elastic and inelastic reactions whereas impurities - such as He, H, T-are only produced in materials by inelastic reactions, i.e. transmutation reactions. Hence, it is very important to predict/simulate the defects created by neutron and PKA spectrum induced by neutron, and the long-term evolution of defects and impurities. Therefore, the calculation of damage generated by fusion neutrons requires a dedicated methodology which can combine the effects of fusion neutron and ion irradiations (PKA spectrum induced by neutron).

The methodology used in this work to calculate the damage generated in materials due to the combined effects of fusion neutron irradiation and the damage generated by ion displaced by neutron irradiation was developed in previous works [5,6] by the authors, and it is based on a methodology previously developed by Vladimirov et al. [7,8]. This methodology allows us to calculate several damage parameters (PKA spectrum, database of displacement cascades, damage profile, damage function, and damage dose (dpa-new concepts)) for the materials analysed.

This methodology consists on a combination of Nuclear Data Libraries Processing, Neutronic transport and Monte Carlo Binary Collision codes. First, the neutron spectrum for the desired area is obtained using the neutron transport codes (MCNP5 code). This neutron spectrum and the nuclear data libraries (FENDL 3.0) are used as input for the Nuclear Data Libraries Processing code NJOY [9] to obtain the PKA spectrum. The recoil matrices are obtained using the module GROUPR of NJOY code. The Nuclear Data libraries used to obtain the recoils matrices are FENDL-3/SLIB release 2. Then, the recoil matrices are weighted by neutron spectra to get an averaged PKA energy spectrum for each facility and irradiation spot under consideration. The PKA differential cross sections weighted with neutron spectrum defines the Kinetic energy distribution of the PKAs induced by a specific neutron spectrum, that is, the PKA spectrum describes how the damage is actually produced during irradiation, since it defines the probabilities to generate each PKA with a specific kinetic energy.

In the case of ion irradiation PKA spectrum was calculated using Marlowe code. The PKA spectrum is obtained for both codes with a specific subroutine developed by us. But, currently we have developed a subroutine to be able to calculate either the PKA spectrum in the total implantation profile as the PKA spectrum on different depth bins.

Afterwards, to evaluate the fraction of Frenkel pairs generated by PKA with energy T, $N_d(T)$, Marlowe code is used. Marlowe is a displacement damage simulations code based on the Binary Collision Approximation (BCA) [10,11].

In order to quantify the energy lost to electronic excitation in the low energy range, several models are available. In the Lindhard, Scharff and Schiøtt (LSS) theory [12], the electronic system of the material is regarded as a continuum and has the consequence that the energy loss cross-section is proportional to the ion velocity, just as depicted by the Ohm's law.

In addition, the binding energies used to calculate the displacement of atoms is obtained from first-principles calculations or molecular dynamics calculations depend on the material assessed.

This model proved to work remarkably well in a broad range of conditions and may expectedly apply in conditions where electron localization effects on ion trajectories are small. They are not when the ion velocity is comparable or higher than the Bohr velocity, which is typically the case for ion energies above 25 keV per amu. With further increasing velocities, the transition from the Lindhard to the Bethe regime governed by Rutherford scattering is met. The physical understanding of this transition gave rise to an extensive literature that is rationalized in [13]. The whole range of velocities and masses is captured in a semi-empirical way by J. Ziegler [14], which we implemented as a specific module in the Marlowe code. To do so, the so-called heavy ion scaling rule can be used to calculate the stopping power of atoms with energies above 25 keV per amu [15]. However, a more computationally efficient way was found and that consists in using directly the stopping power data found in the SRIM code, where the typical stopping powers in nuclear materials (Fe, W, …) are available. Therefore, MARLOWE now allows to simulate the stopping of ions in materials with energies of MeV (or even GeV), such as those formed by collision with energetic fusion neutrons or transmutation reactions.

One of the advantages of MARLOWE is that it allows simulating displacement of atoms in materials much faster than Molecular Dynamics (due to BCA) and exploring much higher energies (MeV-GeV). In particular, it allows defining the lattice structure of the materials and thus allows simulating displacements in monocrystal, polycrystal and amorphous materials, in contrast to SRIM. Therefore, it is possible to take into accounts effects such as channelling, replacements, linear collision sequence, etc. The binding energies of atoms to their lattice sites used to calculate the displacement of atoms are obtained either from first-principles calculations or from molecular dynamics calculations. In order to take into account the recombination of defects that takes place during thermal spike (like MD) an effective capture radius I-V is assumed. Simply, when two defects are within a distance smaller than the capture radius, it is assumed that these defects would recombine during the cooling phase during the cascade, and are thus eliminated. This capture radius is adjusted on MD results.

In addition, a subroutine to be able to calculate either the PKA spectrum in the total implantation profile as the PKA spectrum on different depth bins was developed by us.

After the number of defects corresponding to a given PKA spectrum are calculated, the PKA spectrum is weighted with the damage profile, that is, the Number of Frenkel pairs versus PKA energy to obtain the damage function and the damage dose.

The damage function (W(T)) is calculated using the following equation:

$$W(T) = \frac{1}{D/t} \int^{T} \sigma_{PKA}(T')N_d(T')dT' \qquad (1)$$

where σ_{PKA} (T) is the PKA spectrum, N_d(T) is the number of Frenkel pairs by PKA of energy T, and D/t is the rate of damage created by the atomic displacement.

It is well known that different primary recoil energy spectra can produce completely different damage morphologies. Hence, the damage function converts the PKA spectra to the total damage in the materials and therefore W(T) indicates the cumulative damage production by all PKAs up to the energy T. It gives us information of how the damage is produced and of the average PKA kinetic energy generating the damage. It is a very useful function as it allows us to compare different neutron sources in a simple manner. This is because it is an integrated function which is normalised to the maximum value of each damage function.

In addition, the damage dose, which is, the concentration of vacancies as a function of neutron dose, [dpa values] is calculated using equation 2:

$$dpa = t\phi_{Total} \int_{T_O}^{T_{MAX}} \sigma_{PKA}(T)N_d(T)dT \qquad (2)$$

where Φ_{Total} is the total neutron fluence rate and *t* is the exposition time.

3. Conclusion

A methodology was developed to calculate the damage due to fusion neutrons in Materials (monocrystal, polycrystal and amorphous systems). This methodology is based on the methodology developed by KIT laboratory. It consists of a combination of Nuclear Data Libraries Processing, Neutron Transport and Monte Carlo Binary Collision codes.

This methodology allows to design irradiation experiments with ions to emulate neutron fusion effects in materials. It is possible because the displacements damage generation have been calculated using the same methodology for both neutron and ion irradiations (starting from PKA spectra).

The resulting damage profile used to calculate damage function and damage dose was calculated using MARLOWE code.

- A dedicated module developed at CIEMAT was used to account for energy loss of Ions in materials at energies in the Bethe regime (>> 25 keV / amu).
- Allows defining the lattice structure and accounts for effects such as channelling, replacements, linear collision sequence, recombination of I-V, etc….
- In order to take into account the recombination of defects, the capture radius is generally adjusted on experiments or MD calculations.
- A subroutine to be able to calculate either the PKA spectrum in the total implantation profile as the PKA spectrum on different depth bins was developed.

Mainly, a methodology to develop displacement damage data libraries using the MARLOWE code is proposed in order to standardize the calculation of primary displacement damage on compound materials.

4. Related Publications

- M. Hou, C.J. Ortiz, C.S. Becquart, C. Domain, U. Sarkar and A. Debacker, "Microstructure evolution of irradiated tungsten: Crystal effects in He and H implantation as modelled in the Binary Collision Approximation" Journal of Nuclear Materials, Volume 403, Issues 1-3, August 2010, pages 89-100
- F.Mota, R. Vila, C. Ortiz, A. Garcia, N. Casal, A. Ibarra, D. Rapisarda. V. Queral, "Analysis of displacement damage in materials in nuclear fusion facilities (DEMO, IFMIF and, Technofusion)" Fusion Engineering and Design 86 (2011) 2425 - 2428
- F. Mota, C. J. Ortiz, R. Vila, N. Casal, A. García, A. Ibarra, "Calculation of damage function of Al2O3 in irradiation facilities for fusion reactor applications", to be published in Journal of Nuclear Materials, (ICFRM-15 - 2011)

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15. M. Hou, C.J. Ortiz, C.S. Becquart, C. Domain, U. Sarkar and A. Debacker, Journal of Nuclear Materials, 403 (2010) 89-100