

Description of the PhD project:

Radiation damage phenomena in monazite and other phosphates

Some minerals such as orthophosphates of the monazite–xenotime group (CePO_4 – YPO_4) incorporate considerable amounts of actinoides in their crystal lattice. The radioactive decay of the unstable nuclei causes long–term self–irradiation of these minerals. However, a permanent self–annealing process works against the accumulation of radiation damage; the relationship of these two counteracting processes is controlled mostly by temperature. The dominance of defect accumulation results in structural damage and finally complete destruction of the crystal structure, a process called metamictization. Metamict minerals generally exhibit reduced physical stability and increased chemical reactivity. Understanding of these processes, and their causes, will support the interpretation of radiometric age determinations. The degree of accumulation of radiation damage is also an important information in reconstructing the sample’s geological (thermal) history. Further, the investigation of radiation–induced changes in phosphates, and their long–term stability, is of interest in material and environmental sciences, because monazite–type minerals are considered as potential host minerals for the long–term storage of radioactive waste.

This research project aims at gaining new knowledge of the changes of phosphate minerals induced by natural radioactivity and the role of their chemical composition in structural reactions as well as of their annealing behaviour. To achieve this, structural, crystal–chemical, and isotopic characterization of the samples with micro–methods will be applied. Spectroscopic analysis methods, such as Raman, luminescence and infrared absorption spectroscopy, are to be used primarily; they will be supplemented by transmission electron microscopy, chemical analysis using electron microprobe and X–ray diffraction. The SHRIMP (Sensitive High Resolution Ion MicroProbe) will be used for U–Th–Pb dating and the investigation of potential secondary lead losses (U–Th–Pb isotopic discordance). Samples to be analysed include natural phosphate minerals, products of annealing experiments, and synthetic monazite and xenotime crystals doped with non–formula elements. In addition to the experimental methods, statistic evaluation procedures as well as mathematical modelling calculations will be done. Among other goals, the dependence of both the chemical composition and the degree of radiation damage on spectra will be studied, to finally allow one to use spectroscopic micro–analyses to estimate the radiation damage in samples with known chemical composition. Also, potential effects of the structural state of orthophosphate minerals on their electron backscatter behaviour (for zircon shown by Nasdala et al., 2006) will be investigated.

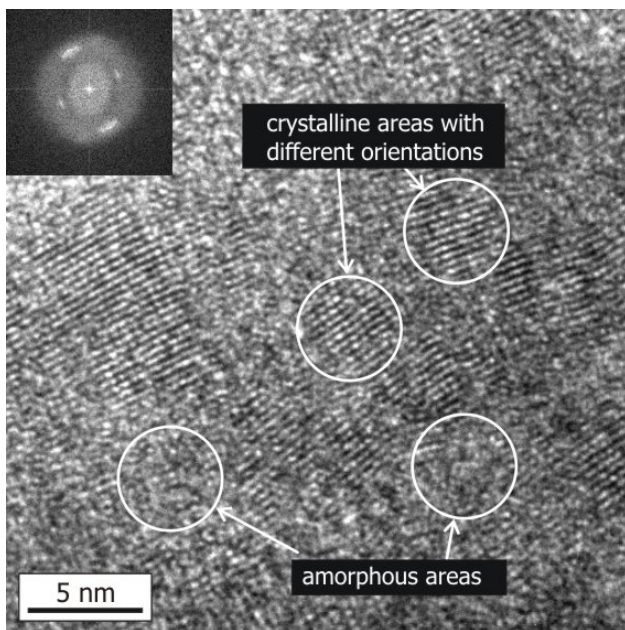


Fig. 1: Transmission electron microscope image of a Th–rich monazite sample from Moss, Norway. As a result of strong self–irradiation, the sample consists of (slightly rotated) crystalline remnants in an amorphous matrix (see Nasdala et al., 2002).

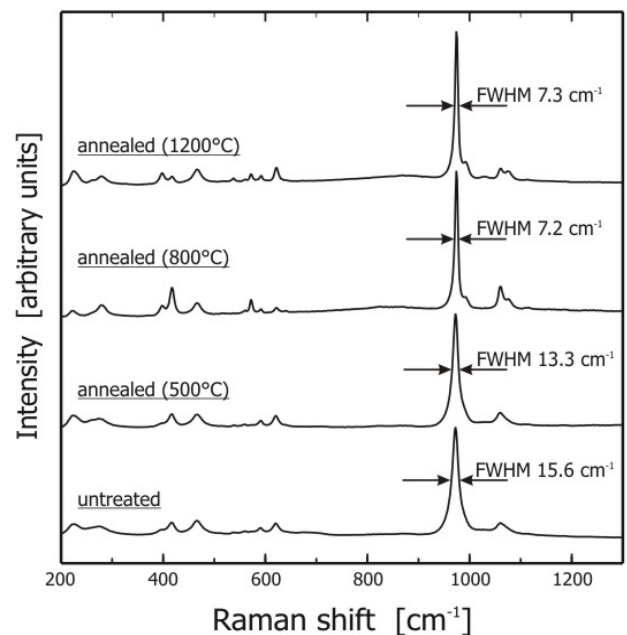


Fig. 2: Raman spectra of a radiation–damaged monazite crystal from Brazil. Annealing has caused partial (500 °C) or complete (≥ 800 °C) structural recovery. See the decreasing width of the $\nu_1(\text{PO}_4)$ band (modified from Seydoux–Guillaume et al., 2002).

References:

- Nasdala, L., Wirth, R. and Finger, F. (2002): TEM and electron microprobe study of a monazite from Moos, Norway. *Ber. Dt. Mineral. Ges., Beih. z. Eur. J. Mineral.*, 14, 107.
- Nasdala, L., Kronz, A., Hanchar, J.M., Tichomirowa, M., Davis, D.W., Hofmeister, W. (2006): Effects of natural radiation damage on back–scattered electron images of single crystals of minerals. *Amer. Min.*, 91, 1739–1746.
- Seydoux–Guillaume, A.M., Wirth, R., Nasdala, L., Gottschalk, M., Montel, J.M., and Heinrich, W. (2002): An XRD, TEM and Raman study of experimentally annealed natural monazite. *Phys. Chem. Minerals*, 29, 240–253.