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Study of uranium in natural and synthetic carbonate apatites using radiochemical, synchrotron radiation and fission track techniques

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Carbonate apatites were found to be present in U-bearing organic-rich phosphatized limestones from Epirus region (NW Greece). The mineralogical investigation of the rock samples revealed, along with organic matter and calcite, the presence of abundant carbonate apatites and in lesser extent fluoroapatites, zircons and pyrite. Electron microprobe (EMPA) and Raman spectroscopy were applied for the characterization of carbonate apatite crystals. Synchrotron radiation micro-X-ray fluorescence (SR μ-XRF) measurements performed using as targets carbonate apatites demonstrated the uranium distribution, while micro-X-ray Absorption Near-Edge Structure (μ-XANES) illustrated that U was present in the crystals tetravalent form (U(IV)).Fission track dating was attempted using both carbonate apatite and fluoroapatite crystals. Fluoroapatites were successfully etched and dated showing various ages. On the other hand, the dating of carbonate apatites was not successful because of the lack of a validated etching procedure. However, the high density of homogenously distributed fission tracks in mica detector was an evidence of their high U content. The mechanisms of U adsorption onto carbonate apatites synthesized in the laboratory were studied and showed that this mineral could act as a significant U sorbent. Sorption experiments performed in solutions of pH 4 to 10 showed a partial dissolution of the synthetic carbonate apatite crystals surface and precipitation of meta-autunite (Ca(UO2)2(PO4)2.6-8H2O). The XRD patters indicated that the formation of meta-autunite gradually decreased with increasing solution pH. Under very alkaline conditions (pH 13) meta-autunite did not appear. Regardless of this the carbonate apatites showed a high uranium uptake. In the latter case the unit cell of carbonate apatite crystals decreased indicating a possible substitution of Ca by U in the crystal lattice. In the current studyalong with a first approach to their etching method, new insights concerning the mineral chemistry of uranium in carbonate apatites are presented.

Authors: Dr TZIFAS, Ioannis (Heidelberg University); Prof. GLASMACHER, Ulrich A. (Institute of Earth Sciences, Heidelberg University); Prof. MISAELIDES, Panagiotis (iSchool of Chemistry, Aristotle University of Thessaloniki); Prof. GODELITSAS, Athanasios (Faculty of Geology and Geoenvironment, National & Kapodistrian University of Athens); Prof. KANTIRANIS, Nikolaos (School of Geology Faculty of Sciences Aristotle University of Thessalonik); Dr KARYDAS, Andreas (Institute for Nuclear and Particle Physics National Center for Scientific Research "Demokritos"); Prof. TSIKOS , Harilaos (Department of Geology, Rhodes University)

Presenter: Dr TZIFAS, Ioannis (Heidelberg University)

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