The new K, Pb-bearing uranyl-oxide mineral kroupaite: Crystal-chemical implications for the structures of uranyl-oxide hydroxy-hydrates

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ABSTRACT

Kroupaite (IMA 2017-031), ideally K Pb [(UO2)3(OH)]·10H2O, is a new uranyl-oxide hydroxyl-hydrate mineral found underground in the Svornost mine, Jáchymov, Czechia. Electron-probe microanalysis (WDS) provided the empirical formula (K1.28Na0.07Pb0.23Cu0.14Ca0.05Bi0.03Co0.02Al0.01)·10H2O, on the basis of 40 O atoms apfu. Sheets in the crystal structure of kroupaite adopt the fourmariereite anion topology, and therefore kroupaite belongs to the schoepite-family of minerals with related structures differing in the interlayer composition and arrangement, and charge of the sheets. Uptake of dangerous radionuclides (90Sr or 131Cs) into the structure of kroupaite and other uranyl-oxide hydroxy-hydrate is evaluated based on crystal-chemical considerations and Voronoi-Dirichlet polyhedra measures. These calculations show the importance of these phases for the safe disposal of nuclear waste.

Keywords: Kroupaite, new mineral species, uranyl-oxide hydroxy-hydrate, crystal structure, Voronoi-Dirichlet polyhedral, 131Cs; 90Sr; Jáchymov

INTRODUCTION

Uranium dioxide, as nuclear fuel or uraninite (Janečzek et al. 1999), UO2+x, readily reacts with oxygen and water to form a fascinating family of uranyl-oxide hydroxy-hydrates (UOHs) (Finch and Ewing 1991; Wronkiewicz et al. 1996; Plášil 2014). These occur in nature as minerals and are among the first alteration products that form during weathering of uraninite in oxidized zones of U deposits worldwide (Finch et al. 1996a, 1996b; Plášil 2018a). They are of interest in forensic studies of intercepted illicit nuclear materials as they provide insights into uranium oxide history. Several studies have focused on their structures, solubilities, and thermodynamic stabilities (Casas et al. 1997; Finch and Murakami 1999; Kubatko et al. 2006a; Klingensmith et al. 2007; Gorman-Lewis et al. 2008; Kirkegaard et al. 2019), due to their general importance in nuclear waste disposal and the environmental chemistry of uranium.

The onset of oxidation and hydration of uranium oxide often yields phases consisting of electroneutral sheets of uranyl pentagonal bipyramids with substantial H2O in the interlayer region, and typically little, if any, additional metal cations (Burns 2005; Krivovichev and Plášil 2013; Lussier et al. 2016; Plášil 2018a). In some cases, uranium oxide hydrates containing mixtures of U(IV), U(V), or U(VI) oxidation states occur as well. Continued alteration of uraninite oxide, alteration in more chemically diverse aqueous fluids, and alteration of geologically old uranium oxide that contains substantial radiogenic lead result in formation of uranyl-oxide hydroxy-hydrates with anionic sheets of uranyl polyhedra charge-balanced by cations in the interstitial regions of the structures.

Here we describe the new mineral kroupaite that was found underground in the Svornost (formerly “Einigkeit” in German) mine in Jáchymov, Western Bohemia, Czechia. Details of the mineralogy, geology, and history of the Jáchymov ore district can be found elsewhere (Ondruš et al. 2003; Hloušek et al. 2014). The specimens studied originate from the Jan Evangelista vein at the Daniel level of the Svornost mine. Associated minerals include fourmariereite, Na-rich metaschoepite, uranoplite, liebigite, ewingite, and gyspum. The new mineral and its assemblage are of supergene origin associated with oxidation-hydration alteration of uraninite in old mine workings. The name honors mining engineer Gustav Kroupa (1857–1935), who was employed by the state mines in Jáchymov beginning in 1886, and who as head of the mining district, approved shipment of 10 tons of leachate obtained from processing pitchblende ore to Marie-Curie Sklodowska and Pierre Curie in 1898. They went on to isolate three grams of the new substance radium chloride, and subsequently the new element radium. The Commission on New Minerals, Nomenclature and Classification of the International Mineralogical Association (IMA) approved the new mineral and name (IMA 2017-031). The description is based upon the holotype specimen deposited in the mineralogical collection of the National Museum in Prague (catalog number no. PIP 16/2017).