

Towards a business case for CO₂ mineralisation in the cement industry

Till Strunge (✉ tms4@hw.ac.uk)

Heriot-Watt University

Phil Renforth

Heriot-Watt University <https://orcid.org/0000-0002-1460-9947>

Mijndert Van der Spek

Heriot-Watt University

Article

Keywords: CO₂, cement, mineralization

Posted Date: June 17th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-478558/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Version of Record: A version of this preprint was published at Communications Earth & Environment on March 14th, 2022. See the published version at <https://doi.org/10.1038/s43247-022-00390-0>.

Towards a business case for CO₂ mineralisation in the cement industry

Article - Draft for submission to Communications Earth & Environment

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 EH14 4AS, UK.

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

Abstract

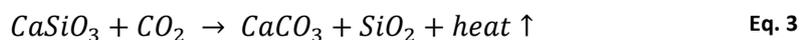
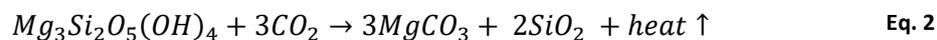
The cement industry is responsible for approximately 7% of anthropogenic CO₂ emissions with low margins and the highest carbon intensity of any industry per unit of revenue. To encourage complete decarbonisation of the cement industry, strategies must be found in which GHG emission reductions are incentivised. Here we show through integrated techno-economic modelling that CO₂ mineralisation using silicate minerals results in emission reductions of 6-31% while generating an additional profit of up to €28 per tonne cement. In order to create positive CO₂ mineralisation business cases three conditions are paramount: the resulting products must be used as a supplementary material in cement blends, the storage of CO₂ in minerals must be eligible for ETS credits or similar, and the feedstock minerals must be available in close proximity (< ~550km, transported by truck and train).

Introduction

The cement industry is responsible for approximately 7% of anthropogenic CO₂ emissions^{1,2} with the highest carbon intensity of any industry per unit of revenue³. Given the use of cement is fundamental to economic development with a projected global market size of \$463 billion⁴ (6.08 Gt/a cement⁵) in 2026, reducing its embodied emissions is essential⁶⁻⁸. Approximately 60% of the cement industries' emissions are process-inherent, resulting from the calcination reaction of limestone⁹. These emissions are particularly challenging to mitigate since either the entire process must be replaced by low emission alternatives^{3,6,10-13} or the emissions have to be captured^{1,3,6,8,14,15}. While the replacement of cement and concrete by alternative building materials like wood or calcinated clay would require an unrealistically rapid change of the entire value-chain, carbon capture and storage technologies present an alternative for decarbonisation, but incur additional production cost^{16,17}. Preferably, strategies must be found in which greenhouse gas (GHG) emission reductions are likely to render additional revenue. Some have suggested that CO₂ can be captured and reacted with activated minerals or industrial wastes to form stable carbonate minerals (also known as 'CO₂ mineralisation')¹⁸⁻²⁰, the products of which could be subsequently valorised. These reactions are exothermic, leading to long-term storage of CO₂¹⁹. Early findings suggest that in addition to CO₂ storage the products may potentially be used in a range of applications, including as fillers, polymer additives, for land reclamation or as supplementary cementitious materials (SCM)^{19,21-24} potentially creating revenues of €14-700 per ton of CO₂ captured¹⁹. Depending on the feedstock material for the reaction, additionally metal oxides such as iron oxides can be separated as a valuable by-product which could be used as pigments or as iron ore^{19,21}.

Multiple feedstocks for CO₂ mineralisation have been proposed, especially rocks containing magnesium- or calcium-rich silicate minerals^{18,20}. For instance, forsterite (Mg₂SiO₄), present in olivine-bearing rocks, lizardite (Mg₃Si₂O₅(OH)₄) present in serpentine-bearing rocks and wollastonite (CaSiO₃).

Rocks can be composed of between 50% and 100% of these minerals, depending on the host geology of the extraction site^{22,25}. The general CO₂ mineralisation reactions for these example minerals are described in Eq. 1 to Eq. 3.



Previous work on CO₂ mineralisation has shown that CO₂ storage cost when using virgin minerals as feedstocks could be in the range of 65-443€/t_{CO₂} avoided²⁶ (excluding CO₂ capture)²⁷ (Supplementary Table 1). However, these studies neglect the value added from the sale of the resulting products, which may be critical to successful adoption by players in an industry characterised by strong competition and high pressure on price. Therefore, we here move beyond mineralisation for storage purposes only, and critically investigate under which conditions there is a positive business case for the *use* of mineral carbonation products in the cement industry. We show that, given the right circumstances, such business cases exist when revenue can be created via the use of mineralisation products as SCM. We created integrated techno-economic models of two carbonation processes to produce supplementary cementitious material that allow for in-depth analysis of the interactions of process and economic performance. By using these models to test the potential business case under different future scenarios we found cost-optimal production processes and scales, and global uncertainty analysis elucidated the main drivers of costs *and* benefits.

Results

Here, we investigate a large volume market, i.e., cement replacement as SCM, since other markets like magnesium carbonate, calcium carbonate and silica fume do not match with the scale of CO₂ emissions from cement production (Supplementary Note 1). We investigated cement production with integrated mineral carbonation, where the CO₂ is captured from the cement kiln and immediately carbonated on-site (Figure 1 and Supplementary Note 2). Our investigations found that a maximum of 25% of ordinary Portland cement can be replaced by SCM (Supplementary Note 3). To this end, we designed improved process flowsheets for direct and indirect mineral carbonation systems. The proposed direct process route uses increased pressure and temperature to achieve carbonation in a single step, while the indirect process route uses multiple steps and additives to extract MgSO₄ from feedstock mineral, which have to be reactivated using heat.^{18,28,29} We incorporated gravity separation to ensure cement replacement quality of the carbonated product (Supplementary Note 4). Material that cannot be used as SCM is stored in the close by limestone quarry (Figure 1).

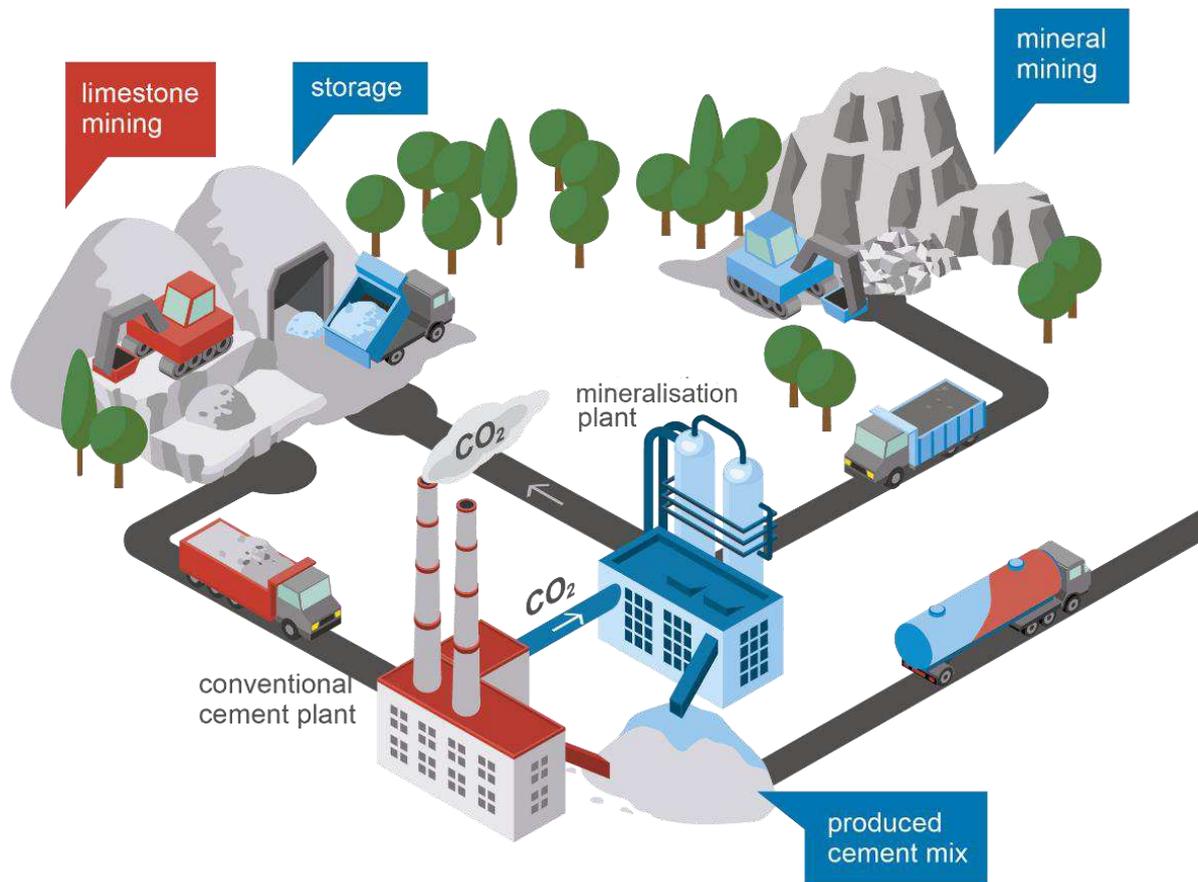


Figure 1: CO₂ mineralisation integrated in the cement production process.

The costs and revenues of CO₂ mineralisation

For this study we defined three scenarios (pessimistic, mid, and optimistic) for factors that have large impacts on CO₂ capture and utilisation through mineralisation (hereafter, CCU_M) (see Table 1). As silica is the reactive ingredient in SCMs, we include the *silica content* to capture multiple possible compositions for the produced supplementary cementitious material, hereafter *SCM_{CCU}*, that can be used as cement replacement (Supplementary Note 3). Higher silica contents in the *SCM_{CCU}* require an increased purification effort because mineralisation processes produce more carbonates than silica. Consequently, the optimistic scenario features the lowest silica content. We included the *Share of cement replaced*, with the highest share presented in the optimistic scenario. As potential revenue streams we included *cement price* and *ETS price*. The cement price determines the revenue from replacing cement with the produced *SCM_{CCU}*, while additional revenue can be expected from lowering the burden for CO₂ allowances under the European emission trading system (ETS). ETS eligibility has been granted by the European supreme court for producing precipitated calcium carbonate from CO₂³⁰ but not yet for magnesium carbonates. Nevertheless, it can be expected that the argumentation of long lasting storage will also apply to magnesium-based carbonates, due to the similar CO₂ storage properties in stable minerals³¹. In the pessimistic scenario, no ETS eligibility was assumed.

Table 1: Scenario definition for techno-economic evaluation.

Description	pessimistic	mid	optimistic
Silica content in SCM_{CCU}¹	50%	40%	30%
Share of cement replaced²	10%	20%	25%
Cement price [€/t_{cement}]³	93	102	116
ETS price [€/t_{CO2}]⁴	0	32	89

¹ Silica is widely accepted as SCM and is the active ingredient of the SCM_{CCU}. Our investigations show that contents of 30-50% of silica in the SCM_{CCU} can be expected to be feasible (Supplementary Note 3).
²The use of SCMs in cement is limited due to potential increased water requirements or strength limitations of resulting mortars, our investigations concluded 10-25 % to be a realistic use case for applying SCM_{CCU} in cement (Supplementary Note 3).
³Comparison of multiple sources: the values used here are the lowest and highest numbers expected for 2030, which was found in reports. In the pessimistic scenario no ETS eligibility is assumed. (Supplementary Table 13).
⁴The numbers represent the lower quantile, median and third quantile of cement prices in Europe (Supplementary Table 13)

Figure 2a shows that both the direct and the indirect process route provide a positive business case under the ‘optimistic’ scenario. The implementation of CCU_M creates a profit of approx. 111€/t_{SCM_{CCU}} (direct) and 92€/t_{SCM_{CCU}} (indirect) leading to additional profit of €38M and €31M per year per plant, which translates into an additional *profit* of €28 and €23 per tonne of cement sold. In the mid scenario the CCU_M processes breaks even with a slight profit of 8€/t_{SCM} (direct) and 11€/t_{SCM} (indirect). In the pessimistic scenario, where CCU_M is assumed not to be recognised by the ETS, both process routes generate a loss for the cement producer of 78€/t_{SCM_{CCU}} (direct) and 50€/t_{SCM_{CCU}} (indirect) resulting in additional *costs* of €8 and €5 per tonne of cement, highlighting the importance of including CCU_M as means of GHG emission reductions in the European ETS. Figure 2a also shows that economics limit the maximum economic capacity of CCU_M plants in cement production, also limiting the amount of CO₂ that can be mineralised: optimal plant capacities for the mineralisation plant that maximise profit for the pessimistic, mid and optimistic scenarios are 136kt_{SCM/a}, 272kt_{SCM/a} and 340kt_{SCM/a} (both for the direct and indirect process, the ‘knee’ in the revenue curve); in the optimistic scenario, a positive business case can be made up to a cement replacement capacity of 900kt_{SCM/a} (direct) and 640 kt_{SCM/a} (indirect). Since the maximum economic capacities of CCU_M plants occur when the capacity of the CCU_M matches the share of cement replaced (e.g. 10% of total cement production is 136kt), it shows that revenues of ETS certificates alone do not cover the costs of the mineralisation. The here calculated values for cost and carbon footprint reductions are consistent with previous studies, when taking into account that a blend of inert and silica is used to produce SCM_{CCU}^{19,27}.

When investigating the GHG emission reductions provided by CCU_M (Figure 2b) it becomes clear that, depending on the scenario, the implementation of CO₂ mineralisation would lead to 14-31% (direct) and 6%-19% (indirect) carbon footprint reduction of a cement plant at the economically optimal plant capacities. This value can move beyond 31% for the direct process in the optimistic scenario, but at the expense of less favourable economics (although still positive). The indirect process suffers from a high electricity and natural gas demand for the additive regeneration, as a result, the carbon footprint reduction deteriorates when moving beyond the optimal capacity.

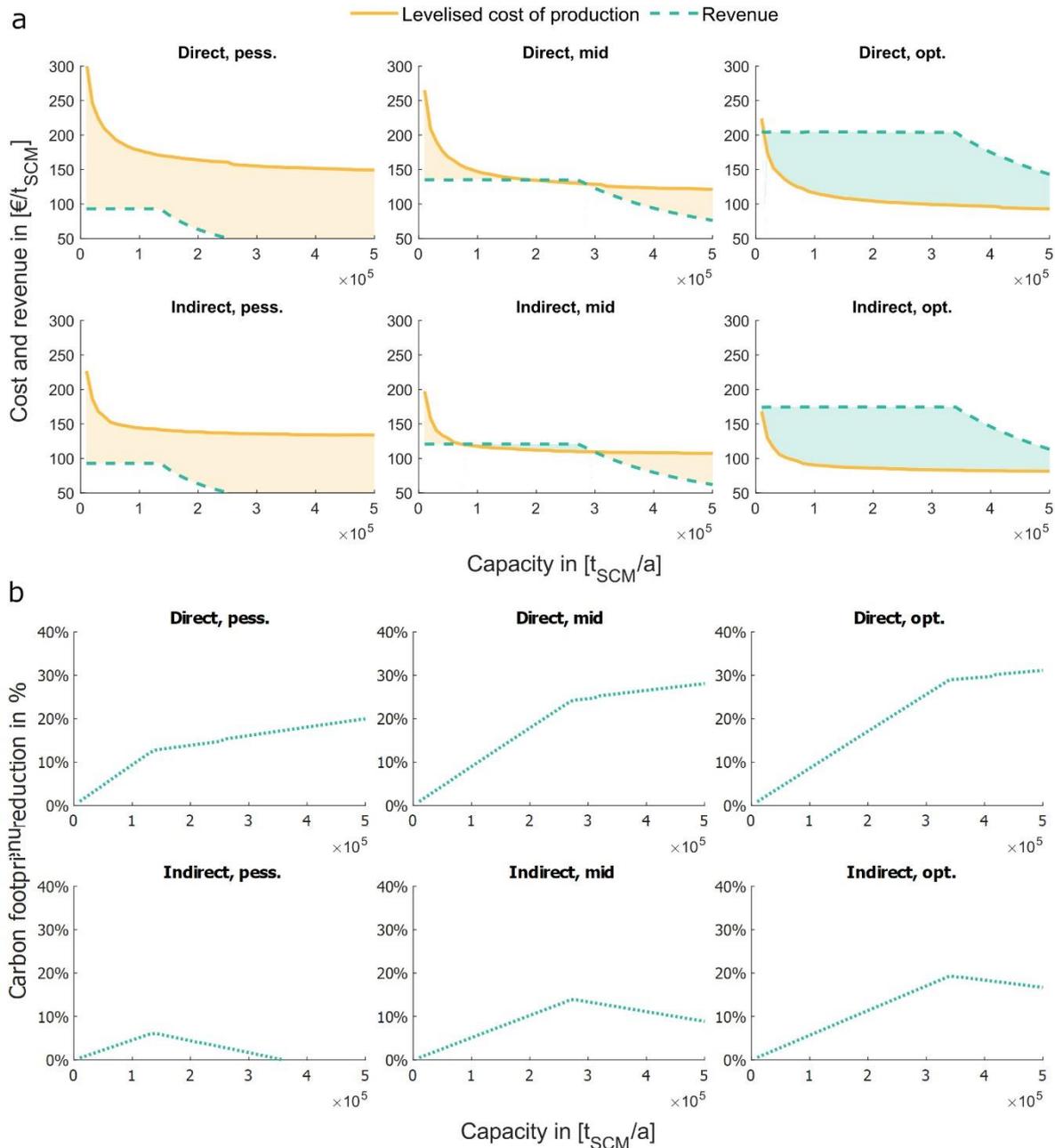


Figure 2: Cost and revenues for different scenarios. Panel a: cost and revenue comparison; Panel b: carbon footprint reduction. Assumptions for the calculations are shown in Supplementary Tables 3-13, additional results shown in Supplementary Figures 5-7.

The worse carbon footprint performance of the indirect process is also reflected in a lower revenue from ETS certificates. A closer investigation of the impact of these certificates reveals that depending on the scenario, the direct process will break even at ETS prices ranging from 67 €/t_{CO2} (pessimistic scenario) to as low as no ETS support needed (optimistic scenario), while the indirect approach will require 91€/t_{CO2} (pessimistic scenario) to no ETS support (optimistic scenario) (Figure 3a).

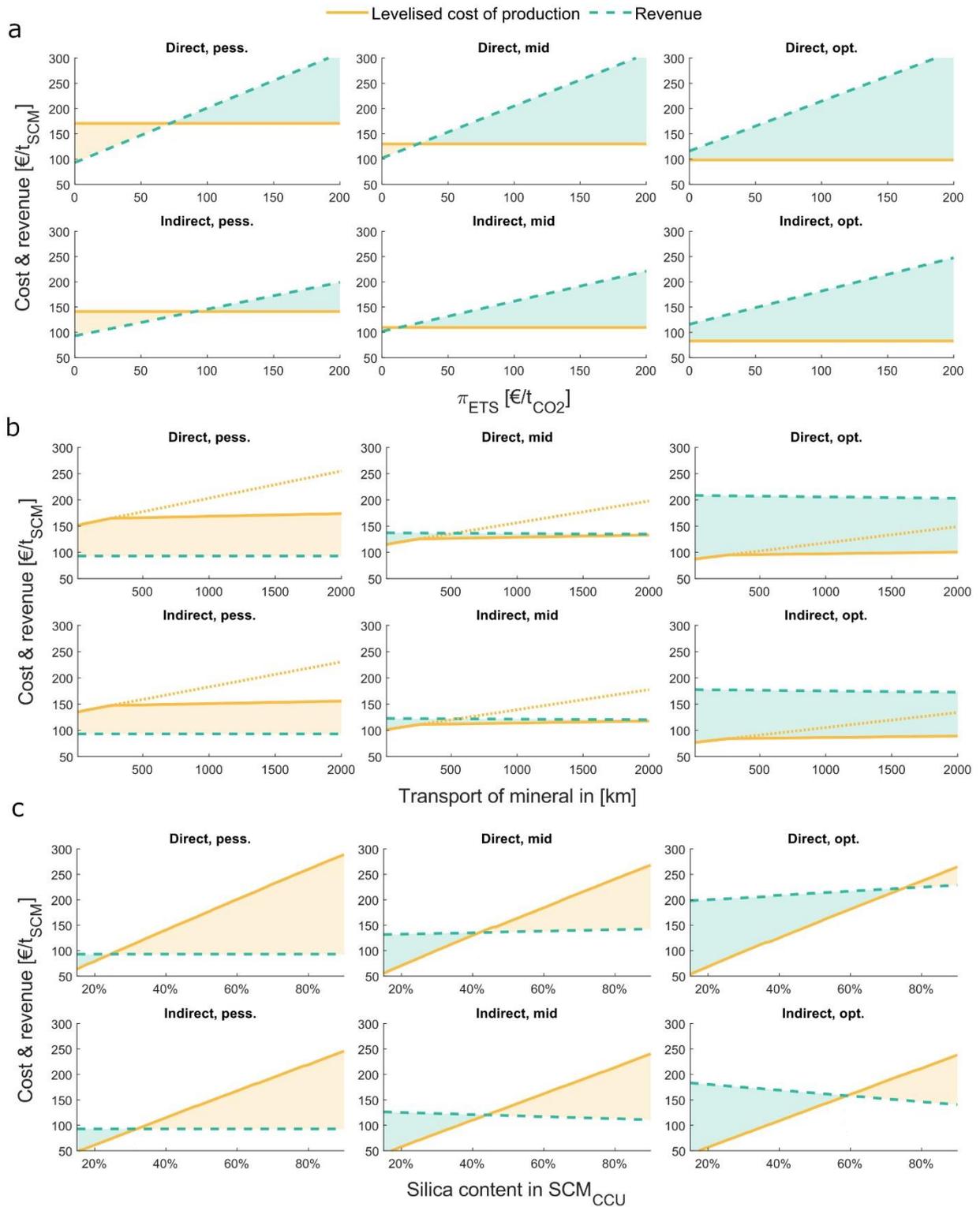


Figure 3: Sensitivities of cost and revenues for different scenarios. Optimized capacities are assumed: low 136 kt/a, mid 272 kt/a, high 340 kt/a. Panel a: effect of ETS price; Panel b: Effect of transport distance of mineral feedstock - dotted line indicates the use of truck (max. 60 km) and train, solid line indicates the use of truck (max. 60km), train (max. 200km) and ship for transport; Panel c: effect of silica content CCU_M product. Only one parameter varied at the time, all other parameters kept constant.

Analysing the significance of mineral feedstock transport elucidates that the revenues gained could offset the costs above 2000km in the optimistic scenario and up to 550km (when transported by truck and train) or 2000km (when transported by truck, train and ship) in the mid scenario (Figure 3b). This

suggests that sufficient revenues from cement replacement and ETS certificates might make CO₂ mineralisation economically viable even when minerals are not mined in very close proximity to the cement plant. It also becomes clear that ship transport is inevitable for longer distances (Figure 3b), which might hinder the implementation of CO₂ mineralisation for some locations.

The share of SiO₂ used in the SCM_{CCU} presents an unexpected trade-off (Figure 3c): as the carbonation reactions produce more carbonate than SiO₂ (Eq. 1 and Eq. 2), achieving lower concentrations of SiO₂ in the SCM_{CCU} require less feedstock mineral to be carbonated. This leads to low concentrations being less costly to produce (smaller plant size and/or smaller separation effort), but also to less CO₂ being stored, creating a trade-off between the revenue from ETS certificates and cost of production for the SCM_{CCU}.

The model shows that lower concentrations in the desired product will improve the economic viability of CO₂ carbonation. In the pessimistic scenario, silica shares below 24% (direct) and 28% (indirect) would be needed in the CCU_M product to break even. In the optimistic, economies of scale as well as higher revenues from increased cement price and ETS will offset the costs of carbonation up to silica shares of 81% (direct) and 58% (indirect). The differences between direct and indirect process can be in part explained by the compositions of the costs (Supplementary Figure 6), which display trade-offs for each process route. While the direct process relies on a separate CO₂ capture plant and compression with intensive pre- and post-treatment, leading to higher capital costs, the indirect process requires more utilities, mainly for the additive regeneration step. Therefore, depending on a cement producer's preference for higher capital costs or higher operational costs, either of the routes could be deemed preferred.

Sensitive factors to lower costs

CO₂ mineralisation processes are still under development and many physico-chemical mechanisms may not yet be fully understood, leading to uncertainty in techno-economic performance. Using a global uncertainty analysis, we investigated which uncertain factors have the largest impact on economic performance, also highlighting directions for cost reductions. Figure 4 shows that the direct process shows a smaller range in the calculated levelised cost of product, with an overall lower mean value compared to the indirect process. The scatter plots reveal that for both processes the price of electricity and the overall interest rate are among the most influential factors: both processes require large amounts of energy, either for grinding of mineral feedstock and compression of CO₂ (direct process), or additive regeneration (indirect process). It needs to be pointed out that the second energy carrier used by these processes, natural gas has a smaller impact on the economic viability, because of the smaller price variance for natural gas than for electricity (Supplementary Figure 9 and Table 12). As discussed above, both carbonation concepts are very capital intensive making them sensitive towards interest on capital as well as the expected learning rate, which will drive down the cost through, e.g., learning by doing when multiple plants are built. These effects are stronger for the direct process due to an overall higher capital intensity. The indirect process shows its highest sensitivity towards the price of the additive ammonium sulphate, which is directly connected to the possible recycling rate of additives (the more can be recycled, the less feed is required). Due to lower concentrations of additives used in the direct process, cost of additives as well as how often they can be recycled play a smaller role in its cost structure in that of the indirect process. Interestingly, the reaction kinetics, represented by the reaction constant, seem to play a smaller role in the cost of the carbonation processes, because they mainly influence the capital costs of the carbonation reactors, which even in the more capital intensive direct process make up approx. 30% of the total capital costs, translating to approx. 6% of the overall levelised cost of product (Supplementary Figure 6). Finally, both processes reduce costs significantly when higher concentrations

of mineral (i.e. solid/liquid ratio) are used, since excess water needs to be separated using centrifuges and requires larger equipment sizes.

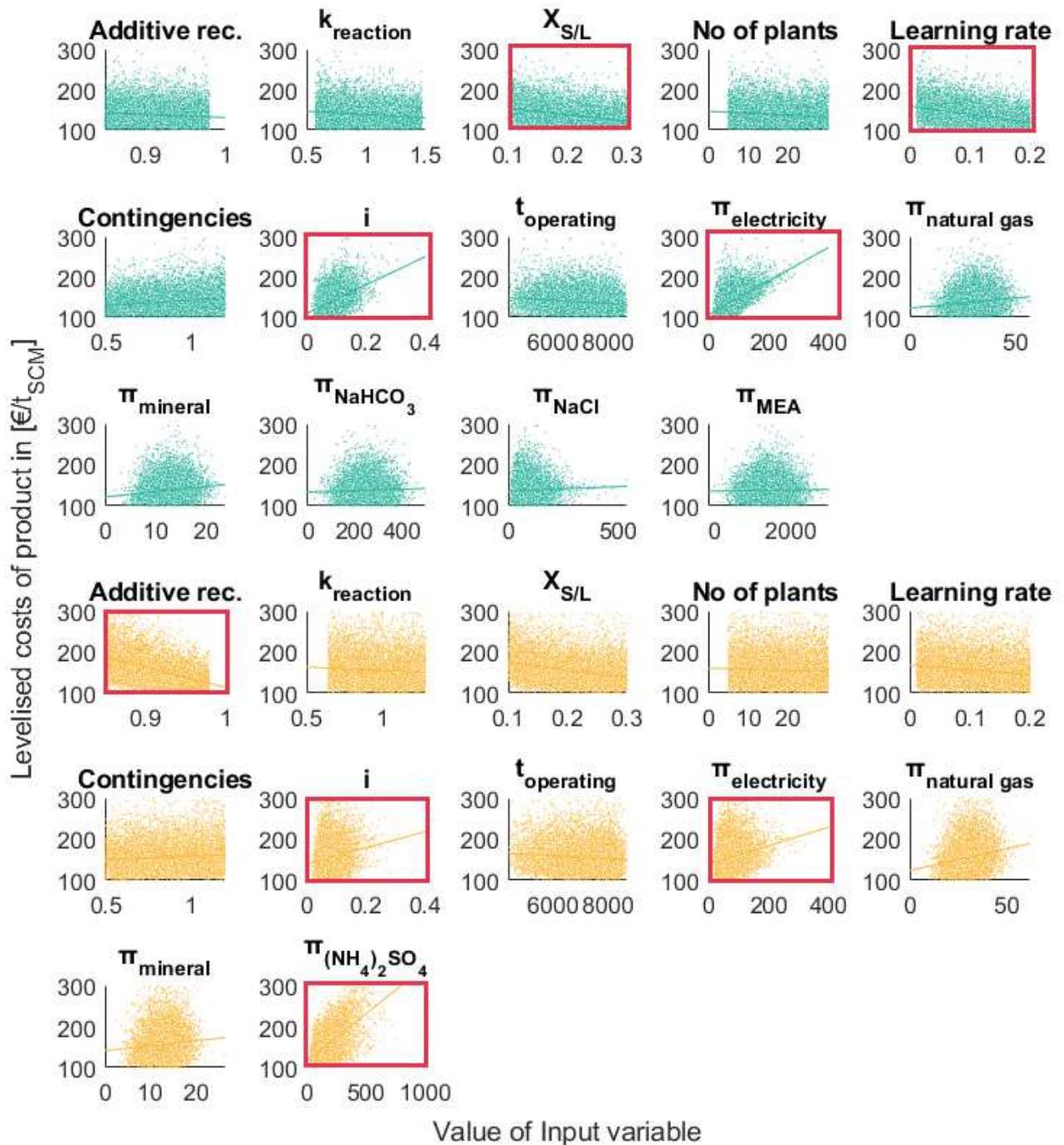


Figure 4: Distribution of LCOP for mid scenario. Top: Direct process; bottom: Indirect process. Frequencies derived by Monte Carlo simulation using 10,000 runs with changing input parameters: Additive recovery (Additive Rec.), reaction constant (k_{reaction}), solid-liquid ratio in reactor ($X_{S/L}$), Number of plants to reach NOAK (No of plants), Learning rate on 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 ($\pi_{\text{(NH}_4)_2\text{SO}_4}$). Input variables with highest impact on the LCOP are marked with box. The frequencies of the sampled input variables are shown in Supplementary Figure 8.

Mineralisation plant cost decline through technological learning

We showed that it is necessary to generate sufficient revenue from using the produced SCM_{CCU} as cement replacement and that in many cases ETS support is needed to create a positive business case. For all capital cost calculations we followed the recently developed hybrid approach by Rubin, et al.^{32,33} that provides a methodologically consistent method to calculate the cost of the N^{th} -of-a-kind (i.e., commercial) plant. We assumed 20 plants must be built to reach this state. After the construction of the first plant, learning effects will drive down the costs with every additional plant being built. This means that the first plants will be significantly more expensive, a development cost that needs to be recovered. In the mid scenario the first 8 plants (direct) and 3 plants (indirect) need additional support to reach a positive business case, while in the pessimistic and optimistic scenarios all first 20 plants will be economically unviable, respectively viable (see Figure 5). This result underpins the recent argument³⁴ that in many cases ETS, or CO_2 taxes, alone are not sufficient to help the market move to low carbon solutions, but that other mechanisms need to be in place, for example government support programmes, like has been the case for wind and solar power. This is a critical observation, as it means that governments may need to invest heavily in first-mover low-carbon plants.

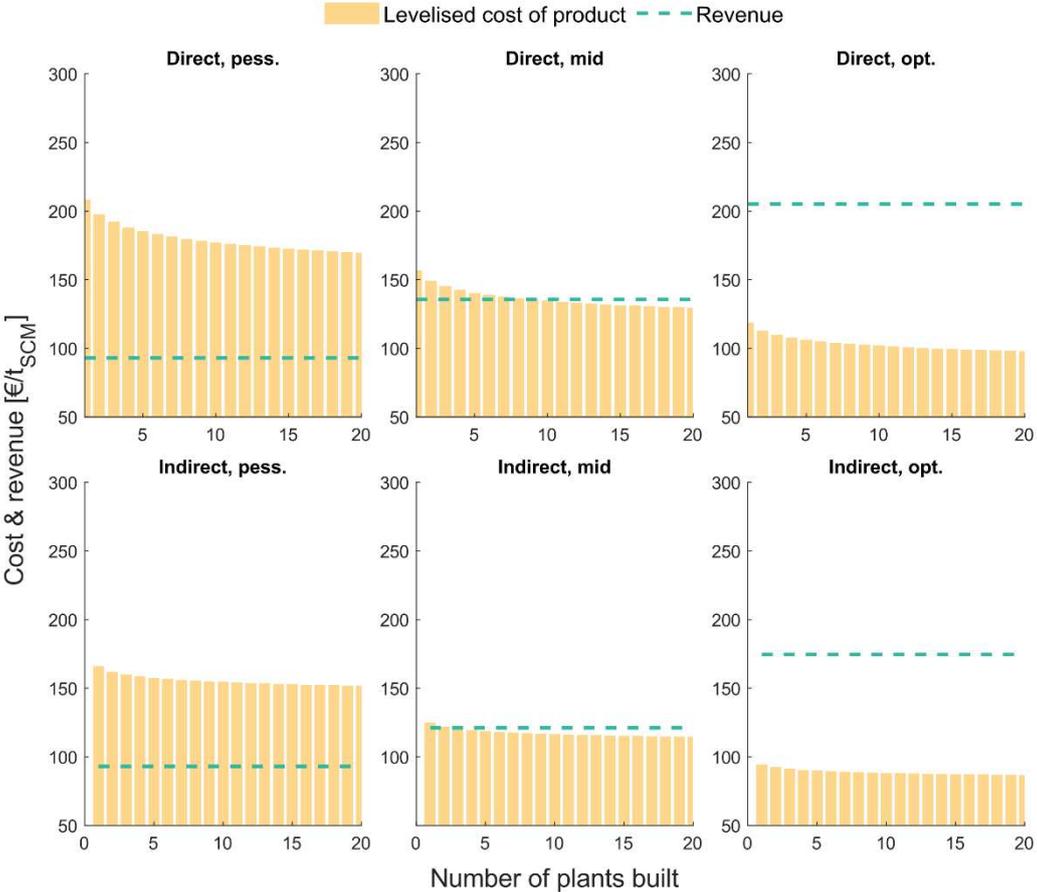


Figure 5: Impact of number of plants build to reach NOAK on the levelised cost of product.

Discussions & conclusion

This study showed that CO₂ mineralisation can reduce the CO₂ emissions from cement production by 6%-31% while generating a positive business case if at least three conditions are met: 1) SCM_{CCU} must be widely accepted and standardised as cement replacement; 2) the production of SCM_{CCU} must be eligible for ETS credits or similar; and 3) the feedstock minerals must be available in relatively close proximity (< ~550 km without ship transport or 2000 km when ship transport is available). SCM_{CCU} thus has a competitive advantage over many other GHG emission reduction measures that cost money, rather than generating it.

Considering the first condition, while initial studies suggest the use of SCM_{CCU} blended with ordinary Portland cement to be feasible (Supplementary Note 3), exact blends that satisfy the requirements of the construction industry (e.g., compressive strength, water demand, curing time) need to be formulated, tested and standardised. We showed that the suggested process routes will be able to provide a variety of blends, which should allow flexibility in producing exactly the formulations required. Regarding the third condition, while we showed that the costs of mineral transport can be counterbalanced by the generated revenue, the economic viability deteriorates when minerals are not available in proximity, limiting the SCM_{CCU} deployment to especially these locations where minerals can be mined regionally, e.g., close to Norway, Italy, or Spain^{18,22,35}, among others.

To reach complete decarbonisation of the cement industry (i.e., beyond what SCM_{CCU} can economically achieve), multiple approaches may have to be implemented in parallel^{6,8}. This may lead to synergies with, or barriers for, SCM_{CCU}. A major synergy can be created by combining CCU_M with CO₂ capture and geological storage by sharing the CO₂ capture and compression plants, leveraging economies of scale, and lowering the specific capital cost burden for the CCU_M. Conversely, a potential barrier for SCM_{CCU} might arise from the simultaneous introduction of other SCMs as means of emission reduction, such as calcined clay. The silica phases in the other SCM^{6,36} could compete with the ones existing in SCM_{CCU}, potentially limiting the effectiveness of these combinations. This may however not always be true, for the CO₂ curing of concrete, a recent study³⁷ showed that in most cases using SCMs with similar silica contents as SCM_{CCU} might increase the probability of achieving emission reductions of the cured concrete, suggesting that SCM_{CCU} could actually be applied in tandem with CO₂ curing approaches. Similarly the GHG emissions reductions explored here can be expected to be complementary to strategies that consider CO₂ reaction the end of the life cycle³⁸. Whether these combinations would lead to overall emission reductions is yet to be analysed by rigorous life cycle analysis. The other critical barrier is standardization and acceptance of cement blends using SCM_{CCU}, as already stated, where experience from the introduction of novel cement blends with the purpose of emission reduction (e.g., limestone-cements) in the past suggests that it might take years to decades to reach a wide market penetration,^{39,40} all the more reason to start formalising these blends now.

On the economics, the analysis showed that the technical performance of the process, especially the carbonation kinetics, is a lesser determinant of final production costs, meaning we can start scaling up the current processes now, without needing to wait for, e.g., improved reactor configurations. Additional research should therefore be focused on the least mature areas of the process (i.e. the separation of products): while the pre-treatment of the minerals (crushing and grinding) can be seen as mature (technology readiness level (TRL) 9) and the mineralisation processes are based on conceptual process designs validated in laboratory settings^{28,29,41,42} (TRL 4), we suggested a new product separation process based on limited laboratory⁴³ and basic research (TRL 2-3, Supplementary Note 2 and 3).

Other economic parameters, e.g. high electricity costs, could however become a barrier for the deployment of CO₂ mineralisation processes in the cement industry. One to bear in mind specifically

might be the interest rate, to which our results were very sensitive: for new technologies, companies (through return on equity) as well as lenders tend to request higher rates, meaning that the first few plants (until completely de-risked) may be more expensive than our analysis finds. Government guaranteed loans may circumvent this, as may direct government subsidies, in addition to the aforementioned ETS eligibility.

Methods

The techno-economic assessment (TEA) and revenue model were specified in MATLAB, allowing us to easily compare different scenarios and rigorously assess ranges of assumptions: Low TRL technologies, including CO₂ mineralisation, are inherently uncertain in nature, a detailed process design is often yet to be formalised and physico-chemical mechanisms may not yet be fully understood, requiring large parametric and/or sensitivity studies to determine possible economic designs and operating conditions⁴⁴. The MATLAB model first solves the processes' mass and energy balances after which the equipment is sized automatically in order to derive the capital costs (CAPEX) or total capital required (TCR) followed by calculations of the operational expenditures (OPEX) and levelised cost of product (LCOP) as well as the expected revenue (R). This sequence was repeated several times for a new set of assumptions to derive cost and revenue hotspots in an iterative manner.

Deriving the levelised cost of product (LCOP)

We calculated the levelized cost of production with discounting capital costs using the expected lifetime of the plant L and the overall interest i incorporating interest on debt i_{debt} , the debt to equity ratio DER as well as the return on equity ROE , reflecting the interest that need to be paid for loans as well as the interest expected by the shareholders of the company (see Eq. 4 to Eq. 6). Note the levelisation factor α , which allows easy annualization of capital costs, assumes the plant is built in one year. This is a simplification often used⁴⁵, and we use it here to allow analytical calculation of annualised TCR.

$$LCOP = \frac{\alpha \cdot TCR + OPEX}{\dot{m}_{SCM}} \quad \text{Eq. 4}$$

$$\alpha = \left(\frac{i}{1 - (1 + i)^{-L}} \right) \quad \text{Eq. 5}$$

$$i = i_{debt} \cdot \left(\frac{DER}{1 + DER} \right) + ROE \cdot \left(\frac{1}{1 + DER} \right) \quad \text{Eq. 6}$$

Calculating capital expenditures

In order to evaluate the economic viability of a mature technology we aim to estimate TCR on the basis of a nth of a kind plant following the approach recently postulated by Rubin, et al.^{32,33} adhering to guidelines for techno-economic evaluations of carbon capture and utilisation technologies⁴⁶. We start with estimating the costs of the first of a kind (FOAK) plant bottom up and used learning rates to determine the nth of a kind (NOAK) cost. This approach is required here, because we aim to answer a 'what will' type of question, i.e., what *will* the costs be of a mature mineralisation technology³², such that we can compare this to expected revenues.

We used the sum of the total direct costs of all process units as a basis to derive the total plant costs (TPC) on which the TCR is based (see Eq. 7).

$$TPC = \sum TDC \cdot (1 + f_{indirect}) \cdot (1 + f_{process}) \cdot (1 + f_{project}) \quad \text{Eq. 7}$$

Where $f_{indirect}$, $f_{process}$, $f_{project}$ factor in indirect costs, process contingencies and project contingences. We calculated the total capital requirement (TCR, including owner's costs and interest during construction) for the NOAK plant based on the TPC of the FOAK plant (see Eq. 8):

$$TCR = \left(\frac{TPC}{\dot{m}_{SCM}} \right) \cdot N^{-E} \cdot \dot{m}_{SCM} \cdot ((1 + i)^{t_{construction}} + f_{owner}) \quad \text{Eq. 8}$$

Here N represents the number of plants necessary to reach NOAK, E the experience factor, i the interest during construction, $t_{construction}$ the estimated time for construction and f_{owner} factors in the owners' cost. The plant's production capacity of SCM is represented by \dot{m}_{SCM} . As the one factor learning rate (LR) is defined as the cost reduction (or increase) through doubling the cumulative production capacity, we derived the experience factor as follows ⁴⁵ (see Eq. 9):

$$E = \frac{\ln(1 - LR)}{\ln(2)} \quad \text{Eq. 9}$$

We derived learning rates for novel processes such as the CO₂ carbonation from a comparable process where learning curves from historical data or literature estimates were available. We selected integrated gasification combined cycle and pulverized coal combustion with CCS (both use solvent-based CO₂ capture and include substantial solids processing). Rubin, et al. ⁴⁷ reported a learning rate for these processes to be between 1.1 and 20%. An assumption needed to be made on when NOAK is reached, i.e., how many plants will actually deploy the technology until we can assume the technology to be mature. Following Greig, et al. ⁴⁸ we use 20 plants as an estimate for reaching maturity, which translates to a market share of 10% in Western Europe, as there are 193 integrated cement plants in Western Europe⁴⁹, which produce clinker themselves and could therefore be suitable for using CO₂ mineralisation.

TDC cost curve estimation

Two approaches were used to estimate the total direct cost: a bottom-up approach, where cost functions for a piece of equipment (pumps, heat exchangers, etcetera) were derived from the Aspen Capital Cost Estimator. This was done by running many different design value combinations (pressure, temperature, flow, etcetera) and fitting the resulting data points to a curve that could be implemented into our TEA model; and second a top-down approach, using factorial methods with TCR estimates from existing plants or open literature. As there are several detailed cost estimates for CO₂ capture and compression, we used the top-down approach for these operations. For all other unit operations, we calculated the TPC using the bottom-up approach. The top down approach is shown in Eq. 10, where \dot{m}_i represents the plant's capacity, n the scaling factor and I capital cost index for a certain year to account for inflation⁵⁰:

$$TDC = TDC_{old} \cdot \left(\frac{\dot{m}_{new}}{\dot{m}_{old}}\right)^n \cdot \left(\frac{I_{new}}{I_{old}}\right) \quad \text{Eq. 10}$$

For all unit operations calculated using the bottom up approach we used the Aspen Capital Cost Estimator to derive consistent, comparable and up to date estimates of the capital costs. Because the Aspen Capital Cost Estimator can only evaluate a discrete set of design value combinations, we created a set of data points for each of the proposed equipment and used regressions to derive cost curves that can be implemented in the TEA model. The goal of these cost curves is to be able to predict costs for data points that are in-between the discrete points calculated in Aspen. For extrapolation, these function should only be used very carefully, as it can be assumed that if the input is out of the boundaries in the Aspen Capital Cost Estimator, there might be technical limitations in order to build the equipment (for example: reactor size and wall stability issues). We based regression approaches on the widely used cost curve approach that can be found in such textbooks as Towler and Sinnott ⁵⁰ which we expanded where necessary. We used Eq. 11 when the design parameters can be assumed independent, Eq. 12 when the design parameters are not independent and Eq. 13 when the influence of one variable is scaling in the opposite direction of the other.

$$TDC = a \cdot x^n + b \cdot y^m + c \quad \text{Eq. 11}$$

$$TDC = a \cdot x^n + b \cdot y^m + c + d \cdot x \cdot y \quad \text{Eq. 12}$$

$$TDC = a \cdot x^{-n} + b \cdot y^m + c \quad \text{Eq. 13}$$

Sizing of equipment

As a basis for the TDC estimate the model sizes the equipment.. While for some equipment this is undemanding (e.g. for a ball mill), some equipment needed to be described in more detail (e.g. heat exchangers). The number of identical equipment pieces is selected by the model with a simple iterative heuristic: First the equipment is sized; afterwards it is assessed whether the equipment exceeds the maximum size for this unit (taken from Aspen Capital Cost Estimator). If the calculated design exceeds the criteria, the task is divided by identical equipment units until no single piece exceeds the maximum size.

Sizing crusher and grinder

The mineral pre-treatment steps crushing and grinding have been investigated by several researchers. An in-depth analysis was published by Gerdemann et al. who report 2 kWh/t_{mineral} for crushing and between 81 and 97 kWh/t_{mineral} for grinding to be necessary in order to activate the minerals for the carbonation reaction²⁵ (Supplementary Table 6). For the process jaw crushers and ball mills are considered due to their low costs and the widely usage in the cement industry to this point. We size the crusher according to material throughput and ball mill by energy requirement (Supplementary Table 7).

Sizing reactors

The TEA model is based on literature values for the reaction rate for different processes and conditions. All studies used reported data for an autoclave run in a batch process^{25,28,29,51,52}. As full-scale processes with their large amount of feedstock requirement are expected to be performed as a continuous reaction, the model uses continuously stirred tank reactors (CSTR). We used space time τ to derive the size of CSTR reactors. Under the assumption that the density of the system does not change significantly during the reaction, the volume of CSTR and space time is defined as follows (see Eq. 14 and Eq. 15)⁵³:

$$V = \frac{\dot{v}_o \cdot c_{Ao} \cdot X}{-r_A} \quad \text{Eq. 14}$$

$$\tau = \frac{V}{\dot{v}_o} \quad \text{Eq. 15}$$

We calculated the volume for the reactors under a 1st order reaction assumption (see Eq. 16 and Eq. 17).

$$\tau = \frac{c_{Ao} \cdot X}{-r_A} = \frac{c_{Ao} \cdot X}{k \cdot c_{Ao} \cdot (1 - X)} = \frac{X}{k \cdot (1 - X)} \quad \text{Eq. 16}$$

$$V_{x,CSTR} = \dot{v}_{slurry} \cdot \tau \quad \text{Eq. 17}$$

After the total reaction volume is calculated, it was needed to design each individual reactor unit. The equipment costs for the carbonation reactors were estimated on the basis of a pressure vessel. First, we derived the reactor height and radius, with the minimal surface area (closest to the sphere), to reduce material costs.

$$V_{CSTR} = 2 \cdot \pi \cdot r^3 \quad \text{Eq. 18}$$

$$r = \left(\frac{V_{CSTR}}{2\pi} \right)^{\frac{1}{3}} \quad \text{Eq. 19}$$

$$h = 2r \quad \text{Eq. 20}$$

Second, we calculated the wall thickness following Towler and Sinnott⁵⁰:

$$t_w = \frac{P_i \cdot 2r_i}{2 \cdot S \cdot E - 1.2 \cdot P_i} \quad \text{Eq. 21}$$

To determine the size and number of reactors, we use the criteria minimum wall thickness according to typical maximal allowable stress⁵⁴, maximum wall thickness and maximum size of reactor to derive the number and sizes of CSTRs.

Separation via hydro cyclones

For the separation of unreacted mineral and the reaction products in the direct process route we propose to use hydro cyclones. The separation process was simulated in Aspen Plus, the assumptions are shown in the supplementary information. To size the hydro cyclones, we performed parametric studies in Aspen Plus to obtain points of the design space on which we fitted curves. The number of cyclones N depends on the mass flow (\dot{m}) of separated mineral and the separation efficiency (E), while the diameter (d) is a function of E . The best fit was found with the following relationship:

$$N_{cyclones} = a + b \cdot \dot{m}_{slurry}^n \cdot E^m \quad \text{Eq. 22}$$

$$d_{cyclone} = a \cdot E^n + b \quad \text{Eq. 23}$$

The Aspen simulation showed that small changes of the mineral distribution caused by different conversion rates (e.g. 0.5 instead of 0.7 conversion) will not change the design or efficiency of the separation significantly (between 0.07% - 0.3% difference), thus were neglected in this model.

Dewatering using centrifuges

For the dewatering we used solid bowl centrifuges. As reported in literature, solid bowl centrifuges might be viable for ultra-fine separation, here high rotation speeds need to be considered (> 3000rpm)⁵⁵. The dewatering was simulated in Aspen Plus and a parametric study was performed using different diameters, length and rotation speeds. We selected an optimal design for each point where according to the simulation no material is lost in the wet stream. The derived functions can be described as follows, with d being the diameter, rpm the centrifugal speed, and l the length (see Eq. 24 and Eq. 25):

$$d_{dewatering} = a + d \cdot rpm^o \cdot \dot{m}_{water\ removed}^m \quad \text{Eq. 24}$$

$$\frac{l_{dewatering}}{d_{dewatering}} = a + c \cdot rpm^o + d \cdot \dot{m}_{water\ removed} + b \cdot rpm^n \cdot \dot{m}_{water\ removed}^m \quad \text{Eq. 25}$$

Separation via classification centrifuges

The classification of product is used in the direct carbonation process. The classification aims to separate Silica (0-1 μ m) from MgCO₃ (1-5 μ m). Aspen Plus simulation showed disc centrifuge to be sufficient for this task. The sizes for different throughputs were derived using parametric studies in Aspen Plus. A regression was used to determine a design function (see Eq. 26).

$$d_{classification} = a + b \cdot \dot{m}_{slurry}^n \quad \text{Eq. 26}$$

Sizing heat exchangers and furnaces, crystallisers and rotary dryers

The heat exchanger and furnace sizes were calculated using the transferred heat ($\dot{Q}_{HE,i}$), which was derived using the Shomate equation. The general approach can be described as follows (see Eq. 27):

$$\begin{aligned} \dot{Q}_{HE,i} &= \dot{H}_{HE,out} - \dot{H}_{HE,in} \\ &= \sum \left(\Delta H_{f,j,out}^{25^\circ C} + \int_{25^\circ C}^{T_{out}} cp \, dT \right) \cdot \dot{n}_{j,out} - \sum \left(\Delta H_{f,j,in}^{25^\circ C} + \int_{25^\circ C}^{T_{in}} cp \, dT \right) \cdot \dot{n}_{j,in} \end{aligned} \quad \text{Eq. 27}$$

For the design of furnaces, the duty and volumetric flow rate are sufficient input for the TEA model. For heat exchangers, crystallisers and rotary dryers the area was derived in order to size the equipment (see Eq. 28).

$$A = \frac{\dot{Q}}{k \cdot \Delta T_{ln}} \quad \text{with } \Delta T_{ln} = \frac{(T_{He} - T_{Ka}) - (T_{Ha} - T_{Ke})}{\ln\left(\frac{T_{He} - T_{Ka}}{T_{Ha} - T_{Ke}}\right)} \quad \text{Eq. 28}$$

CO₂ capture

Of the two processes studied here, only the direct carbonation process uses separate CO₂ capture and compression equipment. An MEA post-combustion capture plant was considered for the direct process route. Following the top down approach shown in Eq. 10, the MEA capture costs were calculated using the data published by Anantharaman, et al. ⁵⁶, which have been estimated specifically for the case of capturing CO₂ at a cement production site. The indirect process uses an integrated ammonia capture process. Following the top down approach shown in Eq. 10, the ammonia capture costs are calculated using the data published by Li, et al. ⁵⁷. As the indirect process does not use the CO₂ stripper because ammonium bicarbonate is used as the CO₂ carrier directly, only relevant units of the published data were selected; the CO₂ stripper was replaced by the additive regeneration step in the indirect process.

CO₂ Compression

For deriving the capital cost functions for compression, recent literature has shown that the estimations can differ largely (i.e. some estimates show differences of over 1000%) ⁵⁸. We utilize calculations from van der Spek, et al. ⁵⁹ following the top down approach shown in Eq. 10. Here, a five-stage compression is simulated after the MEA post-combustion capture plant. The desired final pressure in this paper was set to 110bar. The pressures used for the direct carbonation process are in a similar range (100-150bar), thus a scaling approach is used for estimating the total direct costs for the compressors in a top down manner. We selected a scaling factor of 0.76 following IEAGHG ⁶⁰.

Calculating operational expenditures

OPEX were split into fixed and variable OPEX. Fixed OPEX covers salaries for employees C_{Labour} to run the plant, insurance and local tax $C_{insurance}$, maintenance $C_{maintenance}$ as well as administration C_{admin} and support. Variable OPEX include costs of utilities $C_{electricity \& \text{ natural gas}}$ and feedstocks $C_{feed, total}$ and costs of transport. We used pricing data from different publications as well as web-based platforms such as Statista or Alibaba. The basis for the OPEX calculations is shown in Supplementary Table 12.

Fixed Operational Expenditures

We derived fixed OPEX using the approach from Peters, et al. ⁶¹ for cost of labour and for all other fixed OPEX following Anantharaman, et al. ⁵⁶ (see Eq. 29 to Eq. 32).

$$C_{labour} = 10.447 \cdot \left(\frac{\dot{m}_{SCMCCU}}{t_{working}} \cdot 24 \right)^{0.2485} \cdot \pi_{salary} \quad \text{Eq. 29}$$

$$C_{insurance} = f_{insurance} \cdot TPC \quad \text{Eq. 30}$$

$$C_{maintenance} = f_{maintenance} \cdot TPC \quad \text{Eq. 31}$$

$$C_{admin} = f_{admin} \cdot (C_{labour} + C_{maintenance}) \quad \text{Eq. 32}$$

Variable Operational Expenditures

The cost calculation of feedstocks and utilities used the mass and energy balances as a basis. The costs for the feedstocks and utilities were multiplied by the market price π (see Eq. 33 to Eq. 35).

$$C_{electricity \& \text{ natural gas}} = \sum w_i \cdot \pi_i \quad \text{Eq. 33}$$

$$C_{feed, i} = \dot{m}_{feed, i, total} \cdot \pi_i \quad \text{Eq. 34}$$

$$C_{feed, total} = \sum C_{feed, i} \quad \text{Eq. 35}$$

The cost of transport was taken into account as well. Here, it was assumed that the minerals are transported no further than 60km by truck and 200km by train; distances beyond 260km will be done by ship transport ¹⁹. As some material will not be used in the SCM_{CCU} it has to be stored. The storage quarry was assumed to be 10 km away from the cement plant and it is accessed by truck. The costs were calculated using different prices for transport π_i and distances $dist_i$:

$$C_{transport} = \dot{m}_{mineral,in} \cdot (\pi_{truck} \cdot dist_{truck} + \pi_{train} \cdot dist_{train} + \pi_{ship} \cdot dist_{ship}) \quad \text{Eq. 36}$$

Calculating the revenue

For the calculation of the revenue, first we determined how much of the produced material must have to be stored in a quarry and what share can be used to displace cement production (\dot{m}_{used} and material being stored \dot{m}_{stored}). There are two reasons why material needs to be stored: either too little silica is produced, so that some of the inert material needs to be separated and disposed; or silica and inert material need to be stored when the capacity of the carbonation plant $\dot{m}_{SCM_{CCU}}$ is larger than the material that could be blended with the cement $\dot{m}_{cementblend}$ (See Eq. 37 and Eq. 38 with X_i being share of material i in the cement blend and \dot{m}_{cement} the total capacity of the cement plant.

$$\dot{m}_{used} = \begin{cases} \dot{m}_{SiO_2,out} + \dot{m}_{SiO_2,out} \cdot \frac{X_{inert}}{X_{SiO_2}}, & \dot{m}_{SCM_{CCU}} \leq \dot{m}_{cementblend} \\ \dot{m}_{cement} \cdot (X_{SiO_2} + X_{inert}), & \dot{m}_{SCM_{CCU}} > \dot{m}_{cementblend} \end{cases} \quad \text{Eq. 37}$$

$$\dot{m}_{stored} = (\dot{m}_{SiO_2} + \dot{m}_{inert}) - \dot{m}_{blend} \quad \text{Eq. 38}$$

To assess the CO₂ emissions displaced by SCM, calculations from Ostovari, et al. ¹⁹ were adapted. We accounted for the minerals' mining emissions and additive production, e_{feed} , the added emissions for transport of the mineral to the plant, $e_{t,min}$, and carbonated material to the storage site, $e_{t,stor}$, added emissions for electricity, e_{el} , and natural gas, e_{ng} , as well as for the construction of the plant, e_{constr} , as additional CO₂ burden. From that burden we subtracted the emission reductions through CO₂ that is bound in the product, e_{bound} , and emissions that are avoided by replacing clinker production with SCM_{CCU}, $e_{replace}$ (see Eq. 39).

$$e_{SCM_{CCU}} = (e_{feed} + e_{t,min} + e_{t,stor} + e_{el} + e_{ng} + e_{constr}) - (e_{bound} + e_{replace}) \quad \text{Eq. 39}$$

Using the mass and the net emissions of SCM_{CCU} and the cement selling price π_{cement} , the overall profit was derived by subtracting the production costs from revenue r .

$$r = \frac{\dot{m}_{used} \cdot \pi_{cement} - \dot{m}_{stored} \cdot c_{storage} + e_{SCM_{CCU}} \cdot \pi_{ETS}}{\dot{m}_{SCM_{CCU}}} \quad \text{Eq. 40}$$

$$profit = r - LCOP \quad \text{Eq. 41}$$

$$\Delta\pi_{cement} = \frac{profit \cdot \dot{m}_{SCM_{CCU}}}{\dot{m}_{cement,out}} \quad \text{Eq. 42}$$

Uncertainty analysis

Uncertainties are inevitable in early stage techno economic assessments such as this CCU_M study. Such technologies are inherently uncertain in nature, requiring large sensitivity studies to determine possible economic designs and operating conditions ^{37,44}. In order to perform thorough techno-economic

assessments, Monte Carlo simulation following a first screening with single factor sensitivity analysis has been suggested, to reduce the overall computational effort for the Monte Carlo simulation^{44,46,62-64}. We followed that approach here to identify potentially influential parameters on model results, in which each input parameter was changed by the same amount from the nominal value (-50% to +50%), except if clear natural limits were reached (such as reaction yield that cannot be larger than 1) (Supplementary Figure 8). As many input parameters' influence on the results might have interdependencies to other input parameters and themselves carry a probability of reaching a certain value⁶², a probabilistic uncertainty analysis in the form of a Monte Carlo simulation was performed for the input parameters that showed a high sensitivity in the single factor analysis. For each selected parameter we determined a probability density function following the approach by Hawer, et al.⁶⁴ and performed a Monte Carlo simulation with 10,000 runs using the open-source tool UQLAB⁶⁵. The used probability density functions are described in Supplementary Figure 9 and Supplementary Table 13.

Acknowledgements

Till Strunge has been funded by the German Federal Ministry of Education and Research (BMBF) as part of the project "CO2MIN" (033RC014). Phil Renforth is funded by the UK's Greenhouse Gas Removal Programme, supported by the Natural Environment Research Council, the Engineering and Physical Sciences Research Council, the Economic & Social Research Council, and the Department for Business, Energy & Industrial Strategy under grant no. NE/P019943/1. We want to thank the "CO2MIN" project partners at RWTH Aachen and Heidelberg Cement for their support, in particular we want to dearly thank Mr. Hesam Ostovari and Mr. Dario Kremer for their detailed feedback.

Data availability

Data generated as part of this study have been made available in the supplementary information. Upon request by the editor or reviewer these can also be provided in form of Microsoft Excel files for the final submission.

Code availability

The MATLAB code developed as part of this study can be provided to the editors and reviewers upon request. The MATLAB code will be made accessible (e.g. via github) before publication.

References

- 1 IEA. Cement. (Paris, 2020).
- 2 Le Quéré, C. *et al.* Global carbon budget 2018. *Earth System Science Data* **10**, 2141-2194 (2018).
- 3 Czigler, T., Reiter, S., Schulze, P. & Somers, K. Laying the foundation for zero-carbon cement. (McKinsey & Company, 2020).
- 4 Fortune Business Insights. *Cement Market Size, Share & Industry Analysis, by Type, by Application and Regional Forecast 2019-2026*, <<https://www.fortunebusinessinsights.com/industry-reports/cement-market-101825>> (2019).
- 5 Expert Market Research. *Global Cement Market to Reach 6.08 Billion Tons by 2026*, <<https://www.expertmarketresearch.com/pressrelease/global-cement-market#:~:text=According%20to%20a%20new%20report,6.08%20Billion%20Tons%20by%202026>> (2020).
- 6 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).
- 7 Hoddinott, P. The role of cement in the 2050 low carbon economy. *Proceedings of the European Cement Association* (2013).
- 8 The European Cement Association. Cementing the European Green Deal. (The European Cement Association,, Brussels, 2020).
- 9 Andrew, R. M. Global CO₂ emissions from cement production. *Earth System Science Data* **10**, 195 (2018).
- 10 Maddalena, R., Roberts, J. J. & Hamilton, A. Can Portland cement be replaced by low-carbon alternative materials? A study on the thermal properties and carbon emissions of innovative cements. *Journal of Cleaner Production* **186**, 933-942, doi:<https://doi.org/10.1016/j.jclepro.2018.02.138> (2018).
- 11 Gartner, E. & Sui, T. Alternative cement clinkers. *Cement and concrete research* **114**, 27-39, doi:10.1016/j.cemconres.2017.02.002 (2018).
- 12 Shubbar, A. A., Sadique, M., Kot, P. & Atherton, W. Future of clay-based construction materials – A review. *Construction and Building Materials* **210**, 172-187, doi:<https://doi.org/10.1016/j.conbuildmat.2019.03.206> (2019).
- 13 D'Amico, B., Pomponi, F. & Hart, J. Global potential for material substitution in building construction: The case of cross laminated timber. *Journal of Cleaner Production* **279**, 123487, doi:<https://doi.org/10.1016/j.jclepro.2020.123487> (2021).
- 14 Bellmann, E. & Zimmermann, P. Climate protection in the concrete and cement industry - Background and possible courses of action. (Berlin, 2019).
- 15 Deolalkar, S. P. in *Designing Green Cement Plants* (ed S. P. Deolalkar) 83-86 (Butterworth-Heinemann, 2016).
- 16 Roussanaly, S. *et al.* Techno-economic Analysis of MEA CO₂ Capture from a Cement Kiln – Impact of Steam Supply Scenario. *Energy Procedia* **114**, 6229-6239, doi:<https://doi.org/10.1016/j.egypro.2017.03.1761> (2017).
- 17 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).
- 18 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).
- 19 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).
- 20 Romanov, V. *et al.* Mineralization of Carbon Dioxide: A Literature Review. *ChemBioEng Reviews* **2**, 231-256, doi:10.1002/cben.201500002 (2015).
- 21 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).

- 22 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).
- 23 Fagerlund, J., Nduagu, E., Romão, I. & Zevenhoven, R. CO₂ fixation using magnesium silicate minerals part 1: Process description and performance. *Energy* **41**, 184-191, doi:10.1016/j.energy.2011.08.032 (2012).
- 24 Woodall, C. M., McQueen, N., Pilorgé, H. & Wilcox, J. Utilization of mineral carbonation products: current state and potential. *Greenhouse Gases: Science and Technology* **9**, 1096-1113 (2019).
- 25 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).
- 26 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).
- 27 Narahariseti, 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).
- 28 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).
- 29 Wang, X. & Maroto-Valer, M. M. Optimization of carbon dioxide capture and storage with mineralisation using recyclable ammonium salts. *Energy* **51**, 431-438 (2013).
- 30 Siwior, P. & Bukowska, J. Commentary on European Court of Justice judgement of 19 January 2017 in case C-460/15 Schaefer Kalk GmbH & Co. KG v Bundesrepublik Deutschland. *Environmental Protection and Natural Resources; The Journal of Institute of Environmental Protection-National Research Institute*. **29**, 25-30 (2018).
- 31 Porterfield, W. W. *Inorganic chemistry*. (Academic press, 2013).
- 32 Rubin, E. S. *et al.* in *Towards improved guidelines for cost evaluation of carbon capture and storage* (eds Simon Roussanaly, Edward S. Rubin, & Mijndert Van der Spek) (2021).
- 33 Rubin, E. S. Improving cost estimates for advanced low-carbon power plants. *International Journal of Greenhouse Gas Control* **88**, 1-9 (2019).
- 34 Patt, A. & Lilliestam, J. The Case against Carbon Prices. *Joule* **2**, 2494-2498, doi:<https://doi.org/10.1016/j.joule.2018.11.018> (2018).
- 35 Park, A.-h. A., Kelemen, P., Matter, J. & Gadikota, G.
- 36 Nguyen, Q. D., Afroz, S. & Castel, A. Influence of Clay Calcination Method on the Mechanical Properties and Chloride Diffusion Resistance of Limestone Calcined Clay Cement (LC3) Concrete. *Journal of Marine Science and Engineering* **8**, 301 (2020).
- 37 Ravikumar, D. *et al.* Carbon dioxide utilization in concrete curing or mixing might not produce a net climate benefit. *Nature Communications* **12**, 855, doi:10.1038/s41467-021-21148-w (2021).
- 38 Renforth, P. The negative emission potential of alkaline materials. *Nature Communications* **10**, 1401, doi:10.1038/s41467-019-09475-5 (2019).
- 39 Dewald, U. & Achternbosch, M. Why more sustainable cements failed so far? Disruptive innovations and their barriers in a basic industry. *Environmental Innovation and Societal Transitions* **19**, 15-30, doi:<https://doi.org/10.1016/j.eist.2015.10.001> (2016).
- 40 Manns, W., Thielen, G. & Laskowski, C. *Evaluation of the results of tests for building inspectorate approvals for Portland Limestone cements.*, (Verein Deutscher Zementwerke e.V./Forschungsinstitut der Zementindustrie (Eds.), 2000).
- 41 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).
- 42 Sanna, A., Gaubert, J. & Maroto-Valer, M. M. Alternative regeneration of chemicals employed in mineral carbonation towards technology cost reduction. *Chemical Engineering Journal* **306**, 1049-1057 (2016).
- 43 Kremer, D. & Wotruba, H. Separation of Products from Mineral Sequestration of CO₂ with Primary and Secondary Raw Materials. *Minerals* **10**, 1098 (2020).

- 44 van der Spek, M. *et al.* Unravelling uncertainty and variability in early stage techno-economic assessments of carbon capture technologies. *International Journal of Greenhouse Gas Control* **56**, 221-236 (2017).
- 45 Rubin, E. S. *et al.* A proposed methodology for CO₂ capture and storage cost estimates. *International Journal of Greenhouse Gas Control* **17**, 488-503 (2013).
- 46 Zimmermann, A. *et al.* Techno-Economic Assessment & Life Cycle Assessment Guidelines for CO₂ Utilization (Version 1.1). (2020).
- 47 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).
- 48 Greig, C., Garnett, A., Oesch, J. & Smart, S. Guidelines for scoping and estimating early mover ccs projects. *Univ. Queensl., Brisbane* (2014).
- 49 CemNet. *The Global Cement Report - Online Database of Cement Plants*, <<https://www.cemnet.com/global-cement-report/>> (2021).
- 50 Towler, G. & Sinnott, R. *Chemical engineering design: principles, practice and economics of plant and process design*. (Elsevier, 2012).
- 51 Wang, X. & Maroto-Valer, M. M. Dissolution of serpentine using recyclable ammonium salts for CO₂ mineral carbonation. *Fuel* **90**, 1229-1237 (2011).
- 52 Wang, X. & Maroto-Valer, M. M. Integration of CO₂ capture and mineral carbonation by using recyclable ammonium salts. *ChemSusChem* **4**, 1291-1300 (2011).
- 53 Smith, R. *Chemical process: design and integration*. (John Wiley & Sons, 2005).
- 54 Towler, G. & Sinnott, R. K. *Chemical engineering design: principles, practice and economics of plant and process design*. (Elsevier, 2012).
- 55 Pinkerton, A. P. & Klima, M. S. Evaluation of a solid-bowl centrifuge for ultrafine size separations. *Mining, Metallurgy & Exploration* **18**, 162-166 (2001).
- 56 Anantharaman, R. *et al.* CEMCAP Framework for Comparative Techno-economic Analysis of CO₂ Capture From Cement Plants-D3. 2. (Zenodo, 2018).
- 57 Li, K. *et al.* Technoeconomic Assessment of an Advanced Aqueous Ammonia-Based Postcombustion Capture Process Integrated with a 650-MW Coal-Fired Power Station. *Environmental Science & Technology* **50**, 10746-10755, doi:10.1021/acs.est.6b02737 (2016).
- 58 Knoope, M., Ramírez, A. & Faaij, A. A state-of-the-art review of techno-economic models predicting the costs of CO₂ pipeline transport. *International journal of greenhouse gas control* **16**, 241-270 (2013).
- 59 van der Spek, M., Ramirez, A. & Faaij, A. Challenges and uncertainties of ex ante techno-economic analysis of low TRL CO₂ capture technology: Lessons from a case study of an NGCC with exhaust gas recycle and electric swing adsorption. *Applied Energy* **208**, 920-934 (2017).
- 60 IEAGHG. *Assessment of emerging CO₂ capture technologies and their potential to reduce costs*. (IEAGHG, 2014).
- 61 Peters, M. S., Timmerhaus, K. D. & West, R. E. Plant design and economics for chemical engineers. *International edition* (1991).
- 62 van der Spek, M. *et al.* Uncertainty analysis in the techno-economic assessment of CO₂ capture and storage technologies. Critical review and guidelines for use. *International Journal of Greenhouse Gas Control* **100**, 103113, doi:<https://doi.org/10.1016/j.ijggc.2020.103113> (2020).
- 63 Rubin, E. S. Understanding the pitfalls of CCS cost estimates. *International Journal of Greenhouse Gas Control* **10**, 181-190, doi:<https://doi.org/10.1016/j.ijggc.2012.06.004> (2012).
- 64 Hawer, S., Schönmann, A. & Reinhart, G. Guideline for the Classification and Modelling of Uncertainty and Fuzziness. *Procedia CIRP* **67**, 52-57 (2018).
- 65 Marelli, S. & Sudret, B. in *The 2nd International Conference on Vulnerability and Risk Analysis and Management (ICVRAM 2014)*. pp. 2554–2563.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SupplementaryInformationTowardsabusinesscaseforCO2mineralisation.docx](#)