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Evaluation of the primary displacement damage in the neutron irradiated RBMK-1500 graphite

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ABSTRACT

Numerical simulations of the neutron induced primary displacement damage in the RBMK-1500 reactor graphite were performed using the Monte Carlo technique based on the Norgett-Robinson-Torrens (NRT) model. The sequence of the displacement evaluation consists of the reactor core modelling using MCNP6 and GEANT4 codes and output parameters, such as recoil ion and neutron flux distribution used by SRIM2013 and NJOY2016 codes, respectively, to evaluate displacements per atom. The comparisons were made between these codes in order to evaluate the accuracy of each approach. The neutron flux in the spent RBMK-1500 graphite was sufficiently correctly represented by both MCNP6 and GEANT4 models. The comparison between SRIM and GEANT4 codes shows that GEANT4 predicts fewer vacancies by more than 16% than SRIM in the ion energy range from 1 keV to 1 MeV. The neutron energy required to produce recoils due to scattering is around 80 eV. Below this energy displacements occur mostly due to neutron capture reactions. The average displacement rate was found to be around 0.51 displacements per atom per full power year, which corresponds to the $1.36 \cdot 10^{14}$ n/cm^{2}s neutron flux.

1. Introduction

Graphite is a primary material for the gas-cooled (UNGG, Magnox), RBMK and VHTR type nuclear reactor cores, serving as a low absorption neutron moderator and reflector [1]. The RBMK-1500 reactor is the water-cooled graphite moderated boiling water fission reactor, whose core consists of fuel, the control rod, and other purpose channels surrounded by the graphite stack [2]. There are two such units at the Ignalina NPP site not operating anymore (unit 1 was shut down in 2004 and unit 2 was shut down in 2009), and more than 3600 metric tonnes of irradiated and contaminated graphite waste is present in both reactors [3]. During the operation period, high energy neutrons continuously interact with the graphite matrix leading to the production of energetic recoil atoms as well as neutron activation products. The production of radionuclides depends on the presence of their precursors, which are mainly found in graphite as impurities [4,5]. Some radionuclides, such as ^{14}C or ^{10}Be, may originate during neutron interaction with ^{13}C, which is naturally abundant in the graphite matrix. The initial structural state of the graphite matrix and disordering effects of irradiation determine the locations, chemical bonds and mobility of radionuclides. It has been shown that ^{14}C tends to stabilize in the graphite matrix into sp2 or sp3 structures, but it can be removed through radiolytic corrosion when located close to open pores [6]. Thus, the evaluation of structural changes of graphite as well as their evolution under particular irradiation conditions may provide with insights about radiological characteristics of graphite waste, which needs to be considered prior to the treatment and disposal. Previous studies regarding the neutron induced primary displacement damage in graphite of RBMK reactors are solely based on displacement cross sections and the neutron spectrum, along with only comparative results [7]. Therefore, the need for this type of research comes from the lack of studies regarding the primary displacement damage evaluation in the RBMK-1500 reactor graphite.

Under the nuclear reactor conditions the majority of displacements in the graphite matrix occur due to elastic and inelastic scattering of the neutrons. Part of the kinetic energy of the neutrons is transferred to a lattice atom and, if the energy is high enough to displace the atom from its original lattice position, the recoil atom, known as the primary knock-on atom (PKA), is formed. The PKA then loses the energy through electronic excitations of material atoms (electronic energy loss) and elastic atomic collisions (nuclear energy loss) creating additional recoil atoms. The latter part is termed as the damage energy or the ballistic damage [8,9]. Depending on the energy, one PKA can create groups of 5–10 secondary knock-on atoms (SKA), thus producing a displacement cascade. The displaced atom leaves a vacancy in its original lattice position and, as recoil atoms are mobile, it can occupy a

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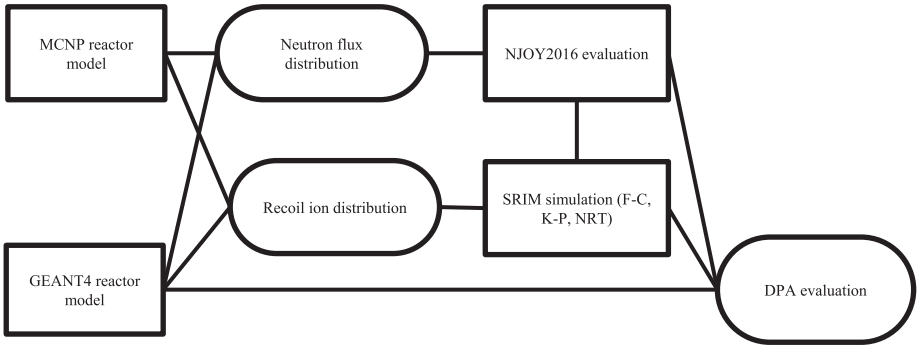

Fig. 1. Primary displacement damage evaluation sequence scheme.

vacant site in the lattice restoring its structure. The recoils that do not recombine are forming the stable point defects known as Frenkel pairs (consisting of a single vacancy and interstitial). Large quantity of point defects may agglomerate and this leads to more advanced defects (di-vacancies, line defects, etc.) and defect clusters. The quantity of point defects is often measured in DPA (displacement per atom) units, meaning the average number of times an atom is displaced from its lattice. The DPA value allows comparing different particle irradiation (protons, neutrons, electrons, etc.) damage effects on the material.

In an attempt to study the effects of the material exposure to the neutrons under the reactor conditions the ion beam modification as well as high energy ion implantation is widely used. This technique allows covering a broad range of the neutron recoil spectrum, while the rate at which atoms are displaced can be much higher ($\sim 10^4$ times) compared to the one in the nuclear reactor [10]. By varying irradiation conditions (e.g. ion nature, energy, fluence) both the ballistic damage and electronic excitation effects can be studied in coupled or decoupled manner [9]. Thus, when considering the irradiation regime in order to study the certain effects the theoretical simulation should be performed. The understanding of radiation damage processes as well as the subsequent evolution of the interaction induced phenomena forms the basis to study the properties of targeted material.

Most of the displacement simulations are performed using two distinct methods: molecular dynamics (MD) and binary collision approximation (BCA) simulations. The first method uses realistic interatomic potential functions to describe the forces on the atom as a function of the distance between it and the atoms in the system, thus yielding the equations of motions for the atoms. These equations are then numerically solved over small time steps [11]. By varying the time length different stages of the displacement cascade can be studied and a thorough evaluation of the primary displacement damage can be performed [12,13]. This method is useful when the dynamics of the defect creation and evolution of the system are in mind as it gives more detailed results. This approach demands much of computing time, so large scale systems are not very suitable for simulations. The binary collision method considers only the interactions between two colliding atoms at a time, thus ignoring the many-body interactions and it is more time efficient. Using this method, only the particles with significant energies are tracked. Two distinct classes of BCA models exist: those for crystalline targets (termed BC or binary crystal models) and the ones for amorphous targets (termed MC or Monte Carlo models). The first one is somewhat similar to MD models as atoms have well-defined initial positions and collision parameters are calculated from the particle locations [11]. In MC models stochastic methods are used to locate target atoms and to determine collision parameters [14]. Previous studies used MCNP [15], GEANT [16], SRIM [17], NPRIM [18] and other applications to evaluate the neutron induced displacement damage. In this paper, the simulation setup based on the Monte Carlo (BCA) approach is described to obtain the neutron induced primary displacement damage in the RBMK-1500 graphite by MCNP6, GEANT4, SRIM and NJOY2016 applications. The aim of this work is to provide the theoretical evaluation of the neutron induced point defect generation in the RBMK-1500 graphite and to use this data for further researches.

2. Methodology

2.1. Simulation of neutron interactions with graphite

The simplified sequence of simulation used in this research is shown in Fig. 1. MCNP and GEANT4 were used for the evaluation of neutron and recoil ion distributions in the reactor core model. The neutron and recoil ion distributions evaluated by MCNP were used in NJOY2016 and SRIM applications, respectively, for the displacement evaluation. GEANT4 allowed a direct displacement evaluation. The steps of the sequence are discussed in detail below.

MCNP 6.1 (Monte Carlo N-Particle) code [19] was used for simulation of the neutron interaction with graphite. MCNP is based on the technique where individual particles are tracked starting from their creation in the particle source to their disappearance by absorption or escape. Particle parameters and the type of interactions it undergoes depend on the probability distributions, which are randomly sampled using transport data. MCNP uses cross-sections from nuclear data libraries, such as ENDF, JENDL, etc. In this research ENDF-VII.1 [20] reaction cross-section library was used.

A single column of the RBMK-1500 reactor graphite stack with 2.4% ^{235}U fuel enrichment and 0.41% erbium absorber fuel assembly was modelled as it corresponds to the typical load of Unit 1 of the Ignalina NPP. The column consisted of the 686 cm height fuel assembly surrounded with graphite and the 50 cm height top and bottom neutron reflectors (see Fig. 2).

The fuel assembly consisted of 18 fuel rods with the zirconium alloy cladding surrounded by coolant water and the fuel channel tube. The whole assembly was surrounded with the graphite sleeve and the column. The density of graphite in this model was 1.675 g/cm^3, the temperatures of the graphite column and the graphite sleeve were 700 K and 620 K, respectively. The cooling water density was 0.5 g/cm^3 and the temperature was 550 K. Although the fuel enrichment and burnup vary along the reactor, in this research a fresh fuel combination was used, which gave the neutron multiplication factor $k_{eff} = 1.38$. There was no significant difference in the neutron flux distribution between fresh and irradiated fuel configurations. For simplification and acceleration of simulation, reflective surfaces were used on the side surfaces of the column. In a simple manner, the use of reflective surfaces simulates the infinite lattice without the need to simulate the whole reactor.

Spatial distributions of neutron and recoil ion fluxes are obtained using F4 and F4Mesh tallies, and PTRAC (*Particle Track Output*) function, which is used for tracking individual ions. MCNP PTRAC card generates a separate output file of user-defined events and parameters, i.e., the type of event, energy and location of the particle, etc. In this case, only the bank events of carbon recoil ions are recorded within the graphite. The PTRAC output file is later used in SRIM for modelling of