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Effect of pH on crystallization of lead bearing phases on gypsum at ambient conditions.

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Pollution of surface waters and groundwaters by Pb2+ and other heavy metals is a common feature in a variety of geological settings due to both, natural processes and human activities. The bioavailability of metal contaminants is strongly dependent on the type of minerals that are most abundant in those settings because the interaction of their surfaces with polluted waters give raise to sorption processes that sequester metals and reduce their concentration in the aqueous phase. Calcium sulphate minerals, namely gypsum and anhydrite, are highly abundant in sedimentary basins. Dissolution-crystallization processes occurring as a result of the interaction between these sulphates and Pb-bearing waters are involved in the sequestration of this pollutant and their development has been previously studied in the pH range between 4.75 and 6.8 [1]. Much less attention has been paid so far to the development of these processes in highly acidic environments, where the weathering of Pb-bearing minerals is accelerated and the availability and mobility of dissolved Pb is likely to be increased. Here, we present the results of interaction experiments between dissolved Pb and gypsum surfaces at acidic to slightly basic pHs (2.5, 5.5 and 7.9) during increasing reaction times (from 5 minutes to 24 hours) at atmospheric conditions. The main goals of this work are (a) to analyse the physicochemical evolution of the system and (b) to identify and characterize the neo-formed phases that can effectively reduce Pb bioavailability.

SEM imagining of gypsum surfaces after short interaction with dissolved Pb2+ evidence the simultaneous development of etch pits and the formation of newly formed secondary phases. The chemical evolution of the aqueous phase, which is characterised by a rapid decrease of Pb2+ concentration and a parallel increase of Ca2+ concentration, is consistent with the concomitance of gypsum dissolution and Pb-bearing phases precipitation. The nature of the Pb-bearing phases is strongly influenced by the initial pH of the aqueous solution. Thus, anglesite ($PbSO_4$) is the only phase that precipitates under acidic conditions (pH = 2.5 and 5.5) as deduced from both XRD and Raman spectroscopy analyses. Anglesite crystals initially nucleate randomly oriented on gypsum surface, quickly carpeting large areas. Anglesite crystals initially appear as tabular rhombus-shaped single crystals that evolve towards elongated along [100] crystals with a "double arrow" habit as growth proceeds. In contrast, under alkaline conditions Pb-bearing precipitates consist of nanometric to micrometric sized flake-like particles. The nature of these particles is unclear. We discuss the influence of pH in the mineralogy of neo-formed Pb-bearing phases as well as the morphological evolution of anglesite on the basis of physicochemical and crystal chemical considerations.

[1] Astilleros, J.M., Godelitsas, A., Rodríguez–Blanco, J.D. Fernández–Díaz, L., Prieto, M, Lagoyannis, A. and Harissopulos, S. (2010). Applied Geochemistry, 25, 1008-1016.