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Evaluation of damage in neutron irradiated boron carbide

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ABSTRACT

When irradiated in a reactor, neutron absorber materials undergo two different damages induced, first, by the elastic scattering of neutrons; second by the neutron absorption reactions. In boron carbide irradiated in a fast neutron flux, neutron scattering and the slowing-down of He and Li arising from the (n,α) absorption reaction both lead to the displacement of B and C atoms which energy ranges up to a few MeV. Simulating the neutron damage by ion irradiation requires the calculation of the damage produced in both cases. In this paper we propose an estimation of the actual defect production rate resulting from the fast neutron irradiation. For this, we first estimate the energy spectra of both the primary knocked-on atoms and atoms created by the absorption reactions, here in a Phenix-like neutron spectrum. We then deduce from SRIM calculations the actual energy distribution of all the atoms displaced along the displacements cascades induced by those primary projectiles. At last, we obtain an estimation of the number of displaced atoms per produced helium, about 305, most of them resulting from neutron scattering. This is far from a Kinchin-Pease or NRT estimation, of the order of several thousands, this arising from the fact that most of the energy is dissipated through electronic interactions. Such results are then used in order to perform ion irradiations aiming at a realistic simulation of synergetic effects of helium implantation and damage production.

1. Introduction

Ion implantations are widely used to simulate the damage and the composition change arising in materials irradiated in nuclear plants [1,2]. However, some parameters can hardly be accounted for; for example, a reactor irradiation most often last up to a few years to be compared to a few hours for an ion implantation, this making kinetics effects difficult to handle. Another parameter is the way the energy is dissipated into the material. The slowing-down of the particles happens according to two different processes [3]; either an interaction with the electrons (electronic slowing-down, associated with the electronic stopping power, Se) or with the nuclei (nuclear stopping power, Sn). The former is of low consequences in metals (energy dissipation in the electron sea) but can induce important effects in insulators or semiconductors [4]. Moreover, in some conditions, nuclear reactions happen (fuel, neutron absorber, tritium blankets in ITER, spallation sources…). In those cases, first the composition of the material is modified, but second the reaction products can be energetic enough to produce defects.

We are here interested in the behavior of helium in boron carbide. Boron carbide is widely used as a neutron absorber, either in thermal or fast neutron flux [5]. The main absorption reaction is the $^{10}B(n,\alpha)^{7}Li$ one. In fast neutron reactor, helium can be produced in large quantities, about $10^{22}/cm^3$ (i.e. concentration about 0.1), leading to swelling and cracking of the material. ^{4}He and ^{7}Li are emitted with energies in the MeV range (mass defect of the main absorption reaction). Owing to the low atomic mass of the constituents of the material, they can then produce structural defects. On the other hand, in a fast neutron flux, the primary knocked-on atoms (pka) produced by neutron scattering have an energy spectrum up to a few MeV, high enough to displace the atoms of the material. In both cases, neutron absorption or scattering, the nuclei emerging from the reaction are light ones (B, C, Li, He). They will slow-down in a matrix also constituted of light nuclei. In that case, it is known that most of the pka energy is dissipated by electronic interaction. However, some atomic displacements arise, mostly at the end of the ion paths. The questions which arise are then the ratio of produced helium to the total number of displaced atoms, the way energy is dissipated, and the consequence of the damage on the kinetics of helium, e.g. nucleation and growth of clusters.

Simulating the behavior of helium in boron carbide with ion implantations then leads to wonder if the associated damage in reactor can be reproduced as well as the gas concentration. For that, it is necessary to perform ion irradiations which parameters such as the damage to helium concentration ratio or the Se/Sn ratio are precisely known and

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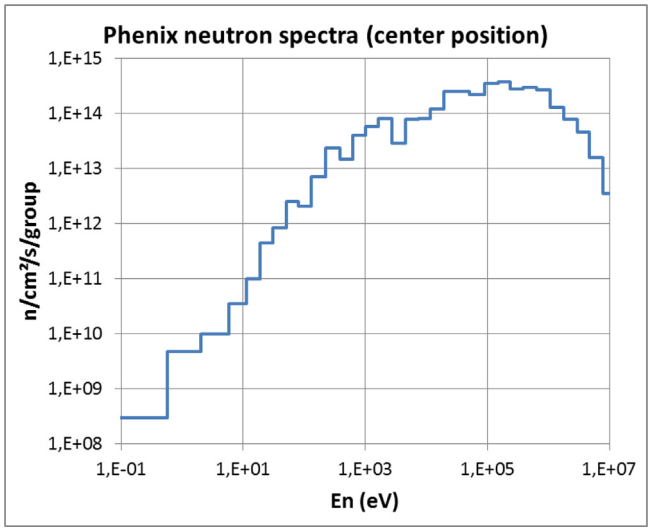

Fig. 1. Neutron spectrum at the center of the late French Phenix LMFBR [10].

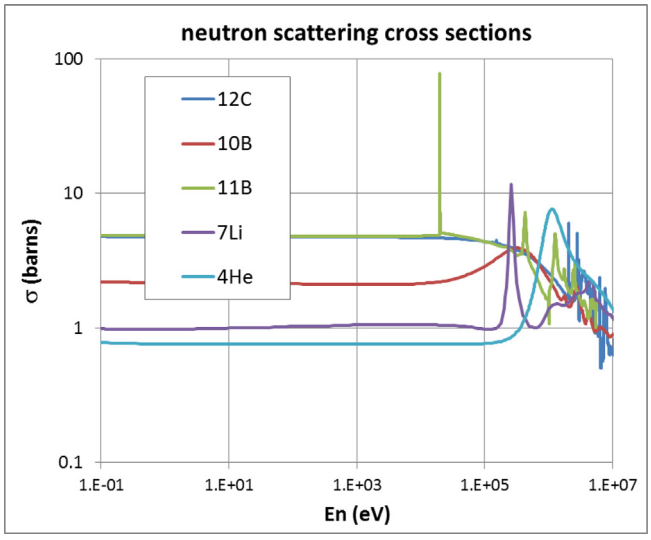

Fig. 2. Neutron scattering cross sections on the isotopes to be considered, ^{10}B, ^{11}B, ^{12}C, ^{7}Li, ^{4}He, from [11].

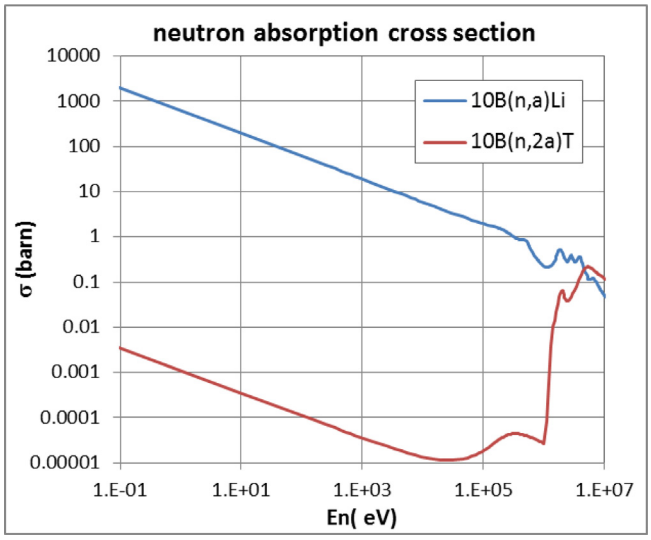

Fig. 3. Neutron absorption cross section for the ^{10}B(n,α)^{7}Li and ^{10}B(n,2α)^{3}H reactions, from [11].

representative. Such estimations can be done e.g. with tools such as SRIM [6]. But the comparison with the reactor conditions requires making this estimation with the actual reactor parameters. The available data are the material characteristics, the neutron flux in the reactor and the interaction cross sections. Here, we detail the route we have followed from those nuclear data to the atoms displacement rate (obtaining the helium production rate is straightforward, by convoluting the neutron spectrum and the absorption cross section). The first step is calculating the so-called primary damage spectrum [7–9], i.e. the energy distribution of the pka and of the created atoms by the absorption reactions. We then use the SRIM software to estimate the number of displaced atoms as a function of the pka's energy and the way energy is dissipated. At last, those results allow us to estimate the helium to atomic displacements ratio and the electronic stopping power range.

2. Data

We consider a neutron spectrum equivalent to the one in the center of the late French Phenix LMFBR (sodium cooled fast neutron reactor) [10]. The spectrum is divided in 36 groups (Fig. 1). The total flux is about 3.10^{15} n.cm^{-2}.s^{-1}.

The neutron interaction cross sections (neutron scattering and absorption) are taken from the ENDF data base [11]: Figs. 2 and 3. For the absorption reactions, we consider only the ^{10}B(n,α)^{7}Li one. We then neglect the ^{10}B(n,2α)^{3}H reaction. As a matter, the cross section for this reaction is about 10^{-6} lower than the (n,α) one, excepted in the fast neutron range (where it is rather poorly known [12]), where the neutron flux significantly decreases, leading to a production rate about one thousand lower than the (n,α) one. Second, it has a lower mass defect, about 0.23 MeV to be compared to about 2.8 MeV for the (n,α) reaction, leading to a much lower energy of the pka's. Excepted in the fast neutron range, the (n,α) absorption cross section varies as $\sigma = 611/\sqrt{E}$ (σ in barn, E in eV). Convoluting the neutron spectrum and the (n,α) absorption cross section leads to the helium production rate:

$$\tau_{He} = 4,01 \times 10^{-9}\ \text{at}^{-1}.\ \text{s}^{-1}$$

where 'at' stands for an equivalent boron carbide atom (initially full density $B_{0.8}C_{0.2}$, 48% ^{10}B enriched, 10^{22}/cm^3 burnup: see here under).

In order to have a good estimation of the damage on all the atoms, including the neutron capture products, we consider a highly irradiated boron carbide sample. We have chosen a 10^{22}.cm^{-3} capture density (most often called burnup). The boron concentration in non-irradiated B_4C boron carbide is about $1.1x10^{23}$.cm^{-3}. As a result, the mean atomic composition of the irradiated material is taken as $B_{0.72}C_{0.2}He_{0.08}Li_{0.08}$.

The initial ^{10}B enrichment of boron is taken as 48%, which reduces to 39% for a 10^{22}.cm^{-3} burnup.

Estimating the atom displacement rates requires the knowledge of the displacement energy threshold (TDE) for all the atoms of the material. Here we consider only mean values. For B and C, we have considered the values proposed in SRIM, which are coherent with the estimations performed by Zuppiroli et al. [13]. For He, we have used a most recent estimation of the thermal diffusion energy [14]. For Li, we have used an arbitrary, intermediate value: the only available value is for lithium aluminate, about 22 eV, i.e. in a quite different electronic environment with strong ionic bondings we have supposed higher than in B_4C [15]. Now, on a formal point of view, this value is of low importance: Li emitted with energy close to the TDE is unable to displace B or C and is of low efficiency on He. The values are then the followings:

- B: 25 eV
- C: 28 eV
- He: 2 eV
- Li: 15 eV