1532 Goldschmidt Conference Abstracts

Reaction of SO₂ and SO₂ with O₂ after dissolution during benchtop experiments of CO₂ storage at elevated temperature and pressure

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Captured CO_2 streams from the burning of fossil fuels can contain impurities like SO_2 and O_2 as well as others. The presence of these impurities in CO_2 that is being geologically stored can result in significant impacts on the chemistry of the aqueous phase. Understanding how these changes in chemistry may affect the physical and chemical behaviour of the storage system is a vital component of the risk assessment process. Fundamental to that task is being able to correctly predict how the impurities react once dissolved in the water because the formation of strong acids and changes in redox state are expected to occur. When SO_2 dissolves in water there are several possible reactions that can occur:

$SO_{2(aq)}$ +	- H ₂ O E	$HSO_3 + H^+$	
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1)

2)

3)

- $SO_{2(aq)} + H_2O + \frac{1}{2}O_{2(g)} \Box H_2SO_4$
- $4SO_{2(aq)} + 4H_2O \square 3H_2SO_4 + H_2S$

1) is a simple hydration reaction that is reversible and can result in low pH. The second reaction involves oxidation (by O_2 or other oxidents) and results in lower pH than 1). 3) is a disproportionation reaction that will produce both oxidized and reduced sulphur species and will result in pH higher than 2) but lower than 1). The H_2S in 3) can be substituted for by elemental S as a product. A series of batch experiments were conducted to evaluate the effects of impurities on CO2 storage in sedimentary rocks from the Surat Basin, Australia (Farquhar et al., 2015; Pearce et al., 2015). The experiments were run using pure CO₂, CO₂ + 0.16% SO₂ and CO₂ + 0.16% SO₂ + 2% O2 at 12 MPa and 60°C. Here we report the observations regarding how SO_2 reacted during the experiments. With 0.16% SO₂ and no additional oxidant, disproportionation did occur with either S₀ or H_2S or both and very low pH. In the presence of O_2 , the SO2 was oxidized giving sulphuric acid and typically a lower pH than the experiments without O₂. During the 15 days of the experiment runs, it was clear there was a rate control on the SO2 oxidation or disproportionation reactions.

Farquhar et al. (2015) Chem. Geol. 399, 98-122; Pearce et al. (2015) Chem. Geol. 399, 65-86.