MODELLING AND SIMULATION OF ^{14}C-TRANSPORT

WERNER VON LENSA

Forschungszentrum Jülich: presented on behalf of: Institut für Nuklearen Brennstoffkreislauf *and* The University of Aachen, Germany

ABSTRACT

This work represents a progress report on the CARBODISP project, examining the molecular dynamics and the transport mechanism of ^{14}C in irradiated nuclear graphites and, in particular, the effect of grain boundaries.

1. MOLECULAR DYNAMICS STUDY OF ^{14}C TRANSPORT IN NUCLEAR GRAPHITE

The work in the field of modelling and simulation has been focused on the description of the formation of the radionuclide ^{14}C and its transport in the nuclear graphite as a result of recoil process. In view of a potential decontamination of the irradiated graphite, the knowledge of the binding state of the ^{14}C-atoms and their local distributions in the graphite structure as well as the release mechanism is of particular importance.

As nuclear material graphite is characterized by a well defined crystalline structure as given in Figure 1 (hexagonally shaped). The individual ^{14}C-atoms are located in layers with a distance of 3.5 A which are shifted against each other. In the case of irradiation elastic and inelastic interaction with fast neutrons take place- in parallel to the formation of ^{14}C by activation leading to the creation of Frenkel-pairs (interstitials and vacancies). Due to the high cross section for this process the concentration of the defect pairs are higher than ^{14}C concentration by orders of magnitude. However as a consequence of low energy for potential recombination and temperature dependent self diffusion the concentration vacancy-interstitial takes place resulting in a reduction of the concentration (0.15 eV).

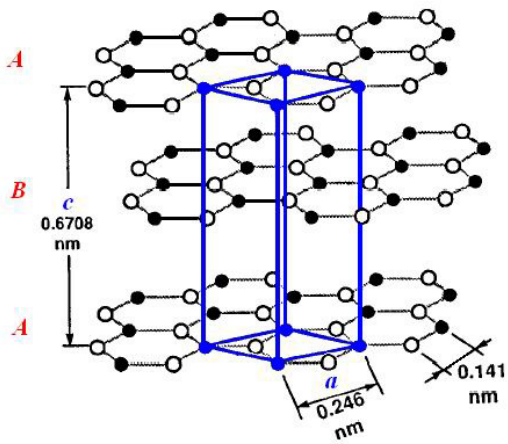

FIG. 1: Crystalline Structure of Graphite

^{14}C is formed as a result of the activation of captured trace elements like oxygen and nitrogen existing in nuclear graphite and particularly by activation of carbon-isotopes. The formation by activation of ^{13}C is less than over the ^{14}N activation which is embedded in graphite during the manufacturing process or reactor operation. The reason for the higher activation rate is the higher cross section as depicted in Figure 2.

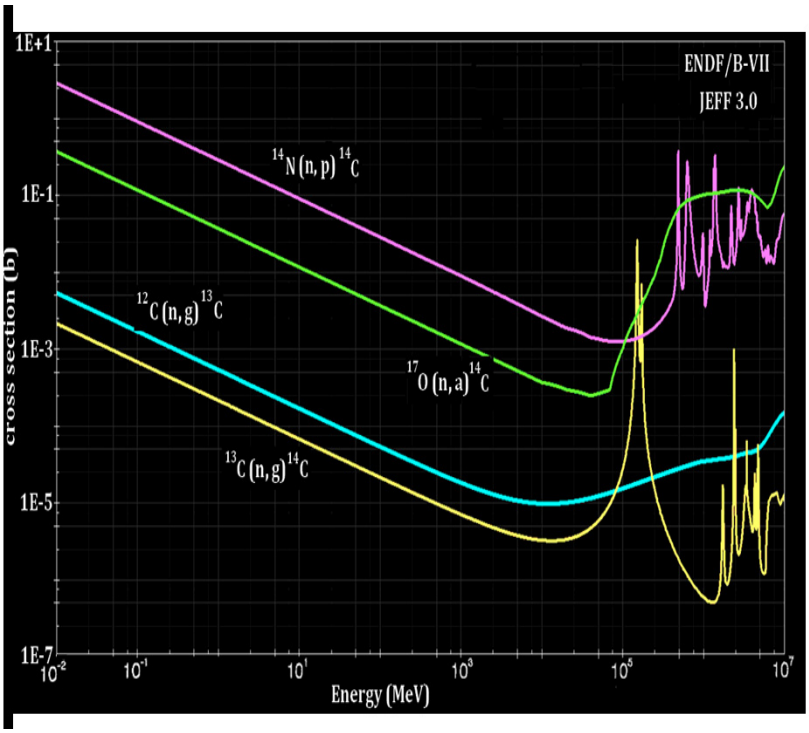

FIG. 2: Cross Sections for Production of ^{14}C

2. TRANSPORT MECHANISM OF ^{14}C

The mechanism of the transport/ distribution and release of ^{14}Cinside the irradiated graphite structure is a complex issue and is studied by the application of molecular dynamic (MD) method which allows the simulation of the interaction mechanisms at atomistic level in the lattice structure of graphite. The result of such a simulation is given in Figure 3, which shows the configuration and state of the graphite lattice at end of recoil atom transport (^{14}C). Accordingly the ^{14}C atom is located in an interstitial position accompanied with a deformation of the lattice structure due to the cascade process. For this aim the program LAMMPS was used to investigate the behaviour and the configuration of the defects (^{14}C). The MD works with the solution of Newton's equation of motion for atoms and molecules, coupled together by certain atomic interaction potentials. The transport process and the binding of ^{14}C were initially investigated in pure graphite with a crystalline structure under various initial and boundary conditions; in addition, a possible effect of the grain boundaries, pores and impurities was studied. The formation process of ^{14}C is associated with the recoil of ^{14}C atoms with a kinetic energy in the range of keV (2.56 keV for the ^{13}C activation and 42 keV by ^{14}N) leading to the removal from the lattice site. This transfer of ^{14}C is accompanied by various interaction and ionization processes which result in a disorder of the lattice structure as well as in the formation of further displacements (interstitial atoms and vacancies).

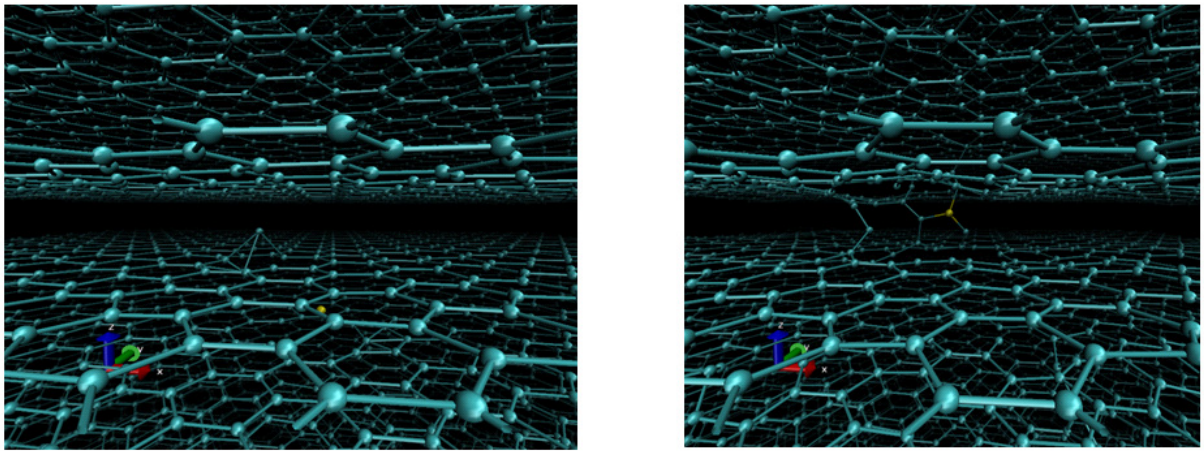

FIG. 3: Initial and Final State of Single Cascade in Perfect Lattice Structure (yellow: C14)

The MD simulation showed that at an initial energy of 10 keV, for example, the recoil atom is displaced after the generation by a mean distance of 60-130 angstrom in the crystal structure being influenced by the interacting potentials (see Figure 4). However the result depends on the type of the potentials governing the interaction between the individual atoms in the short range and recoil direction relative to the orientation of the crystal lattice.

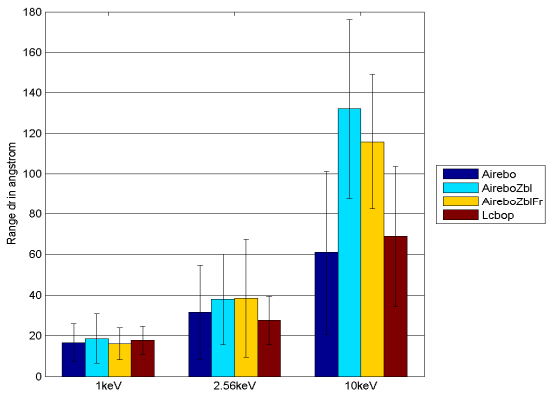

FIG. 4: Range of ^{14}C Dependence on Recoil Energy and f or Different Potentials

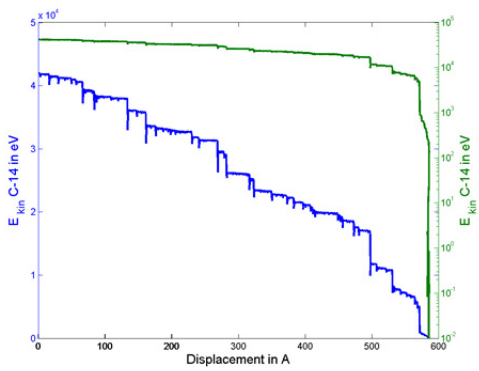

FIG. 5: Displacement and Energy Loss of ^{14}C in Crystalline Graphite for 40 keV

Resulting from the symmetrical character of the individual atoms at the c-layers, the mean range and the transport of the primary ^{14}C is not significantly influenced by the temperature variation. This result, however, needs to be verified by detailed modelling of the boundary conditions.

In addition, specific and stepwise energy loss of the ^{14}C atoms taking place during the transport process was studied as given in Figure 5 for an example of 42 keV (^{14}C produced by ^{14}N). Accordingly a smooth energy loss takes place with a specific value of approx. 100 eV per atom. After getting lower energy (5 keV), the energy loss shows a sudden drop probably caused electronic interaction and ionization.

3. EFFECT OF GRAIN BOUNDARIES

Due to the complex manufacturing process the reactor graphite consists of polycrystalline configuration and no specific alignment which lead partly to the formation of grain boundaries. To answer the question of the grain boundaries on the ^{14}C transport a polycrystalline model consisting of individual monocrystaline grains was generated for MD simulation. For the model different orientation and angle respectively between the individual crystalline grains were assumed as given in Fig. 6. In this configuration the recoil ^{14}C-Atom was started under various angles with recoil energy of 40 keV (^{14}C by activation of ^{14}N). Due to the fact that no particular potentials are acting at the interface (representing the pores) the ^{14}C atoms cross the grain boundaries and pores without any deflection and path change. The results of the simulation have been summarized and depicted in Figure 7 for different orientation of individual crystalline grains. Accordingly the starting ^{14}C atom undergoes a transport without being influenced by the adjacent boundary layers but by the orientation of the subsequent grain layers. The figure shows that a significant accumulation of ^{14}C in the vicinity of grain boundaries or surfaces (pores) caused only by the stopping processes (potentials) is not expected (Figure 7).

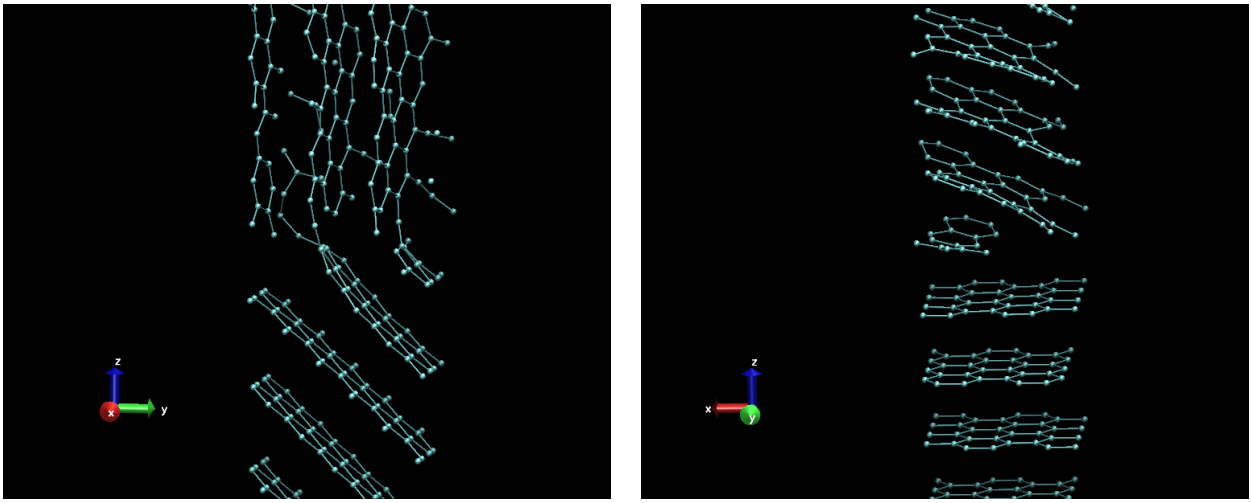

FIG. 6: Configuration of crystalline grains for Md simulation of the boundary effect

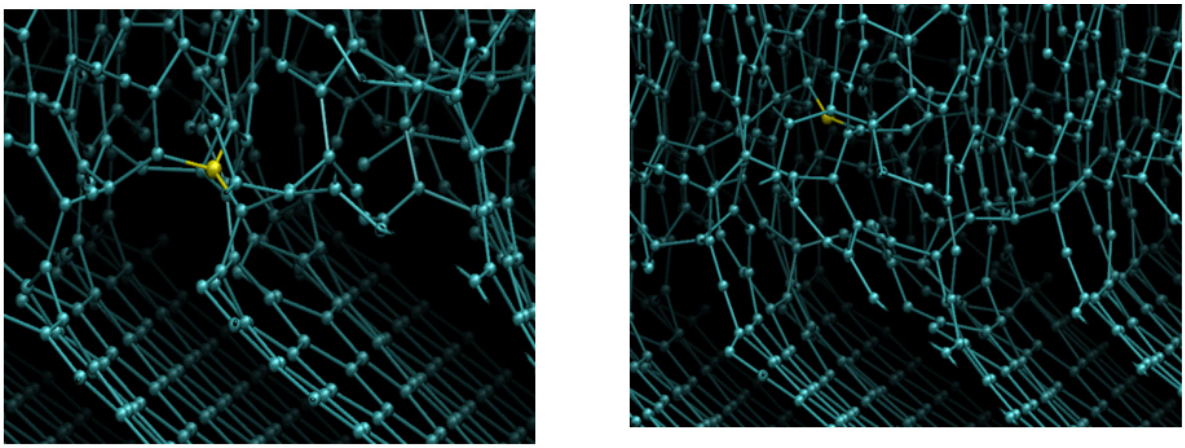

FIG. 7: Final states of ^{14}C near Grain Boundaries

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