Cu-Ba hydrated uranyl phosphates from the Nisa deposit: chemical and petrographic evidences for mineral reequilibration processes

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Uranyl phosphates are a mineral group which include a wide range of different species, each containing specific cations within the hydrated interlayer between the uranyl and phosphate layer, and often display a geochemical/mineralogical relationship with Fe(III) oxy-hydroxides. The environmental relevance of these U-phases arises from their low solubility at most natural conditions found in surface and groundwater, where they can ultimately control aqueous U levels.

In the present work, samples of naturally occurring uranyl phosphates from the Tarabau occurrence in the Nisa deposit, located in central-eastern Portugal, are studied with X-ray diffraction, Electron Microprobe and Scanning Electron Microscopy, with the purpose of i) identifying uranyl phosphate mineral paragenesis, ii) assessing chemical homogeneity and stoichiometry of the most relevant phases and iii) unraveling possible textural features of mineral reequilibration processes. The observations here presented follow prior field mapping and ore assessment works undertaken in the area of occurrence.

The obtained results allowed us to decipher mineral reequilibration reactions affecting the studied samples. Thus, reactive paths involving the replacement of Cu-bearing by Ba-bearing uranyl phosphates, cation-bearing uranyl phosphate by cation-depleted uranyl phosphate and cation-bearing uranyl phosphate by Fe, Al oxy-hydroxides have been defined. However, the studied textural features point towards two different mechanisms of mineral replacement, with superimposed expressions. On one hand, the replacement of Cu by Ba uranyl phosphate phases, and later by oxy-hydroxides, takes place by coupled dissolution-precipitation reactions. On the other hand, cation depletion affecting uranyl phosphates occurs by a cation exchange process, possibly giving rise to increasing mineral porosity. From the gathered information we also conclude that the widely accepted reactive path models concerning the UO_2-Fe(III)-H_2O system need to be reconsidered, aiming at a more accurate understanding of U(VI) mobility in the environment.

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