Clarkeite: New chemical and structural data

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ABSTRACT

Clarkeite crystallizes during metasomatic replacement of pegmatitic uraninite by latestage, oxidizing hydrothermal fluids. Samples are zoned compositionally: Clarkeite, which is Na rich, surrounds a K-rich core (commonly with remnant uraninite) and is surrounded by more Ca-rich material; volumetrically, clarkeite is most abundant. Clarkeite is hexagonal (space group $R\bar{3}m$) a = 3.954(4), c = 17.73(1) Å (Z = 3). The structure of clarkeite is based on anionic sheets of the form $[(UO_2)(O,OH)_2]$. The sheets are bonded to each other through interlayer cations and H₂O molecules. The empirical formula for clarkeite from the Fanny Gouge mine near Spruce Pine, North Carolina, is:

 $\{Na_{0.733}K_{0.029}Ca_{0.021}Sr_{0.009}Y_{0.024}Th_{0.006}Pb_{0.058}\}_{\Sigma 0.880}[(UO_2)_{0.942}O_{0.918}(OH)_{1.082}](H_2O)_{0.069.}$

Na predominates and the Pb is radiogenic. The general formula for clarkeite is

 $\{(Na,K)_{p}M_{q}^{2+}M_{r}^{3+}M_{s}^{4+}Pb_{x}\}[(UO_{2})_{1-x}O_{1-y}(OH)_{1+y}](H_{2}O)_{z}$

where Na >> K and p > (q + r + s). The number of O²⁻ ions and OH groups in the structural unit is determined by the net charge of the interlayer cations (except Pb): y = 1 - (p + 2q + 3r + 4s). This suggests that the ideal formula for ideal end-member clarkeite is Na[(UO₂)O(OH)](H₂O)₀₋₁. The structural sheets are destabilized as U decays to Pb (increasing *x*), and Pb enters vacant interlayer cation sites. Clarkeite eventually recrystallizes to lead uranyl oxide hydrates such as wölsendorfite or curite.