Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.35393

Zeolite - hydroxylapatite mineral mixture as a novel geochemical barrier for Th immobilization

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Bentonite is widely used as a backfill material in nuclear waste repositories because of its favorable physicochemical properties. However, it exhibits limitations under certain geochemical conditions. Consequently, alternative or supplementary materials, including zeolites, remain the subject of investigation.

We propose a novel reactive barrier material composed of Pb-modified clinoptilolite and hydroxylapatite ($Ca_5(PO_4)_3OH$). The zeolite serves as a controlled, environmentally safe source of Pb^{2+} [1], whereas hydroxylapatite provides PO_4^{3-} through partial dissolution. These components promote the formation of low-solubility lead phosphate, pyromorphite ($Pb_5(PO_4)_3Cl$). Pyromorphite (Pym) is a structurally tolerant lead apatite capable of incorporating various trace elements, including radionuclides such as U and Th [2, 3].

Pb-modified zeolite was prepared by sorption of Pb^{2+} onto natural clinoptilolite. Sorbed Pb exhibits optimal binding strength. It is sufficiently stable to prevent Pb desorption to solution, yet reactive enough to participate in phosphate-driven precipitation reactions. The Pb-zeolite was mixed with synthetic hydroxylapatite (HAP). We hypothesized that exposure of this mixture to Th- and Cl-bearing solutions would induce precipitation of Th-bearing Pym directly on zeolite surfaces, resulting in Th immobilization:

$$Pb^{2+}_{\text{desorbed from zeolite}} + PO_4^{3-}{}_{\text{HAP dissolution}} + Th^{4+}_{\text{contamination}} + NaCl_{\text{supplied}} = (Pb,Th,Na)_5(PO_4)_3Cl_{\text{precipitated}}$$

Batch experiments were conducted at pH 5 and 8 under both static and dynamic conditions. A mixture consisting of 0.5 g Pb-zeolite (70 mg Pb/g) and 0.02 g of synthetic HAP was reacted with 50 mL of solution containing 50 ppm Th. The reaction proceeded as predicted. Thorium removal was rapid and efficient. Complete Th removal was achieved within hours consuming only one third of supplied Pb and PO_4. The reaction produced acicular crystals of Th-rich Pym on zeolite surfaces as seen on Fig. 1. As a reactive geochemical barrier, the tested material may serve as an effective supplement to bentonite backfill materials.

This research was supported in part by NCN grant no. 2024/55/B/ST10/01958.

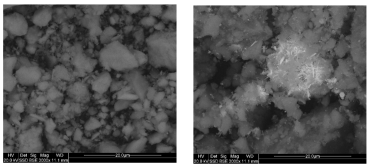


Fig. 1. Mineral mixture prior to reaction (left) and after reaction (right). White acicular crystals of Th-bearing Pym precipitated on the surface. SEM/BSE imaging.

[1] Stępień et al. (2025) *Mineralogia* 56, 44-51. DOI: 10.2478/mipo-2025-0006

[2] Sordyl et al. (2024) *Polyhedron* 252, 116891. DOI: 10.1016/j.poly.2024.116891

[3] Sordyl et al. (2023) *App Geochem* 158, 105780. DOI: 1016/j.apgeochem.2023.105780