**Numerical studies of CO2 capture by enhanced weathering of carbonate minerals**

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**Highlights**

* Weathering of calcite is used to model CO2 capture in an aqueous environment.
* Intrinsic kinetics of calcite dissolution is coupled with mass transfer processes.
* Passive absorption and reinforced bubbling of CO2 were numerically compared.
* Mass transfer of CO2 from air to aqueous solution can be the rate-limiting step.

**1. Introduction**

The target of restricting global mean temperature rise to around 1.5 C will require draw-down of some 810 Gt CO2 from the atmosphere by 2100 according to climate models [1]. The capture and storage of atmospheric carbon dioxide could thus become a very large industry later this century. Various methods and technologies have been suggested for this process, and have been recently reviewed [2,3]. The weathering of natural minerals like carbonate and silicate offers one possible route. The chemistry involves dissolution of CO2 in water and subsequent reaction to bicarbonate and carbonate as balancing cations are solubilised from the rock. However, in order to play a significant role, this very slow natural process needs to be dramatically accelerated, affordably and without causing unacceptable environmental damage. A key aspect of designing and assessing a suitable reaction scheme and equipment is to identify the rate-limiting step in this three-phase system, so that further enhancement measures can be introduced accordingly.

**2. Methods**

In this work, we consider two reactor schemes with respect to gas supply. One uses passive diffusion of CO2 at the air-water interface, the other is by active supply through gas bubbling. The three-phase bubble reactor provides greater gas/liquid contact area, and higher rates of CO2 absorption [4]. The effect of mass transfer rate of CO2 from atmosphere to aqueous solution on the overall reaction rate was numerically studied for the two schemes. Calcite (CaCO3) is used as the model mineral for the calculations due to its relatively fast weathering rate. The reaction scheme and all chemical rate constants are taken from the work of Plummer et al [3].

Calcium ions are formed at the surface of the mineral particles, at a rate which depends on the composition of the bulk water phase. CO2 which is transferred into the water through the gas/liquid interface is ionized to bicarbonate and carbonate. The kinetic scheme accounts for the presence of H+, OH-, HCO3-, CO32-, in addition to physically dissolved CO2 and Ca2+. A charge balance is maintained. Diffusion of CO2 from gas to liquid takes account of resistance in both gas and liquid phase, using the two-film theory.

**3. Results and discussion**

   **Figure 1.** Dependence of CO2(aq) conc on  **Figure 2.** Effect of gas rate on CO2(aq) conc

Fig. 1 shows the concentration of aqueous CO2 in the passive diffusion case. As the mass transfer coefficient increases, the aqueous CO2 concentration approaches the equilibrium level at a steadily faster rate. This suggests that in this case the rate of absorption of CO2 is the limiting factor, and not the rate of dissolution of mineral. The grinding of rock to create small particles, and thus high particulate surface area is an expensive process. It is thus important to establish, for any particular mineral and reactor configuration, what particle size is required. Fig. 2 compares the two CO2 supply modes, passive diffusion and bubbling, and shows, as expected that gas bubbling improves the mass transport rate and leads to a faster process. In practice, given the very large scale of reactors that will be needed, the improved rate of absorption will have to be balanced against the extra cost of air compression for bubble injection. Further calculations, including a trickle-flow scheme that will be matched with experiment, are planned.

**4. Conclusions**

Calculations of CO2 mass transfer rates in the absorption of atmospheric CO2 during mineral weathering show that in passive diffusion it can be the rate-limiting step, rather than chemical reaction or mineral dissolution. An enhanced weathering reactor with air bubbling significantly improves the mass transport, but at the cost of greater cost and complexity. The mass transfer and mineral reaction model developed will be useful for scoping other, novel reactor designs.

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