



## Review article

# Carbon capture, utilization and storage opportunities to mitigate greenhouse gases

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## ABSTRACT

Carbon capture, utilization and storage (CCUS) technologies are utmost need of the modern era. CCUS technologies adoption is compulsory to keep global warming below 1.5 °C. Mineral carbonation (MC) is considered one of the safest and most viable methods to sequester anthropogenic carbon dioxide (CO<sub>2</sub>). MC is an exothermic reaction and occur naturally in the subsurface because of fluid-rock interactions with serpentinite. In serpentine carbonation, CO<sub>2</sub> reacts with magnesium to produce carbonates. This article covers CO<sub>2</sub> mitigation technologies especially mineral carbonation, mineral carbonation by natural and industrial materials, mineral carbonation feedstock availability in Pakistan, detailed characterization of serpentine from Skardu serpentinite belt, geo sequestration, oceanic sequestration, CO<sub>2</sub> to urea and CO<sub>2</sub> to methanol and other chemicals. Advantages, disadvantages, and suitability of these technologies is discussed. These technologies are utmost necessary for Pakistan as recent climate change induced flooding devastated one third of Pakistan affecting millions of families. Hence, Pakistan must store CO<sub>2</sub> through various CCUS technologies.

## 1. Introduction

Pakistan is facing potential risk to the economy, social and environmental development due to the extreme impacts of climate change highlighted by United Nations Framework Convention on Climate Change (UNFCCC) [1,2]. Climate change badly affected Pakistan and Himalayan glaciers are melting faster [3]. Rains, floods and earthquakes occur more frequently [4]. Emissions of greenhouse gases (GHGs) in the atmosphere are constantly increasing with ever-growing energy needs and population. Climate change and global warming are main concerns in 21st century. GHGs especially CO<sub>2</sub> is released into the atmosphere due to world energy dependence on fossil fuels. Many industries are responsible for GHGs emissions such as cement industry emits 5 % GHGs [5,6]. In the coming century, it's not possible to absorb these GHGs and CO<sub>2</sub> concluded by global carbon cycle, so adaptation technologies to

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capture CO<sub>2</sub> are required urgently [7–10].

Paris agreement was a legal binding which is signed in 2015 by 166 countries including Pakistan.

The aim of Paris Agreement was to reduce the GHGs emission to address the issue of global warming as the temperature of earth is higher than the pre-industrial period. Paris agreement does not focus only on the reduction of GHGs emissions but also focus on the management of forests, better use of land and the availability of financial and technical resources for underdeveloped countries to combat the negative impact of climate change especially global warming. It signaled the lower dependence on fossil fuels for membered countries [11]. Global warming must be kept below 1.5 °C as per IPCC recommendations [12].

The intergovernmental panel on climate change (IPCC) directs CO<sub>2</sub> storage and utilization for the mitigation of greenhouse gas concentrations [13]. Carbon dioxide capture and storage refer to the necessary technologies in the immediate future, aiming to store the CO<sub>2</sub> produced from the combustion of fossils fuels in steel and iron making, cement manufacture [14] and power generation and then store it underground into stable geological sites for a very long time [7,15,16]. There are many approaches to sequester CO<sub>2</sub> such as mineral carbonation, geological sequestration, oceanic sequestration, biological sequestration [17–19]. Pakistan has a greater potential and availability of natural minerals, which can be utilized to capture CO<sub>2</sub> through mineral carbonation. Similarly, vast deposits of serpentine exist in the northern region of Pakistan [20]. This study is based on a survey in Pakistan highlighting feedstock minerals availability in Pakistan, detailed characterization of serpentine, its potential utilization for greenhouse gases mitigation and different ways of CO<sub>2</sub> sequestration. Process of carbon mineralization involve the dissolution of magnesium silicates such as serpentine and olivine with successive precipitation of carbonates and silica [21].

## 2. Background and CO<sub>2</sub> mitigation techniques implementation requirement

Prolonged monsoon rains due to climate change affected one third of Pakistan population (Fig. 1). Fig. 1 shows damaged houses per district in different areas throughout Pakistan [22]. Millions of people were affected, and estimated loss is more than 30 billion US dollars (According to Asian Development Bank). Similar climate change induced flooding is also observed in India and China.

Climate change can be mitigated through CO<sub>2</sub> utilization and storage. Ten pathways [23] for CO<sub>2</sub> utilization and storage are given in Fig. 2. Various techniques can be used for carbon capture, utilization and storage [24]. These CO<sub>2</sub> mitigation techniques are chemical production, oceanic storage, geological storage, mineralization and many more (Fig. 2).

Mission Innovation report (2018) has introduced the concept of enhanced metal recovery (EMR) and mineral carbonation is used for recovery of valuable metals along with CO<sub>2</sub> sequestration [25]. Three potential EMR routes exist, in-situ, ex situ EMR as a heap leaching process and ex situ EMR in a dedicated processing plant [26]. Wang et al. recovered 90 % of nickel and cobalt through EMR and convert magnesium into magnesite in an integrated process which used natural olivine [27]. Nickel and cobalt are used in electric vehicles, battery storage, hydrogen production and fuel cells [28]. Through conventional froth flotation process, 30 % of the nickel may waste as mine tailings which is due to nickel locked inside silicate grains. Mineral carbonation especially pH swing process may

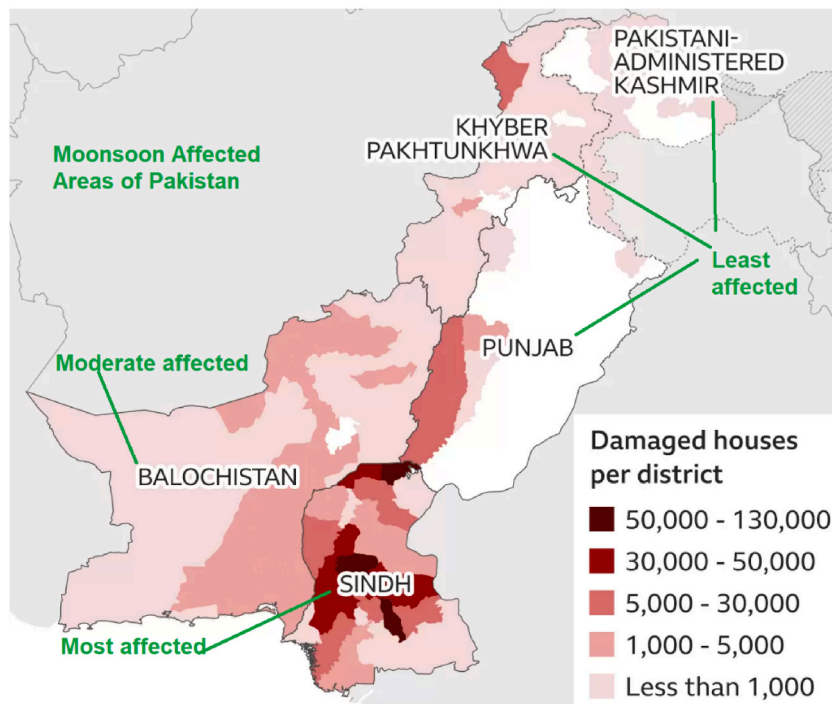


Fig. 1. Climate change affected one third of pakistan population [22].

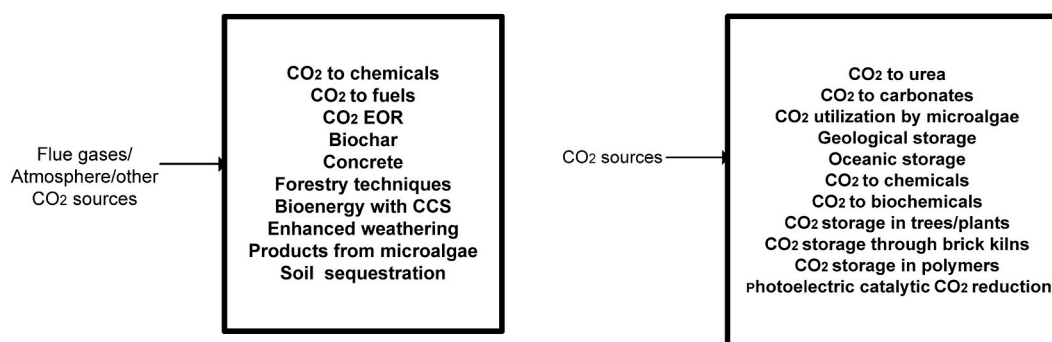


Fig. 2. Ten pathways for carbon capture (left), CO<sub>2</sub> mitigation techniques (right).

use these mine tailings and recover this nickel along with CO<sub>2</sub> sequestration [28]. Wang et al. reacted CO<sub>2</sub> with saprolite and limonite laterites and olivine and carbonate Mg<sup>+2</sup> present in them while nickel and cobalt were leached in aqueous solution using nitrilotriacetic sodium which were recovered through sulphide precipitation [29].

### 2.1. Mineral carbonation

CO<sub>2</sub> is separated from flue gases using different membranes. Aqueous mineral carbonation can be used to transform this separated CO<sub>2</sub> into carbonates with different rocks (dunite) and minerals (Olivine, Antigorite, and Lizardite) using CO<sub>2</sub> separation membranes and its fixation may provide significant contributions in CO<sub>2</sub> mitigation [30]. Mineral carbonation is a mature technology and permanent CO<sub>2</sub> storage. Single stage carbonation [31–36], two stage carbonation [37–39] and acid dissolution [40–42] are all currently being investigated. The by-products and minerals feed can also be used in cement production [40]. Direct carbonation is better than indirect carbonation. Mineral carbonation is a leading technology as it is a permanent storage and can be employed at any place especially in vicinity of CO<sub>2</sub> emissions.

### 2.2. CO<sub>2</sub> to urea

It is best method to store CO<sub>2</sub> permanently on large scale. In this method CO<sub>2</sub> reacts with ammonia to form carbamate which decompose into urea that is used by plants [43,44]. Urea is used as a fertilizer for crops and CO<sub>2</sub> fixed through this process is at large scale and cannot return. This process is normally employed in fertilizer sector where CO<sub>2</sub> is a byproduct of ammonia production process. CO<sub>2</sub> from other CO<sub>2</sub> emitting industries such as cement industry and steel manufacturing may be coupled with fertilizer industries to convert it into urea fertilizer as fertilizer production is dependent on CO<sub>2</sub> availability.

### 2.3. Utilization of CO<sub>2</sub> in microalgae

CO<sub>2</sub> is eaten by microalgae. Numerous microalgae growth characteristics have been observed to increase with rising CO<sub>2</sub> concentrations [45]. The biomass created by microalgae can once more be used in the combustion process. Microalgae perform better than terrestrial plants [46]. A bioprocess can transform the hemicellulose of plants into compounds. Using microalgae impregnation, biodiesel can be made from ethanol [47]. Microalgae plantation can be increased artificially to enhance CO<sub>2</sub> storage. This can be coupled with biodiesel production from microalgae.

### 2.4. Enhanced oil recovery

Although it is not a permanent storage, CO<sub>2</sub> is employed in enhanced oil recovery. Although it is momentarily confined, there is a potential of leakage. Excess CO<sub>2</sub> from gas treatment facilities may be used in fertilizer industries to manufacture urea. Pakistan has significant potential for enhanced oil recovery as it has gas and oil wells for these fuels' exploration. Major issue in this technology is to ship CO<sub>2</sub> from emitting facilities to the enhanced oil recovery sites. Although this is a temporary CO<sub>2</sub> fixation but still it contributes in immediate CO<sub>2</sub> mitigation.

### 2.5. Geological CO<sub>2</sub> storage

There is no doubt that CO<sub>2</sub> is kept in geological storage [48–50], but leaks are possible. However, its significance for the immediate reduction of CO<sub>2</sub> emissions cannot be disputed. It is necessary to determine risks and potential uses for escaping CO<sub>2</sub>. Thousands of people were killed by CO<sub>2</sub> that released after an African lake exploded [51].

2.6. CO<sub>2</sub> to chemicals

It is the method of permanent storage of CO<sub>2</sub> and it can be converted into ethylene carbonate, urea and salicylic acid [52]. CO<sub>2</sub> is fixed permanently in the form of chemicals.

2.7. CO<sub>2</sub> to biochemicals

Different biochemicals like carbon monoxide, methane, glucose, methanol, and synthetic fuel can be produced. Enzymes are necessary for CO<sub>2</sub> fixation, and it is a slow process. Further research on discovering new enzymes that accelerate CO<sub>2</sub> fixation is required.

2.8. Storage of CO<sub>2</sub> in trees and plants

Plants use CO<sub>2</sub> during the process of photosynthesis. It is quick and permanent process of storing CO<sub>2</sub>. There is a dire need to increase the number of trees and plants worldwide to capture the excessive amount of CO<sub>2</sub>. Millions of trees has been planted in Pakistan through Billion Tree Tsunami program.

2.9. CO<sub>2</sub> capture through brick kilns

Major emissions from brick kilns are CO<sub>2</sub> and particles which affect environment [53]. Scrubbers using amines can capture these emissions. Such techniques must be adopted worldwide to mitigate greenhouse gas emissions. Coupling amines systems with brick kilns can significantly reduce CO<sub>2</sub> emissions and particulate matter. Such measures will offset CO<sub>2</sub> emissions.

2.10. Storage of CO<sub>2</sub> in polymers

Polymerization reaction consume CO<sub>2</sub>. Different polymers can store CO<sub>2</sub> by using adsorption and absorption process [54,55]. Binding CO<sub>2</sub> in polymers will enhance CO<sub>2</sub> fixation.

2.11. Oceanic CO<sub>2</sub> storage

It is quite risky because it may cause damage to marine life. There are large number of ocean available worldwide for this process. Pakistan also has oceans which can be used for CO<sub>2</sub> sequestration. Oceanic CO<sub>2</sub> storage is temporary but can be utilized to offset current CO<sub>2</sub> emissions and delay global warming process.

2.12. CO<sub>2</sub> reduction using photoelectric catalysts

This is an emerged technology which has a potential to sequester huge mass of anthropogenic CO<sub>2</sub> emissions [56,57]. Further

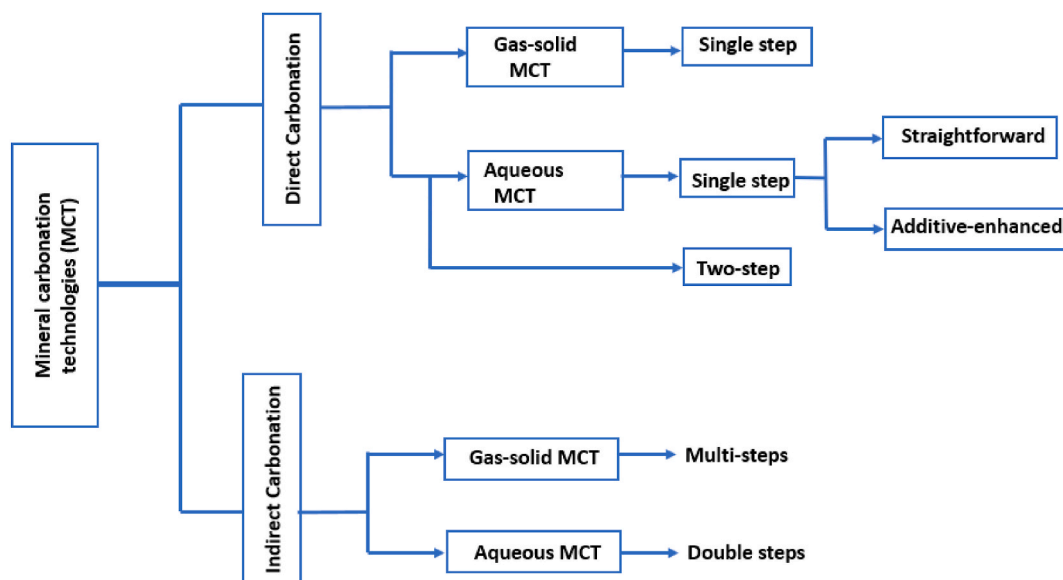


Fig. 3. Mineral carbonation processes for CO<sub>2</sub> sequestration [64].

research is required to develop efficient photoelectric catalysts.

### 3. Mineral carbonation by natural and industrial materials

Mineral carbonation (proposed in 1990s) is an alternate of silicates weathering [58]. CO<sub>2</sub> mineral storage or mineral carbonation permanently fix anthropogenic CO<sub>2</sub> without having chances of leakage [59]. Ex-situ mineral carbonation and in-situ mineral carbonation are widely used. Mineral carbonation converts CO<sub>2</sub> into magnesite (MgCO<sub>3</sub>), calcite (CaCO<sub>3</sub>) or dolomite [60]. Magnesium and calcium silicates are used worldwide for this process. Similarly, Pakistan has considerable resources of magnesium and calcium silicate rocks, which can be utilized for potential CO<sub>2</sub> storage and capture [20,61]. Magnesium silicates are peridotite (olivine, forsterite) and serpentinite (lizardite, antigorite, chrysotile). These materials may bind all emitted CO<sub>2</sub> from fossil fuels [62,63]. Mineral carbonation processes are shown in Fig. 3.

### 4. Mineral carbonation feedstock availability in Pakistan

Potential feedstock minerals such as serpentine (lizardite and antigorite) and olivine are widely found in ophiolite belt [63]. The northern region of Pakistan (Fig. 4) have large amounts of these materials. Serpentine is deposited at highest position of the Skardu. Pakistan is quarried from a mountain range around 4000 m above sea level which is 38 km from Shiger (Fig. 5) and it posses Serpentine stone. Serpentine belt is shown in Fig. 5 (a) and serpentine mineral is shown in Fig. 5 (b). Moreover, very high quality olivine exists in Pleo-Island Arc (North Pakistan) [65]. Mineral carbonation technology can be implemented in these areas of Pakistan resulting in CO<sub>2</sub> mitigation. Other option is to transport these mineral and rocks where CO<sub>2</sub> emissions exist and store them via reaction with these minerals and rocks.

X-ray diffraction analysis (Fig. 6) show that serpentinites consist of lizardite, clinochlore, and talc. The most abundant minerals are polymorphs of serpentine (lizardite,  $d = 7.5938 \text{ \AA}$ ; clinochlore  $d = 4.767 \text{ \AA}$ ; talc =  $3.11 \text{ \AA}$ ).

FTIR techniques (Fig. 7) is used to investigate the adsorption of water from atmospheric air, the hydroxyl groups (-OH) in the spectral region of  $3675 \text{ cm}^{-1}$ . Si-O symmetrical stretching vibration silicate range at  $871\text{-}1050 \text{ cm}^{-1}$  [66].

SEM and EDS analysis of serpentine powder is shown in Fig. 8. SEM disclose typical serpentine structure. Layered structure of serpentine is visible in SEM images [Fig. 8 (a,b)]. Fig. 8 (c) shows area selected for EDS analysis. EDS analysis (Fig. 8 (d)) indicate Mg, Si, O, Ca, Al and W elements are present. These elements are typical for serpentine.

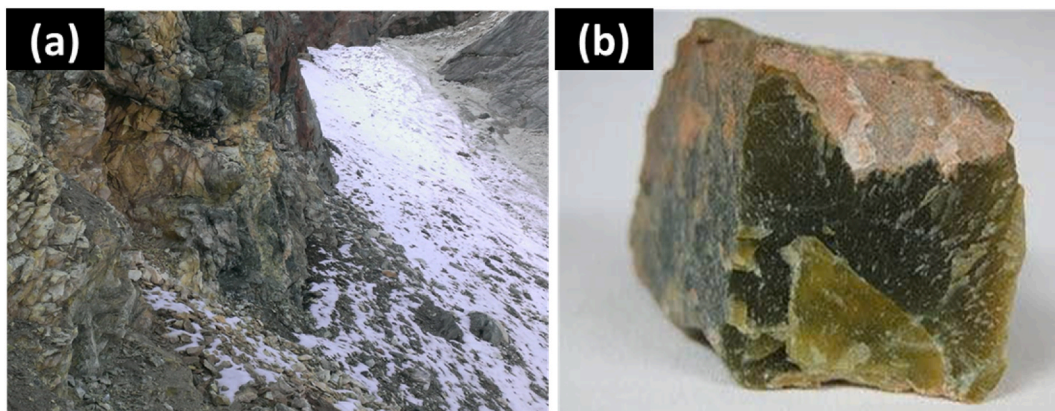
### 5. CCUS opportunities in Pakistan and their suitability

#### 5.1. Geo sequestration and availability of depleted gas and oil reservoirs

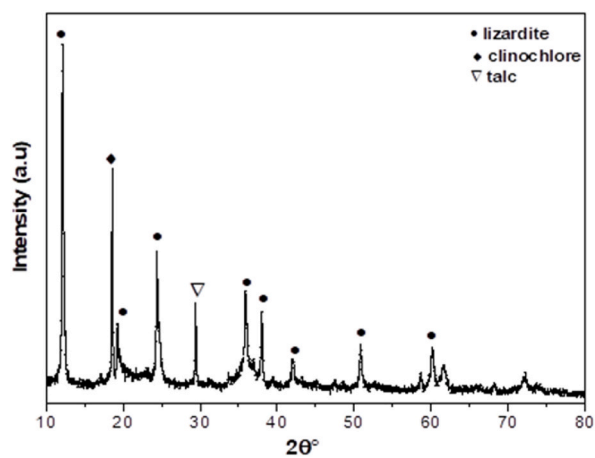
Conventional storage sites for geologic storage include CO<sub>2</sub> storage in underground geological cavities such as depleted gas, oil



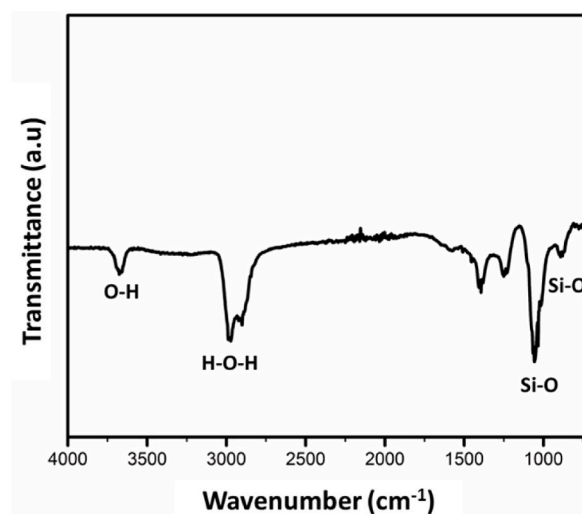
Fig. 4. Geological map of serpentine and olivine formation in Pakistan.



**Fig. 5.** Serpentine belt is a metamorphic rock in Shiger Skardu Pakistan composed of serpentine group minerals (lizardite, talc, and clinochlore) (a) serpentine belt (b) serpentine mineral.

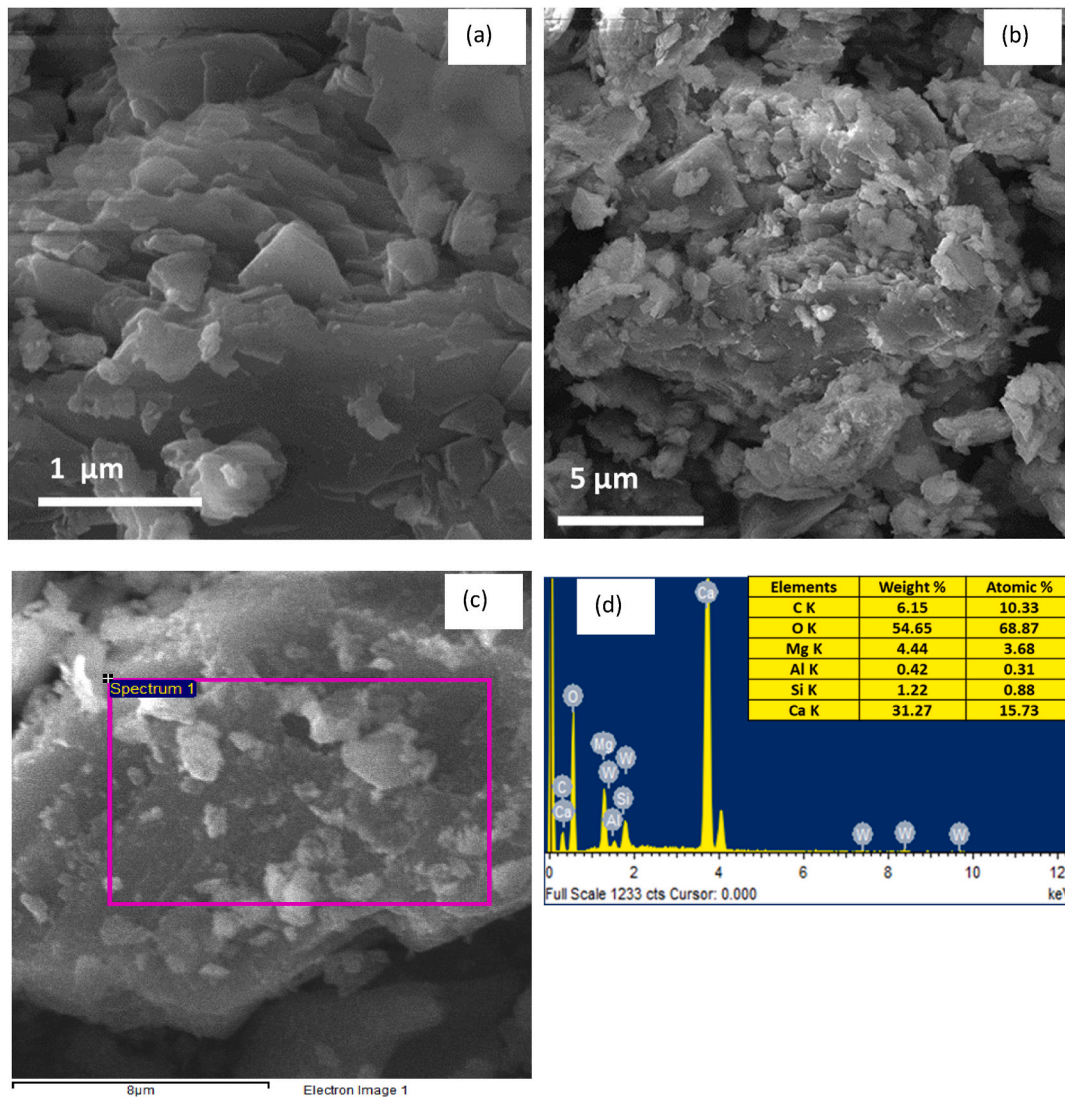


**Fig. 6.** XRD pattern of Serpentine rock from Shiger Skardu Pakistan.



**Fig. 7.** FTIR spectrum of Serpentine rock from Shiger Skardu Pakistan.





**Fig. 8.** SEM at different magnification and EDS analysis of powdered serpentine from Shiger Skardu Pakistan (a) serpentine particles (b) zoom in view of serpentine particles (c) area on serpentine particle for EDS analysis (d) EDS analysis of serpentine particle.

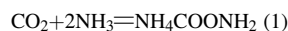
reservoirs, saline aquifers, salt caverns and coal seams. Initially, the  $\text{CO}_2$  is transported to a storage site after separation from a flue gas offshore emitting source which is finally injected into an underground reservoir at high pressure. However, the potential concern with geological storage is the hazard of leakage which require constant monitoring over long term [67,68]. Pakistan has various depleted reservoirs having potential for  $\text{CO}_2$  storage. This technology is a suitable option for Sindh and Balochistan where plenty of depleted gas and oil reservoirs exist. Main limitation of the technology is releasing potential of stored  $\text{CO}_2$ .

## 5.2. Oceanic sequestration

Our planet contributes the largest carbon sinks in the oceans [13]. As ocean storage can be used for injecting  $\text{CO}_2$  into the ocean at a broad depth of around 1500 m [68], where carbonic acid is formed [69]. However, the challenge with this method of storage is decreasing pH and lack of permanency which can cause environmental endangerment having adverse impacts on marine life. The storage below the seabed is another form of  $\text{CO}_2$  sequestration, where  $\text{CO}_2$  is pumped at deeper depth into the sea [70]. At such depths, it eliminates monitoring requirement because  $\text{CO}_2$  become denser than water offering greater permanency of storage [68]. Pakistan has oceans which can be used for  $\text{CO}_2$  storage. This technology is suitable for Sindh especially Karachi city where oceans exist in close vicinity. Main limitations of the technology are availability of oceans and release potential of stored  $\text{CO}_2$ .

### 5.3. CO<sub>2</sub> for urea production

CO<sub>2</sub> can be converted into urea via reaction with ammonia typically in a fertilizer industry. These reactions are shown in equation 1 and equation 2. This reaction occurs at high pressure (3200 psig) and temperature (383 F) with excess ammonia (4.5/1). Excess ammonia is recovered through ammonia recovery condensers and recycled back to reactor.



(Ammonium carbamate) → (Urea) + (Water).

Overall reaction



Various fertilizer industries exist in Pakistan which transforms CO<sub>2</sub> into urea and have capacity typically 1750 metric tonne per year (MTD) to 2000 MTD. List of fertilizer industries in Pakistan is given in Table 1. This technology is suitable for Punjab as oceanic sequestration and geo sequestration does not suit for Punjab. Main benefit of the technology is conversion of huge amount of CO<sub>2</sub> into urea. Main limitation of urea manufacturing technology is availability of natural gas.

### 5.4. CO<sub>2</sub> for methanol and other chemicals production

Methanol is one of the most promising chemicals as well as clean burning fuel. Moreover, it is also useful for obtaining acetic acid, dimethyl ether, methyl tertiary butyl ether, and methylamine [71]. Industrially, large amount of methanol can be produced from the catalytic conversion of synthesis gas such as CO<sub>2</sub>, CO, and H<sub>2</sub> [72]. Other chemicals which can be produced from CO<sub>2</sub> are ethylene carbonate, salicylic acid, formaldehyde, formic acid, di-methyl carbonate, copolymers, cyclic carbonates, polymer building blocks and fine chemicals [52]. Pakistan can use flue gases from recently installed coal power plants or other industries and produce methanol through process shown in Fig. 9. Methanol production can also be used to fulfill domestic methanol requirements. This technology is suitable for Punjab because most of the CO<sub>2</sub> emitting industries such as cement industries and steel industries are in Punjab. These CO<sub>2</sub> emissions can be converted into methanol.

### 5.5. Direct air capture technology

Direct air capture (DAC) is another emerging technology for CO<sub>2</sub> capture. CO<sub>2</sub> concentration in air has reached 419 ppm due to anthropogenic CO<sub>2</sub> emissions into atmosphere [34]. DAC process enables CO<sub>2</sub> capture from atmosphere thus helping in circular economy. According to IEA in a sustainable development scenario, 10 Gt CO<sub>2</sub> emissions must be captured by 2070. From 10 Gt emissions, it is expected that 2 Gt emissions will be captured through DAC technology. Ninety percent of these emissions must be stored underground and 10 % be converted into CO<sub>2</sub> based fuels. In DAC process, absorbents capture CO<sub>2</sub> from air flow using these absorbents. Chemical sorbents are hydroxides of sodium, potassium and calcium and CaO. These hydroxides and oxides bind CO<sub>2</sub> from air. These absorbents can be regenerated through heating. Currently 27 CCUS facilities are capturing 40 Mt CO<sub>2</sub> emissions [73]. DAC is a mature technology which Pakistan can adopt to capture CO<sub>2</sub> emissions and offset climate change affects. This technology is suitable for all provinces of Pakistan and can be installed at any place.

## 6. Conclusions and recommendations

CO<sub>2</sub> mitigation technologies especially mineral carbonation can be employed in Pakistan to offset greenhouse gases in Pakistan and reduce global warming which can reduce devastating flooding caused by monsoon. Based on characterization analysis, calcium oxide and iron oxide have a higher sequestration capacity. The presence of lizardite, talc, and clinocllore were confirmed by XRD analysis in serpentine from Shiger serpentine belt. Therefore, this study has shown that MC can be used in Pakistan for CO<sub>2</sub> sequestration. To line

**Table 1**

Urea manufacturing fertilizer industries in Pakistan and their capacity.

Urea fertilizer industry	Capacity (Metric tonne per day)	Location
FatimaFert Sheikhpura	1750	Sheikhpura
FFC Sadiqabad plant 1	2105	Sadiqabad
FFC Sadiqabad plant 2	1925	Sadiqabad
FFC Mirpur Mathelo plant 3	1740	Mirpur Mathelo
Engro Dharki plant 1	3500	Dharki
Engro Dharki plant 2	2740	Dharki
FFBL Karachi	1670	Karachi
Fatima fertilizer Sadiqabad	3500	Sadiqabad
PakAmerican Mianwali	1050	Mianwali
PakArab fertilizer Multan	300	Multan



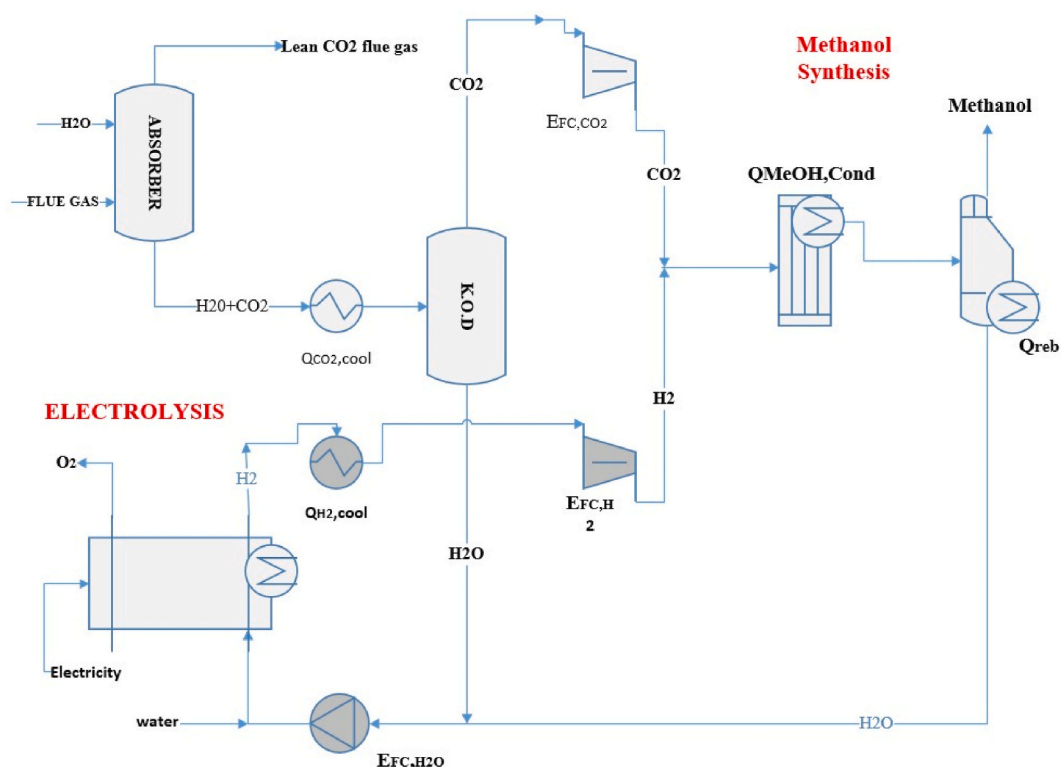


Fig. 9. Methanol production from flue gases.

with the concept of green technology, Pakistan has an indigenous serpentine reserve of Shiger serpentine belt which can be used for environmental sustainability in climate mitigation strategies. Furthermore, CO<sub>2</sub> can be converted into urea, methanol and other chemicals. Depleted oil and gas reserves available in Pakistan can be utilized for enhanced oil recovery. Oceanic CO<sub>2</sub> storage and direct air capture are also viable option which can be used in Pakistan. Coupling amines systems with brick kilns, microalgae plantation, trees plantation and increasing urea and methanol production facilities can also offset CO<sub>2</sub> emissions thus reducing greenhouse gases and monsoon caused flooding.

#### Data availability statement

Data will be provided on demand.

#### CRedit authorship contribution statement

**Muhammad Imran Rashid:** Writing – original draft, Formal analysis, Data curation, Conceptualization. **Zahida Yaqoob:** Writing – review & editing, Data curation. **M.A. Mujtaba:** Writing – review & editing, Software, Resources. **M.A. Kalam:** Writing – review & editing. **H. Fayaz:** Writing – review & editing, Funding acquisition. **Atika Qazi:** Writing – review & editing, Funding acquisition.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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