

Supplementary Information to *Towards a business case for CO₂ mineralisation in the cement industry*

Till Strunge^{1,2}, Phil Renforth¹, Mijndert Van der Spek¹

¹ Research Centre for Carbon Solutions, School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, United Kingdom.

² Institute for Advanced Sustainability Studies e.V., Potsdam, Germany

Content

Supplementary Notes	3
Supplementary Note 1: Market analysis for CO ₂ mineralisation products.....	3
Supplementary Note 2: Scope definition for the assessment	3
Supplementary Note 3: SCM _{CCU} product specification	3
Supplementary Note 4: Improved process designs for SCM production	4
Supplementary Note 5: Comparison of CO ₂ mineralisation to other emission reduction strategies	5
Supplementary Figures	7
Supplementary Figure 1: Current demands in Europe and the potential supply from CO ₂ carbonation..	7
Supplementary Figure 2: Flowsheets of proposed full-scale processes	8
Supplementary Figure 3: Description of the techno-economic modelling sequence.....	9
Supplementary Figure 4: Scope of the assessment for the comparison of different CO _{2e} emission reduction measures	9
Supplementary Figure 5: Cost comparison of different process routes and mid scenario	10
Supplementary Figure 6: Distribution of costs for the mid scenario	11
Supplementary Figure 7: Material use for the mid scenario	12
Supplementary Figure 8: Sensitivity analysis with the assumptions of the mid scenario	13
Supplementary Figure 9: Probability density functions for the input parameters for the MonteCarlo simulations	14
Supplementary Tables.....	15
Supplementary Table 1: Literature review	15
Supplementary Table 2: Review of carbonated material use in standardized cement products	16
Supplementary Table 3: Process assumptions.....	16
Supplementary Table 4: Equations used for mass balance of feed recycling and separation.....	16
Supplementary Table 5: Used Equations for Density and viscosity	17
Supplementary Table 6: Heat capacity calculation for different compounds.....	17
Supplementary Table 7: Equations for electricity consumption.....	18
Supplementary Table 8: Used equations for deriving the total direct costs	18
Supplementary Table 9: Major equipment design assumptions	21
Supplementary Table 10: Assumptions carbon footprint reduction assessment	21
Supplementary Table 11: Capital Cost factor assumptions.....	22
Supplementary Table 12: Operational cost factor assumptions.....	22
Supplementary Table 13: Probability density function for Monte Carlo Simulation and Scenario definition	23

Supplementary Table 14: Equations used for the comparisons of different strategies	24
Supplementary Table 15: Assumptions used for the comparisons of different strategies.....	24
Supplementary Table 16: List of abbreviations and symbols.....	25
Supplementary References.....	27

Supplementary Notes

Supplementary Note 1: Market analysis for CO₂ mineralisation products

The goal of implementing CO₂ mineralisation in a cement production facility is to capture most of the plant's emissions while creating revenue through utilizing mineralisation products. Numerous small volume applications have been proposed to use carbonates (e.g., carbonates used as polymer additives or aggregate feedstock for land reclamation projects)^{1,2}. While at first these seem promising, niche markets offer only limited CO₂ storage potential^{1,2}. Furthermore, these markets may not be located close to the cement production facilities³ or have stringent product specifications which can only be achieved at high costs¹. Carbonating only the process-inherent emissions of cement would lead to a production of magnesite/calcite and silica 60x and 200x respectively greater than the current European market (Supplementary Figure 1). Therefore, targeting niche markets such as selling high purity magnesium carbonate (MgCO₃) or calcium carbonate (CaCO₃) and silica, i.e., silicon dioxide (SiO₂), will not significantly lower emissions from cement production. Instead, we consider the use of these materials in cement products.

Supplementary Note 2: Scope definition for the assessment

A typical European cement production facility has an annual capacity of 1.4 million tonnes of cement per annum (Mt_{cement}/a) emitting 850ktCO₂/a⁴, and will be used as the baseline in this work. We propose retrofitting post-combustion capture onto this facility to obtain a pure CO₂ gas with sufficient capacity to meet the demands of a mineralisation system that will be located on the same site. Suitable feedstock minerals occur in distinct locations in south and northern Europe⁵⁻⁷, we assume a transport of 1200 km via a combination of train, ship and truck, which would allow to transport mineral from a site in Norway (Region of Steinsvik) to northern Germany (Region of Hamburg), where multiple cement plants are located⁸. On the cement site the carbonation products are used as supplementary cementitious material (SCM) to create blended cement. The stoichiometry of the carbonation reaction produces inert material (carbonates) with limited application in cementitious products². Unused material is stored in the limestone quarry, which are assumed to be located within 10 km of the cement production facility.

Supplementary Note 3: SCM_{CCU} product specification

The by-product silica (SiO₂) from mineralisation reactions has been described of being of amorphous nature^{6,9,10}. Amorphous silica is a widely accepted pozzolanic additive in cement production. When added to cement, amorphous silica and calcium hydroxide react to produce additional binding products (e.g., calcium silicate hydrates), leading to a comparable or increased strength to using cement alone^{11,12}. Experimental work suggests that replacing 5-30wt% of cement using silica increases the strength by 5-38 weight percent (wt%), with the strongest effect observed at 10-15wt% replacement^{11,13-15}. Initial applicability studies by Benhelal, et al.⁹ showed that a mixture of at least 5wt% carbonated material could be used as an SCM (hereafter SCM_{CCU}) without significant workability (i.e. curing time, water demand) and strength changes of resulting mortars. As such, SCMs must have high concentrations of small particles of silica as the major drivers for the pozzolanic effect^{9,16}. Fine particle silica increases the surface area of the cement, resulting in increased water requirements or the need for a superplasticizer to maintain workability^{11,16}. Commonly, replacing cement with silica does not exceed 10 wt%¹⁷. As the literature review was not conclusive on the precise compositions (i.e., content of silica and inert material in SCM) and the potential share of a subsequently resulting SCM_{CCU} in cement blends, we contacted two experts from this field who considered 25-35wt% replacement of ordinary Portland cement by carbonated material containing 30-50wt% silica to be the upper limit^{18,19}, while maintaining similar product properties as ordinary Portland cement (e.g., compressive strength, curing time, water

demand). As cement is highly standardized we additionally reviewed the European standard for cement products EN-197¹² showing that all cement types currently allow the usage of up to 5wt% of SCM via CO₂ mineralisation (Supplementary Table 2). Additionally, if carbonated minerals can be classified as natural pozzolanic material they could be used as SCM in certain blends (e.g., Portland pozzolana cement). That would allow displacements of up to 35wt% or 55wt% under current product specifications. For using silica in cement blends, which is the reactive component of the SCM_{CCU}, EN-197¹² additionally limits the share of silica in the cement blend to maximal 10wt%.

Following these investigations, we assume a share of 10-25% of SCM_{CCU} in cement with shares of 50%-30% of silica in the SCM_{CCU}: pessimistic scenario: 10% SCM_{CCU} in cement and 50% silica in SCM_{CCU} (total: 5% silica in cement blend); mid scenario: 20% SCM_{CCU} in cement and 40% silica in SCM_{CCU} (total: 8% silica in cement blend); optimistic scenario: 25% SCM_{CCU} in cement and 30% silica in SCM_{CCU} (total: 7.5% silica in cement blend). Within these ranges we expect a blend to be formulated with similar product properties as ordinary Portland cement while respecting the current the European standard for cement products EN-197¹² (i.e., current limits for shares of silica and SCM of standardized cement blends).

Supplementary Note 4: Improved process designs for SCM production

Two pathways for mineral carbonation are typically distinguished⁵: a direct pathway where ground minerals are reacted with CO₂ in a pressurized stirred tank using an aqueous slurry with additives such as NaCl and NaHCO₃^{10,20-23}; and an indirect pathway where alkaline earth metal oxides are first extracted and in a later step reacted with CO₂ to form carbonates. We selected one direct route and one indirect route to analyse and compare their technical and economic performance. We only selected processes that use natural minerals as feedstock: availability of some industrial wastes²⁴ might be limited in the future due to more sustainable production processes, and heavy metal contents of some wastes might limit their usability²⁵. Due to the availability of the mg-rich silicates in Europe⁵⁻⁷, we focused our assessment on olivine and serpentine-bearing feedstocks.

We compared multiple reaction conditions for the selection of a direct route which either use serpentine or olivine bearing rocks and differ in pressure, temperature, additive concentration as well as mechanical pre-treatment. Post-combustion CO₂ capture (i.e., monoethanolamine (MEA) post-combustion capture) was used to capture the CO₂ from the cement kiln flue gas^{20,22,26}. The minerals are first mined, crushed and grinded. In the case of serpentine, thermal pre-treatment was added to activate the minerals. In a next step the water and the additives sodium bicarbonate (NaHCO₃) and sodium chloride (NaCl) are added, and the mixture is compressed to the desired reaction pressure (100-150 bar) and heated in a heat exchanger, recovering the heat from the reaction solution leaving the reactor. To create a process that resembles a large scale process as close as possible we implemented heat integration proposed by Ostovari, et al.² and developed a separation process based on initial findings from Kremer and Wotruba²⁷ that allows us to increase the efficiency of using mineral feedstock and to produce a SCM with alterable silica concentrations. As the reaction of the direct process are not complete, when leaving the reactor, we here propose a post-processing train; first, cyclones are used to partially separate unreacted mineral via gravity separation (<10-75µm, depending on the grinding size)^{21,22} from the reaction products (3-5µm for magnesite and <1µm for silica)²⁷. Unreacted mineral is cycled back into the process to increase the efficiency of utilizing the mineral feedstock. In a second step a share of the resulting product-rich slurry is fed into a classification centrifuge where magnesite and unreacted mineral are separated from the stream. Afterwards this silica-rich slurry is added to the residual slurry, producing a desired silica content. The SCM_{CCU} is dewatered using solid bowl centrifuges, which have been reported

to be suitable for this task²⁸. The SCM_{CCU} is assumed to be used with a residual humidity of 10%. We selected the conditions with the lowest costs (Supplementary Figure 5) leading to a design for a direct carbonation route based on Eikeland, et al.²² (Olivine, 100bar, 190°C).

As one of the most eminent indirect routes we selected a liquid/liquid approach, also referred to as the Nottingham route^{2,29}. The Nottingham route uses serpentine as a feedstock, which is crushed, grinded and reacted in a three-step aqueous process; First, an ammonium bisulphate solution ($\text{NH}_4\text{HSO}_{4\text{aq}}$) is used to extract magnesium oxide and impurities (e.g., iron oxide), leaving silica behind. Second, impurities are precipitated via pH adjustment. Third, the extracted magnesium reacts with ammonium bicarbonate ($\text{NH}_4\text{HCO}_{3\text{aq}}$) which acts as CO_2 carrier and is recycled in an integrated post-combustion capture process, in which ammonia reacts with the flue gas-inherent CO_2 to ammonium bicarbonate. After the separation of the carbonation products, the remainder of the ammonium sulphate-rich solution ($(\text{NH}_4)_2\text{SO}_4$), is evaporated leading to a thermal treatment to recover NH_3 and NH_4HSO_4 ³⁰⁻³³. Here, a jacketed mechanical scraped-surface crystallizer was selected to evaporate water from the additive (ammonium sulphate) and a indirect rotary drum dryer was selected for the thermal treatment. We selected this route for this study because it is possible to obtain high purity separated products² so no purification step has to be designed. We implemented heat integration proposed by Ostovari, et al.² which also incorporates recompression of steam created during the additive regeneration. To produce the SCM_{CCU}, silica is separated from the slurry after the first reaction and blended with separated carbonates from the third reaction. Solid bowl centrifuges are again used for dewatering. Simplified flowsheets are³⁴ shown in Supplementary Figure 2.

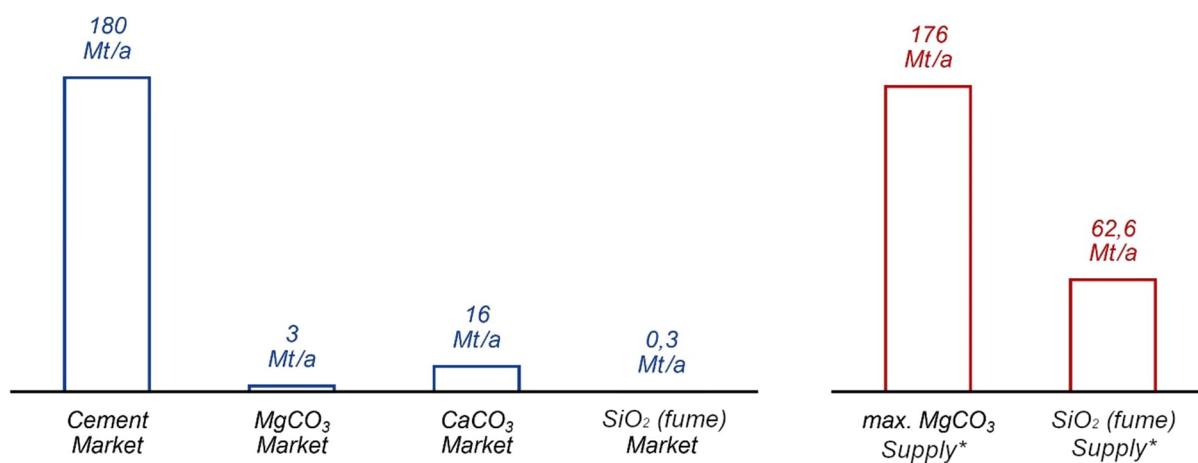
Supplementary Note 5: Comparison of CO_2 mineralisation to other emission reduction strategies

To achieve significant emission reductions, the cement industry is limited in its options due to high process-inherent emissions. A review of academic literature and policy advice reports revealed that a set of strategies will be needed to combat CO_2 emission reduction in the cement industry. Because the cement industry's emissions result from both the high energy demand (i.e., energy-related emissions) and the calcination reaction itself (i.e., process-inherent emissions), we must differentiate between measures to reduce either or both. Tackling energy-related emissions can be achieved using efficiency and substitution measures. While many efficiency gains have been achieved in conventional cement production processes in recent decades³⁵ which tackle energy-related emissions, currently predominantly the use of alternative fuels^{16,35-39} and process electrification⁴⁰ is suggested to further reduce these emissions. Since these measures will at best reduce energy-related emissions to zero, additional measures are needed to tackle the remaining process-inherent emissions (approx. 60 % of total emissions⁴¹). Conventionally, the cement industry used clinker substitutes called supplementary cementitious materials (SCM) to tackle these emissions, which can be industrial wastes (e.g., steel slag, fly ash) or virgin minerals (e.g., limestone, natural pozzolans)^{16,35,39,42,43}. Given overall strength or workability limitations (i.e. increased water requirements and altered curing time) with the use of SCM¹⁶ these measures will not be sufficient to mitigate all process-inherent emissions. An overview of the emission reduction approaches with high emission reduction potentials³⁵ can be summarised as:

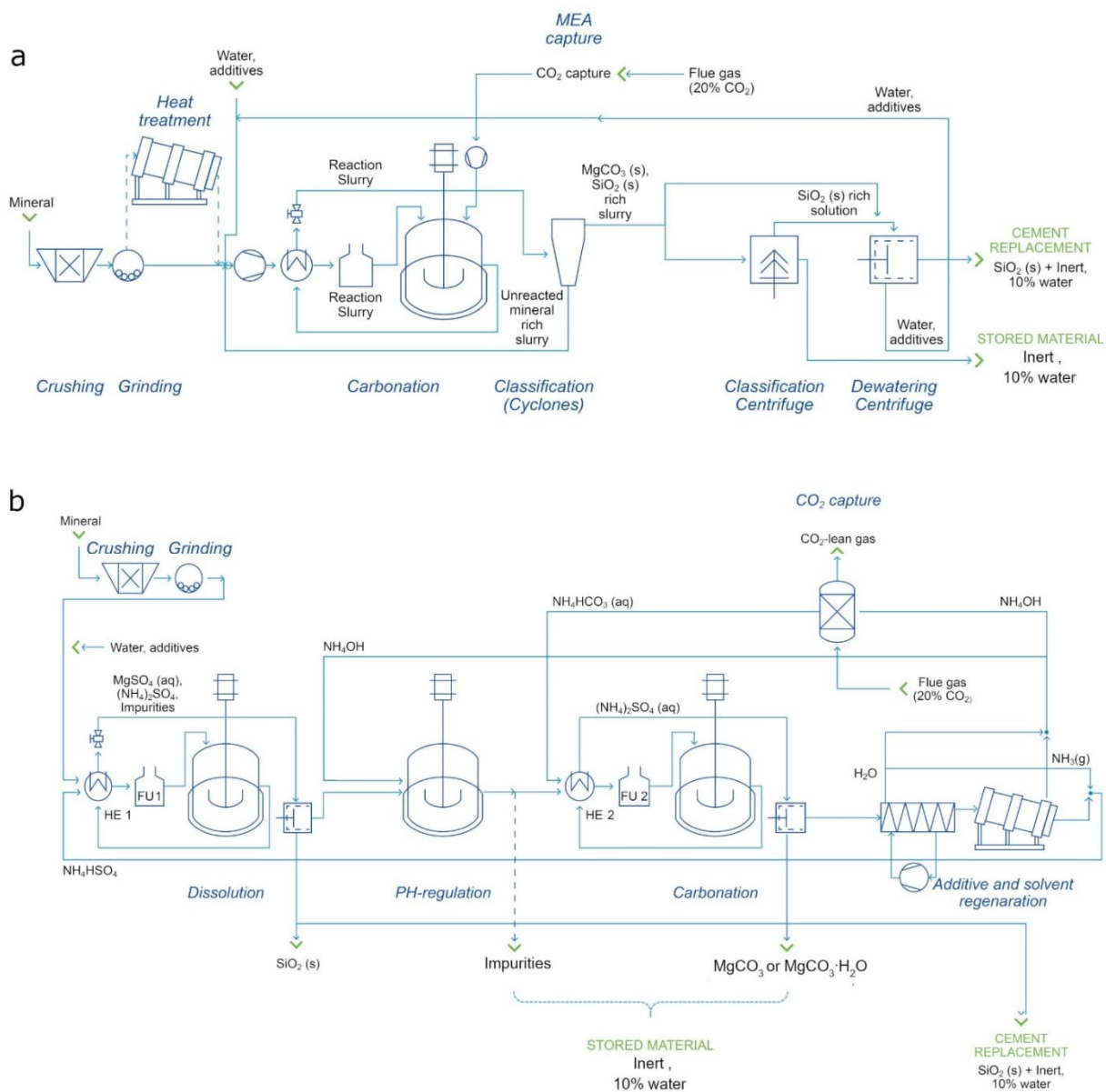
- Alternative fuels or electrification
- Alternative supplementary cementitious materials
- Carbon capture and storage and/or carbon capture and utilisation
- Alternative clinkers

For this assessment we compared CO₂ mineralisation to alternative fuels (i.e., biofuel in form of biomass or biomass by-products), other alternative SCMs (i.e., calcinated clay) and geological storage (i.e., MEA post combustion capture and Oxy fuel combustion with offshore storage). We excluded the use of traditional SCMs from waste streams (fly ash and blast furnace slag) as they have been used extensively to reduce emissions and are unlikely to be able to significantly reduce the emissions further³⁵ as well as alternative clinkers, which are expected to only be used for niche products in the future³⁵. Additionally, we exclude other means of carbon capture and utilisation, where CO₂ is not stored in minerals (e.g., production of fuels) since it does not lead to permanent storage of CO₂. As the baseline we use the production of ordinary Portland cement with a clinker content of 100% (following EN-197¹² this must be >95%, see Supplementary Table 2), as this cement type can be used to produce all other cement blends. All considered processes and scopes are shown in Supplementary Figure 4. The used equations are provided in Supplementary Table 14 and the used assumptions in Supplementary Table 15.

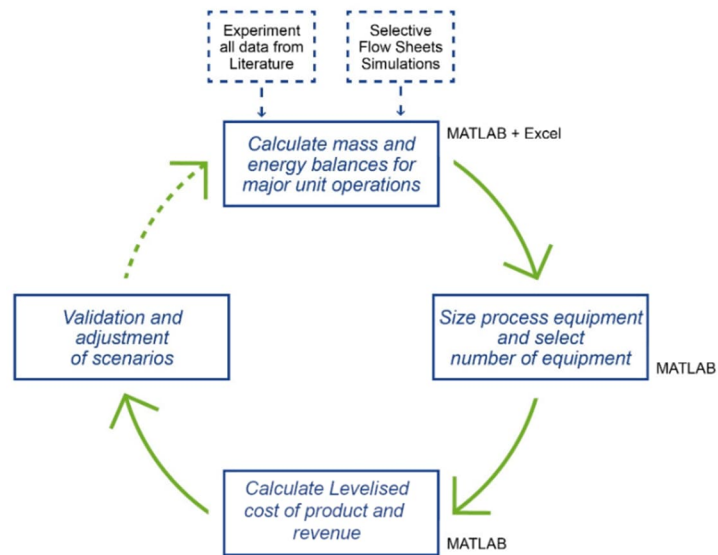
Supplementary Figures



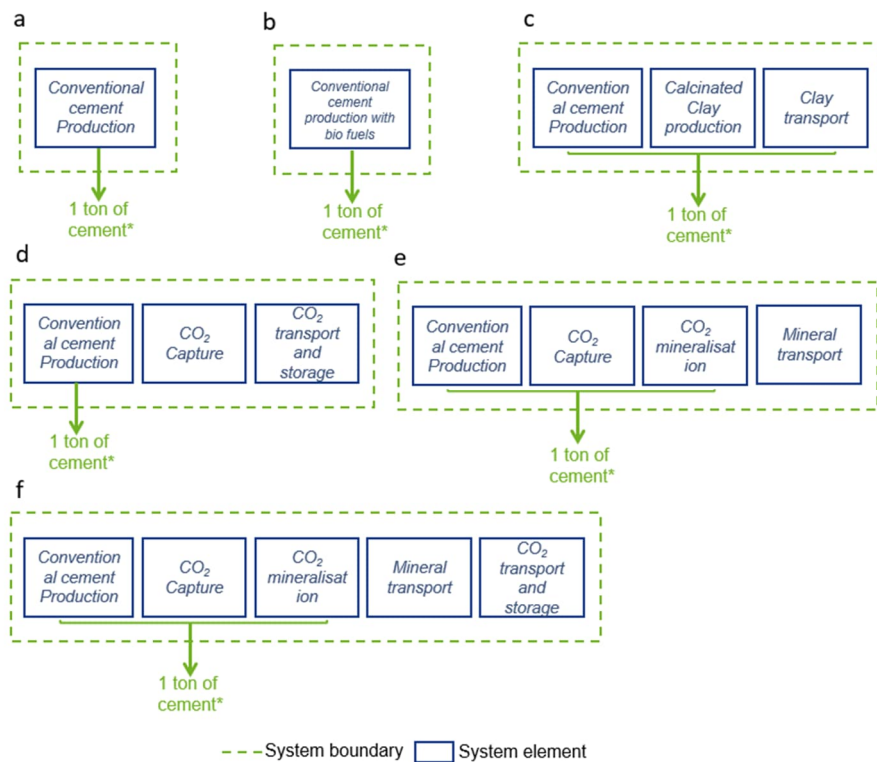
Supplementary Figure 1: Current demands in Europe and the potential supply from CO₂ carbonation; Left, market sizes for cement, magnesium carbonate (MgCO₃), calcium carbonate (CaCO₃) and silica (SiO₂)⁴⁴⁻⁴⁸. Right, supply from potential reaction of CO₂ with forsterite (Mg₂SiO₄); *Stoichiometric reaction yield assumed, only process CO₂ emissions (60% of total emissions) taken into amount. Illustration designed by Wernerwerke, Cordes + Werner GbR ©2021.



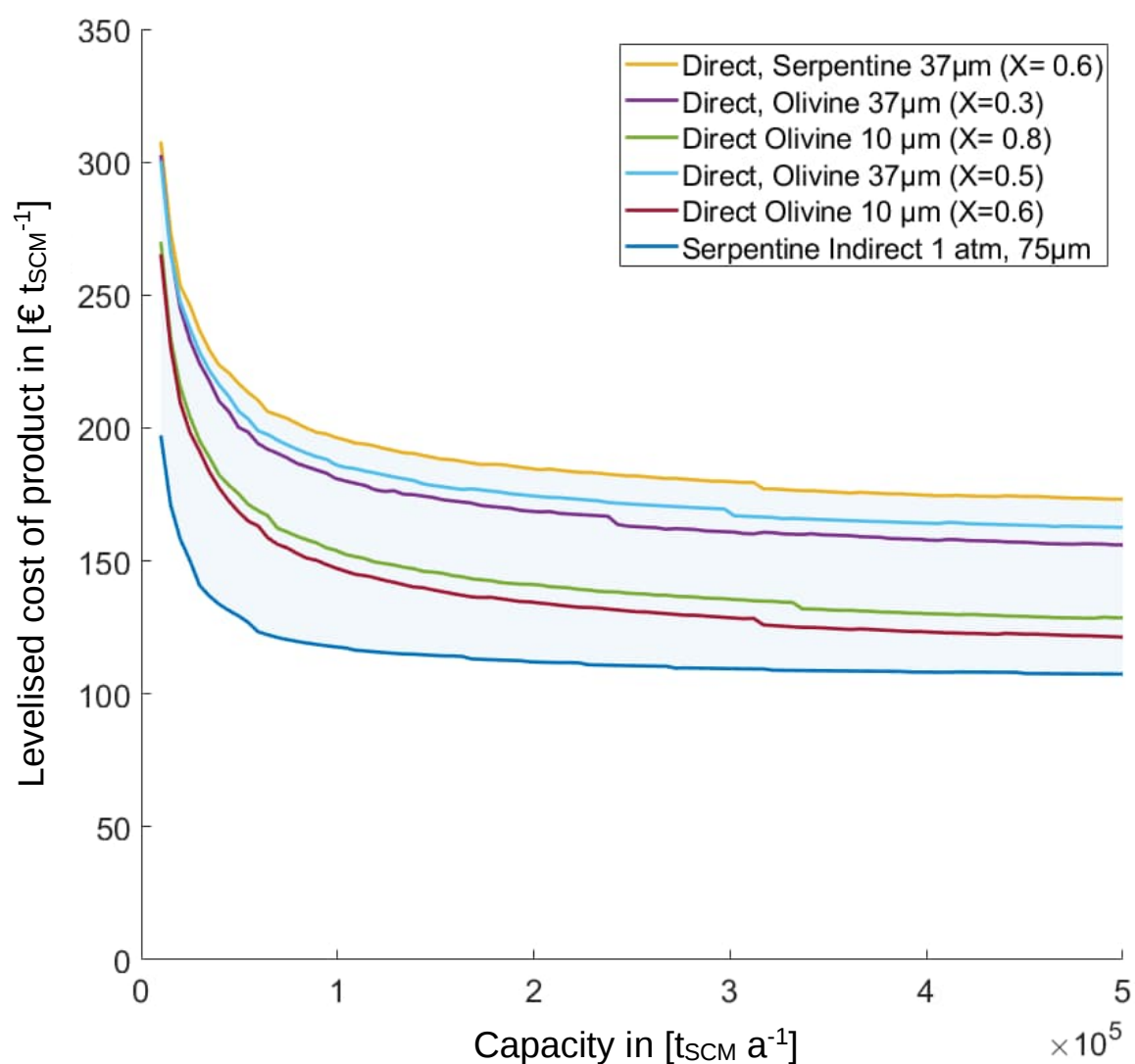
Supplementary Figure 2: Flowsheets of proposed full-scale processes . Panel a :direct process based on Eikeland, et al.²² with heat treatment only being considered when serpentine is used as mineral feedstock. Panel b: indirect process based on Wang and Maroto-Valer³⁰ and Ostovari, et al.². Illustration designed by Wernerwerke, Cordes + Werner GbR ©2021.



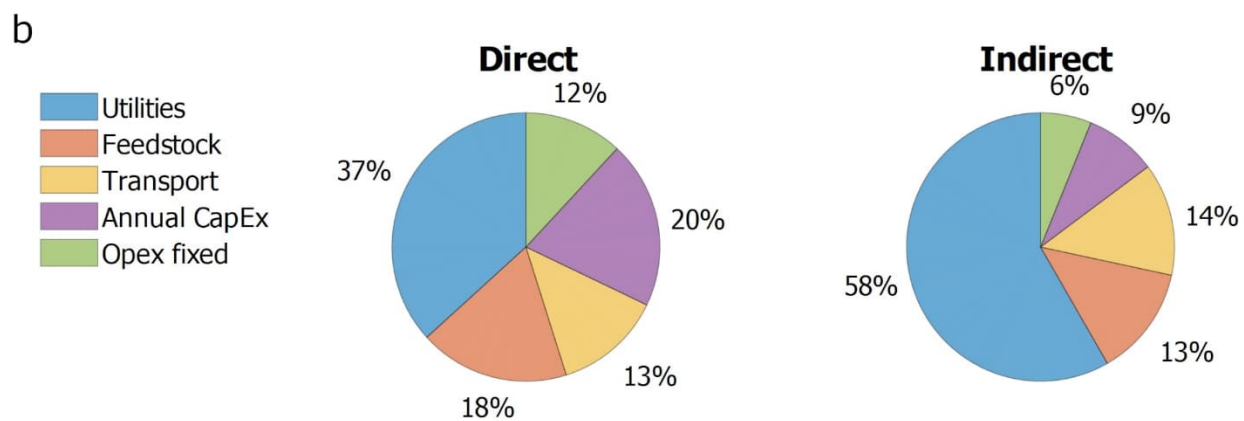
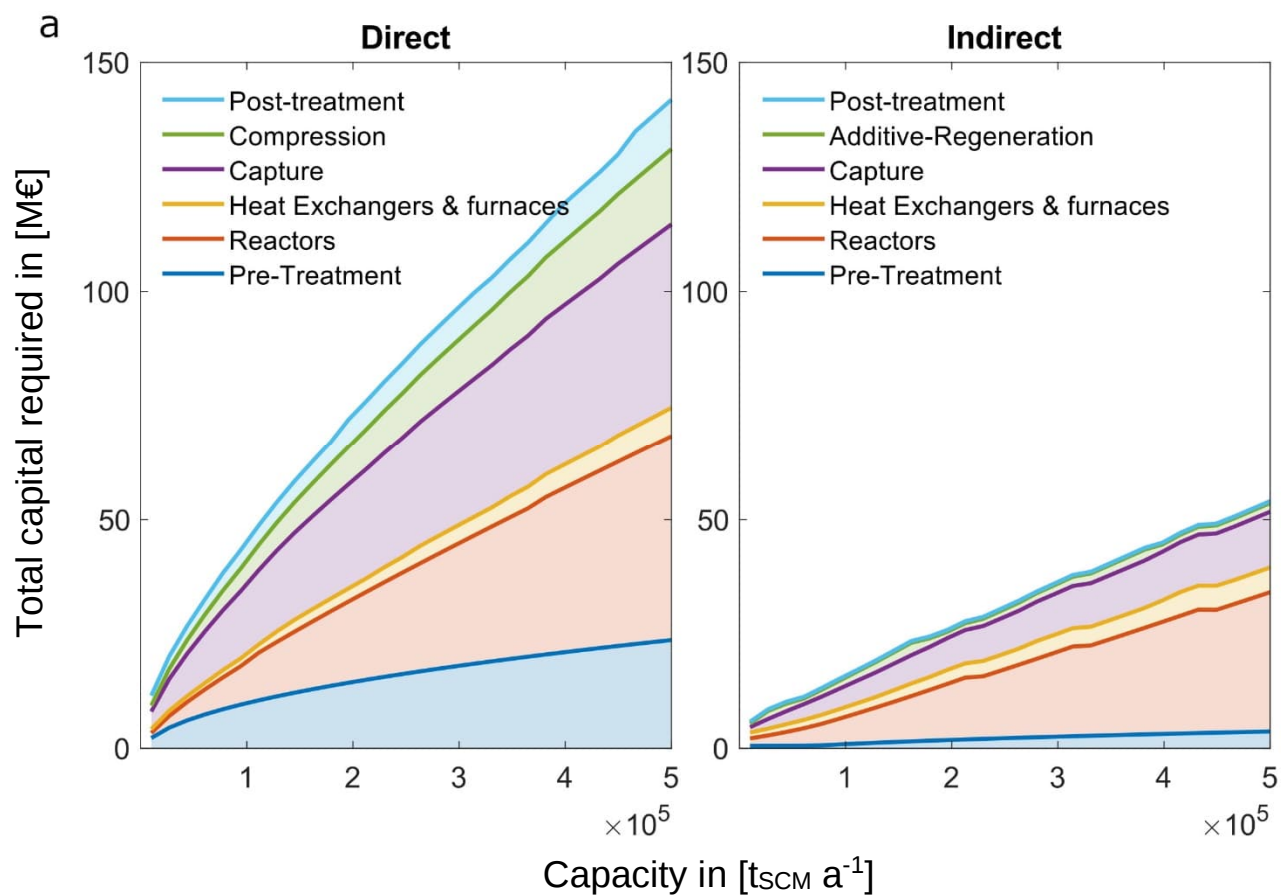
Supplementary Figure 3: Description of the techno-economic modelling sequence used in this study. Illustration designed by Wernerwerke, Cordes + Werner GbR ©2021.



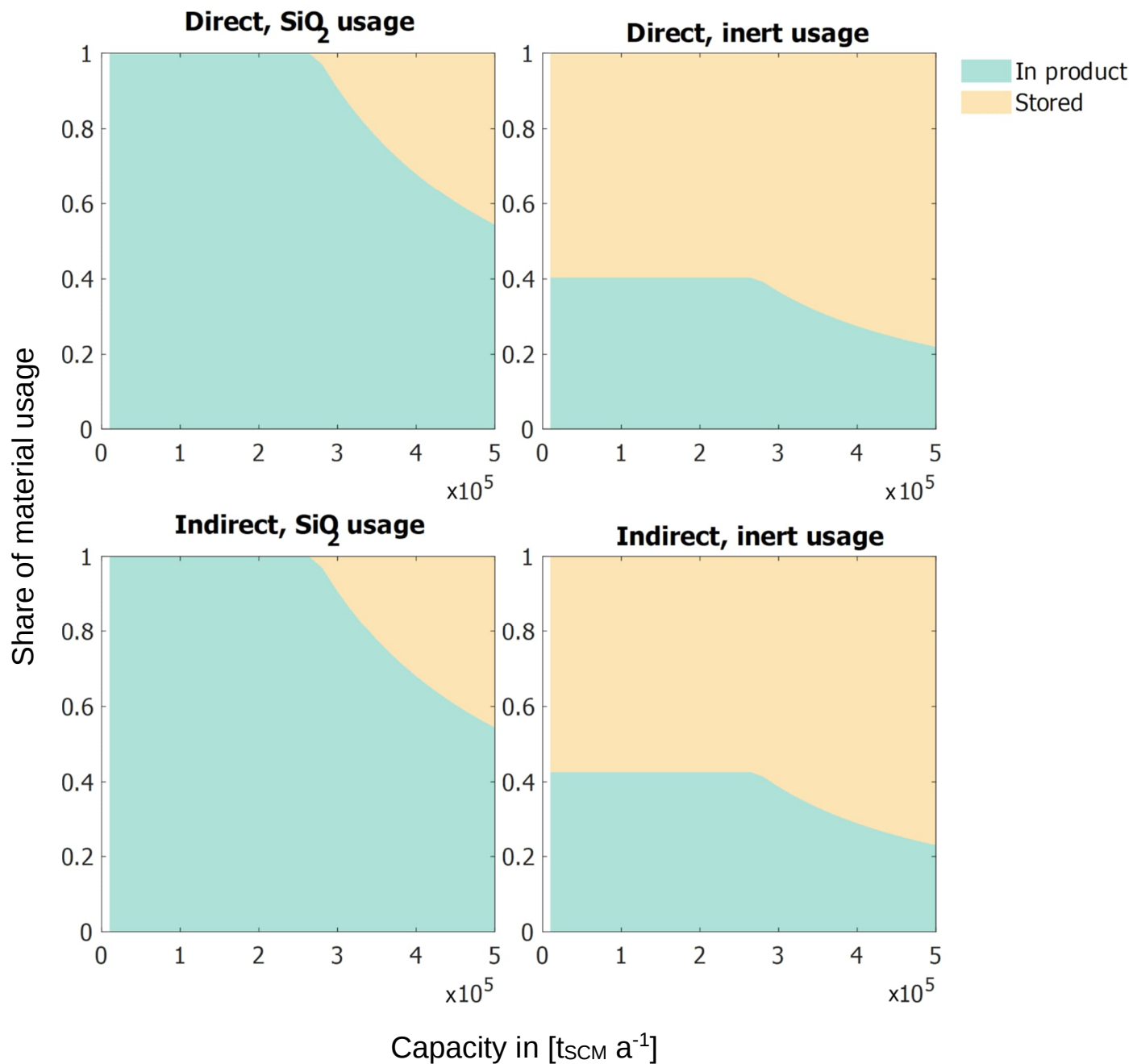
Supplementary Figure 4: Scope of the assessment for the comparison of different CO_{2e} emission reduction measures : panel a: Baseline, panel b: Biofuels, panel c: Calcined clay cement, d: Cement production with CO₂ capture and off-shore geological storage, e: Cement production with CO₂ capture and CO₂ mineralisation, f: Cement production with CO₂ capture and off-shore geological storage and CO₂ mineralisation. *The functional unit is defined as 1 ton of cement with the equivalent performance of 1 ton of ordinary Portland cement.



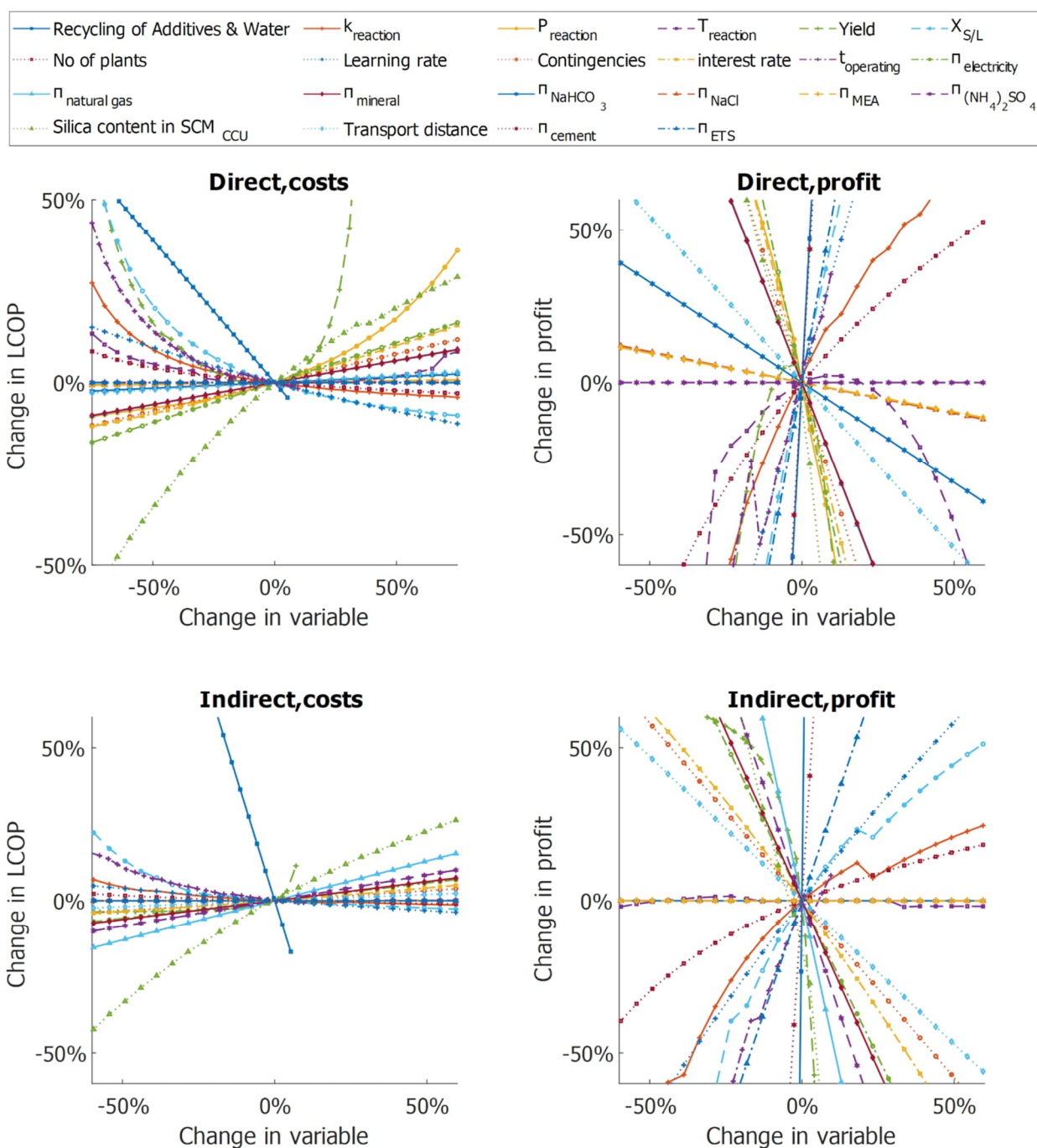
Supplementary Figure 5: Cost comparison of different process routes and mid scenario . Process assumptions are shown in Supplementary Table 3. Note all mentioning in the main article of “direct process” refers to “Direct, Olivine, 10µm (X=0.6)” and “indirect process” refers to “Indirect, Serpentine 75µm”. Adapted from Strunge ⁴⁹.



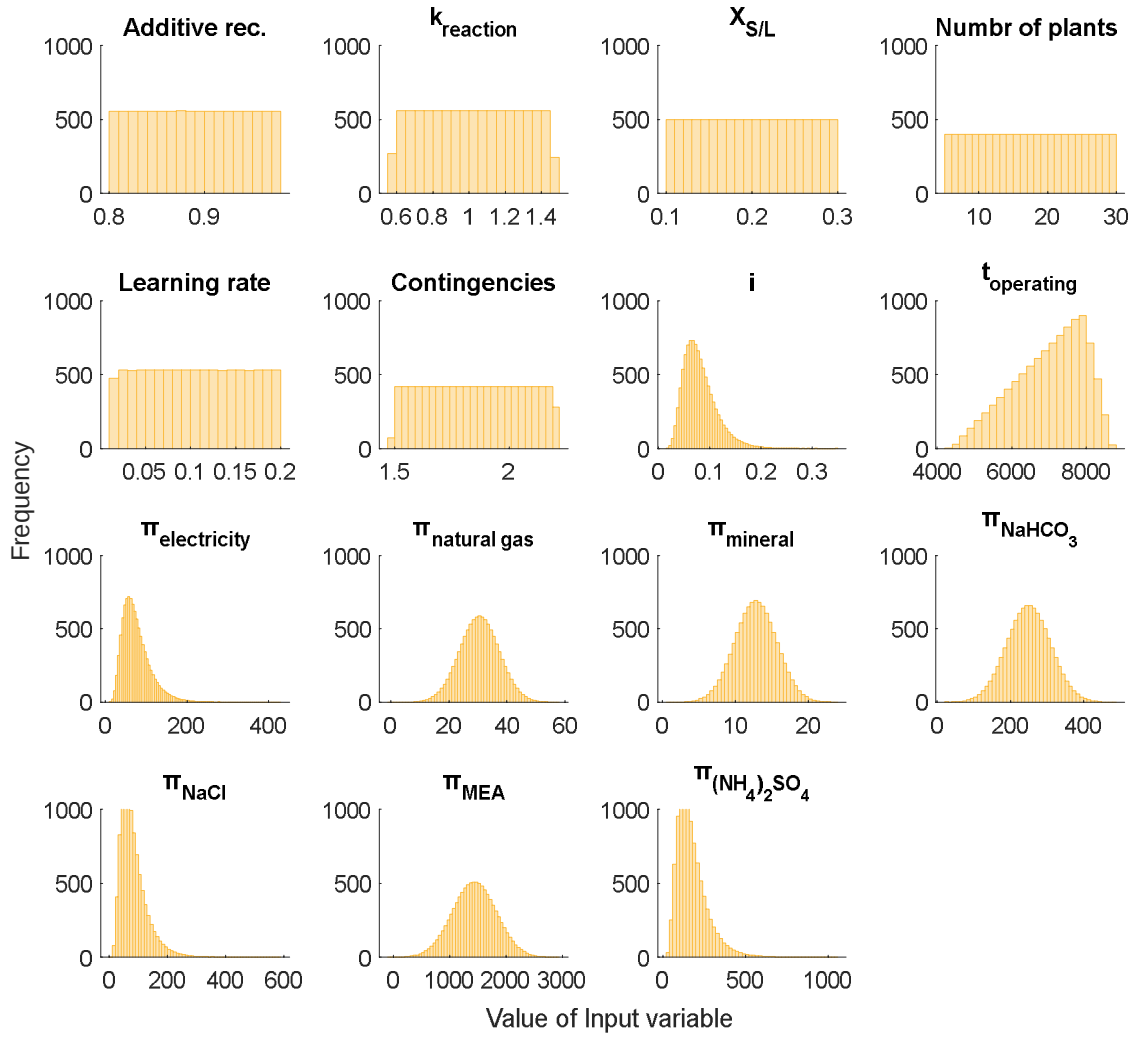
Supplementary Figure 6: Distribution of costs for the mid scenario: Panel a: Total capital required; Panel b: distribution of levelised costs of product.



Supplementary Figure 7: Material use for the mid scenario. Note, this figure displays the amount of material being used for the production of SCM_{CCU} (In product) and how much material has to be stored (Stored). Inert material consists of unreacted mineral, carbonate and impurities.



Supplementary Figure 8: Sensitivity analysis with the assumptions of the mid scenario and changing input parameters: additive recovery (Recycling of Additives & Water), reaction constant (k_{reaction}), reaction pressure (P_{reaction}), reaction temperature (T_{reaction}), reaction yield (Yield), solid-liquid ratio in reactor ($X_{\text{S/L}}$), Number of plants to reach NOAK (No of plants), Learning rate on CAPEX (Learning rate), combined process and project contingencies (Contingencies), interest rate on capital (i), operation hours per year ($t_{\text{operating}}$), electricity price ($\pi_{\text{electricity}}$), natural gas price ($\pi_{\text{natural gas}}$), mineral price (π_{mineral}), Sodium bicarbonate price (π_{NaHCO_3}), Sodium chloride price (π_{NaCl}), monoethanolamine price (π_{MEA}), Ammonium sulphate price ($\pi_{(\text{NH}_4)_2\text{SO}_4}$), content of SiO_2 in product (Silica content in SCM_{CCU}), distance of feedstock minerals (Transport distance), cement price (π_{cement}), ETS price (π_{ETS}). Note that only one factor is changed at a time even when related (e.g. an increase in pressure does not change the yield). Hence, for the Monte Carlo Simulations only the reaction constant is used to cover the reaction conditions (see “Sensitive factors to lower costs” in the main article).



Supplementary Figure 9: Probability density functions for the input parameters for the MonteCarlo simulations using 10,000 runs: additive recovery (Additive Rec.), reaction constant (k_{reaction}), solid-liquid ratio in reactor ($X_{S/L}$), Number of plants to reach NOAK (No of plants), Learning rate on (Learning rate), combined process and project contingencies (Contingencies), interest rate on capital (i), operation hours per year ($t_{\text{operating}}$), electricity price ($\pi_{\text{electricity}}$), natural gas price ($\pi_{\text{natural gas}}$), mineral price (π_{mineral}), Sodium bicarbonate price (π_{NaHCO_3}), Sodium chloride price (π_{NaCl}), monoethanolamine price (π_{MEA}), Ammonium sulphate price ($\pi_{(\text{NH}_4)_2\text{SO}_4}$).

Supplementary Tables

Supplementary Table 1: Literature review of existing economic assessments in the field of CO₂ carbonation

Author	Costs	Unit	Comment	Feedstock
Sanna, et al. ⁵	44-264*	€ tCO ₂ , sequestered ⁻¹	/	mg -silicates
Hitch and Dipple ⁵⁰	73* ⁺	€ tCO ₂ , sequestered ⁻¹	direct	mg -silicates
IEA GHG 2000	95*	€ tCO ₂ , avoided ⁻¹	direct	mg-silicate
IEA GHG 2000	>150*	€ tCO ₂ , avoided ⁻¹	indirect	mg-silicate
Naraharisetti, et al. ⁵¹	410* [#]	€ tCO ₂ , avoided ⁻¹	direct	olivine
Naraharisetti, et al. ⁵¹	443* [#]	€ tCO ₂ , avoided ⁻¹	direct	olivine
W.K. O'Connor and L.R. Penner ²¹	65*	€ tCO ₂ , avoided ⁻¹	direct	olivine
Naraharisetti, et al. ⁵¹	302* [#]	€ tCO ₂ , avoided ⁻¹	direct	serpentine
W.K. O'Connor and L.R. Penner ²¹	258*	€ tCO ₂ , avoided ⁻¹	direct	serpentine
Sanna, et al. ¹	16-75* ⁺	€ tCO ₂ , sequestered ⁻¹	post processing only	serpentine
Naraharisetti, et al. ⁵¹	349* [#]	€ tCO ₂ , avoided ⁻¹	direct	serpentine
Huijgen, et al. ⁵²	77	€ tCO ₂ , avoided ⁻¹	direct	steel slag
Katsuyama, et al. ⁵³	128-284*	€ tCaCO ₃ ⁻¹	/	waste cement
Iizuka, et al. ⁵⁴	25* ⁺⁺	€ tCO ₂ , avoided ⁻¹	indirect	waste cement
Naraharisetti, et al. ⁵¹	304* [#]	€ tCO ₂ , avoided ⁻¹	direct	wollastonite
W.K. O'Connor and L.R. Penner ²¹	93*	€ tCO ₂ , avoided ⁻¹	direct	wollastonite
Huijgen, et al. ⁵²	102	€ tCO ₂ , avoided ⁻¹	direct	wollastonite
Kakizawa et al 2001	57* ⁺⁺	€ tCO ₂ , avoided ⁻¹	indirect	wollastonite
Pérez-Fortes, et al. ⁵⁵	M65.5 [§]	€ tCO ₂ , sequestered ⁻¹	direct	fly ash
Mehler, et al. ⁵⁶	228	€ tCO ₂ , avoided ⁻¹	direct	fly ash
Pedraza, et al. ⁵⁷	33-39*	€ tCO ₂ , sequestered ⁻¹	direct	Cement kiln dust
* Currency adapted to € # Cost of CO₂ capture included + Only operational expenditures assessed § Only capital expenditures assessed ++ Only operational expenditures in the form of power generation assessed and revenue from CaCO₃ included.				

Supplementary Table 2: Review of carbonated material use in standardized cement products following EN 197¹². *
Carbonated material has to be acknowledged as a natural pozzolan and must exhibit a silica content of at least 25 wt%.

Description		Max SCM _{CCU} allowance in current standard
CEM I	Portland cement	5%
CEM II	Portland-slag cement	5%
	Portland-silica fume cement	5%
	Portland pozzolana cement	20-35%*
	Portland-fly ash cement	5%
	Portland burnt shale cement	5%
	Portland limestone cement	5%
	Portland composite cement	20-35%*
CEM III	Blast furnace cement	5%
CEM IV	Pozzolanic cement	35-55%*
CEM V	Composite cement	30-50%*

Supplementary Table 3: Process assumptions

Description	Direct, Serpentine 37µm (X= 0.6) ²¹	Direct, Olivine 37µm (X= 0.3) ²¹	Direct, Olivine 37µm (X= 0.5) ²¹	Direct*, Olivine 10 µm (X= 0.6) ²²	Direct, Olivine 10 µm (X= 0.8) ²²	Indirect ^{30,32,33*}
Yield	0.6	0.3	0.5	0.6	0.8	Overall: 0.87 Mineral diss.: 0.91 PH-Adjus.: 1.0 Carbonation: 0.95
particle size [µm]	37	37	37	10	10	75
P [bar]	115	150	150	100	100	1
T [°C]	155	185	185	190	190	80
C_{NaHCO3} [mol/l]	0.64	0.64	0.64	0.5	0.5	/
C_{NaCl} [mol/l]	1	1	1	0.75	0.75	/
C_{(NH4)2SO4} [mol/l]	/	/	/	/	/	5.96
Reaction rate constant	0.1822**	0.3022**	0.3022**	1.4717**	1.4717**	Mineral diss.:1.2839** Carbonation: 4.186**
* Selected as routes for this study (referred to as “direct process” and “indirect process”).						
**derived regression of published lab data ^{21,22,30,32,33}						

Supplementary Table 4: Equations used for mass balance of feed recycling and separation in direct process.

Use	Formula
Recycling unreacted mineral, direct aqueous process	$\dot{m}_{Mg2SiO4,theo} = \frac{\dot{m}_{CO2,reacted}}{M_{CO2}} * \frac{V_{Mg2SiO4}}{V_{CO2}} * M_{Mg2SiO4}$ $\dot{m}_{Mg2SiO4,real} = \dot{m}_{Mg2SiO4,theo} * \frac{1}{Yield} - \dot{m}_{Mg2SiO4,theo} * \left(\frac{1}{Yield} * (1 - Yield) * recovery\ factor \right)$

**Split of
stream for
classificati
on
centrifuge**

$$z = \frac{1 - \left(\frac{1-x}{x}\right) * \left(\frac{y}{1-y}\right) + \left(\frac{1-y}{y}\right) - \left(\frac{1-x}{x}\right)}{1 + \left(\frac{1-y}{y}\right)}$$

x = content of SiO_2 in final product
 y = content of SiO_2 in cyclone output
 z = split of stream fed to classification centrifuge

Supplementary Table 5: Used Equations for Density and viscosity

Description	Equation	Reference
Density slurry	$\rho_{\text{slurry}} = \frac{100}{\left(\frac{X_{\text{solid,liquid}}}{\rho_{\text{solid}} \cdot 1000}\right) + \left(\frac{100 - X_{\text{solid,liquid}}}{\rho_{\text{H}_2\text{O}} \cdot 1000}\right)}$	Shashi ⁵⁸
Viscosity slurry	$v_{\text{slurry}} = v_{\text{H}_2\text{O}} \cdot \left(1 + 2.5 \cdot V_{\text{fraction}} + 10.06 \cdot V_{\text{fraction}}^2 \cdot 0.00273 \cdot \exp(16.6 \cdot V_{\text{fraction}})\right)$	Shashi ⁵⁸
Density ABS_(aq)	$\rho_{\text{ABS(aq)}} = 0.99671 + (0.604706 \cdot 10^{-1}) \cdot c_{\text{ABS}} - (0.108981 \cdot 10^{-2}) \cdot c_{\text{ABS}}^2 + (0.106394 \cdot 10^{-4}) \cdot c_{\text{ABS}}^3$	Stelson and Seinfeld ⁵⁹
Density AS_(aq)	$\rho_{\text{ABS(aq)}} = 0.0771 \cdot c_{\text{ABS}}^{0.8463} + 0.9945$	Adapted from The Engineering ToolBox ⁶⁰
Density MS_(aq)	$\rho_{\text{MS}} = 0.9943 + -0.001323 \cdot c_{\text{wt\%,MS}} + 0.003697 \cdot T$	handymath.com ⁶¹

Supplementary Table 6: Heat capacity calculation for different compounds : The integral of the heat capacity is

derived using the Shomate equation: $\int_{T_0=25^\circ\text{C}}^T \text{cp } dT = a \cdot (T - T_0) + b \cdot \frac{T^2 - T_0^2}{2} + c \cdot \frac{T^3 - T_0^3}{3} + d \cdot \frac{T^4 - T_0^4}{4} - e \cdot ((T)^{-1}) - ((T_0)^{-1})$

Description	a	b	c	d	e	Reference
H₂O_(l)	-203.606	1523.29	-3196.41	2474.455	3.855326	Chase ⁶²
Fe₂(SO₄)_{3(s)}	85.69794	803.9765	-695.757	215.6099	-0.396145	Chase ⁶²
CO_{2(g)}	24.99735	55.18696	-33.6914	7.948387	-0.136638	Chase ⁶²
NH_{3(g)}	19.99563	49.77119	-15.376	1.921168	0.189174	Chase ⁶²
SiO₂	-6.076591	251.6755	-324.796	168.5604	0.002548	Chase ⁶²
Fe(OH)_{3(s)}	56.70701	132.1357	-83.5997	17.25465	0.711267	Chase ⁶²
MgCO_{3(s)}	44.937	149.7085	-74.1827	11.9767	-0.629261	Chase ⁶²
H₂O_(g)	30.092	6.832514	6.793435	-2.53448	0.082139	Chase ⁶²
MgO	47.25995	5.681621	-0.872665	0.1043	-1.053955	Chase ⁶²
Fe₂O₃	93.43834	108.3577	-50.86447	25.58683	-1.61133	Chase ⁶²
NH₄OH_(l)	9.8	-	-	-	-	Southard and Green ⁶³
Mg₃Si₂O₅·(OH)_{4(s)}	1.09	-	-	-	-	Engineering Toolbox ⁶⁰
MgSO_{4(l)}	111.713	-	-	-	-	Southard and Green ⁶³

NH_4HSO_4 (l)	1.423	-	-	-	-	National Center for Biotechnology Information ⁶⁴
$(\text{NH}_4)_2\text{SO}_4$ (l)	1.419	-	-	-	-	MatWeb ⁶⁵
NH_4HCO_3 (l)	1.119	-	-	-	-	Rademaekers, et al. ³⁴

Supplementary Table 7: Equations for electricity consumption

Description	Equation	Reference
Crushing	$w_{\text{crushing}} = 2 \cdot \dot{m}_{\text{mineral}}$	Gerdemann, et al. ²⁰
Grinding	$w_{\text{grinding}, \leq 75\mu\text{m}} = 10 \cdot K_{\text{Bond}} \cdot \left(\frac{1}{\sqrt{75}} - \frac{1}{\sqrt{d_{\text{mineral, feed}}}} \right) \cdot \dot{m}_{\text{mineral}}$	Schwister and Leven ⁶⁶
	$w_{\text{grinding}, 75\mu\text{m to } 37\mu\text{m}} = 70.31 \cdot \dot{m}_{\text{mineral}}$	Gerdemann, et al. ²⁰
	$w_{\text{grinding}, 37\mu\text{m to } 10\mu\text{m}} = 150 \cdot \dot{m}_{\text{mineral}}$	Hangx and Spiers ⁶⁷
Slurry Pumping	$w_{\text{pump}} = \left(\frac{\left(\left(\frac{V_{\text{slurry}}}{\tau \cdot 3600} \right) \right) \cdot g_{\text{earth}} \cdot \text{pump}_{\text{head}} \cdot \frac{\rho_{\text{slurry}}}{\eta_{\text{pump}}}}{1000} \right) \cdot t_{\text{operating}}$	Costa, et al. ⁶⁸
Slurry stirring	$w_{\text{stirring}} = \frac{V_{\text{slurry}} \cdot \text{rpm}^2 \cdot t_{\text{operating}} \cdot \nu_{\text{slurry}}}{1000}$	Costa, et al. ⁶⁸
CO₂ compression (5 stage)	$w_{\text{comp}} = (17.501 \cdot \ln(p) + 12.876) \cdot \dot{m}_{\text{CO}_2}$	Aspen Plus Simulation
Steam compression	$w_{\text{compression}} = (52.897 \cdot p - 52.668) \cdot \dot{m}_{\text{steam}}$	Aspen Plus Simulation
Solid bowl centrifuge	$w_{\text{centrifuge}} = (P_{\text{solid}} + P_{\text{water}} + P_{\text{empty}}) \cdot \eta_{\text{motor}} \cdot t_{\text{operation}}$ $P_{\text{solid}} = m_{\text{solid}} \cdot (2 \cdot \pi \cdot u_{\text{bowl}} \cdot r_{\text{exit, solid}})^2$ $P_{\text{liquid}} = m_{\text{solid}} \cdot (2 \cdot \pi \cdot u_{\text{bowl}} \cdot r_{\text{exit, water}})^2$ $P_{\text{loss}} = m_{\text{shell}} \cdot f_{\text{drag}} \cdot g_{\text{earth}} \cdot 2 \cdot \pi \cdot u_{\text{bowl}} \cdot r_{\text{shell}}$	Adapted from Bell ⁶⁹
Disc centrifuge	$w_{\text{centrifuge}} = 1.9339 \cdot \exp(0.0039 \cdot \text{diameter})$	Regression from specification sheets

Supplementary Table 8: Used equations for deriving the total direct costs , derived from Aspen capital cost estimator.

Equipment	Limits	Function	Coefficients	R ²
Centr. Pump	V = 10-150L/s P= 10-150bar	$\text{TDC} = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = -5.534e+04 b = 3.169e+04 c = 9.356 d = 19.64	98.81%

			m = 2.306 n = 0.409	
Ball Mill	Power = 290- 21540 kW	$TDC = a \cdot x^n + b$	a = 1.233e+05 b = -9.006e+05 n = 0.4977	99.89%
Furnace	V = 50- 150L/s Duty = 100- 1000MW	$TDC = a + b \cdot x^n + c \cdot y^m$	a = 2.37e+05 b = 6686 c = 7.102 m = 1.988 n = 0.5224	99.91%
Cone Crusher	m = 75- 300t/h	$TDC = a \cdot x^n + b$	a = 5040 b = 3.818e+04 n = 0.7435	99.83%
Agitator	Power = 10-75kW	$TDC = a \cdot x^n + b$	a = 909 b = 1.753e+04 n = 1.084	99.84%
Reactor	V = 50- 200m3 P = 20- 140bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 3.153e+05 b = 0.2124 c = 437.7 d = 90.18 m = 1.347 n = 2.698	99.85%
Heat exchanger max 10 bar	A = 25- 75m2	$TDC = a \cdot x^n + b$	a = 462 b = 7.943e+04 n = 1	90.05%
	A = 100- 200m2	$TDC = a \cdot x^n + b$	a = 169.6 b = 1.261e+05 n = 1	99.24%
	A = 225- 500m2	$TDC = a \cdot x^n + b$	a = 164.5 b = 1.577e+05 n = 1	98.64%
	A = 550- 900m2	$TDC = a \cdot x^n + b$	a = 114.2 b = 2.074e+05 n = 1	98.71%
	A = 950- 1200m2	$TDC = a \cdot x^n + b$	a = 115 b = 2.423e+05 n = 1	100.00%
Heat- exchanger 10-100 bar	A = 100- 200m2 P = 10- 100bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 1.257e+05 b = 4.525 c = 0.02784 d = 1.000 m = 3.284 n = 1.705	99.42%
	A = 225- 500m2 P = 10- 100bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 1.441e+05 b = 44.82 c = 16.96 d = 1.000 m = 1.999 n = 1.242	97.97%
	A = 600- 900m2	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 1.915e+05 b = 14.7 c = 23.76	98.40%

	P = 10-100bar		d = 1.000 m = 2.000 n = 1.318	
	A = 950-1200m2 P = 10-100bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 9.173e+07 b = -9.052e+07 c = -2.057e+05 d = 6.274 m = 0.1789 n = 0.0009788	98.21%
Heat-exchanger 125-150bar	A = 100-200m2 P = 125-150bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 2.474e+05 b = 14.09 c = 31.46 d = 4.678 m = -19.47 n = -5.865	99.03%
	A = 225-500m2 P = 125-150bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 2.417e+05 b = 999.2 c = 5.187 d = 2.141 m = 2.000 n = 0.8516	99.93%
	A = 600-900m2 P = 125-150bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = -1.478e+05 b = 1081 c = 1.899e+05 d = 2.302 m = 0.2334 n = 0.7631	95.76%
	A = 950-1200m2 P = 125-150bar	$TDC = a + b \cdot x^n + c \cdot y^m + d \cdot x \cdot y$	a = 4.067e+05 b = 308.1 c = 3.595e+04 d = 2.465 m = 0.3395 n = 0.9353	99.85%
Mechanical scraped-surface crystalliser	l = 8-150m	$TDC = a + b \cdot x^n$	a = 2238 b = 9615 n = 0.8106	100.00%
Indirect rotary dryer	A = 10-185m ²	$TDC = a + b \cdot x^n$	a = 69.7 b = 6513 n = 0.9392	100.00%
Hydro-cyclones	d = 100-290mm pcs per manifold = 10-100	$TDC = a + b \cdot x^n + c \cdot y^m$	a = -0.4077 b = 27.36 c = 0.0003486 m = 1.616 n = -0.3809	99.80%
	d=300-370mm pcs per manifold = 10-100	$TDC = a + b \cdot x^n + c \cdot y^m$	a = 6.806 b = 1880 c = 0.0001596 m = 1.741 n = -2.428	99.72%
	d=375-500mm pcs per	$TDC = a + b \cdot x^n + c \cdot y^m$	a = -47.25 b = 77.37 c = 0.001123	99.24%

	manifold = 10-100		m = 1.443 n = -0.06914	
Solid bowl centrifuge	d = 450-1250mm l = 750-3000mm	$TDC = a + b \cdot x \cdot y + c \cdot y^m$	a = 4.192e+04 b = 0.1509 c = 13.29 m = 1.384	99.75%
Highspeed disc centrifuge	d = 37.5-125cm	$TDC = a + b \cdot x$	a = 1.848e+05 b = 4.784e+05	99.98%

Supplementary Table 9: Major equipment design assumptions

Equipment	Description	Reference
Reactor	Max size 500 m ³ (for p > 50 bar) Max size 1000 m ³ (for p < 10 bar) Max wall thickness 0.2m Gas phase in reactor: 0.3 (direct), 0.05(indirect) Joint efficiency 1 Vessel security factor 0.1	Own estimation, Towler and Sinnott ⁷⁰
Centrifugal Pump and compressor	Pump efficiency 0.80 Isentropic compressor efficiency 0.85	Anantharaman, et al. ⁴
Heat exchanger	Minimal Temp difference 10K Heat transfer coefficient liquid-liquid 675 W/m ² K Heat transfer coefficient liquid-gas 40 W/m ² K Heat transfer coefficient steam evaporation 1950 W/m ² K Heat transfer coefficient indirect dryer 293 W/m ² K	Njeng, et al. ⁷¹ , The Engineering ToolBox ⁷²
Hydro Cyclone	Separation efficiency unreacted mineral 0.6 Max pressure drop 0.3 bar Max number per manifold 100	Parametric study in Aspen Plus
Classification Centrifuge	Separation efficiency Silica 0.85 Purity of Silica stream 0.99 Speed 4000 rpm	Parametric study in Aspen Plus
Dewatering Centrifuge	Wall thickness 0.02m Speed 3000 rpm	Own estimation, Parametric study in Aspen Plus

Supplementary Table 10: Assumptions carbon footprint reduction assessment : the assessment was adapted from Ostovari, et al.². Multiple values are not displayed, as they belong to propriety datasets. Some propriety data shown here has before been published in Ostovari, et al.² with the approval of the GaBi.

Description	Name of data set	Database / Reference	Year	Value	
Electricity grid emission	Electricity grid mix [EU-28]	GaBi ts ⁷³	2016	417	kg _{CO2e} MWh ⁻¹
Natural gas emission	Thermal energy from natural gas [EU-28]	GaBi ts ⁷³	2016	241.0	kg _{CO2e} MWh ⁻¹
Transport train	Rail transport, average train, gross tonne weight 1000t / 726t payload capacity [EU-28]	GaBi ts ⁷³	2016	0.0258	kg _{CO2e} km ⁻¹ t ⁻¹
Transport truck	Transport, small truck (up to 14 t total cap., 9.3t payload) [EU-28]	GaBi ts ⁷³	2016	0.0791	kg _{CO2e} km ⁻¹ t ⁻¹

Transport ship	/	Psaraftis and Kontovas ⁷⁴	/	0.033	kg _{CO2e} km ⁻¹ t ⁻¹
Water	Process water [EU-28]	GaBi ts ⁷³	2016	/	kg _{CO2e} t ⁻¹
NaHCO₃	Sodium bicarbonate	GaBi ts ⁷³	2017	/	kg _{CO2e} t ⁻¹
NaCl	Sodium chloride (rock salt) [EU-28]	GaBi ts ⁷³	2016	/	kg _{CO2e} t ⁻¹
(NH₄)₂SO₄	Ammonium sulfate production [RoW]	Ecoinvent 3.71 ⁷⁵	2019	/	kg _{CO2e} t ⁻¹
Monoethanol-amine	/	Pehnt and Henkel ⁷⁶	/	/	kg _{CO2e} t ⁻¹
Construction	/	Ostovari, et al. ²	/	0.04	kg _{CO2e} t _{CO2stored} ⁻¹
Mining Mineral	/	Ostovari, et al. ²	/	5.23	kWh t ⁻¹
Cement production	/	Voldsund, et al. ⁷⁷	/	850	kg _{CO2e} t _{clinker} ⁻¹

Supplementary Table 11: Capital cost factor assumptions

Description	Value	Basis	Reference
Indirect costs	14%	TDC	Anantharaman, et al. ⁴
Process contingencies	40%	EPC	EPRI ⁷⁸ , AACE ⁷⁹
Project contingencies	30%	EPC	EPRI ⁷⁸
Owner's costs	7%	TPC	Grande, et al. ⁸⁰
Learning rate	10.55%	/	Rubin, et al. ⁸¹
Number of plants for NOAK	20	/	Greig, et al. ⁸²
Lifetime of the plant	30 years	/	/

Supplementary Table 12: Operational cost factor assumptions

Description	Value	Basis	Reference
Insurance and local tax	2%	TPC	Anantharaman, et al. ⁴
Maintenance	2.5%	TPC	Anantharaman, et al. ⁴
Administration and support	30%	Operating and Maintenance	Anantharaman, et al. ⁴

Supplementary Table 13: Probability density function for Monte Carlo Simulation and Scenario definition

Variable	Min	Max	Assumpti on in mid scenario [#]	Probability density function	Reference
Recycling of Additives & Water	0.8	0.98	0.95	Uniform	own estimations, Wang and Maroto-Valer ³⁰ , Eikeland, et al. ²²
Mineral Purity	0.5	0.8	0.8	Uniform	Kremer, et al. ⁶
Reaction constant [§]	0.5759* 0.6420**	1.4717* 1.2839**	1.4717* 1.2839**	Uniform	**Wang and Maroto-Valer ³⁰ , *Eikeland, et al. ²²
Solid content in Reactor*	0.125	0.3	0.125* 0.3**	Uniform	Wang and Maroto-Valer ³⁰ , Eikeland, et al. ²² W.K. O'Connor and L.R. Penner ²¹
Number of plants to reach NOAK	5	30	20	Uniform	own estimations, Greig, et al. ⁸²
Learning rate	1.1%	20%	10.55%	Uniform	Rubin, et al. ⁸¹
Process and project contingencies	30%, 15%	70%, 30%	40% 30%	Uniform	EPRI ⁷⁸ , AACE ⁷⁹
Interest rate on debt	1.40%	3.72%	/	/	European Central Bank ⁸³
Return on equity	5%	20.20%	/	/	Gurufocus ⁸⁴
Debt/equity	31.00%	73.00%	/	/	Macrotrends ^{85,86}
Overall interest [#]	4%	17 %	7.69%	Lognormal	/
Working hours	4380	8688	8000	Triangular	own estimations, Deolalkar ³⁹
Electricity price (in 2030)	38	200	62	Lognormal	European Commission ⁸⁷
Natural gas price (in 2030)	16.18	40.94	32	Normal	Duić, et al. ⁸⁸
Extraction Costs Mineral	3.52	22.95	12	Normal	Brown, et al. ⁸⁹ , Béarat, et al. ⁹⁰ Huijgen, et al. ⁵² et al.
Price NaHCO ₃ *	100*	400*	209	Normal	Comparison of vendor prices ⁹¹
Price NaCl*	20*	300*	61.6	Lognormal	Comparison of vendor prices ⁹¹
Price MEA*	600*	2050*	1320	Normal	Comparison of vendor prices ⁹¹
Price (NH ₄) ₂ SO ₄ **	90**	740**	140	Lognormal	Comparison of vendor prices ⁹¹
Cement price ^{§§}	78	172	102	Lognormal	de Vet, et al. ⁹² , Statista ⁹³
ETS price (in 2030) ^{§§}	40	89	32	Triangular	Schjølset ⁹⁴ , Watson ⁹⁵ , Carbon Tracker Initiative ⁹⁶ , Carbon Pulse ⁹⁷
* for direct process only ** for indirect process only § Note the reaction constant for the indirect process is taken from the slowest step, the mineral dissolution. # derived from Monte Carlo simulation on interest on debt, ROE and debt/equity ratio §§ used for scenario definition. ## when multiple assumptions could be used (>5 references), the median was chosen.					

Supplementary Table 14: Equations used for the comparisons of different strategies to mitigate CO_{2e} emission reductions.

Description	Equation	Variable description
Baseline	$LCOP_{\text{cement}} = LCOP_{\text{clinker}} + e_{\text{clinker}} \cdot \pi_{\text{ETS}}$	
Biofuels	$LCOP_{\text{cement}} = LCOP_{\text{clinker}} + MAC_{\text{bio fuel}} \cdot (e_{\text{clinker}} \cdot \Delta e_{\text{cement}}) + (e_{\text{clinker}} \cdot (1 - \Delta e_{\text{cement}})) \cdot \pi_{\text{ETS}}$	$MAC_{\text{bio fuel}}$: Marginal abatement costs Δe_{cement} : Carbon footprint reduction.
Calcinated clay cement	$LCOP_{\text{cement}} = LCOP_{\text{calcinated clay cement}} + e_{\text{clinker}} \cdot (1 - \Delta e_{\text{cement}}) \cdot \pi_{\text{ETS}} + \dot{m}_{\text{feedstock}} \cdot d_{\text{Transport}} \cdot \pi_{\text{Transport}}$	
Cement production with off-shore CCS	$LCOP_{\text{cement}} = LCOP_{\text{clinker with CCS}} + e_{\text{clinker}} \cdot (1 - \Delta e_{\text{cement}}) \cdot \pi_{\text{ETS}} + \dot{m}_{\text{CO}_2 \text{ stored}} \cdot d_{\text{Transport}} \cdot \pi_{\text{Transport}}$	
Cement production with CO₂ mineralisation	$LCOP_{\text{cement}} = LCOP_{\text{clinker}} \cdot (1 - X_{\text{replaced}}) + LCOP_{\text{SCM}_{\text{CCU}}} \cdot X_{\text{replaced}} + (e_{\text{clinker with CCS}} + e_{\text{SCM}_{\text{CCU}}}) \cdot \pi_{\text{ETS}}$	X_{replaced} : Share of cement replaced
Cement production with off-shore CCS and CO₂ mineralisation	$LCOP_{\text{cement}} = LCOP_{\text{clinker}} \cdot (1 - X_{\text{replaced}}) + LCOP_{\text{capture}} \cdot \dot{m}_{\text{CO}_2 \text{ captured}} + (LCOP_{\text{SCM}_{\text{CCU}}} - LCOP_{\text{capture}} \cdot \dot{m}_{\text{CO}_2 \text{ stored, CCU}}) \cdot X_{\text{replaced}} + (e_{\text{clinker with CCS}} + e_{\text{SCM}_{\text{CCU, direct}}}) \cdot \pi_{\text{ETS}}$	$e_{\text{SCM}_{\text{CCU, direct}}}$: Direct process emissions of SCM _{CCU} production.

Supplementary Table 15: Assumptions used for the comparisons of different strategies to mitigate CO_{2e} emission reductions.

	Carbon capture and storage	Calcinated clay cement	CO ₂ mineralisation	Biofuels
Reservoir locations in Europe	Reservoirs for CO ₂ storage are located in the North Sea and Mediterranean Sea ⁹⁸ .	Reservoirs of kaolinite clays are located in the Czech Republic, Hungary, Poland, South of Germany ⁹⁹ .	Reservoirs of olivine bearing rocks are located in Norway, Portugal, Spain, Greece, Italy ⁶ .	Feedstock locations are included in the used range for costs.
Transport assumptions for the model	Pipeline transport 100-2000km	Mineral transport: 100-2000km (truck and train)	Mineral transport in Scenario: 1200 km (truck, train and ship)	/

Costs	MEA ⁷⁷ : LCOP _{clinker with CSS} = €107.4t _{clinker} ⁻¹	Calcined clay cement ¹⁰¹ : LCOP _{calculated clay cement} = 54.66* €/t _{cement}	LCOP _{SCM_{CCU}} is calculated for each scenario by the model presented in this paper.	Marginal abatement costs ³⁶ : MAC _{bio fuel} = €15 – €50t _{CO2e,avoided} ⁻¹
	Oxy-fuel combustion ⁷⁷ : LCOP _{clinker with CCS} = €93t _{clinker} ⁻¹	m _{feedstock} = 0.5 · m _{cement}	Mineral transport ⁸⁹ : π _{transport,truck} = €0.04t ⁻¹ km ⁻¹	
	CO ₂ transport derived from Zero Emission Platform ¹⁰⁰ for pipeline transport and offshore storage: π _{CO₂,transport} = 0.0099 · d _{Transport} + 1.2185 with d _{transport} being distance in [km].	Mineral transport ⁸⁹ : π _{transport,truck} = €0.04t ⁻¹ km ⁻¹ π _{transport,train} = €0.032t ⁻¹ km ⁻¹	π _{transport,train} = €0.032t ⁻¹ km ⁻¹ π _{transport,ship} = €0.0032t ⁻¹ km ⁻¹	
Carbon footprint reduction[#]	MEA ⁷⁷ : Δe _{cement} = 0.64 Oxy-fuel combustion ⁷⁷ : Δe _{cement} = 0.82	Calcined clay cement ¹⁰¹ : Δe _{cement} = 0.34	Carbon footprint reductions are calculated for each scenario by the model presented in this paper.	Alternative fuels ^{§102} : Δe _{cement} = 0.25
* Currency adapted to €. # Reductions calculated in comparison to ordinary Portland Cement. § 70% of fossil fuels are replaced by alternative fuels.				

Supplementary Table 16: List of abbreviations and symbols.

Abbreviation	Description	Unit
ABS	Ammonium bisulphate	/
Additive Rec.	Additive recovery	[fraction]
AS	Ammonium sulphate	/
α	Levelisation factor	[fraction]
c _{A0}	Initial concentration of educt A	[mol l ⁻¹]
C _i	Cost of i	[€ a ⁻¹]
CaCO ₃	Calcium carbonate	/
CAPEX	Capital expenditures	/
CaSiO ₃	Wollastonite	/
CCS	Carbon capture and storage	/
CCU	Carbon capture and utilisation	/
CCU _M	Carbon capture and utilisation through mineralisation	/
CO ₂	Carbon dioxide	/
CO _{2e}	Carbon dioxide equivalent	/
CSTR	Continuously stirred tank reactors	/
d	Diameter	[m]
DER	Dept to equity ratio	[fraction]
dist	Distance	[km]
Δ _i	Change of i	/
E	Experience factor	[fraction]
e _i	CO _{2e} emissions of i	[tCO _{2e} a ⁻¹]
ETS	European emission trading system	/
η	Efficiency	[fraction]
f _i	Costing factor i	[fraction]
FOAK	First-of-a-kind	/
h	Height	[m]
Ĥ	Enthalpy stream	[KW]
i	Interest rate on capital	[fraction]

I	Capital cost index	[real number]
k	Heat exchange coefficient	[W m ⁻² K ⁻¹]
k_{reaction}	Reaction rate constant	[1 s ⁻¹]
l	Length	[m]
LCOP	Levelised cost of product	[€ t ⁻¹]
LR	Learning rate	[fraction]
$\dot{m}$	Mass flow	[t a ⁻¹]
MEA	Monoethanolamine	/
MEA CCS	Monoethanolamine post combustion capture with offshore geological CO ₂ storage	/
Mg ₂ SiO ₄	Forsterite	/
Mg ₃ Si ₂ O ₅ (OH) ₄	Lizardite	/
MgCO ₃	Magnesium carbonate	/
MS	Magnesium sulphate	/
No of plants	Number of plants to reach maturity	[natural number]
NOAK	N th -of-a-kind	/
OPEX	Operational expenditures	/
Oxy-fuel CCS	Oxy-fuel combustion with offshore geological CO ₂ storage	/
$\dot{Q}$	Heat stream	[KW]
ρ	Density	[kg m ⁻³]
π_i	Price of i	[€ t ⁻¹] or [€ MWh ⁻¹]
R	Revenue	[€ t ⁻¹]
r	Radius	[m]
r_A	Reaction rate for educt A	[mol l ⁻¹ s ⁻¹]
ROE	Return on equity	[fraction]
rpm	Centrifugal speed	[rpm]
SCM	Supplementary cementitious material	/
SCM _{CCU}	Supplementary cementitious material produced via CO ₂ mineralisation	/
SiO ₂	Silica	/
T	Temperature	[K] or [°C]
TEA	Techno-economic assessment	/
$t_{\text{construction}}$	Estimated time for construction	[years]
TCR	Total capital required	[€]
TDC	Total direct costs	[€]
$t_{\text{operating}}$	Operation hours per year	[h a ⁻¹]
TPC	Total plant costs	[€]
TRL	Technology readiness level	/
t_w	Wall thickness	[m]
τ	Space time	[s]
V	Volume	[m ³]
$\dot{v}$	Volume flow	[m ³ a ⁻¹]
ν	Viscosity	[10 ⁻⁶ Pa s]
w	Electrical work	[KWh]
X	Extend of reaction	[fraction]
X_i	Share of material i in cement blend	[fraction]
$X_{S/L}$	Solid-liquid ratio in reactor	[fraction]

Supplementary References

- 1 Sanna, A., Hall, M. R. & Maroto-Valer, M. Post-processing pathways in carbon capture and storage by mineral carbonation (CCSM) towards the introduction of carbon neutral materials. *Energy & Environmental Science* **5**, 7781, doi:10.1039/c2ee03455g (2012).
- 2 Ostovari, H., Sternberg, A. & Bardow, A. Rock 'n' use of CO₂: carbon footprint of carbon capture and utilization by mineralization. *Sustainable Energy & Fuels*, doi:10.1039/D0SE00190B (2020).
- 3 Mario, M.-A., Vicente, N., José María del, C., José Santos, L.-G. & Esteban, M. D. Review of coastal Land Reclamation situation in the World. *Journal of coastal research* **75**, 667-671, doi:10.2112/SI75-133.1 (2016).
- 4 Anantharaman, R. *et al.* CEMCAP Framework for Comparative Techno-economic Analysis of CO₂ Capture From Cement Plants-D3. 2. (Zenodo, 2018).
- 5 Sanna, A., Uibu, M., Caramanna, G., Kuusik, R. & Maroto-Valer, M. M. A review of mineral carbonation technologies to sequester CO₂. *Chem Soc Rev* **43**, 8049-8080, doi:10.1039/c4cs00035h (2014).
- 6 Kremer, D. *et al.* Geological Mapping and Characterization of Possible Primary Input Materials for the Mineral Sequestration of Carbon Dioxide in Europe. *Minerals* **9**, doi:10.3390/min9080485 (2019).
- 7 Park, A.-h. A., Kelemen, P., Matter, J. & Gadikota, G.
- 8 CemNet. *The Global Cement Report - Online Database of Cement Plants*, <<https://www.cemnet.com/global-cement-report/>> (2021).
- 9 Benhelal, E. *et al.* The utilisation of feed and byproducts of mineral carbonation processes as pozzolanic cement replacements. *Journal of Cleaner Production* **186**, 499-513 (2018).
- 10 Stopic, S. *et al.* Synthesis of Nanosilica via Olivine Mineral Carbonation under High Pressure in an Autoclave. *Metals* **9**, 708 (2019).
- 11 Wong, H. S. & Abdul Razak, H. Efficiency of calcined kaolin and silica fume as cement replacement material for strength performance. *Cement and Concrete Research* **35**, 696-702, doi:<https://doi.org/10.1016/j.cemconres.2004.05.051> (2005).
- 12 European standard. EN 197-1:2000: Cement. Composition, specifications and conformity criteria for common cements. (British Standards Institute, 2000).
- 13 Khedr, S. A. & Abou-Zeid, M. N. Characteristics of silica-fume concrete. *Journal of Materials in Civil Engineering* **6**, 357-375 (1994).
- 14 Bhanja, S. & Sengupta, B. Investigations on the compressive strength of silica fume concrete using statistical methods. *Cement and Concrete Research* **32**, 1391-1394, doi:[https://doi.org/10.1016/S0008-8846\(02\)00787-1](https://doi.org/10.1016/S0008-8846(02)00787-1) (2002).
- 15 Hooton, R. D. Influence of Silica Fume Replacement of Cement on Physical Properties and Resistance to Sulfate Attack, Freezing and Thawing, and Alkali-Silica Reactivity. *ACI Materials Journal* **90**, doi:10.14359/4009 (1993).
- 16 Lothenbach, B., Scrivener, K. & Hooton, R. Supplementary cementitious materials. *Cement and concrete research* **41**, 1244-1256 (2011).
- 17 Mindess, S. in *Developments in the Formulation and Reinforcement of Concrete (Second Edition)* (ed Sidney Mindess) 3-17 (Woodhead Publishing, 2019).
- 18 Blaum, P. (ed Till Strunge) (Personal Communication).
- 19 Strunge, J. (ed Till Strunge) (Personal Communication).
- 20 Gerdemann, S. J., O'Connor, W. K., Dahlin, D. C., Penner, L. R. & Rush, H. Ex situ aqueous mineral carbonation. *Environmental science & technology* **41**, 2587-2593 (2007).
- 21 W.K. O'Connor, D. C. D., G.E. Rush, S.J. Gerdemann, & L.R. Penner, a. D. N. N. *Aqueous Mineral Carbonation : Mineral Availability, Pretreatment, Reaction Parametrics, And Process Studies*. (National Energy Technology Laboratory, 2005).
- 22 Eikeland, E., Blichfeld, A. B., Tyrsted, C., Jensen, A. & Iversen, B. B. Optimized carbonation of magnesium silicate mineral for CO₂ storage. *ACS applied materials & interfaces* **7**, 5258-5264 (2015).
- 23 Huijgen, W. J. J. *Carbon dioxide sequestration by mineral carbonation*, S.n., (2007).

- 24 Wörtler, M. *et al.* Steel's contribution to a low-carbon Europe 2050: Technical and economic analysis of the sector's CO₂ abatement potential. *London: BCG. Retrieved April 20*, 2015 (2013).
- 25 Yu, Q., Nagataki, S., Lin, J., Saeki, T. & Hisada, M. The leachability of heavy metals in hardened fly ash cement and cement-solidified fly ash. *Cement and concrete research* **35**, 1056-1063, doi:10.1016/j.cemconres.2004.03.031 (2005).
- 26 Nduagu, E., Bergerson, J. & Zevenhoven, R. Life cycle assessment of CO₂ sequestration in magnesium silicate rock – A comparative study. *Energy Conversion and Management* **55**, 116-126, doi:10.1016/j.enconman.2011.10.026 (2012).
- 27 Kremer, D. & Wotruba, H. Separation of Products from Mineral Sequestration of CO₂ with Primary and Secondary Raw Materials. *Minerals* **10**, 1098 (2020).
- 28 Pinkerton, A. P. & Klima, M. S. Evaluation of a solid-bowl centrifuge for ultrafine size separations. *Mining, Metallurgy & Exploration* **18**, 162-166 (2001).
- 29 Geerlings, H. & Zevenhoven, R. CO₂ mineralization-bridge between storage and utilization of CO₂. *Annu Rev Chem Biomol Eng* **4**, 103-117, doi:10.1146/annurev-chembioeng-062011-080951 (2013).
- 30 Wang, X. & Maroto-Valer, M. M. Optimization of carbon dioxide capture and storage with mineralisation using recyclable ammonium salts. *Energy* **51**, 431-438 (2013).
- 31 Sanna, A., Dri, M. & Maroto-Valer, M. Carbon dioxide capture and storage by pH swing aqueous mineralisation using a mixture of ammonium salts and antigorite source. *Fuel* **114**, 153-161 (2013).
- 32 Wang, X. & Maroto-Valer, M. M. Dissolution of serpentine using recyclable ammonium salts for CO₂ mineral carbonation. *Fuel* **90**, 1229-1237 (2011).
- 33 Wang, X. & Maroto-Valer, M. M. Integration of CO₂ capture and mineral carbonation by using recyclable ammonium salts. *ChemSusChem* **4**, 1291-1300 (2011).
- 34 Rademaekers, K. *et al.* Atmospheric Gas-Aerosol Equilibrium: IV. Thermodynamics of Carbonates. *Aerosol Science and Technology* **23**, 131-154, doi:10.1080/02786829508965300 (1995).
- 35 Favier, A., De Wolf, C., Scrivener, K. & Habert, G. A sustainable future for the European Cement and Concrete Industry: Technology assessment for full decarbonisation of the industry by 2050. (ETH Zurich, 2018).
- 36 Czigler, T., Reiter, S., Schulze, P. & Somers, K. Laying the foundation for zero-carbon cement. (McKinsey & Company, 2020).
- 37 Bataille, C. Low and zero emissions in the steel and cement industries. doi:doi:<https://doi.org/10.1787/5ccf8e33-en> (2020).
- 38 The European Cement Association. Cementing the European Green Deal. (The European Cement Association,, Brussels, 2020).
- 39 Deolalkar, S. P. in *Designing Green Cement Plants* (ed S. P. Deolalkar) 83-86 (Butterworth-Heinemann, 2016).
- 40 Tokheim, L.-A. *et al.* in *TCCS–10. CO₂ Capture, Transport and Storage. Trondheim 17th–19th June 2019. Selected papers from the 10th International Trondheim CCS Conference*.
- 41 Hoenig, V. & Schneider, M. in *VDZ Congress*.
- 42 Scrivener, K. L., John, V. M. & Gartner, E. M. Eco-efficient cements: Potential economically viable solutions for a low-CO₂ cement-based materials industry. (2016).
- 43 Gartner, E. & Sui, T. Alternative cement clinkers. *Cement and concrete research* **114**, 27-39, doi:10.1016/j.cemconres.2017.02.002 (2018).
- 44 European Association of Mining Industries Metal Ores & Industrial Minerals. The European Magnesite/Magnesia Industry: enabler in the transition to a low-carbon economy. (Brussels, 2020).
- 45 European Cement Association. *Key Facts & Figures*, <<https://cembureau.eu/about-our-industry/key-facts-figures/>> (2017).
- 46 Markets and Markets. Silica Fume Market by Application, by Geography - Global Analysis and Forecast to 2020. (2015).

- 47 Industry ARC. *Silica Fume Market - Forecast(2021 - 2026)*,
 48 <<https://www.industryarc.com/Report/15138/silica-fume-market.html>> (2020).
- 49 Markets and Markets. *Calcium Carbonate Market by Type (GCC and PCC), End-Use Industry (Paper, Plastic, Paints & Coatings, Adhesive & Sealants), and Region (APAC, North America, Europe, South America, Middle East & Africa) - Global Forecast to 2024*,
 50 <<https://www.marketsandmarkets.com/Market-Reports/calcium-carbonate-market-86344547.html>> (2018).
- 51 Strunge, T. in *TCCS-11 - Trondheim Conference on CO2 Capture, Transport and Storage*. (SINTEF).
- 52 Hitch, M. & Dipple, G. Economic feasibility and sensitivity analysis of integrating industrial-scale mineral carbonation into mining operations. *Minerals Engineering* **39**, 268-275 (2012).
- 53 Naraharisetti, P. K., Yeo, T. Y. & Bu, J. New classification of CO2 mineralization processes and economic evaluation. *Renewable and Sustainable Energy Reviews* **99**, 220-233 (2019).
- 54 Huijgen, W. J., Comans, R. N. & Witkamp, G.-J. Cost evaluation of CO2 sequestration by aqueous mineral carbonation. *Energy Conversion and Management* **48**, 1923-1935 (2007).
- 55 Katsuyama, Y. *et al.* Development of a process for producing high-purity calcium carbonate (CaCO3) from waste cement using pressurized CO2. *Environmental Progress* **24**, 162-170, doi:10.1002/ep.10080 (2005).
- 56 Iizuka, A., Fujii, M., Yamasaki, A. & Yanagisawa, Y. Development of a new CO2 sequestration process utilizing the carbonation of waste cement. *Industrial & Engineering Chemistry Research* **43**, 7880-7887 (2004).
- 57 Pérez-Fortes, M., Bocin-Dumitriu, A. & Tzimas, E. Techno-economic assessment of carbon utilisation potential in Europe. *Chemical Engineering Transactions* (2014).
- 58 Mehleri, E., Bhawe, A., Shah, N., Fennell, P. & MacDowell, N. in *Fifth International Conference on Accelerated Carbonation for Environmental and Material Engineering (ACEME 2015)*, New York.
- 59 Pedraza, J., Zimmermann, A., Tobon, J., Schomäcker, R. & Rojas, N. On the road to net zero-emission cement: Integrated assessment of mineral carbonation of cement kiln dust. *Chemical engineering journal (Lausanne, Switzerland : 1996)* **408**, doi:10.1016/j.cej.2020.127346 (2021).
- 60 Shashi, E. *Piping calculations manual*. (McGraw-Hill, 2005).
- 61 Stelson, A. W. & Seinfeld, J. H. Prediction of the density of ammonium bisulfate solutions. *The Journal of Physical Chemistry* **85**, 3730-3733, doi:10.1021/j150624a042 (1981).
- 62 The Engineering ToolBox. *Density of aqueous solutions of some inorganic substances*, <https://www.engineeringtoolbox.com/density-aqueous-solution-inorganic-salt-acid-concentration-d_1958.html> (2017).
- 63 handyath.com. *The Complete Aqueous Magnesium Sulfate Solutions Density-Concentration Calculator*, <<https://www.handyath.com/cgi-bin/mgsulfble.cgi?submit=Entry>> (n. d.).
- 64 Chase, M. W. J. NIST-JANAF Thermochemical Tables. *J. Phys. Chem. Ref. Data, Monograph* **9**, 1-1951 (1998).
- 65 Southard, M. Z. & Green, D. W. *Perry's chemical engineers' handbook*. (McGraw-Hill, 2018).
- 66 National Center for Biotechnology Information. *PubChem Compound Summary for CID 6097028, Ammonium sulfate.*, <<https://pubchem.ncbi.nlm.nih.gov/compound/Ammonium-sulfate>> (2004).
- 67 MatWeb.

Material	Property	data,
< http://www.matweb.com/search/datasheet.aspx?matguid=71feb59d7edd41e798c032c1d63de8dd&ckck=1 > (n. d.).		
- 68 Schwister, K. & Leven, V. *Verfahrenstechnik für Ingenieure: Ein Lehr-und Übungsbuch (mit umfangreichem Zusatzmaterial)*. (Carl Hanser Verlag GmbH Co KG, 2020).
- 69 Hangx, S. J. & Spiers, C. J. Coastal spreading of olivine to control atmospheric CO2 concentrations: A critical analysis of viability. *International Journal of Greenhouse Gas Control* **3**, 757-767 (2009).

- 68 Costa, G., Poletti, A., Pomi, R., Stramazzo, A. & Zingaretti, D. Energetic assessment of CO₂ sequestration through slurry carbonation of steel slag: a factorial study. *Greenhouse Gases: Science and Technology* **7**, 530-541 (2017).
- 69 Bell, G. R. A. Analysis and Development of a Decanter Centrifuge
- 70 Towler, G. & Sinnott, R. *Chemical engineering design: principles, practice and economics of plant and process design*. (Elsevier, 2012).
- 71 Njeng, A. S. B., Vitu, S., Clausse, M., Dirion, J.-L. & Debaq, M. Wall-to-solid heat transfer coefficient in flighted rotary kilns: Experimental determination and modeling. *Experimental Thermal and Fluid Science* **91**, 197-213 (2018).
- 72 The Engineering ToolBox. *Heat Exchanger Heat Transfer Coefficients*, <https://www.engineeringtoolbox.com/heat-transfer-coefficients-exchangers-d_450.html> (2003).
- 73 Gabi, S. 9.2 Software-System and Databases for Life Cycle Engineering. *Sphera: Stuttgart, Echterdingen, Germany* (2019).
- 74 Psaraftis, H. N. & Kontovas, C. A. CO₂ emission statistics for the world commercial fleet. *WMU Journal of Maritime Affairs* **8**, 1-25 (2009).
- 75 Swiss Centre for Life Cycle Inventories. ecoinvent Data V 3.71. (2019).
- 76 Pehnt, M. & Henkel, J. Life cycle assessment of carbon dioxide capture and storage from lignite power plants. *International Journal of Greenhouse Gas Control* **3**, 49-66, doi:<https://doi.org/10.1016/j.ijggc.2008.07.001> (2009).
- 77 Voldsund, M. et al. D4. 6: CEMCAP Comparative Techno-Economic Analysis of CO₂ Capture in Cement Plants. *H2020 Project: CO₂ Capture from Cement Production* (2018).
- 78 EPRI. Technical Assessment Guide Volume 1: Electricity Supply—1993, TR-102276-V1R1. *Electric Power Research Institute, Palo Alto, California* (1993).
- 79 AACE. *Cost estimate classification system – as applied in engineering, procurement, and construction for the process industries*, AACE International Recommended Practice No. 18R-97 (Rev. November 29, 2011). (2011).
- 80 Grande, C., Roussanaly, S., Anantharaman, R. & Lindqvist, K. CO₂ Capture in Natural Gas Production by Adsorption Processes for CO₂ Storage, EOR and EGR. *IEAGHG, editor* (2016).
- 81 Rubin, E. S., Azevedo, I. M., Jaramillo, P. & Yeh, S. A review of learning rates for electricity supply technologies. *Energy Policy* **86**, 198-218 (2015).
- 82 Greig, C., Garnett, A., Oesch, J. & Smart, S. Guidelines for scoping and estimating early mover ccs projects. *Univ. Queensl., Brisbane* (2014).
- 83 European Central Bank. *Cost of borrowing for corporations - Euro area*, <<https://sdw.ecb.europa.eu/browseSelection.do?type=series&q=MIR.M.U2.B.L22.A.R.A.2240.EUR.N+MIR.M.U2.B.A21.AM.R.A.2240.EUR.N&node=SEARCHRESULTS>> (2021).
- 84 Gurufocus.

ROE	%	Sector	Distribution,
< https://www.gurufocus.com/term/ROE/OTCPK:HDELY/ROE-Percentage/HeidelbergCement%20AG > (2020).			
- 85 Macrotrends.

Holcim	Debt	to	Equity	Ratio	2010-2020,
< https://www.macrotrends.net/stocks/charts/HCLMY/holcim/debt-equity-ratio > (2020).					
- 86 Macrotrends.

HeidelbergCement	AG	Debt	to	Equity	Ratio	2008-2020,
< https://www.macrotrends.net/stocks/charts/HDELY/heidelbergcement-ag/debt-equity-ratio > (2020).						
- 87 European Commission. Communication from the commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the regions - Energy prices and costs in Europe (COM (2019) 1 final). (Brussels, 2019).
- 88 Duić, N. et al. Heat Roadmap Europe: EU28 fuel prices for 2015, 2030 and 2050 - Deliverable 6.1: Future fuel price review. (University of Zagreb, Zagreb, 2017).
- 89 Brown, T. J. et al. Underground mining of aggregates. Main report. (2010).
- 90 Béarat, H. et al. Carbon sequestration via aqueous olivine mineral carbonation: role of passivating layer formation. *Environmental science & technology* **40**, 4802-4808 (2006).
- 91 Alibaba. *Product search at Alibaba.com*, <<https://www.alibaba.com/>> (2020).
- 92 de Vet, J.-M. et al. Competitiveness of the European Cement and Lime Sectors. (2018).

- 93 Statista. *Forecast in percentage change in cement prices worldwide from 2013 to 2016 with a forecast from 2017 to 2020*, <<https://www.statista.com/statistics/248359/change-in-global-cement-prices/>> (2017).
- 94 Schjølset, S. The MSR: Impact on market balance and prices. (Thomson Reuters,, 2014).
- 95 Watson, F. *Analysts see EU carbon prices at Eur56-Eur89/mt by 2030*, <<https://www.spglobal.com/platts/en/market-insights/latest-news/coal/120320-analysts-see-eu-carbon-prices-at-eur56-eur89mt-by-2030>> (2020).
- 96 Carbon Tracker Initiative. *EU carbon prices could double by 2021 and quadruple by 2030*, <<https://carbontracker.org/eu-carbon-prices-could-double-by-2021-and-quadruple-by-2030/>> (2018).
- 97 Carbon Pulse. *EU carbon prices to double on raised 2030 climate target, over €100 possible from other reforms -analysts*, <<https://carbon-pulse.com/109931/>> (2020).
- 98 Kjärstad, J., Morbee, J., Odenberger, M., Johnsson, F. & Tzimas, E. Modelling Large-scale CCS Development in Europe Linking Techno- economic Modelling to Transport Infrastructure. *Energy Procedia* **37**, 2941-2948, doi:<https://doi.org/10.1016/j.egypro.2013.06.180> (2013).
- 99 Dill, H. G. A geological and mineralogical review of clay mineral deposits and phyllosilicate ore guides in Central Europe – A function of geodynamics and climate change. *Ore Geology Reviews* **119**, 103304, doi:<https://doi.org/10.1016/j.oregeorev.2019.103304> (2020).
- 100 Zero Emission Platform. *The Costs of CO2 Transport - Post-demonstration CCS in the EU*. (European Technology Platform for Zero Emission Fossil Fuel Power Plants, Brussels, 2011).
- 101 Cancio Díaz, Y. *et al.* Limestone calcined clay cement as a low-carbon solution to meet expanding cement demand in emerging economies. *Development Engineering* **2**, 82-91, doi:<https://doi.org/10.1016/j.deveng.2017.06.001> (2017).
- 102 Wojtacha-Rychter, K., Kucharski, P. & Smolinski, A. Conventional and Alternative Sources of Thermal Energy in the Production of Cement—An Impact on CO2 Emission. *Energies* **14**, 1539 (2021).