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Interaction of dissolved lead with calcite in the pH range between 2.5 and 7.9.

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Fresh water pollution by heavy metals is an important threat to human health in many regions in the world. The concentration of heavy metals in fresh water, as well as their mobility, can be reduced as a result of the development of dissolution - crystallization reactions that take place as fresh waters interact with mineral surfaces. Pb is one of the heavy metals that is most widespread in aqueous environments worldwide and whose effects on human health is most negative. Calcium carbonate minerals, namely calcite and aragonite, are main constituents of chemical sedimentary rocks. The interaction of calcite and aragonite crystal surfaces with Pb-bearing aqueous solutions plays a major role in controlling both, Pb migration and accumulation and, as result, contribute to define Pb bioavailability in natural setting where sedimentary carbonates are abundant.
Godelitsas et al [1] studied the dissolution-crystallization reactions that result from the interaction of calcite and aragonite with aqueous phases that had Pbaq contents lower than 1000 ppm and whose pH varied between 4.75 and 6.8. In comparison, Pb sorption processes by calcium carbonate minerals that take place when pH conditions are more acidic, and/or the concentration of this pollutant is significantly higher have hardly been explored. Aiming to contribute to fill this gap, in this work we focus on calcite and investigate its interaction with Pb-highly concentrated aqueous solutions in the pH range between 2.5 and 7.9 by conducting macroscopic batch-type experiments at ambient conditions. Analysis of the chemical evolution of the aqueous phase indicates that its interaction with calcite results in a rapid removal of Pb when the aqueous solution initial pH was higher than 2.5. SEM observations on calcite crystal surfaces evidence that under these pH conditions Pb removal is the consequence of the coupling between calcite dissolution and the precipitation of secondary phases precipitation. Crystals of these newly formed phases show two types of morphologies, prismatic and platy. Raman spectroscopy analysis confirms the former as cerussite ($PbCO_3$) and the latter as hydrocerussite ($Pb3(CO_3)2(OH)2$). Interestingly, during calcite interaction with Pb- bearing aqueous solutions at pH 2.5 the formation of secondary phases is only detected at the latter stages of the experiment. This is consistent with the chemical evolution of the aqueous solution, characterized by a progressive increase of Ca concentration that runs parallels to a pH increases during the entire period of interaction, while Pb remains constant over most of the experiment to only decrease at the end of reaction time. This evolution points to a continuous dissolution of calcite over the whole duration of the experiment at pH = 2.5, eventually accompanied by the precipitation of secondary phases after prolonged interaction. The different chemical evolution trends as well as the changes in the morphological characteristics of secondary phases observed as a function of the initial pH of the aqueous solution are interpreted taking into consideration thermodynamic and kinetic arguments.

[1] Godelitsas, A., Astilleros, J.M., Hallam K, Harissopulos, S. and Putnis A. (2003). Envion. Sci. Technol., 37, 3351-3360.