

Aqueous mineral carbonation as a possible CO₂ sequestration process

Energetic efficiency and costs

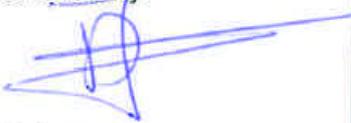
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Abstract

Aqueous mineral carbonation is a potentially attractive sequestration technology to reduce CO₂ emissions. In this paper, the energy consumption and costs of this technology were assessed using either wollastonite (CaSiO₃) or steel slag as feedstock. The major energy-consuming process steps were found to be the grinding of the feedstock and the compression of the CO₂. Within ranges of experimentally investigated process conditions, optimum energetic CO₂ sequestration efficiencies were 79 and 74% for wollastonite and steel slag, respectively. It was shown that the energetic performance for both feedstock might be improved up to >90% by e.g. further grinding of the feedstock and reducing the amount of process water applied. At energetically optimized process conditions, a preliminary cost estimate was made of 93 and 66 €/ton CO₂ avoided for wollastonite and steel slag, respectively (sequestration costs excluding possible capture). For wollastonite, major costs were associated with the feedstock and the electricity consumption (51 and 20 €/ton CO₂ avoided, respectively). A sensitivity analysis showed that additional influential parameters with regard to the sequestration costs include the liquid-to-solid ratio applied in the carbonation reactor and the possible commercial value of the carbonated product.

Keywords

Carbon dioxide, sequestration, mineral carbonation, energy consumption, costs

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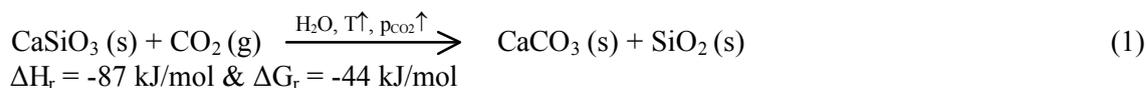
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1. Introduction

Mineral carbonation has been recognized as a potentially promising route for permanent and safe storage of carbon dioxide. Both the potentially large CO₂ sequestration capacity and the exothermic nature of the carbonation reactions involved have contributed to an increasing amount of research performed on mineral carbonation in recent years [1,2]. A number of different carbonation process routes has been reported, of which the one-step aqueous mineral carbonation route was selected as the most promising in a recent review [1], e.g. for wollastonite (CaSiO₃):



In earlier work, we have studied the aqueous carbonation mechanisms of both the industrial residue steel slag [3] and the mineral ore wollastonite [4]. It was shown that carbonation rates, sufficiently fast for industrial implementation, can be obtained by grinding the feedstock and performing the process at elevated temperature and CO₂ pressure (typically, <38μm, 200 °C and 10-40 bar CO₂, respectively). However, these measures to increase the carbonation rate consume energy and increase the sequestration costs. In this paper, we present a system study of an aqueous mineral carbonation process to determine the energetic performance and the CO₂ sequestration costs and to identify energetic and financial bottlenecks.

2. Methodology

A flowsheet of an aqueous mineral CO₂ sequestration process was designed (Figure 2.1) and simulated with ASPEN Plus flowsheet simulation software [5] to determine the composition and physical properties of the streams as well as the power and heat consumption of the process. The mineral carbonation plant was assumed to be located at the source of the solid feedstock. The wollastonite carbonation process was designed to sequester 60 ton/h CO₂ (i.e., the CO₂ emission of a 100 MW power plant, assuming 0.6 kg CO₂/kWh [6]). For steel slag, a significantly smaller carbonation process was assumed taking into account the relatively limited availability of steel slag (15 ton/h CO₂).

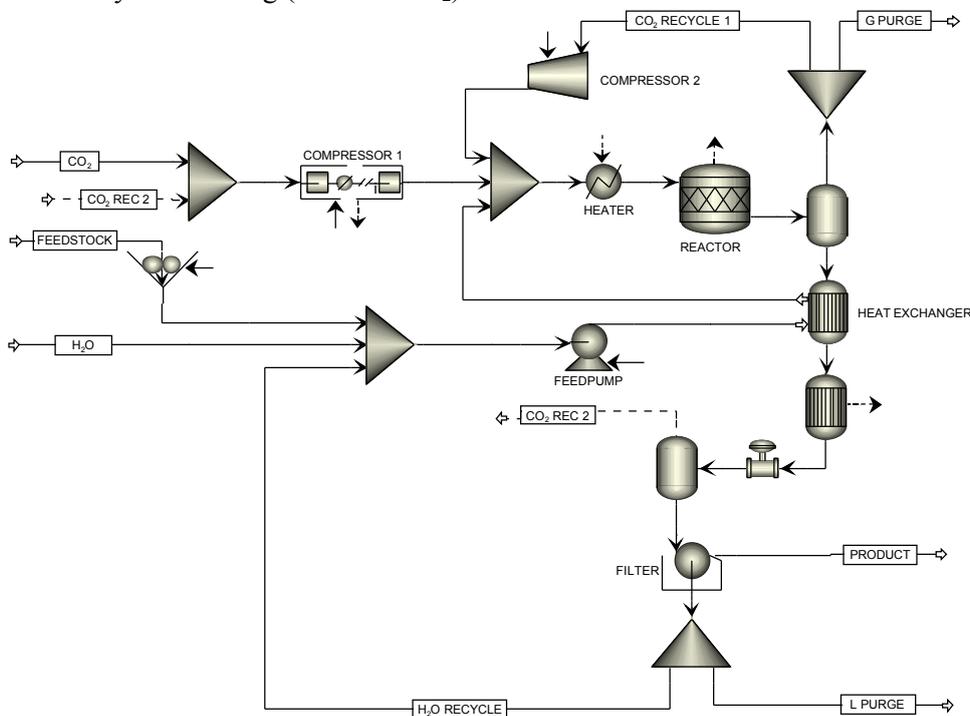


Figure 2.1 Simplified ASPEN flow diagram of an aqueous mineral carbonation process including power (\rightarrow) and heat ($- \rightarrow$) streams [6]

The energetic CO₂ sequestration efficiency (η_{CO_2}) (i.e., the fraction of the CO₂ that is sequestered effectively) was determined by correcting the amount of CO₂ sequestered in the carbonation reactor ($\text{CO}_{2\text{sequestered}}$) for the extra CO₂ emission caused by the energy consumption of the mineral carbonation process:

$$\eta_{\text{CO}_2} = \frac{\text{CO}_{2\text{avoided}}}{\text{CO}_{2\text{sequestered}}} = 1 - \frac{E_{\text{power}} \cdot \varepsilon_{\text{power}} + E_{\text{heat}} \cdot \varepsilon_{\text{heat}}}{\text{CO}_{2\text{sequestered}}} \quad (2)$$

The power consumption of the mineral carbonation process (E_{power}) consisted of power consumption for compression, pumping and grinding. The (possible) power consumption of the reactor and filter were neglected. The net heat consumption of the process (E_{heat}) consisted of the heat required for heating the reactants minus the reaction heat. As conversion factors of the power ($\varepsilon_{\text{power}}$) and heat ($\varepsilon_{\text{heat}}$) consumption into CO₂ emissions, 0.6 and 0.2 kg CO₂/kWh were used, respectively [6].

The energetic efficiency was calculated for various sets of process conditions, including particle size of the feedstock (d), temperature (T), CO₂ pressure (p_{CO_2}) and liquid-to-solid ratio (L/S) in

the carbonation reactor. The carbonation degree of the feedstock measured in a lab-scale autoclave reactor as reported earlier [3,4] was used as an estimate of the conversion in the full-scale continuous carbonation reactor. Thus, the influence of the process variables on the sequestration efficiency was studied and the energetic feasibility of *e.g.* grinding and heating was determined.

For the cost evaluation of the mineral carbonation process, the energetically optimum set of process conditions was taken and a basic design was made of the major process equipment within the carbonation process (Figure 2.1). The investment costs were estimated on the basis of publicly available literature with a factorial cost estimation method [7]. The sequestration costs were determined on the basis of the depreciation costs of the investments, the variable costs (*e.g.*, feedstock, utilities) and other fixed costs (*e.g.*, labour, maintenance). Subsequently, the sequestration costs were corrected for the energetic efficiency of the process to determine the net sequestration costs per ton CO₂ avoided.

Finally, a sensitivity analysis was performed in order to assess the accuracy of both the determined costs and energetic sequestration efficiency, as well as to identify routes for further improvement of the economic and energetic performance. For more details on the methodology and the assumptions used, see Huijgen et al. [6] (process design and assessment energetic efficiency) and Huijgen et al. [8] (estimation CO₂ sequestration costs).

3. Energetic Efficiency

Figure 3.1 shows an example of the calculated CO₂ sequestration efficiency (η_{CO_2}) and also illustrates the contribution of the reaction heat and the energy consumption for heating, grinding and compressing, for both wollastonite and steel slag. At these process conditions, steel slag shows a slightly higher efficiency than wollastonite due its higher carbonation degree [4,6]. The major energy-consuming process steps are the grinding of the feedstock and the compression of the CO₂.

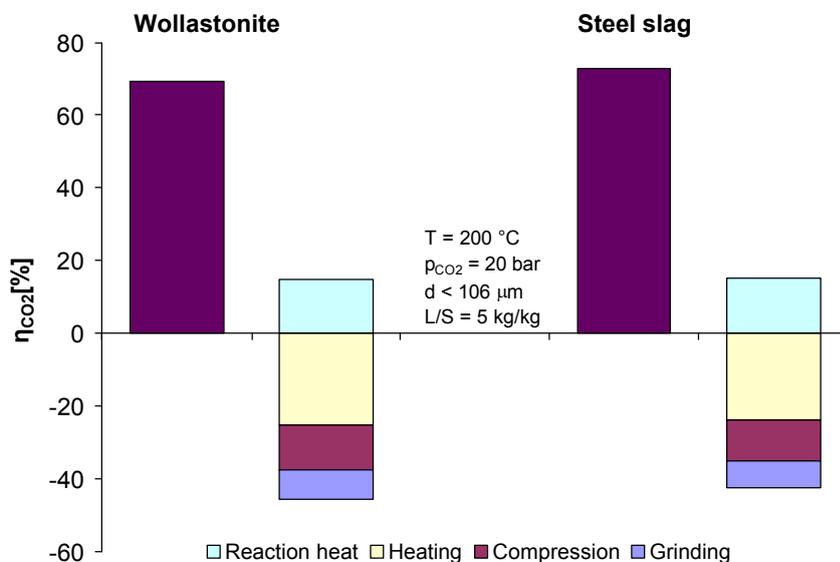
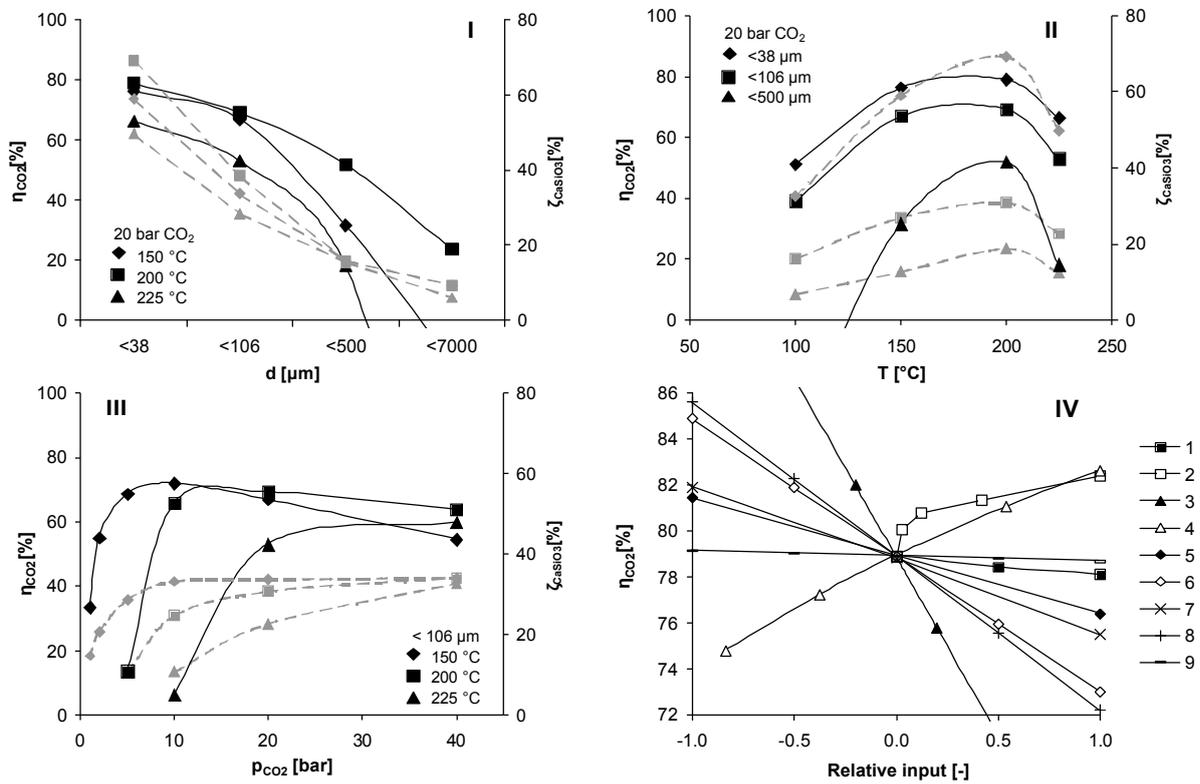


Figure 3.1 Energetic CO₂ sequestration efficiency (η_{CO_2}) for both a mineral ore (wollastonite) and an industrial residue (steel slag) at a selected set of process conditions [6]

The sequestration efficiency was optimised, within the ranges of process conditions studied, for the particle size, reactor temperature and CO₂ pressure. For wollastonite, the effect of these process variables on the sequestration efficiency is shown in Figure 3.2. It can be concluded that the additional amount of CO₂ sequestered at smaller particle sizes has a larger effect on the sequestration efficiency than the extra CO₂ emission caused by the grinding of the particles. Thus, grinding results in a net higher CO₂ sequestration efficiency. For both heating and compressing, an optimum reactor temperature and CO₂ pressure were found (i.e., 200 °C and 20 bar CO₂, respectively). The maximum sequestration efficiencies found within the range of experimentally investigated process conditions were 79 and 74% for wollastonite and steel slag, respectively.



#	Variable	Standard	Min	Max
1	Temperature H ₂ O recycle [°C]	40 (0)	40 (0)	80 (1)
2	Pressure H ₂ O recycle [bar]	1 (0)	1 (0)	34.5 (1)
3	L/S-ratio reactor feed [kg/kg]	5 (0)	2 (-0.6)	10 (1)
4	Carbonation degree (ζ_{CaSiO_3}) [%]	69 (0)	60 (-0.83)	80 (1)
5	Heat of reaction (ΔH_r) [kJ/mol]	84 (0)	64 (-1)	104 (1)
6	$\Delta T_{\text{heat-exchanger}}$ [°C]	20 (0)	10 (-1)	30 (1)
7	Working index for grinding [kWh/ton]	14 (0)	10 (-1)	18 (1)
8	ϵ_{power} [kg/kWh]	0.60 (0)	0.40 (-1)	0.80 (1)
9	ϵ_{heat} [kg/kWh]	0.20 (0)	0.15 (-1)	0.25 (1)

Figure 3.2 Carbonation degree (ζ_{CaSiO_3} ; ---) and CO₂ sequestration efficiency (η_{CO_2} ; —) as a function of particle size, temperature and CO₂ pressure ($L/S = 5$ kg/kg) for wollastonite. IV: Sensitivity analysis of CO₂ sequestration efficiency by wollastonite carbonation for selected parameters shown in the table at energetically optimum reactor conditions ($\zeta_{CaSiO_3} = 69\%$, $T = 200$ °C, $p_{CO_2} = 20$ bar, $d < 38$ μm) and $L/S = 5$ kg/kg. Numbers between brackets are the relative input values used for the graphs [6]

A sensitivity analysis (Figure 3.2) shows that the energetic performance might be optimised even further by, e.g., increasing the solid content of the slurry that is processed (e.g., $\eta_{CO_2} = 88\%$ for wollastonite at $L/S = 2$ kg/kg or 33 wt% solids). In addition, the amount of energy required to heat the reactor feed is reduced by lowering the liquid to solid ratio, which may result in a net surplus of process heat. Further research on carbonation in a continuous pilot-scale reactor is required to determine, among other parameters, the minimum L/S-ratio and the actual conversion in a continuous carbonation reactor.

4. Carbon Dioxide Sequestration Costs

Table 4.1 shows the results of a preliminary cost estimate made for both wollastonite and steel slag at the energetically optimized process conditions. Sequestration costs for steel slag are substantially lower than for wollastonite mainly due to the absence of costs for the feedstock. For wollastonite, major costs are associated with the feedstock and the electricity consumption. In the case of steel slag, the depreciation costs for the investments and the fixed costs are significantly higher than in the case of wollastonite due to the smaller scale of the designed process.

Table 4.1 *CO₂ sequestration costs of mineral carbonation (excluding possible costs of capture) [8]*

Costs [€/ton CO ₂]	Wollastonite		Steel slag	
	Sequestered	Avoided	Sequestered	Avoided
Depreciation investments	8	10	20	23
Feedstock	45	51	0	0
Utilities (electricity, water)	18	21	15	17
Fixed costs (labour, maintenance, etc.)	10	11	23	27
Sequestration costs	82	93	58	66

Figure 4.1 shows the influence of selected parameters on the CO₂ sequestration costs for wollastonite carbonation. The most influential parameters are the assumed feedstock price, the liquid-to-solid ratio applied in the carbonation reactor and the possible commercial value of the carbonated product.

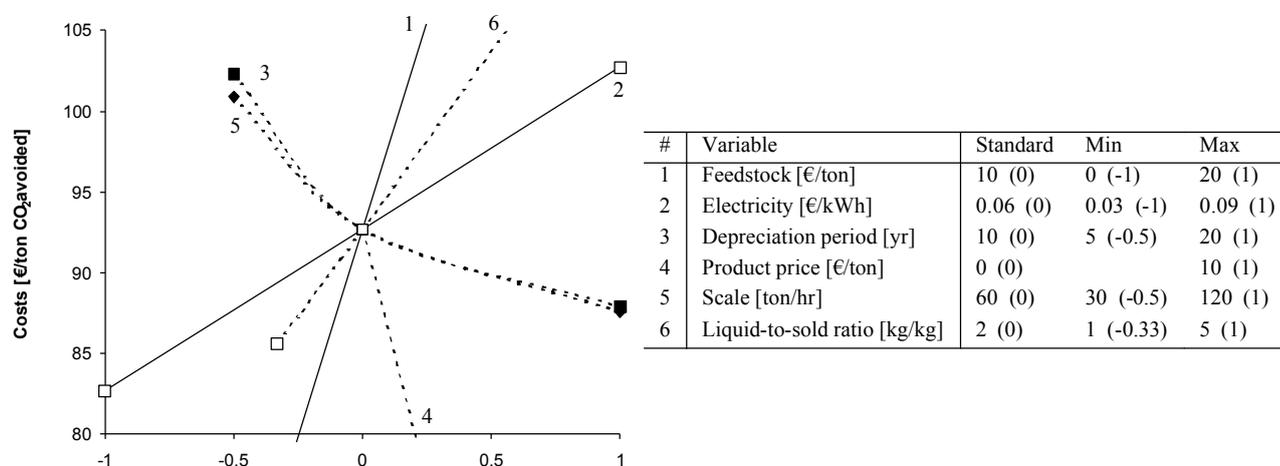


Figure 4.1 *Sensitivity analysis of CO₂ sequestration costs for wollastonite carbonation. The selected parameters are shown in the table [8]*

On the basis of the presented cost evaluation, large-scale CO₂ sequestration by state-of-the-art aqueous mineral carbonation seems relatively expensive compared to both other CO₂ storage technologies and (current) CO₂ market prices. However, niche applications of mineral carbonation with either a low-cost feedstock (e.g., a solid residue) or a carbonated product with a commercial value are economically more attractive.

5. Conclusions

The presented system study of an aqueous mineral carbonation process has shown that the measures taken to increase the carbonation rate (i.e., grinding, heating and compression of CO₂) were, in principle, energetically favourable. Within ranges of experimentally investigated carbonation conditions, the optimum energetic CO₂ sequestration efficiencies were found to be 79 and 74% for wollastonite and steel slag, respectively. The sequestration efficiency can be further improved to >90% by e.g. further grinding of the feedstock and increasing the solid content in the carbonation reactor.

At energetically optimized process conditions, sequestration costs of 93 and 66 €/ton CO₂ avoided were estimated for wollastonite and steel slag, respectively. State-of-the-art aqueous mineral carbonation seems a relatively expensive CO₂ sequestration technology compared to both other CO₂ storage technologies and (expected) CO₂ market prices. Further research on cost reduction should focus on the major costs within the process (e.g., in the case of wollastonite, the feedstock and electricity consumed with 51 and 20 €/ton CO₂ avoided, respectively). A sensitivity analysis showed that additional influential parameters on the sequestration costs include the liquid-to-solid ratio applied in the carbonation reactor and the possible commercial value of the carbonated product.

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