



Molecular dynamics modelling of radiation damage in zircon

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Zircon (ZrSiO_4) is among actinide-bearing phases which has been proposed as a crystalline confinement matrix for nuclear waste management, especially for weapon-grade plutonium and UO_2 spent fuel in the USA. Zircon is also widely used in geochronology. But, with accumulating α -decay damage, zircon undergoes a radiation induced transition to an amorphous (or metamict) state. So, in the present work molecular dynamics simulations (MD simulations) of zircon structure have been performed to study radiation damage in zircon. In this technique, one simulates the propagation of an energetic particle in a system of atoms interacting via model potentials, by integrating the Newton equations of motion.

Author has used version 3.09 of the DL_POLY molecular simulation package. Zircon structure containing 181944 atoms (19x19x21 unit cells) was equilibrated at 300 K for 10 ps, and one Zr atom (usually called the primary knock-on atom, PKA) was given a velocity corresponding to an implantation energy of about 20 keV. MD simulations were performed in the microcanonical ensemble that is under conditions of constant particle number, volume and energy. Results of the MD simulations show that the number of interstitials is equal to 840 atoms. This is very close (4000-5000 atoms for 70 keV recoil atom ^{234}Th) to what is measured in the diffuse x-ray scattering and NMR experiments on amorphous metamict samples (damaged by natural irradiation) of geological age. It has been shown that the damaged structure contains several depleted regions with characteristic sized up to 2,5 nm after single event and up to 4,5 nm after three overlapping events. Furthermore, these events produce channels of depleted matter between the overlapping damaged regions. These channels provide a high-diffusivity path for radiogenic Pb (percolation effect). Loss of radiogenic Pb may result in to incorrect dating of rocks.