

Olivine for CO₂ capture and safe storage

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July 20, 2025

Abstract

Several methods have been proposed in recent years to counteract climate change and ocean acidification by removing CO₂ from the atmosphere. The process of Carbon Dioxide Removal can be achieved by storing CO₂ in minerals, among whose olivine is a promising candidate. The processes that allow to store CO₂ in olivine minerals are weathering and peridotite carbonation. An area with a large amount of peridotite and where carbonation occurs rapidly is the Samail ophiolite in Oman. Naturally peridotite carbonation takes thousands of years to realize so, to make it faster and wider, mineral olivine rocks should be fragmented in nanoparticles. In an experiment, olivine nanoparticles were fabricated with a gas aggregation magnetron sputtering at room temperature and carbonation occurred in a time scale of minutes. Therefore the process of weathering can be enhanced but there is the misunderstanding that it could not be a safe process.

1 Introduction

It is called **Carbon Dioxide Removal** (CDR) the process with which CO₂ is removed from the atmosphere. The most versatile and widely applicable of these methods is **weathering of olivine** which is capable of removing billions of tons of CO₂ from the atmosphere at moderate cost compared to the high cost of Carbon Dioxide Capture and Storage (CCS). Despite its disadvantage, CCS is still the favored solution of many governments [1].

2 Carbon Dioxide Capture and Storage

CO₂ utilization is receiving increasing interest from the scientific community [2]. This is partly due to climate change considerations and partly because using CO₂ as a feedstock can result in a cheaper or cleaner production process compared with using conventional hydrocarbons [3]. CO₂ utilization is often promoted as a way to remove carbon dioxide from the atmosphere [4]. CO₂ is first captured from the atmosphere, then concentrated and finally used.

The term *CO₂ utilization* refers to the **use of CO₂ directly or as a feedstock, in industrial or chemical processes, to produce valuable carbon containing products** [5].

The **CO₂ is supplied from fossil-derived waste gases**, and then it is **captured** from the atmosphere either **by an industrial process**, or biologically **by land-based processes**. Biological or land-based forms of CO₂ utilization can generate economic value in the form of, for example, wood products for buildings or increased plant yields from enhanced soil carbon uptake [6].

CCS has the disadvantage that it is expensive to perform, although it brings to economic achievements. There are **alternative methods to capture CO₂**, provided by nature and less expensive.

A promising approach is the **storage of CO₂ in minerals**, of which mineral olivine is a good candidate [1].

3 Olivine for atmospheric CO₂ capture

The process of Carbon Dioxide Removal, **CDR can be achieved** by different techniques or devices, for example **by storing CO₂ in minerals**.



Figure 1: Olivine gemstones. Picture from [23].

Mineral olivine (fig. 1, fig. 3 and tab. 1) is a **magnesium iron silicate** with chemical formula: $(\text{Mg,Fe})_2\text{SiO}_4$. It is the most abundant mineral in Earth's upper mantle. **Olivine is a promising candidate** for the storage of CO_2 in minerals **because of its Earth abundance and its high CO_2 absorption capacity**, which is of the order of 50% wt.

4 Weathering process

Each year the **Earth emits** in the order of 0.5 Gt of CO_2 , **mainly by volcanoes**. **If all that CO_2 remained in the atmosphere**, it would require little time to **create an atmosphere dominated by CO_2** . Apparently there is an efficient **process by which it is removed and safely stored**. That process is the **weathering of basic silicates, mainly Mg- and Ca-silicates** [1].

Weathering is the process by which rocks, soil and minerals are broken down at the Earth's surface due to physical, chemical or biological agents. Once the rock falls on the Earth's surface, it can **interact with the fluid**, either the atmosphere or the sea, **that brings the CO_2** .

The magnesium and calcium bicarbonate solutions that are formed in such reaction are carried by rivers to the sea, where corals, shellfish and foraminifera convert them to limestone and dolomites (fig. 2 and tab. 1). These are the sustainable CO_2 stores of the Earth. This weathering process has kept the **CO_2 levels of the atmosphere in check** during the whole history of life.



Figure 2: Limestone. Picture from [24].

Humankind is now causing a spike in CO_2 **emissions by burning** in a few hundred years the **fossil fuels** that have formed during hundreds of millions of years. Present emission of CO_2 far exceeds the normal emission by volcanoes, and the weathering process cannot cope with this sudden rise. **By increasing the rate of weathering, it is possible to achieve a new balance** between input and output [1].

5 Mineral carbonation

Mineral carbonation is the **reaction of silicate or oxide minerals with CO_2 in fluid to form solid carbonate minerals**. Mineral carbonation has been proposed as a **method for safe and permanent CO_2 storage**. In mineral carbonation **gas CO_2 is converted to solid, stable carbonate mineral**.

The **fastest** mineral carbonation rates known are for the mineral olivine, which is also the **most abundant** mineral in Earth's upper mantle. For these two reasons, olivine is used to perform mineral carbonation.

To perform mineral carbonation is used **magnesium olivine** which has chemical formula: Mg_2SiO_4 . **Carbonation** of mineral olivine often occurs **with hydration**, a process involving the addition of water.

Carbonation of the mineral olivine occurs via the following simplified reactions. In all of the three chemical reactions [7], **one of the products is always magnesite** (fig. 4 and tab. 1).



Figure 3: Olivine. Picture from [25].



Figure 4: Magnesite. Picture from [26].

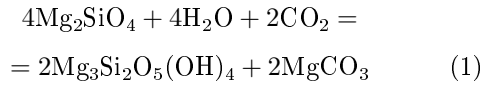
Table 1: Glossary of minerals.

Mineral	A naturally occurring solid that has a well-defined chemical composition and a crystal structure.
Olivine	The most abundant mineral in Earth's upper mantle, with chemical formula $(\text{Mg,Fe})_2\text{SiO}_4$.
Limestone	A type of carbonate sedimentary rock, composed mostly of calcite, a different crystal form of calcium carbonate CaCO_3 .
Dolomite	The most common calcium magnesium carbonate mineral, with chemical formula $\text{CaMg}(\text{CO}_3)_2$.
Magnesite	The most common magnesium carbonate mineral, with chemical formula MgCO_3 .
Serpentine	A hydrous alteration product of olivine, mainly $(\text{Mg,Fe})_3\text{Si}_2\text{O}_5(\text{OH})_4$.
Talc	A hydrous magnesium silicate, with chemical formula $(\text{Mg,Fe})_3\text{Si}_4\text{O}_{10}(\text{OH})_2$.
Quartz	A mineral composed of SiO_2 .
Tectosilicate	A mineral compound with a structure that has silicate tetrahedrons (central silicon atom surrounded by four oxygen atoms) and chemical formula XSiO_2 .
Feldspar	A group of rock-forming aluminium tectosilicate minerals, also containing cations such as Na, Ca, K, Ba.
Plagioclase	One of the feldspar group of minerals, mainly $\text{NaAlSi}_3\text{O}_8^-$ or $\text{CaAl}_2\text{Si}_2\text{O}_8$.
Peridotite	A rock composed of more than 40% olivine and less than 10% plagioclase.
Pyroxene	The second most abundant group of minerals in Earth's shallow mantle, containing mainly $(\text{Mg,Fe,Ca})_2\text{Si}_2\text{O}_6$.
Dunite	A rock composed of more than 90% of olivine.



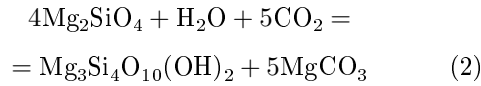
Figure 5: Serpentine. Picture from [27].

1. Reaction with **serpentine** (fig. 5 and tab. 1) as a product.



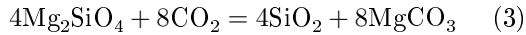
Equation (1) means: olivine + water + carbon dioxide = serpentine + magnesite.

2. Reaction with **talc** (tab. 1) as a product.



Equation (2) means: olivine + water + carbon dioxide = talc + magnesite.

3. Reaction with **quartz** (tab. 1) as a product.



Equation (3) means: olivine + carbon dioxide = quartz + magnesite.

Quartz belongs to the **tectosilicate** group (tab. 1), like other silica minerals that have chemical formulas that contain some multiple of SiO_2 [8].

6 Peridotite carbonation

Mineral olivine in nature is not found alone, but it appears mixed in rocks with some other elements.

Feldspar (tab. 1) is a group of rock-forming aluminium tectosilicate minerals, also containing other cations [9]. The most common members of the



Figure 6: Pyroxene crystals from Afghanistan. Picture from [25].

feldspar group are the **plagioclases** (tab. 1) [10]. Rocks rich in the mineral olivine are known as **peridotites** (fig. 7 and tab. 1). A common constituent of peridotite is **pyroxene** (fig. 6 and tab. 1).

In situ peridotite carbonation is a process in which CO_2 -rich **fluid, air or seawater is transported** to areas where large volumes of **peridotite** are present. It is distinguished from ex situ methods, which involve transport of solid reactants to a concentrated source of CO_2 [7].

Olivine present in a peridotite rock **interacts** with the fluid **by its surface**, then the chemical reaction of mineral carbonation takes place.

7 Peridotite carbonation in Samail Ophiolite in Oman

An area with a **large amount of peridotite** is the **Samail ophiolite in Oman**. Ophiolite is an **uplifted slice** of oceanic crust and upper mantle.

Natural carbonation of peridotite in the Samail ophiolite (fig. 8), in the Sultanate of Oman, is surprisingly **rapid**. Carbonate veins in mantle peridotite in Oman have an average ^{14}C **age of 26,000 years**, and are not 30–95 million years old as previously believed. These data and reconnaissance mapping show that **10^4 to 10^5 tons per year** of atmospheric CO_2 **are converted** to solid carbonate minerals via peridotite weathering in Oman [11].



Figure 7: Peridotite. Picture from [28].

Peridotite carbonation can be **accelerated via drilling hydraulic fractures**, at elevated pressure and at increased temperature at depth [12].

Fractures in peridotite **make larger the contact surface** between the rock and the fluid, so the carbonation chemical reaction is accelerated.

In situ carbonation of peridotite could consume more than 1 billion tons of CO_2 per year in Oman alone, affording a **low-cost, safe, and permanent** method to capture and **store atmospheric CO_2** [11].

Natural peridotite carbonation has two **limitations**. The first is that it is a **very long process**, that takes millions of years to perform at ambient conditions. The second is that we wish that this process happened all over the world. To remove CO_2 from the entire atmosphere, the process of peridotite carbonation should take place not **only in certain regions** of the Earth.

To make peridotite carbonation faster and wider, mineral **olivine rocks should be fragmented** in nanoparticles.

8 Small sized olivine particles

The rate at which olivine carbonation occurs can vary depending on environmental conditions such as temperature, pressure, and the availability of water and CO_2 [13]. In natural geological settings, olivine carbonation at atmospheric conditions is



Figure 8: Veins in Samail Ophiolite in Oman. Picture from [29].

generally a slow process and can take thousands to millions of years to complete, depending on the specific conditions. **Reaction rate can be increased** by supplying CO_2 at extreme pressures and temperatures to force carbonation [14], or by supplying catalytic agents [15].

The rate of carbonation can also be influenced by the **availability of reactive surface area** on the olivine grains. Therefore, **producing smaller sized olivine particles** could make the carbonation process faster and more efficient [16].

Several studies over the last decade, have shown promising results in capturing CO_2 exploiting mineral carbonation with olivine microparticles at high temperatures, 100°C - 200°C , using supercritical CO_2 (pressure of $\text{CO}_2 > 100$ bar), since the olivine carbonation reaches its maximum yield at 185°C and with a CO_2 partial pressure of 150 bar [11].

Although the **fabrication of nanoscale olivine** by high energy ball milling has been reported [17], it is **an energy-intensive** and time-consuming process. Alternative and cost-effective methods are therefore needed to **produce nanoscale olivine** for atmospheric CO_2 capture **at economical conditions** [18].

Since peridotite is composed by many elements (tab. 1), before producing olivine nanoparticles, peridotite must be purified.

9 Olivine nanoparticles: production and experiment

In work [18], **olivine nanoparticles were fabricated** with a gas aggregation **magnetron sputtering** nanoparticle source [19], **operating at room temperature**, which provides well-defined olivine nanoparticle layers.

Magnetron sputtering is a **thin film deposition technique** used in various industrial applications to create coatings with specific properties. It involves **bombarding a target material** with high-energy ions (typically argon ions). The collision dislodges atoms from the target, which then **deposit onto the substrate** [20].

Nanoparticles were produced in gas aggregation magnetron sputtering nanoparticle generators [19], which allows for good control over the nanoparticle properties such as size and composition. The olivine nanoparticles were then deposited with a device at room temperature from a synthetic olivine target. The magnetron power was ≈ 200 W, with argon gas used both as plasma sputter and carrier gas inside the aggregation chamber with a flow rate of 35 sccm (standard cubic centimeters per minute) [18].

The **morphology** of the air-exposed nanoparticles (fig. 9) was investigated by Atomic Force Microscopy (AFM), which were characterized by a **fractal cauliflower structure**: smaller nanoparticles, with a size of ≈ 14 nm, are aggregated to form larger cauliflower nanoparticles, with sizes of ≈ 150 nm [18].

Finally olivine nanoparticles were exposed to CO_2 and air and **carbonation process** was achieved at ambient conditions. The carbonation was measured by X-ray photoelectron spectroscopy (XPS). Nanoscale olivine particles, due to their **high surface-to-volume ratio**, reach a very **high rate of carbonation** at a time scale of **minutes** at ambient conditions [18].

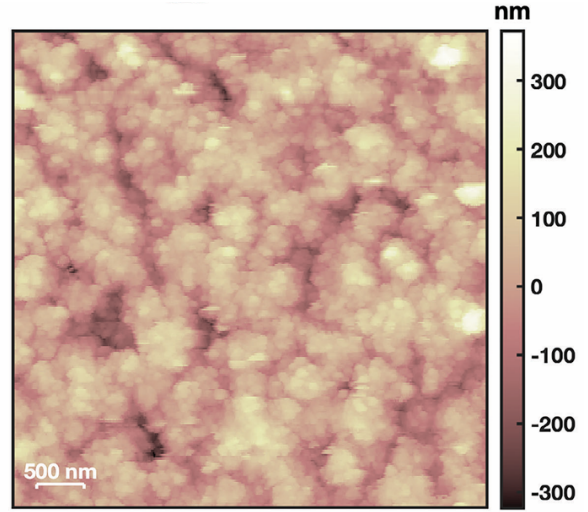


Figure 9: Cauliflower structure made by olivine nanoparticles. Picture from [18].

10 Enhanced weathering of olivine

Olivine nanoparticles were made to enhance the weathering process. By **increasing the rate of weathering**, it is possible to achieve a new balance between CO_2 produced and CO_2 removed from the atmosphere.

This means that the weathering crust on top of many dunite massifs must be removed. Dunites (tab. 1) occur in many countries on all continents. The first step will be reactivate the weathering potential of these massifs. In order to achieve it, the process of **enhanced weathering of olivine** must be implemented [1].

First it is necessary to **mine the olivine** in a number of locations, strategically distributed over the globe. After mining, the **olivine is milled**, reduced in powder and then the resulting **olivine sand is spread** over forest land, deserts, beaches and shallow, energetic seas.

This concept of enhanced weathering has brought forth a lot of misunderstanding. A factor is that CCS is wrongly called geological storage, because it just borrows the storage box from geological formations, whereas **weathering is the main natural process which takes care of geological storage of CO_2** [21].

11 Examples of misunderstanding

A common misleading statement is that olivine rocks are dangerous because they contain asbestos (fig. 10). The truth is that **only some olivine massifs contain asbestos**, but this is rare, and no mining company will run the risk of selling asbestos-containing dunites, because they would quickly lose their clients. There are plenty of asbestos-free dunite massifs around to choose from.

A second argument that is often heard is that dunites (or olivine rocks in general) are dangerous because **they contain nickel**. It is true that these rocks contain more nickel than most other rock-types, but that argument doesn't make these rocks dangerous. Olivine-rich rocks occupy more than 1 million km² on Earth. Hundreds of millions of people live in these areas already for many generations, they grow their crops, they graze their animals, and they drink its water, without any negative consequences for their health [1].

12 Conclusion and perspective

Due to climate considerations, several methods to counteract climate change have been investigated. A promising method is **Carbon Dioxide Removal, CDR, performed with minerals**. Among minerals, **Mg-olivine**, Mg₂SiO₄, is a **good candidate** to perform CDR because of its high CO₂ absorbance rate and its Earth abundance.

Mineral carbonation, after weathering of rocks, is the process with which gas **CO₂ is converted to a solid, stable mineral**. Mg-olivine, CO₂ and sometimes H₂O, are involved in the chemical reaction that transforms them in **magnesite**, MgCO₃, plus another mineral.

Naturally olivine is bound with other elements to form a rock called **peridotite**. An area with a large amount of peridotite and of its carbonation occurrence is the Samail Ophiolite in Oman.

Two limitations for peridotite carbonation are: first it is a slow process (it takes millions of years), second it is not expanded all over the world. From here comes the idea to make olivine nanoparticles.

A simple and economical method of fabricating olivine nanoparticles was achieved in experiment [18], by magnetron sputtering in a gas phase

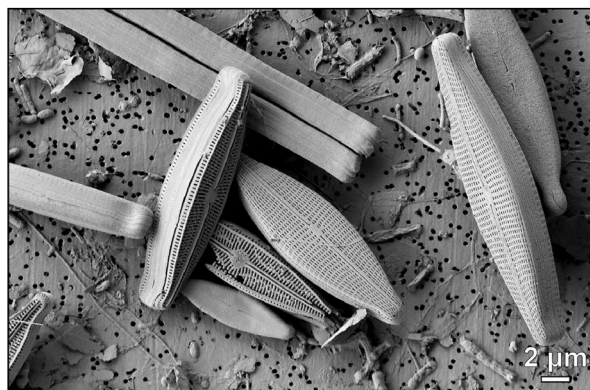


Figure 10: Diatoms (microalgae) from the mine pit lake of a former asbestos mine. Picture from [22].

aggregation reactor, yielding well-defined and uniform olivine nanoparticle assembled layers. Due to their **high surface-to-volume ratio**, carbonation occurred very **rapidly** and almost completely in a matter of minutes.

By fractioning olivine in nanoparticles, it is possible to achieve an **enhanced weathering process**. The procedure to enhance the weathering process is: first **mine olivine** in various location all over the globe, then **mill olivine** to make from it a powder made by olivine nanoparticles, finally **spread fine-grained olivine powder** over agricultural and forest land or along beaches.

There is the misunderstanding that olivine could be dangerous for human beings, for animals and for the environment, because it contains some small quantities of asbestos and nickel. This is not a problem because these quantities are too small to be dangerous. The fact that some people live with their animals, in areas near peridotite, without any health consequences, is a **proof for olivine safety**.

Lack of understanding of geological processes led to false assumptions regarding enhanced weathering as a tool against climate change and ocean acidification. Removing CO₂ from the atmosphere and storing it safely can be possible.

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