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CO_2 mineral sequestration studies in the ultramafic rocks of northern Pakistan

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Abstract

One of the greatest threats to environment caused by anthropogenically induced climate change is the production of CO_2 as a result of industrialization. How to get rid of, or at least reduce it in the atmosphere to save the environment, is one of the biggest challenges of the day. Under controlled experimental conditions with optimized reaction kinetics, mineral carbonation has considerable potential for the safe disposal of CO_2 in the form of environmentally benign carbonates. Pakistan has many major ultramafic complexes with varying thickness and lithologies containing Ca and/or Mg rich silicate minerals, such as pyroxene, olivine, amphibole, serpentine, etc. Our present work is about the mineral carbonation potential of ultramafic rocks of Pakistan, including Chilas and Jijal complexes, Alpuri serpentinites and Dargai ultramafics. The calc-silicates exist in abundance in all of these ultramafic bodies and may act as possible sinks to sequester CO_2 in the form of magnesium and calcium carbonates. In this study, approximate amounts of CO_2 that can be sequestered in all the ultramafic rocks of Chilas, Jijal, Alpuri and Dargai are estimated using equation formulated by Zevenhoven and Kohlmann, (2001). The approximate covered area of the Chilas complex is 7318 km^2, Jijal complex 551 km^2, Alpuri serpentinite 16 km^2 and that of Dargai ultramafic belt is 153 km^2. Taking into consideration the depths, densities and MgO contents, the estimate shows that an ultramafic proportion of 1 million ton (mt.) in the Chilas, Jijal, Dargai, and Alpuri ultramafic can store ~ 01268, 93.79, 47.82 and 1.79 mt of CO_2, respectively.

Keywords: CO_2 Sequestration; Chilas; Jijal; Dargai; Alpuri; Ultramafics

1. Introduction

The carbon dioxide (CO_2) level has been increasing in the atmosphere by different human activities, including burning of fossil fuels and other anthropogenic activities. The atmospheric CO_2 concentration has been increased (US Department of Commerce report, 2006). Disturbance in the global carbon cycle over the past century has influenced the global climate very badly; leading to warmer temperatures, increased ice melts, especially in the polar regions and rise in sea level (Energy Information and Administration USA report, 2010). It is now evident that the rise in CO_2 concentration within the atmosphere is one of the main causes for the apparent rise in the average global temperature. In order to address this problem, a research was conducted under the sponsorship of US Department of energy in order to find an industrially and environmentally worthwhile and improved carbonation method to allow minerals sequestration to be implemented economically (Philip et al., 2000). The team made meaningful success in looking for faster reaction techniques by using magnesium silicates, supercritical CO_2, water, and additives for pretreatment methods to increase mineral reactivity, and in analyzing the structural changes to identify reaction paths and probable barriers. This geochemical trapping system for storing the CO_2 underground is based on the weathering/alternation processes occurring in nature, wherein CO_2 reacts with Ca, Mg and/or Fe-bearing silicate-rich rocks such as ultramafics and mafics to form the respective carbonates.

Pakistan, being a developing country is suffering from rise in fossil-fuel CO_2 emission levels. For example, Sheikh (2010) reported in his work that in the fiscal year 2002-2003, the consumption of coal in cement industry and brick kilns in Pakistan was 19.6% and 53.3%, respectively and the percentages in the fiscal year 2007-08 was changed to 56.6% and 37.2%, respectively.

Pakistan needs to address this problem on urgent basis in order to save its agriculture on which relies its 70 % population. Partial solution to this problem can be obtained by disposing the excess CO_2 by capturing it from the point sources, separating it from flue gases and storing into potential reservoirs other than the atmosphere, which is referred to as carbon sequestration. The potential reservoirs include terrestrial biosphere, oceans and geological formations for storing the CO_2 underground. There are many geological formations, like deep saline aquifers, depleted oil and gas fields, unmineable coal seams, oil-bearing shales, mafic/ultramafic rocks and especially the continental flood basalts that offer better and promising receptors for the long-term sequestration of CO_2 compared to other reservoirs. This study deals with the geological formations within Pakistan that can act as reservoirs for carbon sequestration (Fig. 1).

2. Carbon dioxide storage in geological formations

There are three different ways for carbon dioxide sequestration in geologic formations, i.e. hydrodynamic trapping, solubility, and mineral trapping. (Mani et al., 2008).

2.1. Hydrodynamic trapping

It involves the storage of free CO_2 as gas or as supercritical CO_2 in the pore spaces of the sedimentary layers overlying oil and gas reservoirs, un-mineable coal seams, and deep saline reservoirs. These structures serve as a natural storage of for hydrocarbons, brine, and CO_2. In the developed countries, power plants and other large emitters of CO_2 are commonly installed near these geological strata with potential for CO_2 storage (USDE, 2010). In many cases, the recovery of hydrocarbons is increased by penetrating CO_2 into the geologic formation, thus providing value-added byproducts that make the capture and sequestration, of CO_2 cost effective too (USDE, 2010). This technique ensures that sequestration does not impair the geologic integrity of an underground formation and that CO_2 storage is secure and environmentally acceptable.

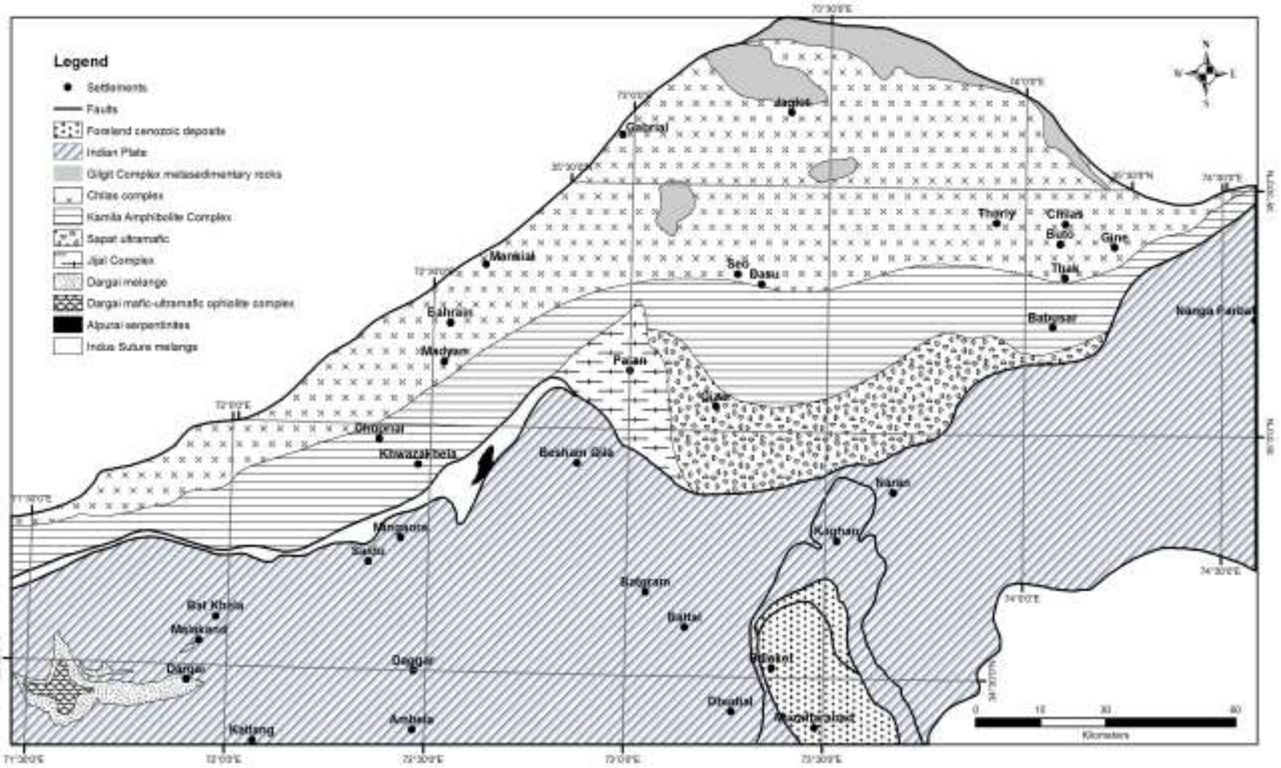

Fig. 1. Map showing location and extension of ultramafic bodies and the tectonic zones of northern Pakistan (modified after Searle et al, 1996; Anczkiewicz et al., 2001; DiPietro et al., 2008).

2.2. Solubility

In this type of trapping, CO_2 is dissolved in fluid phase comprising of aqueous brines and oils. The solution has density greater than brine in order to prevent buoyant escape of CO_2 (Weir et al., 1995). The solubility of CO_2 varies as a function of pressure, temperature, total salinity and brine composition.

2.3. Mineral carbonation trapping

It is a permanent mechanism of sequestration in which silicate minerals are converted to secondary carbonates due to reaction with CO_2 (Mani et al., 2008). Detailed background of this technique is given below.

3. Mineral CO_2 sequestration: Brief review

The basic principal of mineral CO_2 sequestration is the acceleration of weathering/alteration processes occurring in nature, wherein CO_2 reacts with Ca, Fe and or Mg containing minerals, especially silicates. For industrial applications, the process is largely to be completed in hours compared to the natural weathering reactions, which take considerable time. Thus optimization of reaction kinetics is of prime importance in mineral. The permanent sequestration of CO_2 in the form of carbonates is shown by this reaction.

$(Ca, Mg)O + CO_2 \rightarrow (Ca, Mg)CO_3$

$(Ca, Mg)SiO_4 + CO_2 \rightarrow (Ca, Mg)CO_3 + SiO_2$.

Nature stores CO_2 predominantly in carbonates, mainly limestone, dolomite. Listwanite (carbonated serpentinite) represents a fossil mineral carbonation system, serving as a repository of CO_2 in the form of carbonates during the reaction of serpentine with CO_2-rich fluids (Kump et al., 2000; Kojima et al., 1997). CO_2 is made to react with mafic/ultramafic rocks which are the most common source of magnesium, iron or calcium-bearing silicate minerals present in nature (Goff and Lackner, 1998). Examples of such a carbonation reaction with suitable magnesium minerals (Mani et al., 2008) are:

$Mg_3SiO_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + SiO_2 + H_2O$
(Serpentine) (Magnesite) (Silica)

$MgSiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2$
(Olivine) (Magnesite) (Silica)

The reaction of industrial waste acids with olivine on enhanced rates (i.e., industrial time scale) was investigated by Schuiling et al. (1986), Jonckbloedt (1997) and Lieftink (1997). In a subsequent study, Schuiling and Krijgsman (2006) demonstrated that olivine reacts with compressed CO_2 in autoclaves, in the presence of right catalysts, in a matter of hours, which testifies to feasibility of industrial application of CO_2 sequestration. The main advantage of mineral carbonation is the thermodynamic stability of the formed carbonates, which makes the storage permanent and inherently safe (Lackner, et al., 1997). Furthermore, carbonation reactions are exothermic, which enables reduction of energy consumption and costs (Goff and Lackner, 1998; Lackner et al., 1997).

$CaO + CO_2 \rightarrow CaCO_3$ ($\Delta Hr = 179$ kJ/mol)

$MgO + CO_2 \rightarrow MgCO_3$ ($\Delta Hr = 118$ kJ/mol)

Where Hr is the heat released in the reaction.

Finally, the potential of the technology to store appreciable amounts of the CO_2 resulting from fossil fuel combustion is large enough because serpentine, olivine, and pyroxene-rich rocks occur in large amounts in nature (Goff and Lackner, 1998; Lackner, et al., 1997). Olivine is slow to react and serpentines react poorly, unless pretreated to remove chemically bound water. At a high temperature of ~600°C and pressure of about <0.5 kbar, the reaction has favourable conditions for kickstarting the carbonation pathways (Pokrovsky and Schott, 1999).

4. Potential of usage of ultramafic rocks for CO_2 for carbon sequestration in Pakistan

The primary requisite for using mineral carbonation technique for CO_2sequestration is availability of suitable ultramafic rocks containing Mg, Ca and Fe silicate minerals e.g., olivine. Olivine is a common constituent of dunites and peridotites. These rocks are typically abundant in mantle, exposed on Earth's surface as ophiolites. In crustal settings, they form as a part of layered complexes formed in deep-seated basaltic magma chambers as cumulates and are exposed to surface through tectonic exhumation processes. Northern Pakistan is famous for its tectonic evolution as a continent-island arc-continent collision zone involving closure of

Neotethys and collision between the Indian and Eurasian plates with entrapment of the Kohistan fossil-island arc in a broad suture zone between the two colliding plate (Tahirkheli et al., 1979). Ultramafic rocks in northern Pakistan occur both as part of the Kohistan island arc (Jan and Howie, 1981), as well as obduction of ophiolites onto the subducting Indian Plate (DiPietro et al., 2000). In the following we briefly review the occurrence of ultramafic rocks in northern Pakistan for their possible usage in CO_2 sequestration.

4.1. The Chilas complex

Chilas complex is a massive intrusive body occupying the midrib of the Kohistan island arc (Khan et al., 1989). Chilas complex makes a substantial contribution towards forming the mass of the plutonic crust of the island arc, the Kamila amphibolites and Kohistan batholith being the other major contributors. It stretches for over 200 km from western Dir to Nanga Parbat and attains a width of over 40 km in the centre. Much of the complex comprises gabbronorites with lensoide bodies of ultramafic rocks scattered throughout the complex but being most abundant in the vicinity of Chilas covering with a cumulative exposure of ~5 km^2.

4.2. The Dargai (Skhakot-Qila) ophiolite complex

This ophiolites constitutes an arc-shaped body of ultramafic rocks emplaced in the northern end of the Peshwar basin on the southern side of the Malakand range. It was emplaced over metamorphosed pelitic, calcareous and carbonaceous rocks ranging from Precambrian (Manglaur formation) to Triassic (Saidu formation) (Ahmad et al., 1998). Different geophysical studies like gravity and magnetic delineate that it is about less than 1.5 km deep klippe comprising of ultramafic rocks that overlies the Indian plate about 30 km south of the Indus suture zone. It forms part of the vast Indus suture mélange zone (Hussain et al.,1984) and it may be regarded as the western extension of the Indus suture mélange zone and grouped with other melanges of the Indus suture (DiPietro et al., 2000). The ultramafic rocks consist predominantly of harzburgite, followed by wehrlite and dunite.

4.3. The Jijal complex

Jijal complex constitutes the southernmost part of the Kohistan island arc (Jan and Howie, 1981; Coward et al., 1986; Jan and Windley, 1990; Kausar et al., 1998). It occurs in the hanging wall of the Indus suture and covers an area of about 150 km^2. It forms structurally the lowest part of the arc along which it was thrust onto the Indian plate, The complex is marked by a series of (generally layered) granulite facies metamorphosed mafic and ultramafic rocks including dunites, peridotites, wherlites and websterites (Jan and Howie, 1981; Jan and Windley, 1990).

4.4. Alpuri ultramafics

The ultramafic rocks form part of the Indus suture mélange south of the MMT in the Alpurai area of Swat area. There are a number of ultramafic bodies emplaced along the thrust, the largest being the 7×1 km lensoid serpentized mass west of Alpuri. A small lens of serpentinised mass occurs within the schists outcropping along the road just to the south of the main mass and MMT. The trend of the main body is almost north-south. The rocks are brownish to green to grey, hard, massive and medium to fine-grained. Serpentinization is wide-spread. Intense shearing is seen at the contact with other rock units. On the northern end of the body asbestiform serpentine is also noted which imparts a fibrous appearance to the rock (Khan and Humayoun, 1980).

5. Quantification of ultramafic rocks for CO_2 sequestration

In order to quantify various ultramafic rocks of Pakistan for their carbon storage capacity, the weight percentage of MgO (Goff and Lackner, 1998; Zevenhoven and Kohlmann, 2001) has been taken into account. Detailed information about the geology and structure of ultramafics and data on their areal distribution, approximate thickness, chemical composition and mineralogy are utilized to calculate the volume of ultramafic rocks in Chilas, Dargai, Jijal and Alpurai areas. The volume thus calculated was then multiplied by wt% of MgO to assess the quantity of ultramafic rocks needed to sequester CO_2 in these using the following formula of Zevenhoven and Kohlmann, (2001):

$T = 1 \times \rho \times a \times t \times d \times (1-ø)$,

Where T is the amount of CO_2 that can be sequestered, ρ is the % MgO in ultramafics, a is the area, t is the thickness, d is the average density and ø the average porosity of ultramafics.

a. For the Chilas mafic-ultramafic complex, an effective sequestration of about 20% at a depth of 1 km is calculated as:

   Volume of the complex=7318×1=7318 km^3
   Effective volume of the complex for sequestration = 20% of 7318 = 1463 km^3 = 1463 × 10^6 m^3.
   Average density = 3197 kg/m^3.
   Mass of ultramafics = volume × density = $1463 \times 10^6 m^3 \times 3197\ kg/m^3 = 4677 \times 10^9\ m^3$.
   Average % MgO in ultramafics = 27.67%
   Total MgO in ultramafics of Chilas Complex = 27.67% of $4677 \times 10^9 m^3$= 1294 million tons (mt.).
   Since 1 ton of MgO can dispose of approximately 1on t of CO_2 (Zevenhoven and Kohlmann, 2001), with an average porosity of 2% in ultramafic rocks, 1294 mt of MgO in Chilas Complex can sequester 1×1294×(1-0.02) = 1268.12 mt. of CO_2 in the form of magnesium carbonate.

b. For the Jijal Complex, an effective sequestration of about 20% of the complex at a depth of 1 km is calculated as:

   Volume of the complex=551×1=551 km^3
   Effective volume of complex for sequestration = 20% of 551 = 110 km^3 = $110 \times 10^6\ m^3$
   Average density = 3000 kg/m^3
   Mass of ultramafics = volume × density = $110 \times 10^6\ m^3 \times 3000\ kg/m^3$= 330 million tons (mt)
   % MgO in ultramafics = 29% (average) (Jan and Howie, 1981)
   Total MgO in ultramafics of Jijal = 29% of 330 mt = 95.7 million tons (mt)
   Since 1 t of MgO can dispose of approximately 1 t of CO_2 (Zevenhoven and Kohlmann, 2001), with an average porosity of 2% in ultramafic rocks, 95.798 mt of MgO in Jijal Complex can sequester 1×95.7×(1-0.02) = 93.79 mt of CO_2 in the form of magnesium carbonate.

c. For the Dargai Ultramafics, considering an effective sequestration of about 20% at a depth of 1 km:

   Volume of the Dargai Ultramafics= 153 ×1 = 153 km^3
   Effective volume of belt for sequestration = 20% of 153 = 30 km^3= 30 × 10^6 m^3.
   Average density = 4071 kg/m^3.
   Mass of ultramafic bodies = volume × density = 30 × 10^6 m^3 × 4071 kg/m^3= 122 million tons (mt).
   % MgO in ultramafics = 40% (Ahmed, 1988)
   Total MgO in ultramafics of Dargai = 40% of 122 mt = 48.8 million tons (mt).
   Since 1 t of MgO can dispose of approximately 1 t of CO_2 (Zevenhoven and Kohlmann, 2001), with an average porosity of 2% in ultramafic rocks, 48.8 mt of MgO in Dargai Ultramafics can sequester 1×48.8×(1-0.02) = 47.82 mt. of CO_2 in the form of magnesium.

d. For the Alpuri Serpentinites, considering an effective sequestration of about 20% to a depth of 1 km:

   Volume of the serpentinites=16×1=16 km^3
   Effective volume of serpentinites for sequestration = 20% of 16 = 3.2 km^3=3.2 × 10^6 m^3.
   Average density = 3300 kg/m^3.
   Mass of serpentinites = volume × density = 3.2 × 10^6 m^3 × 3300 kg/m^3= 10. 56 million tons (mt).
   % MgO in serpentinites = 17.37%
   Total MgO in Dargai Serpentinites = 17.37% of 10.56mt= 1.834272 million tons.
   Since 1 t of MgO can dispose of approximately 1 t of CO_2 (Zevenhoven and Kohlmann, 2001), with an average porosity of 2% in ultramafic rocks, 1.834272 mt of MgO in Dargai Serpentinites can sequester 1×1.834272 ×(1-0.02) = 1.79 mt of CO_2 in the form of magnesium carbonate.

All the results obtained are presented in Table 1. The estimates, however, can be further improved by including the reaction kinetics and hydrological parameters in the equation.

Table 1. Various properties of samples from different areas and their sequestration potential

Sample Location	**Area (km^2)**	**Average density (kg/m^3)**	**Mass (Million tons)**	**MgO (%)**	**CO_2 Sequestration Potential (Million tons)**
Chilas Complex	7318	3197	4677	27.67	1268.12
Jijal Complex	551	3000	330	29	93.79
Dargai Ultramafics	153	4071	122	40	47.82
Alpuri Serpentinites	16	3300	10.56	17.37	1.79

6. Conclusions

This assessment provides the probable amounts of CO_2 that can be stored as mineral carbonates in ultramafic rocks of Chilas, Dargai, Jijal and Alpuri areas. If the effective sequestration amounts to be 20% of the total, from this computation it is observed that approximately 1268, 94, 48, and 2million metric tons of CO_2 can be sequestered in the ultramafics rocks of Chilas, Jijal, Dargai, and Alpuri ultramafics respectively. For the year 1999, the carbon emission rate of Pakistan was 92 million metric tons, out of which 26 million metric tons had been contributed by the manufacturing units and construction industry (www.asianewsnet.net/climate/pdf/pakistan_profile). The estimates shown here exemplify the sequestration potential capacity of the ultramafic rocks. The low porosity, structural features and chemical compositions of the mafic and ultramafic rocks make them to be the best-suited carbon storage reservoirs in comparison. A similar assessment for mafic and ultramafic rocks exposed elsewhere in Pakistan, based on their geology, structure, mineral and chemical composition for sequestering CO_2 may provide a viable option as another important medium to mitigate the greenhouse gas effect of CO_2.

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