



## Experimental model of cerussite $\text{PbCO}_3$ replacement by mimetite $\text{Pb}_5(\text{AsO}_4)_3\text{Cl}$ at pH 2 – 8

Ewa Stępień and Maciej Manecki

AGH University of Science and Technology, Department of Mineralogy, Petrography and Geochemistry, Kraków, Poland  
(estepien@agh.edu.pl)

The mobility of arsenic in aquatic environments is controlled by oxidation states of arsenic, stability of solid phases, and chemical composition of water (Meng et al., 2002). Binding of arsenic in the environment may occur through precipitation of low-solubility salts (Magalhães, 2002), like mimetite  $\text{Pb}_5(\text{AsO}_4)_3\text{Cl}$ . The aim of this study is to experimentally investigate reactions between cerussite ( $\text{PbCO}_3$ ) and solutions containing  $\text{AsO}_4^{3-}$  at various conditions favouring mimetite formation. These observations may provide a new recognition for As immobilization, which might be relevant in remediation of contaminated natural waters.

The mechanism of cerussite reaction with arsenate solutions (50 mg As/L) was studied at pH 2 – 8 using synthetic cerussite powder and fragments of natural cerussite crystals (Mibladen, Morocco). The reaction was carried out by direct contact of 500 ml of As-containing solution with  $\text{PbCO}_3$ , in presence of  $\text{Cl}^-$  ions. Cerussite was reacted for up to 4 weeks at *in situ* and *ex situ* setups. X-Ray Diffraction (XRD), Scanning Electron Microscopy with Energy Dispersive Spectrometry (SEM-EDS) and Electron Microprobe Analysis (EMPA) were used for analysis of the solid products of the experiments. The solutions were tested for Pb with Atomic Absorption Spectroscopy (AAS) and for  $\text{AsO}_4^{3-}$  using colorimetry.

Precipitation of mimetite on cerussite powder crystals is observed already after 1 day of the reaction with arsenate solutions, at the whole range of pH. Mimetite forms hexagonal rods or needles less than 1  $\mu\text{m}$  in size precipitating in the form of incrustations on  $\text{PbCO}_3$  crystals. Their size depends on the pH: a fine-grained precipitate forms at higher pH. Observations of natural crystals show replacement of cerussite by polycrystalline mimetite crust. The crust made of columnar and needle crystals is porous allowing for solution penetration and progress of the reaction. The replacement features indicate similarity to pseudomorphic reactions, and the mechanism elucidated as interface coupled dissolution - precipitation. Overall, cerussite replacement by mimetite reduces  $\text{AsO}_4^{3-}$  concentration from 50 ppm to below 1 ppm. It also depends on the pH.

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