

Experimental study of the interactions of CO₂-SO₂-O₂ mixtures with saline pore water and reservoir rocks at geological storage conditions

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CO₂-streams from industrial emitters usually contain SO_x, NO_x, O₂, N₂, Ar (among others), which during CO₂ sequestration in parts will be co-injected with CO₂ into the geological storage formation to limit energy consumption and reduce costs of the purification steps. These potential accessory gases in the CO₂-stream can have a significant impact on the chemical reactions and physical processes in the storage formation (e.g. a saline aquifer). Especially SO₂ is highly reactive, since it acidifies the formation water by forming sulfuric acid and initiates precipitation of anhydrite and barite, while other minerals may dissolve (calcite, dolomite, pyrite) [1, 2]. Precise knowledge of these processes is important to predict the extent of reactions on the long term storage capacity in modelling approaches. For this purpose, it is necessary to not only investigate equilibria of the geochemical reactions that take place during CO₂ storage, but also to determine the rates of fluid-mineral reactions and the dissolution rates of CO₂ and accessory gases into the formation water [3].

We have investigated the kinetics of dissolution of CO₂, CO₂-SO₂- and CO₂-SO₂-O₂ mixtures into water and saline brine under geological storage conditions (16 MPa, 333 K) in stirred all-titanium batch reactors. The results confirmed even for these conditions a significantly faster dissolution (or higher mass transfer) of SO₂ into water than CO₂. Analysis of the different sulfur species in the aqueous phase revealed that hydrated SO₂ is the dominant species when oxygen is absent. The disproportionation of sulfur dioxide to sulfate and sulfide is either very slow or did not occur at all at the conditions investigated.

Furthermore, we investigate the reaction rates of the above mentioned fluids and gases with a hematite containing redbed sandstone and size-fractionated hematite crystals, exhibiting a rapid reaction with the fresh hematite surfaces in the mineral separate, but only slow reactions with the hematite in the redbed sandstone.

[1] Pearce et al. (2015): *Chem. Geol.*, 399, 65-86

[2] Renard et al. (2014): *Chem. Geol.*, 382, 140-152

[3] Waldmann & Rütters (2016): *IJGGC*, 46, 116-135