



## **Oriented overgrowth of pharmacolite (CaHAsO<sub>4</sub>·2H<sub>2</sub>O) on gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O)**

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At neutral pH and 25°C the interaction of arsenate-bearing aqueous solutions with gypsum results in surface precipitation of pharmacolite (CaHAsO<sub>4</sub>·2H<sub>2</sub>O) crystals. The crystals grow oriented onto the gypsum surface, forming an epitaxy. Using an *A*-centered unit-cell setting for both pharmacolite (*Aa*) and gypsum (*A2/a*), the epitaxial relationship is found to be (010)<sub>Gy</sub>||(010)<sub>Ph</sub> and [101]<sub>Gy</sub>||[101]<sub>Ph</sub>. Pharmacolite forms thick three-dimensional crystals elongated on [101] with {010}, {111}, and {111} as major forms. Both the crystal morphology and the epitaxial orientation are interpreted on the basis of the bond arrangement in the structure of both phases. The reaction can be envisaged as a sort of solvent-mediated replacement of gypsum by pharmacolite. Under these experimental conditions the process stops at a “pseudo-equilibrium” endpoint in which the reactive solids become completely isolated from the aqueous solution by the epitaxial coating of pharmacolite crystals. The thermodynamic solubility product of pharmacolite was been determined at this stage and found to be  $pK = 4.68 \pm 0.04$ . The reaction paths actually followed by the system and the “true equilibrium” endpoint are modeled using the geochemical code PHREEQC.