

MINERALS IN THE HUMAN BODY

Effect of oxalate and pH on chrysotile dissolution at 25 °C: An experimental study†

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ABSTRACT

The effect of pH on the kinetics of chrysotile dissolution was investigated at 25 °C in batch reactors over the pH range of 1 to 13.5, in oxalic solutions and buffered solutions of inorganic salts. Dissolution rates were obtained based on the release of Si and Mg. Results of the batch with inorganic buffers showed a strong dependence of dissolution rates on pH in the acid range. The logarithm of dissolution rates decreases with the pH with a slope of $n = 0.27$. Around neutral pH, a minimum is reached. From pH 8 to 12, rates increase again when pH increases, and follow a linear dependence with a shallow slope ($n = 0.06$). The Mg/Si ratio shows a non-stoichiometric dissolution reaction with a preferential release of Mg²⁺ at acidic pH; it decreases at neutral pH conditions according to Mg solubility. Our results suggest that the relative ease of the breaking of Mg-O bonds compared with Si-O bonds lead to dissolution via a series of steps involving Si and Mg, where Si release is the rate-limiting step.

In the presence of 15 mmol L⁻¹ oxalate, an intense catalytic effect from pH 1 to 6 is observed because of the capacity of the oxalate anion to form different complexes with Mg. The ratio of the rates derived from Mg and Si concentrations confirm an enhancement of non-stoichiometric dissolution compared with the series without oxalate. The mechanism of catalysis involves different processes depending on pH: At pH 1, XRD analysis confirms the formation of an amorphous silica phase dissolving all the Mg present in the chrysotile structure. At pH 2, XRD and FTIR results also confirm the precipitation of glushinskite, a magnesium oxalate phase. At pH 3 to 6, the presence of oxalate enhances dissolution almost by an order of magnitude compared with the experiments in inorganic buffered solutions. In this case, the mechanism could be due to the formation of aqueous or surface magnesium oxalate complexes. However, dissolution rates at neutral pH in the presence of oxalate are similar to those obtained in inorganic buffered solutions; the pH dependence at pH 8 to 13 is minimal. The increase in saturation and the drastic decrease in Mg solubility at these pH values could lead to precipitation of secondary phases coating the reactive mineral surface and inhibiting the surface.

Results obtained in this study show that chrysotile dissolves faster in acid media and oxalate acts as a strong catalyst increasing the efficiency of magnesium release to solution at ambient temperature. These data may provide an excellent background to design and select optimal conditions in the previous acid treatment for carbon capture processes, as well as help to develop remediation process of asbestos contaminated sites.

Keywords: Chrysotile, asbestos, dissolution, oxalate