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Transformation of abundant magnesium silicate minerals for enhanced CO₂ sequestration

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Global climate change related to anthropogenic CO₂ emissions is one of the most significant challenges for the future of human life on Earth. There are many potential options for reducing or even eliminating atmospheric CO₂ emissions including underground sequestration, carbon mineralization and ocean storage. One of the most promising materials for carbon mineralization is Mg(OH)₂ which is highly reactive and capable of forming stable carbonates. Here we show a novel low-carbon method of producing Mg(OH)₂ from globally abundant olivine-rich silicate rocks. A combination of acid digestion and electrolysis of olivine were used to produce Mg(OH)₂ in a fully recoverable system. The use of Mg(OH)₂ from olivine provides a viable pathway for significant industrial scale reductions in global anthropogenic greenhouse gas emissions.

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The effects of unchecked CO₂ emissions on global climate change are being increasingly seen and felt across the world. Examples of issues include increased land and sea temperatures, glacier and sea ice loss, and sea-level rise¹. Urgent action is needed to limit future emissions and sequester existing atmospheric CO₂ in order to circumvent issues related to global warming². In 2018, ~37 billion tonnes of CO₂ was released into the atmosphere³. Point source emissions such as power generation and industrial production account for ~60% of the total CO₂, but this is expected to decrease to 50% by 2050⁴.

Methods for long-term storage of CO₂ can be classified as either underground sequestration or carbon mineralization, of which underground sequestration in sedimentary formation is considered the most mature technology⁵. Carbon mineralization can be further divided into three approaches: (1) ex situ, where the material is transported to the site and reacted with CO₂ typically at elevated temperatures and pressures, (2) surficial, using dilute or concentrated CO₂, and (3) in situ, where the CO₂ is transported to site with suitable geological formations, typically containing serpentine or olivine-bearing basalts⁵. Ocean storage has also been proposed as a potential means of CO₂ sequestration but comes with a number of environmental consequences⁴.

A number of materials have been proposed for carbon mineralization, including serpentine, olivine, wollastonite, magnesium oxide, and magnesium hydroxide^{6–10}. Of the various materials which may be suited for transport to the emissions source, Mg(OH)₂ is one of the most reactive for carbonization⁵. Aqueous carbonation of Mg(OH)₂ can result in the formation of a hydrated Mg-carbonate such as nesquehonite (MgCO₃·3H₂O), dypingite (Mg₅(CO₃)₄(OH)₂·5H₂O), and hydromagnesite (Mg₅(CO₃)₄(OH)₂·4H₂O) under atmospheric or near-atmospheric conditions¹¹. The formation of magnesite (MgCO₃) is also possible but this typically requires higher temperatures (>100 °C) and pressures above 100 bar. Seeding Mg(OH)₂ slurries with magnesite at elevated pressures and temperatures has been shown to produce a stable anhydrous magnesite¹². One of the benefits of Mg(OH)₂ with regard to overall conversion rates and transportation is that it does not contain significant quantities of unreactive material, such as silica or iron which are typically found in olivine or serpentine.

MgO and Mg(OH)₂ are usually sourced from either the calcination of magnesite or precipitated from Mg-rich brines and seawater¹³. Recovery of Mg(OH)₂ from seawater has the potential to provide virtually limitless supplies of the material; however, it's commercial production typically requires the use of lime, which itself has significant embodied CO₂, resulting in little overall environmental benefit. The high-temperature pyrohydrolysis process of using MgCl₂·6H₂O to form MgO and HCl¹⁴ could potentially provide a low-carbon Mg(OH)₂ alternative; however, concentrations of Mg-chloride in seawater are relatively low which limits the efficiency of the process unless highly concentrated brines are used.

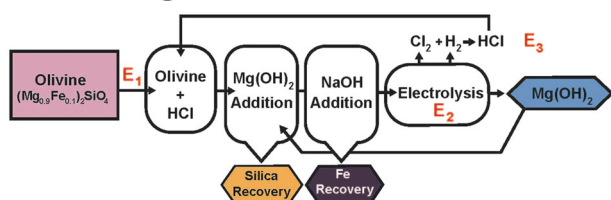
Magnesium-rich silicate minerals, such as olivine ((Mg, Fe)₂SiO₄), in ultramafic and mafic rocks have the potential to produce reactive Mg(OH)₂ with zero CO₂ emissions. Until now, Mg(OH)₂ recovery from olivine has not been used primarily because of the slow rate of hydration (i.e., serpentinization)^{15,16} or ultramafic mineral extraction efforts have been focused on silica recovery¹⁷ rather than Mg(OH)₂ production. Furthermore, a satisfactory energy-efficient industrial process has not been developed yet¹⁸.

Here, we extract Mg(OH)₂ directly from olivine using a combination of acid digestion and electrolysis. The acid used in the digestion process is completely recoverable and provides a source of usable energy. Recovered Mg(OH)₂ was found to have a similar level of reactivity compared to commercially available Mg(OH)₂. A slurry of recovered Mg(OH)₂ and water was shown to effectively sequester CO₂ forming a hydrated Mg-carbonate. In addition to Mg(OH)₂, amorphous silica, which has the potential for use as a partial cement replacement, was also recovered from the olivine.

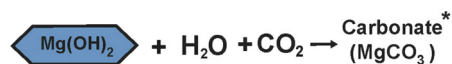
Methods: synthesis of Mg(OH)₂. The synthesis of Mg(OH)₂ uses olivine from ultramafic rocks and/or olivine-rich basalts as the raw material and source of magnesium (Fig. 1). Olivine is sparingly soluble under standard state conditions with a total Mg concentration of less than 6 mg L⁻¹ resulting in excessive energy demands for the recovery process. As the pH of the reaction solution decreases, the solubility of Mg increases considerably. In our investigation, 100 g of finely ground forsterite ((Mg_{0.9}Fe_{0.1})₂SiO₄) olivine¹⁹, with a mean particle size of 28 μm, was combined with 500 ml of 2 M HCl, resulting in the formation of a solution containing MgCl₂ and FeCl₂ and SiO₂. The silica is produced through a process of hydrolysis and subsequent polymerization and condensation of silicic acid (Si(OH)₄). Strong acids such as HCl are known to accelerate the hydrolysis process, whereas, a high pH favors the polymerization and condensation process that is supported by the addition of Mg(OH)₂ in this study. Here, the pH of the solution increased rapidly from less than 0 to ~3 after 4 h of mixing at 20 °C.

The concentration of Mg²⁺ in solution was determined by complexometric titration used to measure the total hardness of water. In total, 1 ml of sample solution was added in a conical flask containing 10 ml of 0.05 M ethylenediaminetetraacetic acid (EDTA) solution and 10 ml of ammonia buffer solution. Eriochrome black T was used as an indicator. The solution was titrated using a 0.025 M magnesium chloride solution until its color changed from blue to pink. The concentration of complexed EDTA solution was used to determine the magnesium ion concentration. Since the concentration of calcium ions in the sample solution was negligible, it was assumed that all the ions complexed with EDTA were magnesium. The Mg concentration was found to be ~24 g L⁻¹, which is more than 19 times the concentration of Mg in seawater and over three orders of magnitude greater than the concentration resulting from

Processing



CO₂ Sequestration



For sequestering 1 tonne of CO₂, 1.3 tonnes of Mg(OH)₂ is required.

Fig. 1 Simplified mineral extraction and carbon sequestration. Energy requirements are shown where E₁ = 0.32, E₂ = 6.6 and E₃ = -0.64 GJ tonne⁻¹ of Mg(OH)₂ produced. Energy consumption details are provided in Supplementary Methods. Note: Direct solid-gas carbon mineralization is also possible and the most likely approach for point source CO₂ sequestration. *Note: The carbonation product is likely to be hydrated Mg-carbonate such as nesquehonite, dypingite, or hydromagnesite, but MgCO₃ formation is also possible depending on reaction conditions²⁷.

serpentinization reactions involving pure water. The potential Mg extraction efficiency, therefore, is considerably higher using an acid digestion solution than it is from seawater.

Once the initial digestion phase was completed the solution was allowed to settle for 1 h after which it was decanted to separate the solution containing the Mg, Fe, and Si ions from any unreacted olivine (Fig. 1). Silica was precipitated from the solution by increasing the pH to >3.5 through the addition of 1 g of $\text{Mg}(\text{OH})_2$. The pH of the remaining solution was further increased to almost 7 to precipitate the dissolved iron using 0.32 g of NaOH. The silica and iron were separated from the solution using a centrifuge in this proof-of-principal testing, whereas, an industrial filtration system would be used in practice. It should be noted that the formation of gel-like products during the precipitation of silica and iron can cause challenges to any filtration system. Regular flushing and cleaning of the membranes would be necessary and likely require a number of crossflow filters, such as those used for silica removal from geothermal waters, to be run in parallel to maintain operations. An alternative approach and potentially more efficient method for the removal and recovery of the silica and iron is by precipitate flotation.

The MgCl_2 solution was electrolyzed in an H-cell with a carbon anode and platinum cathode. A DC power supply was used to generate a current of approximately 100 mA resulting in the formation of Cl_2 gas at the anode and H_2 gas at the cathode. $\text{Mg}(\text{OH})_2$ was formed at the cathode where the pH of the solution increased rapidly to ~9.5. The recovered $\text{Mg}(\text{OH})_2$ was placed in a drying oven at 105 °C for 1 day after which it was analyzed by scanning electron microscope (SEM), thermogravimetric analysis (TGA) and X-ray powder diffraction (XRD). For the commercial production of $\text{Mg}(\text{OH})_2$, H_2 and Cl_2 would be combined to produce HCl for re-use and as an energy resource, as is typically done in the chemical process and manufacturing industry.

Results and discussion

From this process, a total of 35 g of $\text{Mg}(\text{OH})_2$ was produced from 100 g of olivine, with 1 g of $\text{Mg}(\text{OH})_2$ added in the silica precipitation stage. Approximately 5 g of iron oxide was precipitated with the addition of 0.32 g of NaOH, which represented <1% of the total $\text{Mg}(\text{OH})_2$ recovered. In total, 35 g of amorphous silica was also recovered from the precipitation stage. The SEM image and TGA graph provided in Fig. 2a, b show the material recovered after electrolysis was primarily $\text{Mg}(\text{OH})_2$. XRD results provided in Fig. 2c show the recovered silica was predominantly amorphous SiO_2 with some residual unreacted olivine and confirm the material recovered after electrolysis was predominately $\text{Mg}(\text{OH})_2$. The composition of the raw olivine sand, recovered $\text{Mg}(\text{OH})_2$, and recovered silica from olivine, determined by X-ray fluorescence (XRF), are provided in Table 1 and are consistent with the other material characterization analysis.

In addition to the primary recovered $\text{Mg}(\text{OH})_2$, secondary materials of iron hydroxide and silica may also be of use. The iron hydroxide for instance could be used as a high purity raw material for iron production or as an absorbent while the amorphous silica can be used as a partial replacement for Portland cement, in the production of concrete, which is a significant industrial contributor to global CO_2 emissions²⁰. Our approach provides an almost completely closed system for the production of $\text{Mg}(\text{OH})_2$ with the only additions being olivine, as the source of Mg, and minor amounts of NaOH.

CO_2 and energy implications of Mg-hydroxide extraction. Overall, conversion of olivine into $\text{Mg}(\text{OH})_2$ produces no direct CO_2 emissions and the HCl used for digestion was completely

recoverable. The large-scale and industrial use of strong acids such as HCl have serious consequences if released into the environment. However, this is only an issue if there is a loss of containment from the process. The large-scale handling of HCl without loss of containment is well established in the chemical process industries due to its use in commodity materials such as the manufacture of PVC and pickling of steel. What is most important is that neither HCl nor chlorine leaves the process described in our work, as they are recycled within the process.

The total energy required, including quarrying and grinding, to produce $\text{Mg}(\text{OH})_2$ from olivine was calculated to be 6.28 GJ tonne^{-1} (see Fig. 1 and Supplementary Methods for energy determinations and calculations). Further refinement of the process may allow greater use of recovered $\text{Mg}(\text{OH})_2$ for pH control, a decrease in the total energy required, and a reduction in NaOH. The $\text{Mg}(\text{OH})_2$ could be further processed into MgO but for CO_2 sequestration; however, $\text{Mg}(\text{OH})_2$ is known to be a faster reactant than MgO¹⁰.

For every tonne of CO_2 sequestered as a Mg-carbonate, including nesquehonite and hydromagnesite, 1.3 tonnes of $\text{Mg}(\text{OH})_2$ is required, resulting in an energy consumption 8.17 GJ tonne^{-1} of CO_2 (Fig. 1, see Supplementary Methods for calculations). Using the method outlined in this paper would result in a net negative CO_2 emission, even if coal was used to provide the necessary energy, though at ~25% sequestration efficiency. The overall carbon sequestration benefits would improve substantially if a low-carbon energy sources, or even a mixed electrical supply, was used. For example, in California where roughly 50% of the electricity is from non-fossil fuels, ~500 kg of CO_2 would be emitted for every 1000 kg of CO_2 captured and turned into Mg-carbonate.

To confirm the ability of the recovered $\text{Mg}(\text{OH})_2$ to sequester CO_2 , a $\text{Mg}(\text{OH})_2$ water slurry was pressurized to 4 bar with concentrated CO_2 . Over a 48-h period, >50% of the $\text{Mg}(\text{OH})_2$ was converted to a hydrated Mg-carbonate, demonstrating the potential conversion of CO_2 into a solid. The reactivity of the recovered $\text{Mg}(\text{OH})_2$ was also confirmed by the rapid increase in pH to ~10.5 when it was added to de-ionized water. As previously noted, there are a number of other methods for sequestering CO_2 with $\text{Mg}(\text{OH})_2$ using either aqueous^{12,21} or direct solid-gas reaction methods¹⁰. Our proposed process has the potential to provide a substantial source of energy-efficient, low-carbon $\text{Mg}(\text{OH})_2$ for use in various carbon sequestration techniques currently being developed by other investigators.

Olivine resources and feasibility. Ultramafic rocks, enriched in olivine, constitute ~1% of Earth's terrestrial landscape, a high proportion of oceanic crust, and >50 % of the upper mantle²². Olivine-rich deposits (Fig. 3) are primarily present within populated areas of the Circum-Pacific and Mediterranean regions^{23,24}. Olivine is present in many rocks such as basalt and other ultramafics. We would like to note that although olivine was assessed and used in this study, its metamorphic equivalent, serpentine ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$) with its high reactivity in HCl and worldwide deposits, could expand the scope and implications of this study. Our preliminary investigations have shown Mg can also be extracted from basalt; however, we will primarily focus our discussion on two enriched and accessible olivine deposits: the Semail ophiolite (Oman) and the Red Hills Ultramafic Complex (New Zealand) which conservatively contain 1.4×10^5 and 871 billion tonnes of olivine, respectively (see Supplementary Methods for sources and estimate calculations).

Using $\text{Mg}(\text{OH})_2$ to remove and sequester anthropogenic CO_2 estimated for 2020 (40 billion tonnes) in a Mg-carbonate would require ~105 billion tonnes of olivine. Reducing global

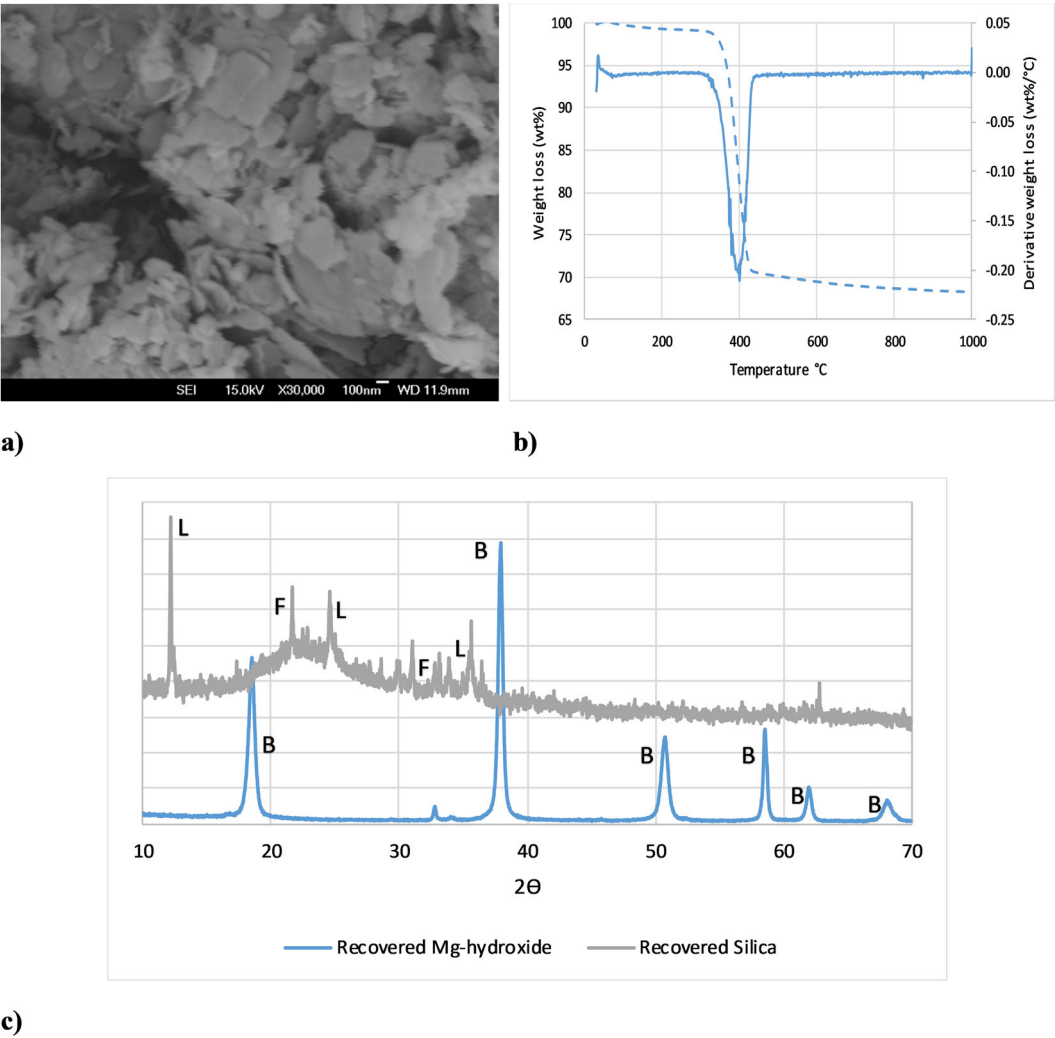


Fig. 2 Characterization of recovered material. **a** SEM image of recovered $\text{Mg}(\text{OH})_2$ from the electrolysis of olivine digestion solution, **b** TGA of recovered $\text{Mg}(\text{OH})_2$ from olivine, **c** XRD of recovered $\text{Mg}(\text{OH})_2$ and of recovered silica (B: brucite, L: lizardite, F: forsterite).

Table 1 Elemental compositions of raw olivine sand and recovered $\text{Mg}(\text{OH})_2$ and silica from olivine, determined by XRF analysis.								
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	Total
	Wt. %							
Raw olivine	39.6	0.38	10.7	0.73	45.0	0.14	0	100.0
$\text{Mg}(\text{OH})_2$ -olivine	0.1	0.11	6.4	0.30	60.4	0.03	<0.01	99.9
Silica-olivine	63.2	0.23	4.7	0.57	13.9	0.03	0.06	99.0

atmospheric CO_2 levels by an additional 10 billion tonnes would necessitate a further 26 billion tonnes of olivine. The sum of olivine required to do all of this would be 131 billion tonnes; 0.1% of the Oman ophiolite or 16% of Red Hills, New Zealand (Fig. 3). To sequester all anthropogenic CO_2 , these two deposits would last on the order of nearly a decade (Red Hills) to 1000 years (Semail), assuming the deposits contained at least 60% olivine. The degree of serpentinization of each deposit and the abundance of other minerals such as ortho- and clinopyroxene would affect the overall extraction efficiency. Smaller deposits, compared to the Semail ophiolite or Red Hills, around the world could significantly aid the global reduction of anthropogenic CO_2 .

Implications. The concept of using ultramafic rocks and its metamorphic equivalents as sources of Mg for CO_2 sequestration

has been considered for at least two decades. However, one limitation with regards to previous and current approaches is related to the distance between the CO_2 gas source and the ultramafic site. Currently, concentrated CO_2 is being transported/piped to ultramafic sites to be directly injected into the subsurface. Although ultramafic sites are around the world, many are not sufficiently close to areas of high CO_2 -producing industries. Our approach allows bulk $\text{Mg}(\text{OH})_2$ to be transported more efficiently to locations/industries where it can be used on-site for point source emissions control. For example, transporting enough $\text{Mg}(\text{OH})_2$ to sequester 1 tonne of CO_2 , from its production location to a point source such as a cement plant 1000 km away by rail, would result in 37 kg of CO_2 emissions²⁵.

In addition, slight modifications to this approach through the further refinement of $\text{Mg}(\text{OH})_2$ to MgO has the potential to

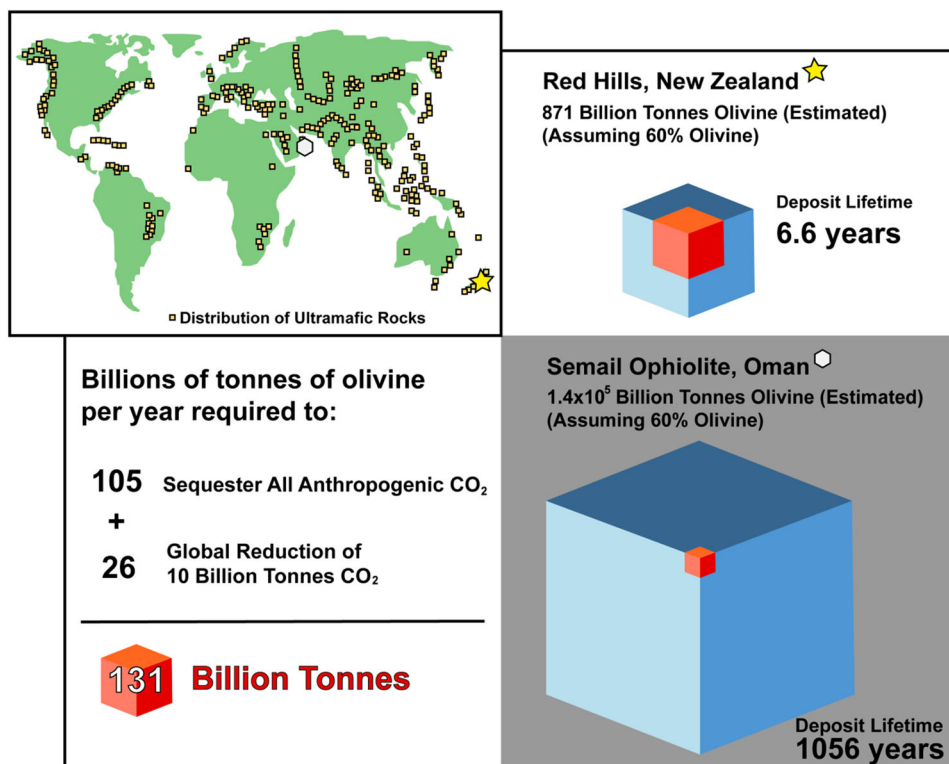


Fig. 3 Distribution of ultramafic rocks and olivine lifetime estimates. The general distribution of ultramafic rocks (including peridotites and serpentinites) is shown worldwide. Ultramafic rock distributions are based on location data from Oze et al.²⁴ and compared to Real and Vishal²³. Please note that more ultramafic rock deposits are present than shown and that the squares do not represent particular sites. The billions of tonnes of olivine per year needed to sequester all anthropogenic CO₂, and reduce global atmospheric CO₂ as well as the lifetime supply of olivine from the Red Hills (New Zealand) and Semail Ophiolite (Oman) is shown and based on calculations provided in Supplementary Methods.

produce construction materials such as Mg-masonry blocks²⁶ rather than simply burying the CO₂. There is considerable interest in the potential use of MgO as an alternative to traditional Portland cement¹³; however, there are a number of issues that must be addressed, including the low pH of the pore solution, which makes its use in steel-reinforced structures challenging, and high water demand which makes handling more difficult. One of the major environmental issues associated Mg-based cements is the vast majority of MgO is produced from MgCO₃ which results in the release of CO₂ at a similar proportion to that of Portland cement. The alternative of recovering MgO from seawater, as previously noted, also has considerable embodied CO₂ due to the use of CaO, sourced from carbonates, in the recovery process. If MgO is to be used as a construction material then a low carbon mineral extraction approach will be needed. The scale and scope to mitigate climate change using Mg-bearing minerals from ultramafic and mafic rocks are massive, but our experiments and others support that this is feasible.

Scaling up and improving efficiencies with regards to the processes described and proven here will require significant development and industrialization efforts. We anticipate that the engineering challenges to produce Mg(OH)₂ for use in CO₂ sequestration, as shown in Fig. 1, are comparable to other well known industrial processes. One of the great challenges, however, is how to use the billions of tonnes of carbonate produced to offset anthropogenic CO₂ emissions and this requires further circumspection. A more realistic and efficient approach to addressing global warming is to reduce CO₂ emissions. Until then, we have demonstrated a process that is feasible and globally communal where many countries and industries can participate.

Overall, the recovery of magnesium hydroxide from olivine-rich (and potentially serpentine-rich) rocks provides the basis for a direct route to reduce global anthropogenic CO₂ and its associated climate change impacts.

Data availability

Data files containing XRD and TGA results of the recovered materials from this investigation are available from the figshare dataset repository: DOI 10.6084/m9.figshare.13543091.

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Author contributions

A.S.: conceptualization, methodology, resources, formal analysis, funding acquisition, and writing—original draft; V.S.: conceptualization; methodology, formal analysis, investigation, and writing—review; C.O.: conceptualization and writing—review and editing; B.S.: investigation and methodology; C.C.: writing—review and editing; N.Y.: investigation and methodology; M.W.: conceptualization and writing—review and editing; A.M.: conceptualization and writing—review and editing.

Competing interests

The authors declare no competing interests.

Additional information

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