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Cost evaluation of CO_2 sequestration by aqueous mineral carbonation

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Abstract

A cost evaluation of CO_2 sequestration by aqueous mineral carbonation has been made using either wollastonite ($CaSiO_3$) or steel slag as feedstock. First, the process was simulated to determine the properties of the streams as well as the power and heat consumption of the process equipment. Second, a basic design was made for the major process equipment, and total investment costs were estimated with the help of the publicly available literature and a factorial cost estimation method. Finally, the sequestration costs were determined on the basis of the depreciation of investments and variable and fixed operating costs. Estimated costs are 102 and 77 €/ton CO_2 net avoided for wollastonite and steel slag, respectively. For wollastonite, the major costs are associated with the feedstock and the electricity consumption for grinding and compression (54 and 26 €/ton CO_2 avoided, respectively). A sensitivity analysis showed that additional influential parameters in the sequestration costs include the liquid-to-solid ratio in the carbonation reactor and the possible value of the carbonated product. The sequestration costs for steel slag are significantly lower due to the absence of costs for the feedstock. Although various options for potential cost reduction have been identified, CO_2 sequestration by current aqueous carbonation processes seems expensive relative to other CO_2 storage technologies. The permanent and inherently safe sequestration of CO_2 by mineral carbonation may justify higher costs, but further cost reductions are required, particularly in view of (current) prices of CO_2 emission rights. Niche applications of mineral carbonation with a solid residue such as steel slag as feedstock and/or a useful carbonated product hold the best prospects for an economically feasible CO_2 sequestration process.

Keywords: Carbon dioxide; Sequestration; Costs; Mineral carbonation; System study; Wollastonite; Steel slag

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1. Introduction

Carbon dioxide sequestration by mineral carbonation is a potentially attractive route to mitigate possible global warming on the basis of industrial imitation of natural weathering processes [1–3]. Potentially suitable feedstocks for mineral carbonation include Ca and Mg silicate ores, such as wollastonite ($CaSiO_3$) and olivine (Mg_2SiO_4), and industrial residues, such as steel slag and municipal solid waste incinerator bottom ash [2,4,5]. For example, the (overall) carbonation process for wollastonite can be given as:

$$CaSiO_3(s) + CO_2(g) \rightarrow CaCO_3(s) + SiO_2(s) \qquad (1)$$
$$\Delta H_r = -87\ \mathrm{kJ/mol}\ \text{and}\ \Delta G_r = -44\ \mathrm{kJ/mol}\ [6]$$

Benefits, compared to other CO_2 capture and storage (CCS) technologies (e.g. storage in depleted gas fields and oceans), include the permanent and inherently safe character of the CO_2 sequestration and its potentially vast storage capacity [2,3]. However, weathering processes are slow at natural conditions, and therefore, various approaches have been studied in order to increase the mineral carbonation rate such that the process can be implemented industrially [2,3]. One of the possible process routes reported is direct aqueous mineral carbonation in which carbonation takes place in a single process step in an aqueous suspension. The presence of water is known to accelerate the carbonation reaction

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Nomenclature

Symbol

A	heat exchanging surface area (m^2)
$CO_{2,avoided}$	net amount CO_2 avoided (kg/h)
$CO_{2,sequestered}$	amount CO_2 sequestered in reactor (kg/h)
$CO_{2,heat}$	extra CO_2 emission due to heat consumption (kg/h)
$CO_{2,power}$	extra CO_2 emission due to power consumption (kg/h)
d	particle size (m)
d	wall thickness (m)
D	diameter (m)
$D[4,3]$	volume based mean particle size (m)
H	height (m)
L/S	liquid-to-solid ratio (kg/kg)
n	stirring rate (rpm)
p	pressure (bar)
p_{CO_2}	CO_2 partial pressure (bar)
Q	heat (W)
T	reactor temperature (°C)
t	reaction time (min)
U	heat transfer coefficient (W/m^2K)

Greek characters

Φ	flow rate (m^3/h)
ΔT_{lm}	logarithmic temperature difference (°C)
ΔG_r	Gibbs energy of reaction (kJ/mol)
ΔH_r	reaction enthalpy (kJ/mol)
ΔP	pressure drop (bar)
ΔT	temperature difference heat exchanger (°C)
ζ_{CaSiO_3}	conversion of $CaSiO_3$ fraction in the reactor inlet (%)
η_{CO_2}	energetic CO_2 sequestration efficiency (%)

Super and subscripts

g	gas
l	liquid

[2]. In previous papers [7,8], we have experimentally studied the aqueous carbonation of the Ca silicates, steel slag and wollastonite, in a laboratory scale autoclave reactor in dependence of the major process conditions, i.e. reactor temperature (T), partial CO_2 pressure (p_{CO_2}), reaction time (t), particle size of feedstock (d), liquid-to-solid ratio (L/S) and stirring rate (n). It has been shown that these Ca silicates can be carbonated substantially within industrially realistic reaction times ($\leqslant$30 min) by finely grinding the feedstock (typically, <38 *μ*m) and moderately elevating the reactor temperature (150–200 °C) and CO_2 pressure (10–40 bar) [7,8]. The potentially costly use of additives and a CO_2 pressure over 100 bar, as typically applied to enhance the carbonation of Mg silicates [9], was found not to be required for Ca silicates [8].

Although CO_2 sequestration by aqueous carbonation of Ca silicates seems technically feasible, the grinding, heating and compression required to increase the carbonation rate cause an energy penalty and increase the sequestration costs. The energy penalty reduces the net fraction of CO_2 sequestered due to the power and heat consumption of the mineral carbonation process. The energetic sequestration efficiency (η_{CO_2}) is defined as:

$$\eta_{CO_2} = \frac{CO_{2,avoided}}{CO_{2,sequestered}} = \frac{CO_{2,sequestered} - (CO_{2,power} + CO_{2,heat})}{CO_{2,sequestered}} \tag{2}$$

with the amount of CO_2 sequestered in the process ($CO_{2,sequestered}$), the net amount of CO_2 avoided ($CO_{2,avoided}$) and the extra CO_2 emissions due to heat ($CO_{2,heat}$) and power consumption ($CO_{2,power}$). In a previous study by Huijgen et al. [10], optimum energetic CO_2 sequestration efficiencies of 84% and 88% have been reported for aqueous carbonation of both steel slag and wollastonite ($T = 200$ °C, $p_{CO_2} = 20$ bar, $d < 38$ μm and assuming a reduced liquid-to-solid ratio of 2 kg/kg). Taking into account the various possibilities identified potentially to increase further the CO_2 sequestration efficiency, it was concluded that aqueous mineral carbonation of Ca silicates is potentially energetically feasible [10]. However, an economic assessment of mineral carbonation is required for final evaluation of the feasibility of this CO_2 sequestration technology.

Various (preliminary) studies on the sequestration costs of mineral carbonation processes have been published [9,11–14]. To the best of our knowledge, only one previous cost evaluation of the aqueous mineral carbonation process has been published in the open literature [9]. In that study, sequestration costs of 93 €/ton CO_2 avoided were reported for wollastonite (conversion rate: 1.2 \$ = 1 €) [9]. These costs seem substantially higher than other possible technologies for carbon capture and storage (e.g. costs of geological storage are typically 0.5–7 €/ton CO_2 injected [3]). Unfortunately, the sequestration costs reported for aqueous wollastonite carbonation were estimated on the basis of a carbonation process designed for olivine [9]. A system study on a dedicated wollastonite carbonation process, fully taking into account its relatively mild process conditions [8], is required to obtain a better insight in the cost breakdown of CO_2 sequestration by wollastonite carbonation. Moreover, industrial residues were not considered as a possible feedstock in the previous study [9], while these materials potentially offer a cost benefit compared to ores, since mining and transportation are no longer required because of their availability in industrial areas near CO_2 sources.

The aim of the present paper is an assessment of CO_2 sequestration costs by aqueous Ca silicate carbonation. For both wollastonite and steel slag as feedstock, a basic design of an aqueous mineral carbonation process will be made to estimate the investment and depreciation costs. Subsequently, the remaining fixed costs (e.g. for labour and maintenance) and the variable costs (e.g. for feedstock and electricity consumption) will be assessed to determine the CO_2 sequestration costs. Finally, a sensitivity analysis will be performed to investigate how the process conditions and the assumptions made affect the overall sequestration costs and to identify routes for cost reduction.

2. Materials and methods

2.1. Mineral carbonation process

Fig. 1 shows a block diagram of a CO_2 sequestration process on the basis of mineral carbonation together with the system boundaries of the present cost evaluation. The process was assumed to be located at the source of the solid feedstock, and therefore, no transport of Ca silicates is required. Within the system boundaries, a flowsheet of a mineral carbonation process was developed (Fig. 2) for simulations with ASPEN Plus flowsheeting software [15]. The wollastonite carbonation process was designed to sequester all CO_2 emitted from a 100 MW power plant (i.e. 60,000 kg/h or 480 kton/yr CO_2 assuming a specific CO_2 emission of 0.60 kg/kWh, as used in an earlier study by Huijgen et al. [10], and an operational time of 8000 h/yr). For steel slag, a significantly smaller carbonation process was designed in view of the relatively limited availability of steel slag; 15,000 kg/h or 120 kton/yr CO_2, consistent with the approximate amount of CO_2 that can potentially be sequestered in the steel slag produced in The Netherlands [16]. Simulations of both the wollastonite and steel slag carbonation processes were performed using the methods and assumptions reported previously [10]. The conversions (ζ_{CaSiO_3}) measured in a laboratory scale autoclave reactor [7,8] were used as input for the continuous carbonation reactor (see also Ref. [10]). In addition, the process conditions corresponding with the maximum energetic efficiency found earlier were used (i.e. $d < 38\ \mu m$, $T = 200\ °C$ and $p_{CO_2} = 20$ bar). The corresponding conversions (ζ_{CaSiO_3}) are 69% and 67% for wollastonite and steel slag, respectively [10]. Finally, simulations were performed at L/S = 2 kg/kg, i.e. the minimum L/S ratio that allowed adequate stirring of the suspension in the laboratory scale autoclave reactor that was used for the carbonation experiments [7] (see also Section 3.1.3 and Ref. [10]). The resulting composition and physical properties of the streams, as well as the power and heat flows, are given in Fig. 2. At these conditions, the total power and heat consumptions in the case of wollastonite are 296 and −295 kWh/ ton CO_2 sequestered, respectively (steel slag: 337 and −167 kWh/ton CO_2, respectively). The corresponding energetic efficiencies (η_{CO_2}) are 88% and 84% for wollastonite and steel slag, respectively (as indicated above; see Ref. [10] for further details).

2.2. Cost evaluation

On the basis of the simulated mineral carbonation process shown in Fig. 2, the variable and fixed sequestration costs were estimated. Costs were calculated for an assumed new plant located in The Netherlands and constructed in the 3rd quarter of 2004 (i.e. corresponding with the major source used for price information [17]). All costs are given in the euro currency using a conversion rate of 1.2 \$ = 1 € for prices given in dollars.

2.2.1. Investment costs

The investment costs were estimated using a detailed factorial cost estimation method as described by Sinnott [18]. First, the price of each major unit operation in the mineral carbonation process (Fig. 2) was calculated as follows:

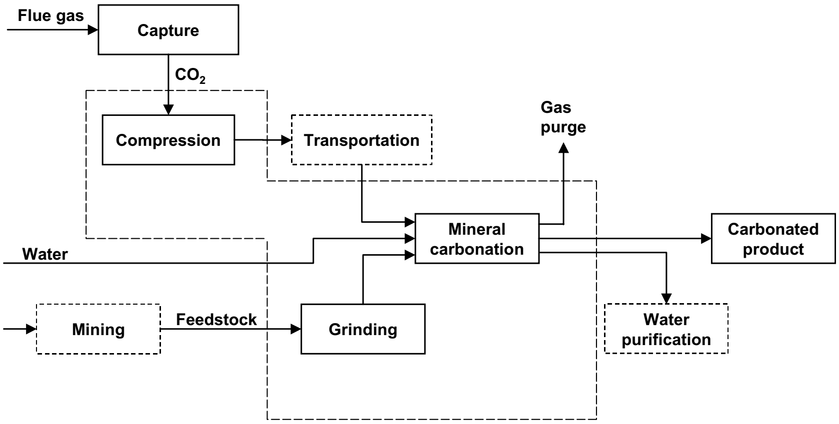
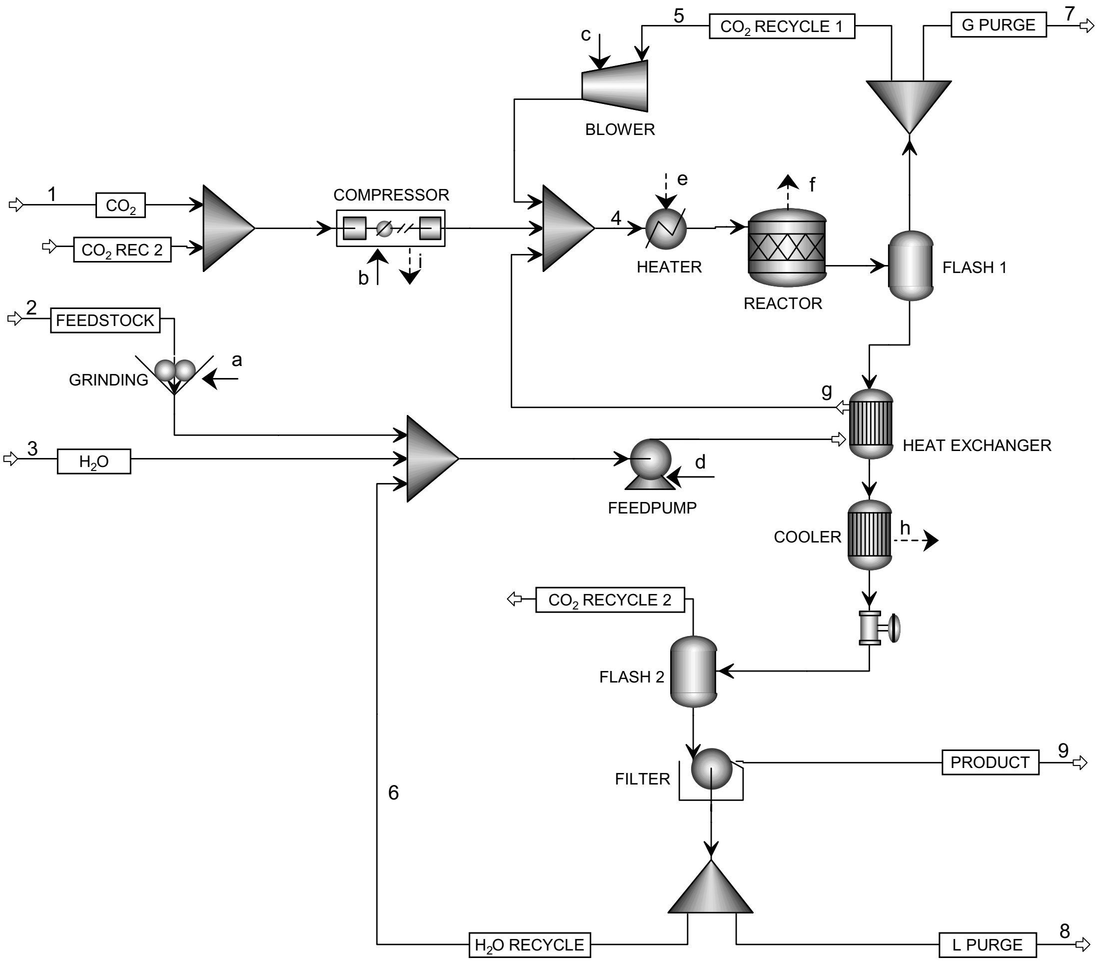


Fig. 1. Block diagram of a CO_2 sequestration process on the basis of mineral carbonation. System boundaries of the present study are indicated by the broken lines.



Stream number		*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*
T [°C]	W	25	25	25	176	200	39	40	39	39
	S	*25*	*25*	*25*	*178*	*200*	*39*	*40*	*39*	*39*
p [bar]	W	1.0	1.0	1.0	35.5	34.5	1.0	1.0	1.0	1.0
	S	*1.0*	*1.0*	*1.0*	*35.5*	*34.5*	*1.0*	*1.0*	*1.0*	*1.0*
Mass flow [ton/h]										
$CaSiO_3$	W		229.2[a]		229.2					70.8
	S		*58.7*		*58.7*					*19.1*
$CaCO_3$	W		4.9		4.9					141.3
	S		*8.0*		*8.0*					*42.1*
SiO_2	W		37.8		37.8					119.7
	S									*20.5*
FeO	W									
	S		*36.7*		*36.7*					*36.7*
H_2O	W			56.8	543.7	2.4	484.3			56.8
	S			*20.3*	*206.8*	*0.6*	*215.3*			*20.3*
CO_2	W	61.8			54.4	6.0	6.1			1.8
	S	*15.6*			*23.0*	*1.5*	*2.6*			*0.6*

Power & heat flows		*a*	*b*	*c*	*d*	*f-e*[b]	*g*[c]	*h*	*i*
E [MW]	W	11.2	5.7	0.0	0.8	-17.7	101.5	-17.0	-3.8
	S	*1.9*	*1.6*	*0.0*	*0.3*	*-2.5*	*38.4*	*-6.1*	*-1.1*

[a] *Excluding gangue* [10]. [b] *Net reaction heat.* [c] *Amount of heat transferred within heat exchanger.*

Fig. 2. Simplified ASPEN flow diagram of an aqueous mineral carbonation process, indicating heat streams (- ➔) and power input (→). The properties and composition of the mass streams and the power and heat flows at the points indicated are given in the table. Reactor conditions: $d < 38$ μm, $T = 200$ °C, $p_{CO_2} = 20$ bar, and $t = 15$ min (wollastonite: $\zeta_{CaSiO_3} = 69\%$ and $\eta_{CO_2} = 84\%$/steel slag: $\zeta_{CaSiO_3} = 67\%$ and $\eta_{CO_2} = 83\%$). Simulations were performed at L/S = 2 kg/kg. W = wollastonite; S = steel slag.

(1) A basic design of the unit operation was made to determine equipment type, main dimensions and possibly wall thickness. Because of possible corrosion due to the presence of carbonic acid at low pH and of dissolved trace elements such as Cl, AISI 316 stainless steel was selected for the construction of all process equipment, except for the grinding equipment and the compressor. It was assumed that carbon steel was a suitable material of construction for the compressor.

(2) Reference equipment was selected for which price information was available in the public literature [17,19]. The size of the reference equipment was chosen as close as possible to the actual designed equipment size.

(3) The cost of the reference equipment was adjusted for the actual size of the equipment type using an apparatus-specific scale factor, which was calculated from the costs data available in the public literature (Table 3) [17,19].

(4) If required, the price of the scaled reference equipment was corrected for the operating pressure as well as the material of construction. On the basis of data presented in Ref. [18], a factor of 2 was assumed for conversion of prices specified for carbon steel equipment into stainless steel equipment (AISI 316). Prices given for equipment operating at atmospheric pressure were corrected by a factor of 1.6 in the case of an operating pressure around 35 bar. Possible corrections for time and location relevant for the price of the reference equipment were made with the ‘chemical engineering plant cost index’ [17].

After calculation of the costs for the major process equipment, these costs were first multiplied individually with specific factors to include direct associated costs (Table 2). The direct associated costs taken into account were equipment erection, instrumentation, piping, process buildings, storages, utilities, ancillary buildings and site development costs. For each major unit operation, the sum of the costs for the equipment and the direct associated investments (the ‘total physical equipment costs’) was determined. Subsequently, these physical equipment costs were extended with the associated indirect costs (i.e. design and engineering costs, contractor’s fee and contingency) (Table 2). For all equipment except the grinding equipment, general numbers available in the literature for a fluid–solid plant were used [18] (Table 2). For the grinding equipment, the costs reported for the reference equipment already included a number of associated investments [19], and the surcharge factors were adjusted accordingly (Table 2). The direct physical equipment costs and the indirect associated investments were added, resulting in the ‘total fixed capital costs’ [18]. Finally, 10% of the total fixed capital costs was reserved as working capital, and the investment costs of the mineral carbonation plant were determined as the sum of the fixed and working capital [18].

2.2.2. Sequestration costs

The sequestration costs consisted of depreciation costs and variable and fixed operating costs. The depreciation costs were determined by linear depreciation of the total fixed capital costs over a period of 10 years. The variable operating costs consisted of costs for feedstock and utilities. Table 1 shows the relevant prices assumed. The price of wollastonite ore mined on a large scale is highly uncertain. For lump wollastonite, which is nowadays mined on a limited scale, a minimum commercial price of 42 €/ton has been reported [12]. If wollastonite would be mined on a significantly larger scale, its price would probably decrease, but to what extent is uncertain. As a first estimate, 10 €/ton was used and the influence of a change in the wollastonite ore price on the sequestration costs was studied in a sensitivity analysis (Section 3.1.3). For steel slag, a price of 0 €/ton was assumed since it is generated as a residue during the production of steel. The utilities required for aqueous mineral carbonation were electricity, e.g. for the crushers, compressors and pumps, (possibly) natural gas to heat the reactants and (cooling) water. In the calculations, the sales price of the carbonated product was set at 0 €/ton for both wollastonite and steel slag, since it is unclear if beneficial application of the product is feasible. Moreover, it was assumed that no costs were required for possible disposal of the carbonated product.

The fixed operating costs consisted of costs for operating labour, maintenance, plant overheads, laboratory, supervision, local taxes and insurance. The assumptions used in determining the fixed operating costs are also shown in Table 1.

In order to determine the sequestration costs, the depreciation costs and the variable and fixed operating costs

Table 1
Assumptions used for estimation of operating costs

Operating costs			Reference
Variable			
Wollastonite	10	€/ton	
Steel slag	0	€/ton	
Water (softened)	0.35	€/m^3	[17]
Cooling water	0.05	€/m^3	[17]
Natural gas[a]	0.12	€/m^3	[17]
Electricity	0.06	€/kWh	[17]
Fixed			
Operating labour	300/yr	k€/operator[b]	
Wollastonite	3	Operators	
Steel slag	2	Operators	
Plant overheads	50	% Operating labour	[18]
Laboratory	20	% Operating labour	[18]
Supervision	20	% Operating labour	[18]
Maintenance	5	% Fixed capital	[18]
Local taxes	1	% Fixed capital	[18]
Insurance	1	% Fixed capital	[18]

[a] Only applicable, if heat flow reactor (*f* in Fig. 2) < heater (e). Not the case at energetically optimum conditions (Fig. 2).

[b] Assumed nominal wages operator: 30,000 €/yr. Full continuous operation = 5 shifts/day. Surcharge factor of 2 is used to include taxes and insurance.

Table 2
Surcharge factors used for various equipment

Surcharges	General equipment[a]	Grinding equipment
Direct associated investments (% equipment cost)		
Equipment erection	45	0[b]
Instrumentation[c]	25	0[b]
Piping	45	20[d]
Process buildings	10	5[d]
Storages	20	10[d]
Utilities	45	20[d]
Ancillary buildings	20	10[d]
Site development	5	5
Sum	*215*	*70*
Indirect associated investments (% total physical equipment cost)[e]		
Design and engineering	25	10[d]
Contractor's fee	5	0[b]
Contingency	10	10
Sum	*40*	*20*

[a] General numbers for fluid-solid plant [18].
[b] Included in the reference equipment price [19].
[c] Includes electrical costs.
[d] Assumed to be partially included in the reference equipment price [19].
[e] Consists of equipment cost and direct associated investment costs.

were added, resulting in the costs per ton CO_2 physically sequestered in the carbonation reactor. Subsequently, the sequestration costs were divided by the energetic CO_2 sequestration efficiency (Eq. (2)) in order to convert the costs per ton of CO_2 sequestered into costs per ton of CO_2 net avoided.

Finally, the sensitivity analysis of the CO_2 sequestration costs comprised both the influence of the assumptions made as well as the effect of the major process parameters (i.e. temperature, pressure, particle size). The latter was done to verify that the economically optimum set of process conditions corresponds with the energetically optimum set (e.g. for wollastonite, 200 °C, 20 bar CO_2, <38 μm [10]), as implicitly assumed in Section 2.1.

3. Results and discussion

3.1. Wollastonite

3.1.1. Investment costs

Table 3 shows the calculation of the investment costs of the wollastonite carbonation process shown in Fig. 2. The costs of the reference equipment selected for the major unit operations and possible scale and other correction factors used are specified. The total investment costs of the wollastonite carbonation plant are 45 M€, of which 41 M€ is fixed capital. Total process equipment costs are 11 M€, of which the most expensive process equipments are the grinding equipment (3.1 M€), the compressor (2.5 M€), the reactor (2.2 M€) and the heat exchanger (2.0 M€). Among the grinding equipment, the fine grinding step in the ball mill is, by far, the most expensive grinding step (2.9 M€). The costs of the second compressor, the feed pump and the flash tanks are negligible compared to the other investment costs.

Below, a short discussion on the selection, design and cost estimation of each major unit operation is given.

3.1.1.1. Grinding equipment. The grinding equipment is required to crush and grind 272 ton/h wollastonite ore (Fig. 2) from an initial particle size of 0.1 m to <38 μm ($D[4,3] = 16$ μm) [8,10]. On the basis of the cost information available [19] and the specifications of various grinding equipment [20], two types of reference grinding equipment were selected for cost estimation of the mineral carbonation process: a cone crusher for initial size reduction, followed by a ball mill. Possible investment costs associated with the impurity (i.e., gangue) of the wollastonite ore [10] have been neglected in this study.

3.1.1.2. Compressor, blower and pump. In the flowsheet simulations, a multi-stage compressor was assumed for compression of the CO_2 feed. Three stages were assumed to be required on the basis of the ratio between the inlet and outlet pressure (isentropic operation with an efficiency of 0.8) [10]. However, price information was only available for a single stage (screw) compressor [17]. Therefore, for cost estimation purposes, three of these single stage compressors were assumed to be placed in series on a single axis and the whole was scaled to the gas volume that had to be compressed. One electro-motor was assumed to be sufficient to drive the axis. The costs of the blower used in the 'CO_2 recycle 1' to compensate for the pressure drop over the reactor (1 bar) (see Fig. 2) were estimated by taking the costs of two blowers in series with a pressure increase of 0.5 bar each [17] (Table 3). The costs for the feed pump of the slurry were estimated on the basis of the costs of a (virtual) number of single stage centrifugal fluid pumps in series. The (virtual) number of pumps in series was determined by the total pressure increase required and the pressure increase of a single centrifugal pump (i.e. 4.3 pumps, 34.5 bar and 8 bar, respectively). Since the pumping of a slurry causes much higher costs for sealing and material of construction compared to the pumping of a fluid a correction factor of 4 was applied. The feed pumps were assumed to be driven by a single electro-motor.

3.1.1.3. Separation equipment. Two types of separation equipment were used: flash tanks to separate gas streams from slurries and a filter to separate solids from the product slurry. The costs of the two flash tanks were estimated on the basis of the available prices for a cylindrical tank. A hold up time of 10 s was assumed to be sufficient for effective separation. The required wall thickness (d) was estimated with [18]:

$$d = \frac{1.1 \cdot \frac{p}{10} \cdot D}{2f - 1.1 \cdot \frac{p}{10}} \quad (3)$$

with $f = 120$ N/mm^2 for stainless steel AISI 316, a safety margin of 1.1, p the operating pressure and $D = 2$ for the height-to-diameter ratio. Thus, a required wall thickness of 16 and 1 mm was determined for the flash tank in the

Table 3
Determination of investment costs for wollastonite carbonation process

Flowsheet	Reference equipment from the literature				Flow-sheeting results		Costs calculation				Price (M€)		
	Equipment type	Price[a] (k€)	Relevant size	Material	Relevant size	Pressure (bar)	Number in series	Correction factors			Equipment	Surcharges	
								Scale	Pressure	Other		Direct	Indirect
Compressor	Single-stage compressor	183	8800 m^3/h	Steel	40353 (10966) m^3/h	1–35.5	3	0.72			1.64 (0.64)		
	Electro-motor	64	630 kW	NA[b]	5742 (1556) kW	NA	1	1.17			0.85 (0.18)		
	Sum										2.49 (0.83)	5.36 (1.78)	3.14 (1.04)
Blower	Blower, 500 mbar	10 (7)	350 (70) m^3/h	Steel	277 (69) m^3/h	34.5–35.5	2	0.41		2[c]	0.04 (0.03)	0.08 (0.06)	0.05 (0.04)
Feedpump	Single-stage centrifugal pump[d]	11	250 m^3/h	AISI 316	724 (274) m^3/h	1–35.5	4.3	0.14		4[e]	0.21 (0.19)		
	Electro-motor	10	132 kW		776 (296) kW	NA	1	1.00			0.06 (0.02)		
	Sum										0.27 (0.21)	0.57 (0.44)	0.34 (0.26)
Flash tank 1	Cylindrical tank, 16 mm	20 (10)	5 (1) m^3	Steel	3.3 (1.1) m^3[f]	34.5	1	0.41		2[c]	0.03 (0.02)	0.07 (0.05)	0.04 (0.03)
Flash tank 2	Cylindrical tank, 3 mm	12 (8)	20 (5) m^3	Steel	17.4 (6.7) m^3[f]	1	1	0.37		2[c]	0.02 (0.02)	0.05 (0.04)	0.03 (0.02)
Filter	Rotating vacuum tumbler filter[g]	206	50 m^2	AISI 316	141 (46) m^2[h]	1	1	0.66	1[i]		0.41 (0.19)	0.88 (0.42)	0.51 (0.24)
Heat exchanger	Shell and tube heat exchanger	342	1000 m^2	AISI 316	4316 (1726) m^2	34.5–35.5	1	0.78	1.6	1.15[j]	1.98 (0.96)	4.25 (2.07)	2.49 (1.22)
Cooler	Shell and tube heat exchanger	252 (134)	700 (300) m^2	AISI 316	695 (255) m^2	34.5/1	1	0.78	1.6	1.15[j]	0.46 (0.22)	0.99 (0.47)	0.58 (0.27)
Grinding equipment	Cone crusher	65	60 ton/h		272 (103) ton/h	NA	1	0.73		1.29[k]	0.25 (0.12)		
	Ball mill	300	8 ton/h		272 (103) ton/h	NA	1	0.57		1.29[k]	2.89 (1.66)		
	Sum										3.14 (1.79)	2.20 (1.25)	1.07 (0.61)
Reactor	Stirred tank with jacket	138	30 m^3	AISI 316	63–120 (49–105) m^3	35.5–34.5	5	0.45	1.6	1.2[l]	2.17 (2.01)	4.67 (4.32)	2.74 (2.53)
Sum											11.0 (6.3)	19.1 (10.9)	11.0 (6.3)
Fixed capital													41.1 (23.4)
Working capital													4.1 (2.3)
Investment													45.2 (25.8)

(*continued on next page*)

reactor outlet and in the water recycle, respectively. The nearest wall thickness for which price information was available was 16 and 3 mm, respectively [17].

3.1.1.4. Heat exchangers. For both heat exchangers (i.e. the heat exchanger between the reactor outlet and inlet as well as the cooler in which the reactor outlet stream is cooled further with cooling water), a U type shell and tube heat exchanger was selected. A heat transfer coefficient (U) of 1000 W/m^2 °C was assumed on the basis of data presented in Ref. [18]. The logarithmic mean temperature difference (ΔT_{lm}) was 23.5 and 24.4 °C for heat exchanger 1 and 2, respectively (cooling water assumed to enter at 15 °C and leave at 40 °C). Thus, the heat exchanging surface areas $\left(A = \frac{Q}{U\Delta T_{\mathrm{lm}}}\right)$ required were 4316 and 695 m^2 on the basis of a heat duty (Q) of 101.5 and 17.0 MW, respectively (Fig. 2). The required amount of cooling water was 585 m^3/h.

3.1.1.5. Reactor. The designed reactor comprised the mixer, heater and reactor in the ASPEN flowsheet (Fig. 2). The final design of the reactor was difficult to determine given the current state of research. As a first approach, a continuous version of a scaled up autoclave reactor was used, i.e. a series of stirred tank reactors with a heating/cooling jacket. The slurry inlet flow was 892 m^3/h, and the residence time was 15 min, resulting in a total slurry volume of the reactor of 223 m^3. The ratio of the gas and slurry volume flows $\left(\frac{\Phi_{\mathrm{G}}}{\Phi_{\mathrm{L+S}}}\right)$ was 1.9 and 0.3 at the reactor inlet and outlet, respectively (gaseous CO_2 was consumed by the carbonation reaction). Assuming a series of five tank reactors and a (simplified) linear development of the conversion (i.e. gas consumption), the volumes of the five reactors were: 120, 106, 92, 78 and 63 m^3 (on the basis of a H:D ratio of 1, D = 5.4, 5.1, 4.9, 4.6 and 4.3 m).

3.1.2. Mineral CO_2 sequestration costs

The net calculated sequestration costs for wollastonite carbonation are 102 €/ton CO_2 avoided (Table 4). These costs consist of the total sequestration costs of 86 €/ton CO_2 sequestered corrected for the energetic CO_2 sequestration efficiency of 84% as determined earlier at L/S = 2 kg/kg [10]. Table 4 shows the depreciation costs and variable and fixed operating costs both per ton of sequestered and avoided CO_2. The largest costs are associated with the feedstock (54 €/ton CO_2 avoided) and the electricity required (26 €/ton CO_2 avoided). The depreciation (10 €/ton CO_2 avoided) and total fixed operating costs (11 €/ton) are relatively limited. The electricity is mainly required for grinding the feedstock (18 €/ton CO_2 avoided) and compression of the CO_2 feed (8 €/ton CO_2 avoided).

3.1.3. Sensitivity analysis and process optimisation

The sensitivity analysis given in Fig. 3 shows the relative effect of selected parameters on the CO_2 sequestration costs of wollastonite carbonation. Within the ranges studied, the largest uncertainties in the reported sequestration costs are

Table 3 (*continued*)

For steel slag, deviating numbers are given between brackets.

[a] All prices were obtained from the DACE price booklet [17], except for the grinding equipment [19].

[b] Not applicable.

[c] Material of construction.

[d] 2900 rpm.

[e] The pumping of a slurry imposes additional requirements for material of construction and sealings.

[f] Hold-up = 10 s.

[g] Although the filter is not operated at vacuum conditions, this is the only continuous filter for which price information was available in Ref. [17].

[h] Estimated by ASPEN assuming a 1 bar pressure drop and a cake resistance of 6.22×10^{10} m/kg for $CaCO_3$ in water [23]. For the compressibility and porosity of the filter cake the standard ASPEN values of 0 and 0.45 were used, respectively. The mass fraction of water within the filter cake was set at 0.85 [10].

[i] Compared to vacuum.

[j] U-type heat exchanger [17].

[k] Price correction 1990–2004 [17].

[l] To include bubble disperser [17].

Table 4
CO_2 sequestration costs for wollastonite and steel slag carbonation (excluding possible costs for CO_2 capture)

Costs (€/ton CO_2)	Wollastonite		Steel slag	
	Sequestered	Avoided	Sequestered	Avoided
Depreciation[a]				
Compressor, blower and pump	2	3	3	4
Reactor	2	2	6	8
Grinding equipment	2	3	6	7
Heat exchangers	2	2	4	4
Other equipment	0	0	1	1
Variable				
Feedstock	45	54	0	0
(Cooling) water	1	1	1	1
Electricity grinding	15	18	13	15
Electricity other	7	8	7	9
Fixed				
Staff	2	2	5	6
Maintenance	4	5	10	12
Other	3	4	8	10
Sum	*86*	*102*	*64*	*77*

[a] Including depreciation of direct and indirect associated investment costs.

associated with the value of the carbonated product (36–168 €/ton CO_2 avoided), the wollastonite ore price (48–156 €/ton), the conversion in the carbonation reactor (88–119 €/ton) and the liquid-to-solid ratio (94–127 €/ton). As discussed above, the sequestration costs for the standard case are 102 €/ton.

The price of wollastonite not only has a large influence on the sequestration costs but also is among the most uncertain parameters (see Section 2.4). Thus, the economic feasibility of CO_2 sequestration by wollastonite carbonation depends largely on the feedstock price. Also, the value of the carbonated product is an important parameter, since the mass of carbonated product is 5.5 or 7.9 times larger than the mass of CO_2 sequestered (in the case of wollastonite and steel slag, respectively). However this parameter is also uncertain, since no thorough assessments of possible markets have been reported so far (possible markets may include calcium carbonate for the paper industry and slaked lime, e.g. for soda and steel production). In the current cost evaluation, it was assumed that the process did not yield a saleable product. In other cost evaluation studies on mineral carbonation processes, prices of 10–15 €/ton of product were assumed [12,13], which would reduce the sequestration costs substantially (e.g. from 102 €/ton CO_2 avoided to 36 €/ton at 10 €/ton product, see Fig. 3). It should be noted that beneficial re-use of the carbonated product may impose the need of a drying step, which is not included in the current process design (Fig. 2). A reduction of the L/S ratio results in smaller streams that have to be processed and, thereby, less expensive equipment. In addition, the energetic efficiency of the process improves [10], which reduces the sequestration costs per ton of CO_2 avoided.

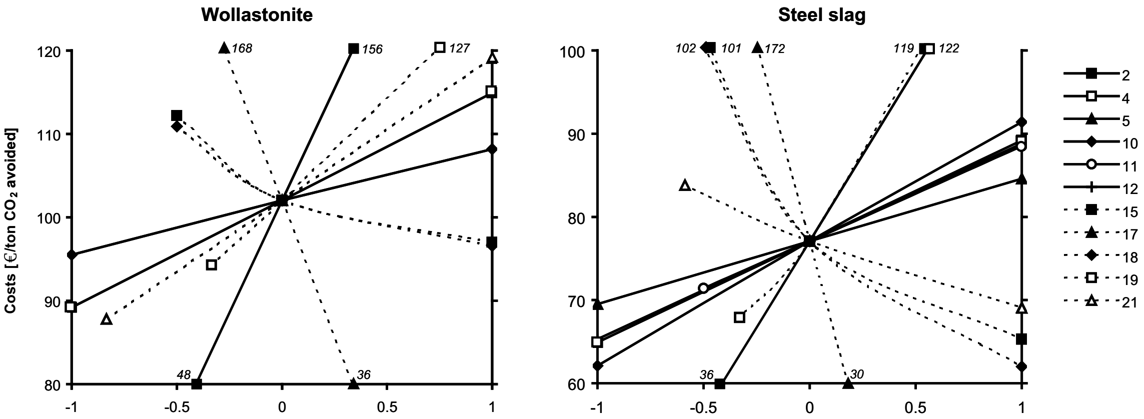
Finally, it should be noted that the temperature of the water recycle does not have a significant influence on the sequestration costs (Fig. 3). In the current process design, the heat duty of the heat exchanger is large, i.e. 101 MW due to, among other factors, the low temperature of the water recycle (40 °C) [10]. This value is of comparable magnitude as the size of the power plant for which the CO_2 emissions are sequestered (100 MW). Although an increase of the temperature of the process water recycle would reduce the deprecation costs for the heat exchanger, the effect on the overall sequestration costs is apparently very limited. Thus, the estimated sequestration costs seem also valid for a mineral carbonation process, which is further optimised with respect to the temperature of the water recycle.

Fig. 4 shows the influence of the temperature, pressure and particle size on, successively, the $CaSiO_3$ carbonation degree measured in a laboratory scale autoclave reactor [8] (I), the energetic efficiency simulated following earlier work by Huijgen et al. [10] (II) and the sequestration costs per ton of CO_2 sequestered (III) and per ton of CO_2 avoided (IV) for wollastonite carbonation. It can be concluded that the economically optimum set of process conditions corresponds with the energetically optimum set, as implicitly assumed in Section 2.1. In fact, optimisation of the aqueous wollastonite carbonation process with respect to the carbonation degree leads more or less directly to minimum sequestration costs, as a result of the dominant effect of the wollastonite feedstock on the overall sequestration costs.

3.2. Steel slag

The total sequestration costs by steel slag carbonation are 64 €/ton CO_2 sequestered or 77 €/ton CO_2 avoided sequestration costs (Table 4). The variable operating costs are substantially lower than for wollastonite because of the absence of feedstock costs. However, these reductions are partially annulled by the higher depreciation costs (23 €/ton CO_2 avoided) and fixed operating costs (28 €/ton) (Table 1). These higher costs are caused by the smaller scale of the process and the larger grinding equipment and reactor relative to the amount of CO_2 sequestered, since more feedstock has to be processed, mainly due to the lower Ca content of the feedstock (23 vs. 30% for wollastonite [7,8]). The total investment costs are 26 M€ lower than for wollastonite due to the smaller scale of the process (the design of the major process equipment and the estimation of the investment costs were performed similarly to wollastonite, see Section 3.1.1).

A potential additional cost benefit of the use of industrial residues as feedstock for mineral CO_2 sequestration is the generation of feedstock and CO_2 at the same location. The cost saving thus obtained is not taken into account in the numbers presented since it is outside the system boundaries, but transportation costs of CO_2 are



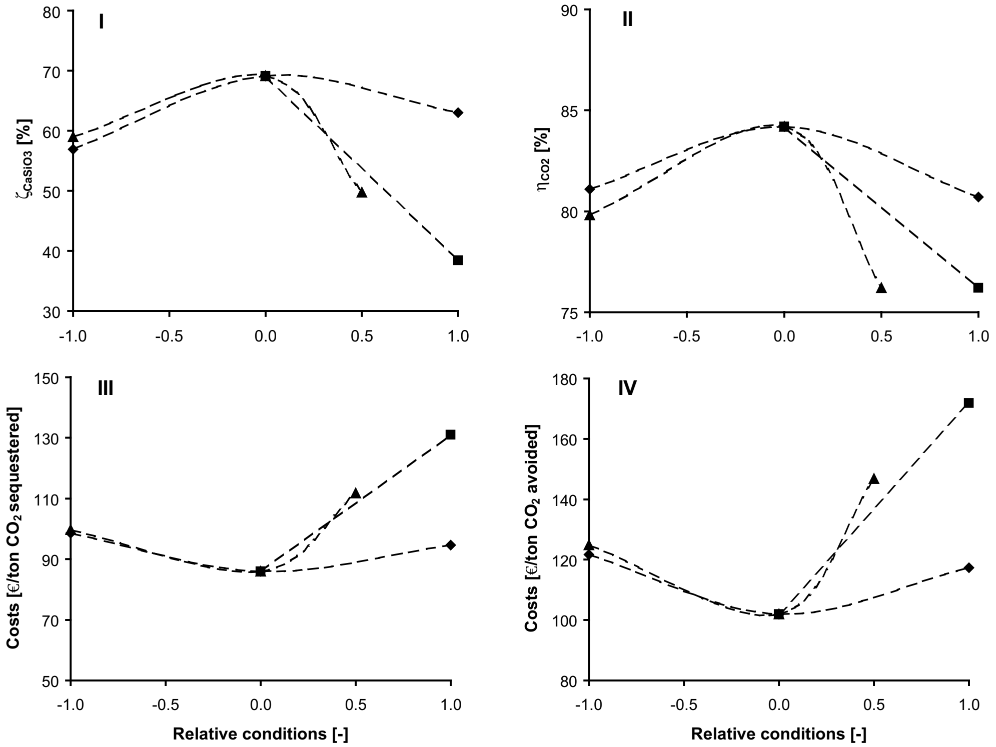
#	Variable	Wollastonite			Steel slag		
	Parameters cost evaluation	*Standard*	*Min*	*Max*	*Standard*	*Min*	*Max*
1	Water [€/m^3]	0.35 (0) [a]	0.2 (-1)	0.5 (1)	0.35 (0)	0.2 (-1)	0.5 (1)
2	Feedstock [€/ton]	10 (0)	0 (-1)	20 (1)	0 (0)	-5 (-1)	5 (1)
3	Cooling water [€/m^3]	0.05 (0)	0.03 (-1)	0.07 (1)	0.05 (0)	0.03 (-1)	0.07 (1)
4	Electricity [€/kWh]	0.06 (0)	0.03 (-1)	0.09 (1)	0.06 (0)	0.03 (-1)	0.09 (1)
5 [b]	Reactor [M€]	2.2 (0)	1.1 (-1)	3.3 (1)	2.0 (0)	1.0 (-1)	3.0 (1)
6 [b]	Grinding equipment [M€]	3.1 (0)	1.6 (-1)	4.7 (1)	1.8 (0)	0.9 (-1)	2.7 (1)
7 [b]	Compressor, blower, and pump [M€]	2.8 (0)	1.4 (-1)	4.2 (1)	1.1 (0)	0.5 (-1)	1.6 (1)
8 [b]	Heat exchangers [M€]	2.4 (0)	1.2 (-1)	3.7 (1)	1.2 (0)	0.6 (-1)	1.8 (1)
9 [b]	Filter and flash tanks [M€]	0.5 (0)	0.2 (-1)	0.7 (1)	0.2 (0)	0.1 (-1)	0.3 (1)
10	Surcharge factors (general & grinding equipment) [-] [c]	4.4 & 2.0 (0)	2.7 & 1.5 (-1)	6.1 & 2.5 (1)	4.4 & 2.0 (0)	2.7 & 1.5 (-1)	6.1 & 2.5 (1)
11	Operating labour [-]	3 (0)	1 (-1)	5 (1)	2 (0)	1 (-0.5)	4 (1)
12	Maintenance [%]	5 (0)	0 (-1)	10 (1)	5 (0)	0 (-1)	10 (1)
13	Staff, Laboratory & Supervision [%]	90 (0)	45 (-1)	135 (1)	90 (0)	45 (-1)	135 (1)
14	Local taxes & Insurance [%]	2 (0)	0 (-1)	4 (1)	2 (0)	0 (-1)	4 (1)
15	Depreciation period [yr]	10 (0)	5 (-0.5)	20 (1)	10 (0)	5 (-0.5)	20 (1)
16	Operational time [hr/yr]	8000 (0)	7500 (-1)	8500 (1)	8000 (0)	7500 (-1)	8500 (1)
17	Product price [€/ton]	0 (0)	-10 (-1)	10 (1)	0 (0)	-10 (-1)	5 (0.5)
	Parameters process simulation [d]						
18	Scale [ton CO_2/hr]	60 (0)	30 (-0.5)	120 (1)	15 (0)	7.5 (-1)	30 (1)
19	Liquid-to-solid ratio [kg/kg]	2 (0)	1 (-0.33)	5 (1)	2 (0)	1 (-0.33)	5 (1)
20	Temperature of H_2O recycle [°C] [e]	40 (0)		100 (1)	40 (0)		100 (1)
21	Carbonation degree (ζ_{CaSiO3}) [%]	69.1 (0)	60 (-0.8)	80 (1)	67.4 (0)	60 (-0.6)	80 (1)

a) Numbers between brackets are the relative input values, defined as: $\mathit{Rel.Value} = \frac{\mathit{Abs.Value} - \mathit{Standard}}{\mathit{Max} - \mathit{Standard}}$*, as used for the graphs. b) Including design parameters, price of reference equipment and price correction factors used (i.e., all assumptions given in Table 3). c) Multiplication factor based on all individual direct and indirect surcharge factors. d) Including possible effect on the energetic efficiency (*η_{CO2}*) [10]. e) For these simulations, the ASPEN flowsheet was extended with a cooler, which cools the second* CO_2 *recycle stream in order to avoid an increase of the gas volume that has to be compressed.*

Fig. 3. Sensitivity analysis of CO_2 sequestration costs for wollastonite and steel slag carbonation for the parameters shown in the table. Table: in case of different values for the wollastonite and steel slag calculations, the values are given as wollastonite/steel slag. Graph: for reasons of clarity, only the parameters with an influence ⩾5 €/ton CO_2 avoided are shown. For variables with a minimum/maximum value outside the range of the graph, the values are indicated at the borders.

typically in the range of 1–8 €/ton CO_2 (transport distance 250 km) [3].

The influences of the various assumptions on the sequestration costs by steel slag carbonation (Fig. 3) show that, since the scale of the process is smaller and the feedstock cost no longer dominates the overall sequestration costs, as in the case of wollastonite, the parameters concerning the estimation of the investment costs now also have a significant influence. Within the ranges studied, the largest uncertainties in the sequestration costs are associated with the (possible) value of the carbonated product (30–172 €/ton), the possible costs of the feedstock (36–119 €/ton CO_2 avoided), the scale of the sequestration process (62–102 €/ton), the depreciation period (65–101 €/ton) and the liquid-to-solid ratio (68–122 €/ton). It is noteworthy that the conversion has a smaller influence on the sequestration costs than in the case of wollastonite. The scale of the process affects the sequestration costs in two ways: (1) scale effect on the investment costs (see also Table 3) and (2) relative change of the number of operators required and, thereby, of the fixed operating costs. The depreciation period only virtually changes the sequestration costs, since the investment costs are spread out over different time periods.



Variable		*Standard* [a]	*Min*	*Max*
Particle size	–■–	<38 (0)	NA	<106 (1)
Reactor temperature	–▲–	200 (0)	150 (-1)	225 (0.5)
CO_2 pressure	–◆–	20 (0)	10 (-1)	30 (1)

[a] Values between brackets are the relative input values used for the graphs.

Fig. 4. Influence of process conditions on $CaSiO_3$ conversion (ζ_{CaSiO_3}) (I), energetic CO_2 sequestration efficiency (η_{CO_2}) (II) and sequestration costs per ton of CO_2 sequestered (III) and per ton of CO_2 avoided (IV). Simulations were performed at L/S = 2 kg/kg.

3.3. Comparison to other cost analyses

For the direct aqueous wollastonite carbonation process studied in this paper, the Albany Research Centre (ARC) calculated sequestration costs of 76 €/ton CO_2 sequestered or 93 €/ton CO_2 avoided $\eta_{CO_2} = 82\%$, 1 € = 1.2$ [9]. Comparison of the ARC results with the results of this study is complicated since: (1) in the ARC study, the wollastonite carbonation costs were estimated on the basis of a process designed for olivine carbonation and (2) the cost methodology itself has not been published [9,21]. In addition, the process design is different with regard to a number of aspects, e.g. (1) the partial recycling of the unconverted feedstock in the ARC process [21] (see also discussion in Ref. [10]) and (2) the scale of the process (i.e. 1100 [9] vs. 60 ton/h CO_2 sequestered in this study). Enlarging the current scale of the process to a scale comparable with that of the ARC study (a 1.3 GW coal fired power plant [9]) would lower the sequestration costs for reasons of 'economy of scale' (Fig. 3). However, given the very large solid material streams involved, a more modest scale for a mineral carbonation plant seems appropriate. In spite of the differences between these studies, the similar sequestration costs for aqueous wollastonite carbonation resulting from the ARC and this study (Table 5) are noteworthy.

In addition to cost estimates for the direct aqueous wollastonite carbonation route, Table 5 shows sequestration costs reported for other mineral carbonation processes. This limited set of cost estimates suggests that: (1) current direct carbonation routes (such as the direct aqueous route discussed in this paper) are less expensive than indirect carbonation routes and (2) mineral carbonation processes using solid residues as feedstock tend to be less expensive than processes using ores (see also a previously published comparison of process routes [2]). Overall, the direct aqueous carbonation processes using either steel slag or olivine (Mg_2SiO_4) seem to have the lowest sequestration costs among the currently available mineral carbonation processes.

Finally, Table 5 also shows the costs of other CCS technologies such as geological storage. In cost estimates of CO_2 storage options other than mineral carbonation, compression is typically omitted from consideration, since compression for transportation purposes is generally already included in the CO_2 capture step. Mineral carbonation costs excluding compression decrease to 85 and 58 €/ton CO_2 avoided for wollastonite and steel slag, respectively, due to the absence of both investment costs for the compressor and electricity costs for compression as well as an increase of the sequestration efficiency to 90%

Table 5
Comparison of sequestration costs reported for mineral carbonation processes and other carbon capture and storage (CCS) technologies (excluding possible costs for CO_2 capture)

Costs mineral carbonation (€/ton CO_2 avoided)	Feedstock	Process route[a]	Extraction agent	Reference
93	Wollastonite	Direct	Water	[9]
102	Wollastonite	Direct	Water	This study
77	Steel slag	Direct	Water	This study
65	Olivine	Direct	Water[b]	[9]
258	Serpentine[c]	Direct	Water[b]	[9]
95[d]	Mg–silicate	Direct	Molten $MgCl_2$	[11]
25[e]	Waste cement	Indirect	Water	[13]
57[e]	Wollastonite	Indirect	Acetic acid	[14]
>150	Mg–silicate	Indirect	HCl	[11]
Other CCS-technologies [€/ton CO_2 injected]	*Storage type*	*Costs type*		
0.5–7	Geological	Storage		[3]
0.1–0.3/yr		Monitoring costs		[3]
4–25	Oceanic			[3]

[a] For more information on process routes and extraction agents see a previously published literature review [2]. Direct route: Ca/Mg-extraction and carbonation takes place in a single process step. Indirect route: Ca/Mg is first extracted and, subsequently, carbonated in a second process step.

[b] Salts added: 0.64 M $NaHCO_3$, 1 M NaCl.

[c] Heat-treated.

[d] Assuming make-up $MgCl_2$ is not produced on-site, but has to be imported.

[e] Comprises only power costs and, in the case of waste cement, a revenue for selling $CaCO_3$.

and 89%. However, even excluding compression, the sequestration costs of current mineral carbonation technologies are high relative to the costs of geological CO_2 capture and storage options. Although the permanent and inherently safe sequestration of CO_2 by mineral carbonation may justify higher costs than those of geological storage, further cost reductions are required, particularly in view of (current) prices of CO_2 emission rights within the EU emissions trading scheme (20–25 €/ton, 2nd half 2005 [22]).

3.4. Discussion

In this paper, we have presented a cost estimate of CO_2 sequestration by aqueous carbonation of Ca silicates. In order to improve the accuracy of the cost estimate, it is recommended to study a number of assumptions made in the economic evaluation of the mineral carbonation process in further detail, in particular, the factors discussed in Section 3.1.3. First, pilot scale experimental research on the carbonation process in a continuous reactor can provide a better insight in the carbonation process under industrial conditions. Research should particularly focus on the carbonation degree under those conditions as well as on the minimum L/S ratio that is required for processing and carbonating. Second, specific cost evaluations should be made for individual potential locations for a mineral carbonation plant taking into account, among other factors, the specific scale of the sequestration process. In addition, assessments of the relevant feedstock price and possible markets for the carbonated product should be made for each specific location.

As noted in Section 3.3, the estimated sequestration costs of state of the art mineral carbonation are high relative to both the costs of other CO_2 capture and storage options as well as (current) CO_2 emission right trade prices within the EU. Research on cost reduction should focus on the major costs identified in this paper. For wollastonite carbonation, measures to increase the carbonation degree further are especially desirable given the large contribution of the ore costs to the overall sequestration costs (e.g. by further grinding [8,10] and/or application of additives [2]). Also, recycling the unconverted feedstock may be considered, although the feasibility of separating the carbonated material from the fresh feedstock seems unclear [10]. An alternative might be the use of a Mg silicate instead of wollastonite, since the costs of Mg silicates are generally relatively low due to their widespread abundance. For residue carbonation processes, optimisation of the carbonation degree is less important since the material is assumed to be available for negligible costs (Fig. 3). Therefore, carbonation at milder process conditions might be considered to reduce, e.g. the electricity consumption and the investment cost. For both types of feedstock, possibilities to increase the energetic performance (e.g. an increase of the pressure at which the water is recycled [10]) of the process may also reduce the sequestration costs. Finally, in order possibly to reduce the overall sequestration costs of mineral carbonation substantially and to create a cost benefit compared to other CCS technologies, research on the possibilities to integrate mineral carbonation processes with the CO_2 capture process step (Fig. 1) is recommended. CO_2 capture costs are substantial in the overall costs of 'carbon capture and storage' technologies. Representative figures reported for different types of power plants range from 19 to 44 €/ton CO_2 on the basis of current technologies [3].

4. Conclusions

A cost estimation of CO_2 sequestration by aqueous wollastonite and steel slag carbonation has resulted in sequestration costs of 102 and 77 €/ton CO_2 net avoided, respectively. For wollastonite, the major costs were found to be associated with the feedstock and the electricity consumption for grinding and compression (54 and 26 €/ton CO_2 avoided, respectively). In addition to the electricity costs, the significantly lower sequestration costs for steel slag were largely determined by the depreciation and fixed operating costs (23 and 28 €/ton CO_2 avoided, respectively) due to the absence of costs for the feedstock and the smaller scale of the process. Sensitivity analyses have shown that influential parameters in the sequestration costs are the possible value of the carbonated product, the price of the feedstock (wollastonite), the liquid-to-solid ratio, the conversion in the reactor (wollastonite) and the scale of the process (steel slag). Although various options for potential cost reduction have been identified, CO_2 sequestration by current aqueous carbonation processes seems expensive relative to other CO_2 storage technologies. The permanent and inherently safe sequestration of CO_2 by mineral carbonation may justify higher costs, but further cost reductions are required, particularly in view of (current) prices of CO_2 emission rights. Niche applications of mineral carbonation with a solid residue as feedstock and/or a useful carbonated product hold the best prospects for an economically feasible CO_2 sequestration process.

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