U^{4+} and Pu^{4+} incorporation in zircon and their effect upon helium diffusion in the host lattice

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Zircon mineral ($ZrSiO_4$) is a potential host matrix for the encapsulation of highly radioactive materials, such as dismantled weapons and nuclear waste from power stations (Ewing, 2001).

We report a detailed computer modelling study, based on interatomic potentials, to address two fundamental issues concerning radiation processes in the zircon mineral: first, the physics and chemistry of U^{4+} and Pu^{4+} inclusion in the crystalline matrix of zircon and, second, the effect of U^{4+}/Pu^{4+} dopants on the incorporation and diffusion behaviour of helium in zircon.

Our results show that U^{4+} and Pu^{4+} can incorporate isomorphously in zirconium sites (Zr^{4+}). The solution energies of U^{4+} and Pu^{4+} are 0.18 eV and 0.03 eV respectively, which indicates that Pu^{4+} is more readily incorporated as a dopant than U^{4+}. However, the positive solution energies show that the inclusion of the two species in the crystalline matrix would occur only under high temperature conditions.

Interstitial incorporation of helium in zircon is an energetically unfavourable process, which causes strong deformation in the host lattice due to movement of atoms in the channel walls to accommodate He. Moreover, the presence of Pu^{4+} and U^{4+} dopants in the zircon lattice has a direct effect upon the energetics of helium diffusion between interstitial sites. For instance, activation energies calculated for He diffusion are dependent upon the distribution of U^4 and Pu^{4+} in the lattice, and can differ by as much as 0.2 eV from those calculated for the perfect zircon.

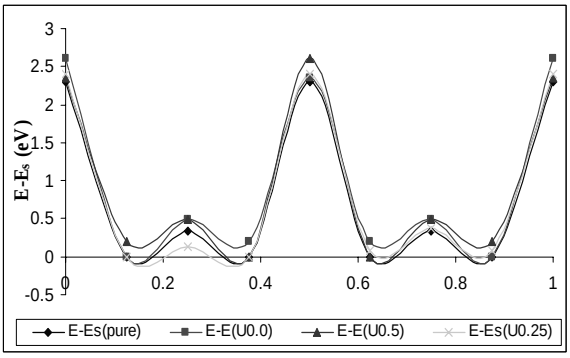

Figure 1: Diffusion pathways for helium movement along the x-direction of U-doped zircon.

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Short-lived naturally occurring radioisotopes (^{234}Th, ^{7}Be, ^{210}Pb) as a tracer for particle transport in the Gironde fluvial-estuarine system (France)

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Human activities release various chemical species to the environment. Many of these substances bind strongly to sediments, which are then delivered to rivers. Thereafter the fate of pollutants in fluvial environments is linked to particle transport. The short-lived, naturally occurring radioisotopes, ^{234}Th ($T_{1/2}$ = 24.1 days), ^{7}Be ($T_{1/2}$ = 53.3 days) and ^{210}Pb ($T_{1/2}$ = 22.3 years), can be used as potential tracers to investigate particle transport from land to sea. However, up to now, their use is still rare and for ^{234}Th mainly devoted to marine environments. Uranium (^{238}U) forms ^{234}Th through radioactive decay in the dissolved phase. However, due to the typically low and invariable concentrations of ^{238}U in riverine environments, the formation of ^{234}Th is often insufficient to be used as a tracer.

The programme ARTTE "Application Radioisotopes as Tracers of particle transport in environment", has been carried out to develop the application of ^{234}Th, ^{7}Be and ^{210}Pb in the polluted (e.g. Cd, Hg (e.g. Schäfer *et al*., 2006)) Gironde fluvial-estuarine system (South-West France). A preliminary investigation in 2005 has revealed adequately elevated and spatiotemporally stable dissolved ^{238}U activities within this system (Saari *et al*., submitted), allowing the ^{234}Th application.

^{234}Th, ^{7}Be and ^{210}Pb activities in suspended materials were measured monthly (2006-2007) by γ-spectrometry within the Garonne and Lot Rivers, and the Gironde estuary. The first results reveal: (1) ^{7}Be/^{210}Pb$_{xs}$ activity ratios decrease from river to estuary, due to aging of sediments and/or to resuspension of old bottom sediments (2) ^{234}Th$_{xs}$ activities show large variabilities with time, from negligible values up to about 300 mBq g^{-1}, probably due to differences of particle residence time in the river channels.

References

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