The Nuclear Fuel Cycle: Role of Accessory Minerals in Problem Solving

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The nuclear fuel cycle involves mining of uranium minerals, e.g., uraninite, coffinite, and brannerite, followed by ore processing, enrichment of the fissile component, and production of fuel pellets that are encapsulated in rods and used in nuclear reactors to generate electricity. Fuel rods are used in either a once-through cycle and removed from the reactor core for storage (awaiting ultimate disposal) or they are reprocessed and used again in reactors.

Historically, studies of Th-U minerals and ore deposits have played a major role in providing background information relevant to the geological disposal of nuclear wastes arising from both domestic electricity generation and national defence programs. As shown in Figure 1 for the example of natural pyrochlore and zirconolite, Th-U minerals may provide useful information on the performance of nuclear waste forms in terms of the crystal chemistry, radiation damage effects produced primarily by alpha decay processes, and their stability when exposed to a range of natural aqueous fluids. This example is discussed in some detail in the companion paper for this conference (Lumpkin et al. 2017). Additional examples are reviewed by Lumpkin and Geisler-Wierwille (2012), including a range of oxide, silicate, and phosphate minerals of interest to the nuclear waste disposal community.

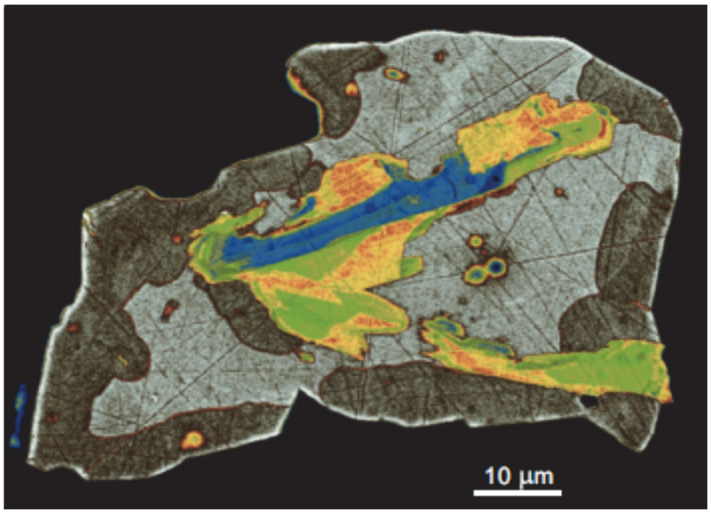

Fig. 1. *False color backscattered SEM image of zirconolite and oxycalciobetafite in Ti-rich hydrothermal veins in the contact aureole of the Adamello massif, northern Italy*

Radiation damage in the minerals and their synthetic analogues occurs on time scales ranging from picoseconds to millions of years and longer in very old rocks. A combination of atomistic modelling, light and heavy ion irradiation, and doping of synthetic samples with short-lived actinides has been very effective in delineating the damage mechanisms and recovery processes. Atomistic simulations using molecular dynamics and density functional theory have been instrumental in understanding the damage and recovery

mechanisms on picosecond time scales and the energetics of some of the processes (e.g., defect formation and migration). These studies range from model systems such as the TiO_2 polymorphs to detailed studies of minerals such as perovskite, pyrochlore, and zircon, among others.

In particular, experimental and geological studies of Th-U minerals have revealed long-term recovery processes occurring at elevated temperature and pressure for minerals including pyrochlore, zirconolite, zircon, perovskite, brannerite, and crichtonite, among others. Studies of natural zircon have been particularly important beginning with the ground-breaking work of Holland and Gottfried (1955) and the work of others in the early 1950s. These studies set the stage for understanding the crystalline to amorphous transformation in zircon and other minerals and had a profound impact on future laboratory studies using samples doped with ^{238}Pu or ^{244}Cm with alpha-decay half-lives of ~ 88 and 18 years, respectively. More recently, it has been pointed out that the thermal history of zircon plays a major role in the amount of radiation damage that is retained today (Nasdala et al. 2004). Additional examples will be illustrated here using thermo-chronology data for specific geological localities.

All of these studies, together with related work have contributed to knowledge about nuclear waste disposal. Furthermore, investigations of meteorites have provided some data on the trapping of noble gases in carbon compounds (amorphous, graphite, diamond, etc.) and this has some relevance to fission gas trapping in nuclear reactor materials (both fission and fusion).

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