

Crystal chemistry and radiation-induced amorphization of P-coffinite from the natural fission reactor at Bangombé, Gabon

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ABSTRACT

Phosphorous-rich coffinite, $U(Si,P)O_4 \cdot H_2O$, from the natural nuclear reactor at Bangombé, Gabon (depth 12.25 m), has been examined as an important primary mineral and alteration product of uraninite under reducing conditions. Based on electron microprobe analyses (EMPA) and textural relationships, two distinct types of coffinite have been identified: (1) P-coffinite-(i) [with P_2O_5 and $(REE+Y)_2O_3$ as high as 9.84 and 8.66 wt%, respectively] replaces uraninite and has a chemical formula of $(U_{0.84 \pm 0.05} Y, REE_{0.12 \pm 0.02} Ca_{0.10 \pm 0.02} Th_{0.003 \pm 0.002})_{\Sigma 1.07 \pm 0.06} (Si_{0.41 \pm 0.07} P_{0.34 \pm 0.03} S_{0.08 \pm 0.05})_{\Sigma 0.84 \pm 0.05} O_4$ and; (2) coffinite-(ii), lacks uraninite inclusions [with P_2O_5 and $(Y+REE)_2O_3$ up to 1.45 and 1.79 wt%, respectively] and has a chemical formula of $(U_{0.78 \pm 0.02} Ca_{0.05 \pm 0.003} Y, REE_{0.03 \pm 0.01} Th_{0.002 \pm 0.001})_{\Sigma 0.87 \pm 0.02} (Si_{1.02 \pm 0.02} P_{0.06 \pm 0.01})_{\Sigma 1.08 \pm 0.01} O_4$. The EMPA elemental maps reveal a homogeneous distribution of P, Si, Nd, and U in P-coffinite-(i). Charge-balance calculations indicate that S substitution for Si in the coffinite structure results in the enhancement of the incorporation of P and REEs. High EMPA totals (95–100 wt%), suggest that water is not an essential component of the coffinite structure. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) reveal the decomposition of single crystals of uraninite into 50 nm grains during the alteration process to P-coffinite-(i). High-resolution transmission electron microscopy (HRTEM) and selected area electron diffraction (SAED) patterns confirm that P-coffinite-(i) has the coffinite structure. However, P-coffinite-(i) has a large amorphous fraction probably due to radiation damage caused by α -decay events. The calculated cumulative dose for both types of coffinite varies from 1.6 – 1.9×10^{18} (α -decay events/mg), which is equivalent to 134.2–161.5 displacement per atom (dpa) that have accumulated most probably during the past 800 million years.

Keywords: Phosphatian coffinite, coffinite, uraninite, natural fission reactor, Bangombé