Controls on the Carbonation of Steel Slag

ELEANOR J. BERRYMAN[1*], ANTHONY E. WILLIAMS-JONES[1],
ARTASCHES A. MIGDISOV[1], SIEGER VAN DER LAAN[2]

[1]McGill University, Montreal, Canada,
berryman.eleanor@gmail.com(* presenting author)
[2]Tata Steel RD&T, IJmuiden, The Netherlands

Goals and Methodology

Mineral carbonation provides a robust method for permanent sequestration of CO_2 that is environmentally inert. Larnite (Ca_2SiO_4), the major constituent of steel slag, reacts readily with aqueous CO_2 [1]. Consequently, its carbonation offers an exciting opportunity to reduce CO_2 emissions at source [2]. A potential added benefit is that this treatment may render steel slag suitable for recycling. This study investigates the impact of temperature, fluid flux and reaction gradient on the dissolution and carbonation of steel slag, and is part of a larger study designed to determine the conditions under which conversion of larnite, and other calcium silicates, to calcite is optimized.

The experiments were conducted on 2 – 3 mm diameter steel slag grains supplied by Tata Steel RD&T. A CO_2-saturated aqueous fluid was pumped through a steel flow-through reactor containing these grains at a temperature between 120°C to 200°C; the fluid pressure was 250 bar. Fluid flux was varied between 0.8 and 6 $mL/min/cm^2$. The duration of experiments ranged from 3 to 7 days.

Results

The steel slag grains reacted with the CO_2-saturated aqueous fluid to form phosphorus-bearing Ca-carbonate phases. At high fluid flux, these phases dissolved at the edges of grains, leaving behind a porous aluminium and iron oxide rind. Increasing temperature increased the rate of this reaction. At low fluid flux, carbonates precipitated on the slag grain surface inhibiting further reaction. In contrast, at intermediate fluid flux, dissolution of the primary Ca-bearing minerals was balanced by precipitation of carbonate phases, thereby optimising carbonation of the steel slag.

Conclusions

These results of this study show that carbonation of steel slag by aqueous CO_2 is feasible using relatively large grains and that it can be optimised by varying fluid flux. Experiments of the type described above will contribute to the eventual global reduction of industrial CO_2 emissions.

[1] Santos et al. (2009) *Journal of Hazardous Materials* **168**, 1397-1403.
[2] Berryman et al. (2011) *Mineralogical Magazine* **73(3)**, 522.

CO_2 attack of a caprock-type argillite: From lab experiments to modeling

GUILLAUME BERTHE[1,2*], SEBASTIEN SAVOYE[3],
CHARLESWITTEBROODT[1], JEAN-LUC MICHELOT[2]

[1]IRSN, DEI/SARG/LR2S,92260 Fontenay-aux-Roses, France
[2]IDES,CNRS-Université Paris-Sud F-91405 Orsay, France
[3]CEA, DEN/DANS/L3MR, F-91191 Gif-sur-Yvette, France
guillaume.berthe@cea.fr (*presenting author)

Introduction

In the case of a CO_2 storage in geological media, the understanding of the behavior of the caprock encountering an acid fluid is crucial. A set of twenty "dynamic" through-diffusion experiments was performed to investigate the changes in chemical and physical properties of a caprock-type argillite.

Methods

The "dynamic" through-diffusion setup enables, during the CO_2 attack, (i) a monitoring of the ion concentrations in the upstream and the downstream reservoirs and, (ii) an estimate of the transport property change, using HTO, HDO, ^{36}Cl and Br^- as tracers [1]. The impact of several parameters on the caprock behavior was studied, such as the amount of carbonate minerals, the sample thickness, the upstream volume, the presence or not of calcite-filled fracture and the bedding plane orientation regarding the diffusion.

Results and Discussion

The results showed that the extent of the reaction front, lowering the caprock confinement properties, would be more related to the initial rock transport properties and the calcite texture than the amount of carbonate minerals.

This large set of experimental data acquired, allowed us to identify from the chemistry-transport modeling some key parameters, such as the kinetic rate equations used for describing the dissolution/precipitation reactions (Figure 1).

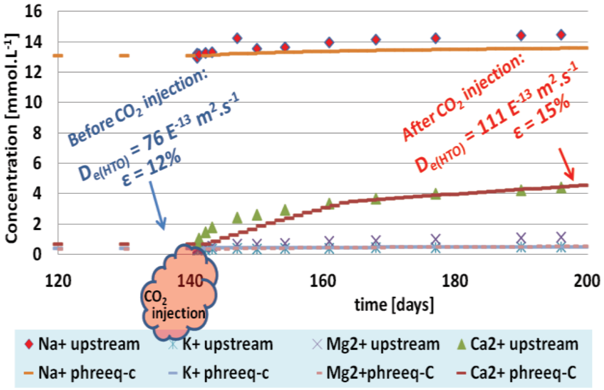

Figure 1: Comparison of experimental concentrations measured in the upstream reservoir with the simulated ones obtained by means of PHREEQC using rate equations from [2].

[1] Savoye *et.al.* (2010) *Environ. Sci. Techn.* **44**, 3698-3704.
[2] Palandri *et al* (2004) USGS, open file report 2004-1068, 64 pp.