université PARIS-SACLAY

ÉCOLE DOCTORALE
Particules, hadrons, énergie et noyau : instrumentation, imagerie, cosmos et simulation (PHENIICS)

Title : Experimental simulation and modeling by ion beams of alpha decay damage in apatite -Applications to thermochronology and to nuclear sciences for energy

Keywords : apatite ; alpha decay ; radiation-induced damage ; ion channeling ; thermochronology ; confinement matrix

Abstract : Apatite is a compound with applications in the fields of earth sciences and nuclear energy. Its natural mineral form can be used to predict thermal histories of rocks and its associated geological processes while a synthesized apatite ceramic form can serve as an alternative confining matrix in the immobilization of actinides of spent nuclear fuel. Under the domain of earth science, an apatite helium (AHe) age, which is an apparent age of a rock, can be determined by measuring the present-day concentrations of U, Th, Sm, and He in an apatite crystal. The measured amount of He is essentially a balance between (1) He production from U and Th radioactive decay series and Sm alpha decay and (2) He that diffused out of the crystal, especially at high temperatures. The He diffusion coefficient is based from the Arrhenius Law but recent studies strongly indicate that the radiation damage in an apatite crystal, which is mostly due to the ballistic damage caused by the slowing down of alpha recoil nuclei, modifies the He diffusion behavior, and consequently the measured amount of retained He in the crystal. The amount of radiation damage in apatite is thus of great importance in the accurate measurement of He ages and modeling of rock thermal histories.

In nuclear energy applications, an apatite crystal will typically be subjected in much higher alpha decay doses than most natural apatite minerals are subjected into. The high damage dose could result in full amorphization and a possible nucleation and growth of He bubbles and both phenomena could destabilize the long-term immobilization performance of apatite nuclear waste forms. While the amorphization mechanism has been studied extensively in the past, the destabilization caused by He bubbles, on the other hand, remained not that much investigated.

The aim of this thesis is to investigate in a fundamental level using *in situ* and *ex situ* ion implantation, Rutherford backscattering Spectrometry (RBS/C), McChasy (an ion channeling Monte Carlo code), and Transmission Electron Microscopy (TEM) the (1) evolution of radiation damage buildup separately-induced in apatite by alpha recoil nuclei and alpha particles and the (2) athermal, ionization-induced recovery of pre-existing defects attributed to the electronic energy loss of alpha particles. The damage buildup experiments were done over a wide ion fluence range to reproduce defects that are representative of what could be found in natural apatite minerals and apatite nuclear waste forms. The irradiation temperatures of damage buildup experiments, in a similar manner, were also varied to correspond with typical earth science and nuclear energy applications (in the rage 293-393 K). Also, ionization-induced recovery experiments were extensively conducted with the objective of determining the dependence of damage recovery kinetics with the initial amount and type of defects present in an apatite crystal.

The experimental and simulation results obtained from this research strongly suggest that the buildup kinetics of radiation damage attributed to both alpha recoil nuclei and alpha particles could be described as a two-step process with each step characterized by specific types of main defects induced by irradiation. Moreover, the alpha particle-induced damage recovery was only found to be significant or efficient in certain types of defects, such as amorphous clusters. Using the kinetics of alpha decay damage buildup and recovery, the long-term microstructural evolution of apatite nuclear waste forms could be predicted and the modeling of rock thermal histories improved.

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