

Integrating CO₂ mineralization in industrial clusters: the benefits of material and heat integration

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Abstract

Curbing and capturing CO₂ emissions is no longer enough to cope with the demanding environmental challenges of the coming years. Long-term storage technologies need deployment, helping industrial sectors to reach ambitious emission standards. Mineral carbonation, a process in which CO₂ reacts with metal-oxides forming stable and insoluble carbonates mimicking the natural weathering process, is a promising avenue to deliver net-negative emissions. In this work, we simulate and optimize the integration of mineralization in industrial clusters. Waste incineration and cement production sectors are used to demonstrate potential synergies. Several mineral ores (serpentine, olivine and wollastonite) are studied and both direct and indirect carbonation reactions are modeled. Results show how mineralization can be successfully integrated in reducing and achieving net-negative emissions. However, the required investment is non-negligible. A CO₂ tax can be used to favor mineralization and was computed for both sectors; values range from 60 to 90 \$/ton CO₂ for cement and between 120 to 200 \$/ton CO₂ for waste incineration.

Keywords

Carbon capture usage and storage, mineralization, industrial symbiosis, heat integration, simulation, optimization

Introduction

The capture, conversion and use of CO₂ is a major topic in both academic and non-academic environments, covering newspapers headlines and filling international political agendas. Once captured, CO₂ can be either directly used for enhancing industrial processes (e.g. enhanced oil recovery, greenhouses) or instead converted to fuels (such as methane, methanol, and formic acid), chemicals (e.g. plastics) or building materials (i.e. cement and concrete). Indeed, carbon capture utilization and storage (CCUS) is a sound technology cluster to stabilize the atmospheric concentration of CO₂. It has found applications in processes like cement, steel making and power generation. However, according to Romão et al. (2012) it presents many challenges such as its high complexity and integration into a single process, alongside the uncertainty of quantifying the storage capacity of geological sites and the potential leakage of CO₂. Moreover, the large majority of industrial processes require high-purity (above 90%) CO₂ concentration, which is often challenging from an energetic and economic perspective.

From a current annual CO₂ bill of 34 Gt to 0 Gt by 2050 IEA (2019b), the road to curbing global CO₂ and greenhouse gases (GHG) emissions is not straightforward. There

is a growing awareness of the need for net-negative emission technologies (NET). Not only are these supposed to compensate for unavoidable CO₂ emitting activities (such as waste incineration or cement production), but also to compensate for less developed economies in which environmental concerns are secondary. Indeed, the industrial sector alone accounts for 24% of current emissions IEA (2019b). Despite the effort to reduce energy-related emissions by renewable deployment, the sector contains hard-to-eliminate emissions, being CO₂ a sub-product in numerous processes. An expansion in NET deployment is thus foreseen in the near future.

Two main NET approaches currently under study are deep-geological storage and mineralization IEA (2019b). The former has gained some traction in the past years and aims at exploring empty oil and gas fields for compressed CO₂ injection. Although promising, projects are still being developed to demonstrate its feasibility, scalability and safety as well as its potential to mitigate GHG emissions (Bui et al. (2018)). Mineralisation, on the other hand, is not new and has been documented for more than a century, with practitioners and engineers trying to circumvent limitations for large-scale deployment.

Mineralization

Carbon capture and usage by mineralization (CCU_m) is a promising avenue to effectively reduce industrial emissions, starting to offer processes that can accommodate typical flue

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gas compositions (CO₂ content between 10 and 15 %) with high conversion. CO₂ mineralization is also recognized as a promising technology in an industrial environment. The International Energy Agency (IEA) issued a technical report IEA (2019a) on potential commercial uses of CO₂. Among these, CO₂-cured concrete is the most mature of carbonation reactions, already reaching industrial application. However, the CO₂ harvesting rate is on the lower spectrum of all CO₂ uses, with values up to 3 % by weight of concrete.

Mineralization is defined as the uptake of CO₂ to form new products through the cleavage of bonds within mineral silicates, promoting and mimicking natural rock weathering. The exothermic reaction of carbon dioxide with minerals/metal oxides forms stable carbonated compounds (equation 1), in which M is an alkaline metal of the 1st or 2nd group of the periodic table. Carbonated products are extremely stable solid compounds (in the geological time dimension according to Lackner et al. (1995)), justified by the thermodynamic feasibility of their formation. Indeed, the Gibbs free energy of formation, which is a measure of the stability and spontaneity of the formation of a substance, is as low as -1,100 kJ/mol for some carbonates, contrasting with -400 kJ/mol for CO₂. Although extremely favorable from a thermodynamic perspective, the reaction has extremely slow kinetics, which has hindered large-scale deployment by requiring energy-intensive activation processes. In this regard, industrial sectors with heat surplus (i.e. waste heat) are suitable for symbiosis.



To date, the (dry) mineralization process is still not suitable for large-scale applications due to its slow reaction kinetics. Aqueous mineralization, on the other hand, has recently proved to be an appealing option to by-pass such limitations (Liu et al. (2021)). It shows considerably faster kinetics at high operating temperature and pressure, and its performance can be further optimized by using catalysts and additives. In fact, water is essential to promote the dissolution of CO₂ (reducing mass transfer limitations) and to obtain high-conversion levels to carbonates (MgCO₃ or CaCO₃) as shown by Ben Ghacham et al. (2015) and Fagerlund et al. (2012), with promising industrial applications reaching a Technology Readiness Level (TRL) between 3 and 9 (Zhang et al. (2020)).

At ambient conditions, the carbonated products (either Calcium- or Magnesium-based) can then be used in different applications such as construction aggregates, road materials, and feedstock supply for iron, steel, cement and glass production (Chiang and Pan (2017)). When using alkaline wastes, it also contributes to decreasing the environmental burden associated with metal leaching. A frequently overlooked feature is the exothermic nature of the process, which allows for heat and system integration in existing industrial clusters thus potentially decreasing heat consumption and associated costs.

Feedstocks

Pan et al. (2020) reviewed the potential of using (industrial) alkaline solid wastes (rich in Ca and Mg) for CO₂ storage via mineralisation. Based on their calculations, 4 Gt of CO₂ can be avoided per year, corresponding to 12.5% of anthropogenic CO₂ emissions. The large majority (3.7 Gt) come from environmental credit substitution (use phase) for the carbonated products, based on an LCA substitution framework.

Nonetheless, alkaline wastes are not enough to neither directly nor indirectly (via substitution policies) eliminate industrial emissions, and the complementary use of mineral ores (such as serpentine, olivine and wollastonite) is necessary. Several studies point that the amount of natural mineral ores available is enough to mineralize $5 \cdot 10^7$ Gt of CO₂, a value far superior to all anthropogenic emissions. Recently, Ostovari et al. (2022) designed a climate-optimal solution for mineralization deployment in Europe, showing that exploring serpentine, olivine and steel slag in Europe is only able to harvest 24% of the 538 Mt of industrial (cement, steel, paper and chemical industries) emissions of the old continent. Therefore, two options are available to complement their work: (i) explore other mineral sources, such as wollastonite, larnite, and alkaline wastes; (ii) import mineral ores to Europe.

Indeed, bearing the consequences of the full supply chain is important when assessing the emission reduction potential of mineralization systems. This is particularly relevant when transporting minerals for long distances (such as in inter-continental trips) or considering harsh pre-treatments. This dimension was studied for Europe in Ostovari et al. (2022). The supply chain dimension reduces by 27% the potential CO₂ offset. Out of these, 72% was allocated to the energy dimension (heat and electricity supply for thermal treatment, grinding, and other pre-treatments), 13% for solid feedstock transport and 2% for CO₂ transport. It seems clear that the two latter pale in comparison to the energy dimension, which is tackled in this manuscript.

Contribution

This work expands the contribution of Ostovari et al. (2020), by not only including different alkaline wastes as described in Pan et al. (2020) but also by integrating and quantifying synergies obtained when integrating mineralization in different industrial clusters. We show how heat integration in different sectors affects economic and environmental indicators of CCUS, contributing to its deployment.

Our work also updates the view on the seminal IPCC special report on mineralization (Mazzotti et al. (2005)), in which mineralization based on natural ores, such as serpentine and olivine, has inherently low rates of carbonation, leading to incredibly large reactors and therefore expensive procedures. Although natural ores still need to undergo energetically demanding activation steps, industrial sector coupling is strategic in the effort to minimize environmental impact and economic burden, further facilitated by industrial clustering and large amounts of waste heat.

At present neither the potential of mineralization for effective CO₂ mitigation is properly understood, nor the symbiosis prompted by integrating mineralization with different industrial sectors. In this work, we develop a platform for competitive mineralization integration with economic, environmental and thermodynamic assessment. Similarly to the approach adopted by Ostovari et al. (2022) we do not consider CO₂ point sources originating from the energy sector, but only hard-to-eliminate industrial CO₂: the intrinsic industrial processes emissions. In addition, we consider that minerals are transported to the industrial site which often contains both waste heat and alkali residues.

Methodology

Our approach couples simulation and optimization in a joint effort to understand and quantify the integration of mineralization in different sectors, leveraging system integration features. A dynamic simulation model, developed in Aspen Custom Modeler (ACM) is embedded in Aspen Plus, which is further employed in an optimization platform that accounts for economic and environmental performance as well as heat integration features; a mixed-integer linear programming (MILP) formulation is used, capable of capturing the discrete and continuous nature of the optimization problem. Similar approaches were developed to handle biowaste (Castro-Amoedo et al. (2021)), or pulp and paper production (Granacher et al. (2022)). The minimization of total cost was chosen as main objective.

In an industrial environment, an *'ex situ'* mineral carbonation takes place, in which alkaline wastes together with mineral ores (serpentine, olivine, wollastonite) are used as feedstock. Two configurations are studied here: (i) Direct carbonation (DC), in which the feedstock enters in the solid phase and the reaction takes place in a one-step CSTR (Figure 1); (ii) Indirect carbonation (IC), which consists of leaching metals (Ca and Mg) from the solid matrix prior to the carbonation reaction (blue section in Figure 1).

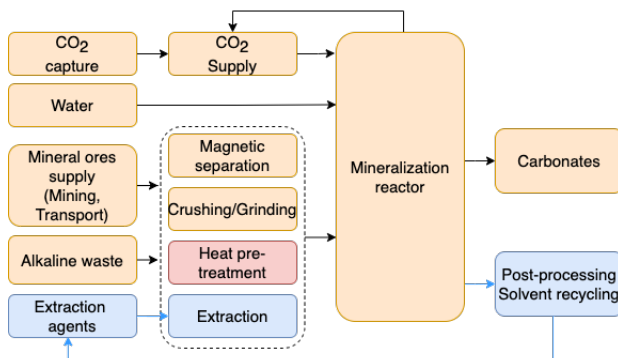


Figure 1: Direct and indirect (blue) carbonation system boundaries. Mineral ores refer to serpentine, wollastonite and olivine. Alkali waste treatment depends on the feedstock source with magnetic separation and heat pre-treatment being optional.

The reaction kinetics (i.e. which reactions take place and how fast) depend on numerous parameters including: particle

size, reaction temperature and pressure, liquid-to-solid ratio, chemical additives and reactor design. Our dynamic simulation includes three fundamental steps: (i) CO₂ dissolution in the aqueous phase (forming carbonic acid (H₂CO₃) and bicarbonate ions (HCO₃⁻)); (ii) dissolution of metal species from the solid matrix (in both direct and indirect carbonation); (iii) ionic reactions, nucleation and carbonate precipitation. The system is modeled as a set of algebraic and differential equations (DAE) (Pan et al. (2012)), considering one vapour, one liquid and two solid phases. Mass and energy balances are given by eq. 2 and 3, respectively; $Flow^{in}$ and $Flow^{out}$ are the inlet and outlet molar flows, respectively; x_i is the molar fraction of specie i , $\nu_{j,i}$ is the stoichiometric coefficient of specie i in reaction j , and R_j is the rate of reaction j ; U is the total energy, h_{in} and h_{out} are the specific enthalpy of the inlet and outlet streams, respectively; Q is the heat transfer with the surroundings. The remaining set of equations and constraints, in particular those governing phase distribution, volume, phase and reaction equilibrium and interactions with the external environment, follow the methodology described in Kakhu and Pantelides (2003), including the reduction to an index-1 DAE system.

$$\frac{dM_i}{dt} = Flow^{in} \cdot x_i - \sum_{k=1}^{n_{exits}} Flow_{k,i}^{out} + \sum_{j=1}^{Nr} \nu_{j,i} \cdot R_j, \quad \forall i \quad (2)$$

$$\frac{dU}{dt} = Flow^{in} \cdot h_{in} - \sum_{k=1}^{n_{exits}} Flow_k^{out} \cdot h_{out,k} + Q \quad (3)$$

Thermodynamic considerations and operating conditions that ensure kinetic feasibility are vital. Among those, temperature and chemical additives were particularly significant. Although high temperature values are known speed up the reaction, it should be kept below 200°C, to ensure a spontaneous reaction and subsequent carbonate precipitation. Model parameters for both direct and indirect aqueous pathways were based on Criado et al. (2014); Kato et al. (1996); Fagerlund et al. (2012).

Each industrial sector is assessed based on 1 ton of (reference) product. For industrial sectors we have used waste incineration and cement production. In both cases, the diluted CO₂ in the flue gases is initially separated and concentrated in an energy- and cost-effective way. Mature post-combustion carbon capture mainly relies on wet scrubbers using monoethanolamine (MEA), given its high reactivity toward CO₂ and its low cost. The process was modeled according to Tock and Maréchal (2012). The feedstock composition is provided in Table 1.

Environmental impacts were retrieved from Ecoinvent database v3.7. Investment costs came from from Turton (2018), annualized using 20 years of expected lifetime and 6% of interest rate; utilities and electricity values were obtained from industrial partners.

To compare and contrast solutions, both normalized total cost and environmental impact (using the widespread IPCC 2013 GWP100a indicator) were used. A CO₂ tax was computed according to eq. 4, using the sector without carbon capture and mineralization as reference. This tax quantifies by

how much should emissions be priced to promote a cleaner industrial sector.

$$\text{Carbon tax} = \frac{\Delta\text{Cost}}{\Delta\text{Emissions}} \left[\frac{\text{EUR}}{\text{tCO}_2\text{eq}} \right] \quad (4)$$

Table 1: Calcium (CaO) and Magnesium (MgO) mass fraction in different alkali waste streams and mineral ores. * average samples value.

Sector/Ore	CaO	MgO	Ref.
Waste Incineration	0.19	0.026	90% BA and 10% FA Pan et al. (2012)
Cement	0.42	0.018	Cement Kiln Dust Pan et al. (2012)
Serpentine	—	0.393	Tebbiche et al. (2021)
Olivine	0.011	0.40	Kremer et al. (2019) *
Wollastonite	0.435	—	Yadav and Mehra (2019)

Results

Venturing to study carbonation reactions requires a prior understanding of thermodynamics and kinetics. As previously discussed, despite a favorable thermodynamic framework, kinetic considerations render *ex-situ* mineralization a difficult-to-deploy technology. When trying to speed up kinetics, the first approach is to increase temperature, which is known to increase the average particle kinetic energy. However, as shown in Figure 2, Gibbs free energy is positive, for certain species, above 400 °C - hence reverting the spontaneity of the reaction. This constrains the process design and is of primary concern when controlling the reactor.

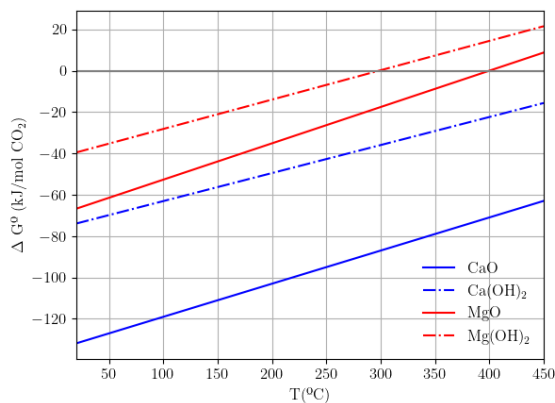


Figure 2: Gibbs free energy variation (ΔG) as a function of Temperature for different metal compounds.

Industrial sectors

Among all industrial sectors, cement and waste incineration are here explored in detail. Both sectors are expected to be part of the future energy system design. Therefore, solutions pertaining to their emissions reduction are particularly relevant. Results are broken between economic and environmental assessment and show the combination of alkaline waste with a set of mineral ores (olivine, wollastonite and serpentine) either using direct or indirect carbonation.

3.1.1 Cement sector

The production of cement without further flue gas treatment, entails emissions close to 0.9 ton CO_2/ton cement (Figure 3). Capture and storage via mineralization (identified by a star) is a net-negative technology, that is able to bring cement production to a NET. In line with Pan et al. (2020), the environmental credit of cement aggregates replacement is crucial in off-setting the system, with the pre-treatment entailing a substantial increase in emissions. The 'Carbonation' label corresponds to the effective amount of CO_2 that is captured in CO_3 form. The direct carbonation of olivine shows the largest potential (-2.5 ton CO_2/ton cement) among solutions, illustrating the double effect of both carbonation and credit substitution. The cost structure (Figure 4) shows the economic impact of mineralisation, with DC options being in general more expensive than IC ones. The burden associated with the heat exchanger network (HEN) is non-negligible as well as that of feedstock supply and energy requirements. In general, a way to offset this additional cost is to sell the carbonated products. The required selling value (green diamonds), ranges between 30 and 70 \$/ton carbonates. If emissions are to be penalized, a CO_2 tax between 60 \$/ton CO_2 (DC Wollastonite) and 90 \$/ton CO_2 (DC Olivine) would be able to incentive a cleaner cement production.

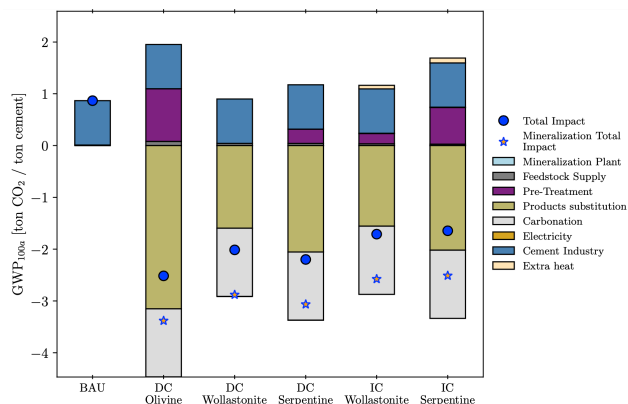


Figure 3: Cement Sector environmental impact assessed using IPCC 2013 GWP100a indicator.

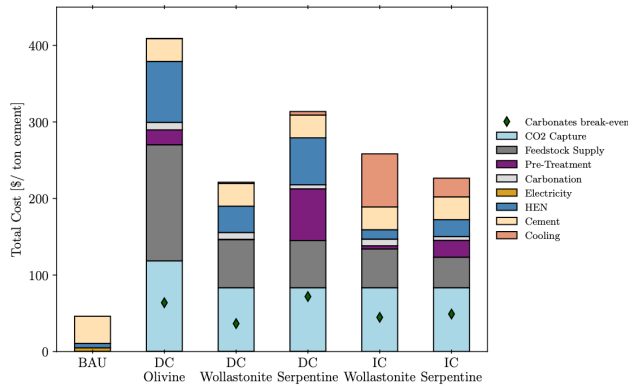


Figure 4: Cement Sector cost partition; Products break-even is the difference between each design and BAU.

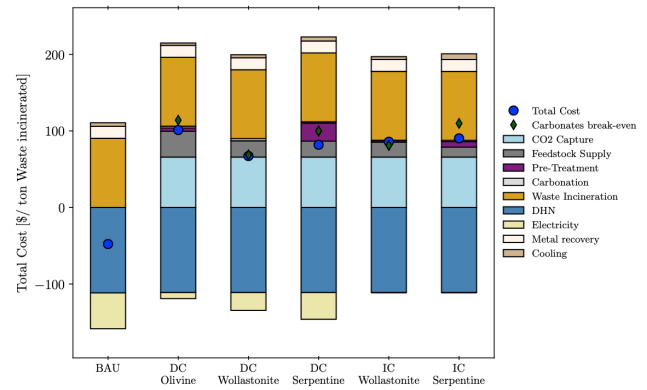


Figure 6: Waste sector cost partition; Products break-even is the difference between each design and BAU.

3.1.2 Waste incineration sector

The waste sector follows a similar trend when coupled with mineralization, being able to integrate the environmental advantages of the latter. The direct carbonation of serpentine is the pathway showing the highest potential to reduce emissions and effectively promote a net-negative sector. Moreover, DC routes seem to be more effective in emission reduction, mainly because IC routes require more steps and chemical additives, burdening the total environmental bill.

Regarding costs (Figure 6), the in-place co-generation of heat and electricity makes this industrial sector profitable. The introduction of mineralization shifts this reality, with values ranging from 65 to 100 \$/ ton of waste. There are several ways to close this economic gap: (i) charging waste producers, with a tax ranging from 110 to 145 \$/ ton of waste; (ii) sell carbonated products with a price (green diamonds) ranging from 80 to 115 \$/ ton of carbonates; (iii) introduce a CO₂ tax, forcing the waste industry to adopt mineralization, ranging from 120 \$/ton CO₂ (DC Wollastonite) to 200 \$/ton CO₂ (IC Wollastonite).

Conclusions

The urgency of climate change and the adoption of sustainable practices across industrial sectors, requires the deployment of technologies that can promote net-negative CO₂ emissions. In this work we study the effect of industrial symbiosis of mineralization in two sectors: cement production and waste incineration. We couple a dynamic simulation model within an optimization framework using a MILP formulation. Our results show the pertinence of integrating mineralization in industrial clusters with an environmental net-benefit, despite a considerable increase in costs due to CO₂ capture and storage. In the best possible scenario, coupling waste incineration with direct carbonation using serpentine results in negative emissions of 0.9 ton CO₂/ton waste, contrasting with the current 0.12 ton CO₂/ton waste. Similarly, the cement sector is able to pass from 0.9 ton CO₂/ton cement to negative emissions in the order of 2.5 ton CO₂/ton cement, when deploying the use of olivine. Overall, taxing emissions will lead to faster deployment of mineralization in the cement sector, with values between 60 and 90 \$/ton CO₂ being enough to shift operation. This compares with values up to 200 \$/ton CO₂ in the waste incineration sector.

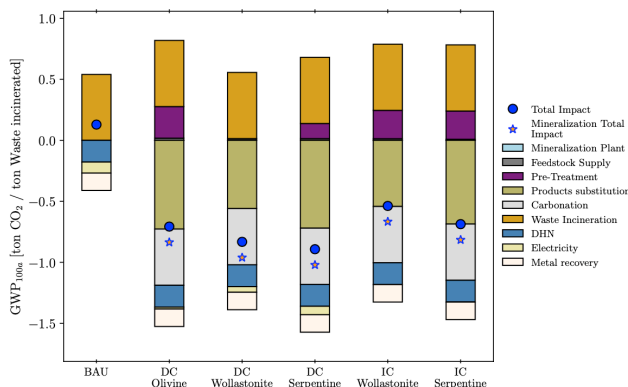


Figure 5: Waste sector environmental impact assessed using IPCC 2013 GWP100a indicator.

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