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Mineralogical and Geochemical Characterization of Jordanian Olivine and Its Ability to Capture CO_2 by Mineralization Process

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Abstract - Olivine rich rocks outcrop in Kharazeh area (KHO) and Jurf Eldaraweesh (JDO) Basaltic rocks southern Jordan. KHO and JDO have been characterized for their mineralogical and geochemical properties investigated for their potential use in CO_2 capture by mineralization process. The effects of particle size, temperature, and contamination time were examined in the mineralization process. Two grain sizes of olivine for each type were used in capture experiments. The first grain size is the separated size fraction between 1 to 0.7 mm (KHO1 and JDO1), while the second grain size is the separated size fraction between 0.7 to 0.3 mm (KHO2 and JDO2). For the mineralization purpose, a chemical reactor was used to determine the olivine CO_2 capture capacity. Thin section studies aided by XRD identification for KHO indicated the presence of olivine crystals as the main mineral with percentage reaches 48% plagioclase, augite, and magnetite. While the mineral content for JDO is (35%) olivine, plagioclase, clinopyroxene, and opaque minerals. Olivine chemical composition results show the high MgO percentage for KHO ranges between 47.5 and 50.13%, while the percentage of MgO varies from 43.6% and 44.6% for JDO. The CO_2 mineralization process results were interpreted using percentage removal curves. CO_2 capture percentage reaches the highest percentage (48%) using KHO2 and 100^o C temperature. In summary, it can be concluded that CO_2 can be removed from atmosphere using Jordanian olivine rich rocks.

Keywords: olivine, Kharazeh, Jurf Eldaraweesh, CO_2, mineralization process

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INTRODUCTION

In these days, global warming and greenhouse effect have great concerns and become one of the most important international environmental problems due to the rise of concentration levels of greenhouse gases. One of the main greenhouse gases is carbon dioxide (CO_2) which increases in the atmosphere, especially with huge increase of CO_2 emissions (Olivier *et al.*, 2017).

CO_2 is naturally released from the respiration of animals, plants, and microorganisms, and is also released by volcanic eruptions and hot springs (EPA, 2008). However, human activities, primarily the combustion of fossil fuels that account for more than 80% of worldwide energy consumption, contribute to the growth of global CO_2 emissions (Kwon *et al.*, 2011). Anthropogenic emission of CO_2, has caused carbon dioxide concentration increases in the atmosphere from 280 ppm in pre-industrial times to 400 ppm currently (ESRL, 2014), and if undiminished, by 2100, these value will rise to 1,100 ppm (White *et al*., 2003; IPCC, 2014).

The amounts of CO_2 released to the atmosphere should be lowered. So, the increase of concentration levels of CO_2 in the atmosphere is one of the most important environmental problems (Olivier *et al*., 2016).

Carbon capture and storage (CCS) technology, and CO_2 mineralization process or mineral carbonation are valuable techniques for the capture of CO_2. It is an effective technology used for reducing greenhouse gas CO_2 from the atmosphere (Qianyi, 2010).

Mineral carbonation is therefore a naturally occurring geological weathering process that involves the reaction of CO_2 with common mineral silicates to form geologically stable solid inorganic carbonates (Doucet, 2011).

One important technique used for CO_2 capturing is by reacting it through a bed of magnesium-based minerals. The idea of applying carbonation reaction for CO_2 storage was proposed by Seifritz (1990) and (Haug *et al*., 2010).

Mineralization is the process of the formation of stable carbonates by reaction of CO_2 with alkaline metal oxides (typically Mg and Ca-based minerals). The process of mineral carbonation is stable over geological time scales, it was discussed further by Dunsmore (1992) and Kwon *et al*., (2011), and subsequently followed by the first detailed exploration of this approach (Lackner, 2002). Significant progress has been made over the last fifteen years and different aspects of CCMC have been reviewed (Lackner, 2002; Huijgen and Comans, 2003, 2005a, 2005b; Sipilä *et al*., 2008; Doucet, 2011a; Kwon *et al*., 2011).

The most common approach for carbonation process is a direct gas-solid carbonation in which gaseous carbon dioxide reacts directly with the solid natural material (Mg and Ca-based minerals) (Huijgen and Comans, 2003).

Olivine (OL) is a natural occurring magnesium-based silicate mineral that can be used as CO_2 capture in the mineralization process (Seifritz, 1990; Kwon *et al*., 2011). Another way of applying this approach is to inject CO_2 into peridotite rocks that are rich in the easily weatherable olivine (Schuiling, 2006; Kelemen and Matter, 2008).

Olivine $[(Mg,Fe)_2SiO_4)]$ is the most abundant mineral in the earth mantle until a depth of 700 km (Dabirian, 2012). Olivine is one of the most minerals of mafic and ultramafic rocks (Dabirian, 2012). It is one of the silicate minerals belongs to isolated (or independent) tetrahedral. Olivine often consists of magnesium (Mg^{2+}), oxygen, and silicon tetrahedral ($SiO_4{}^{2-}$) (Figure 1) (King, 2009).

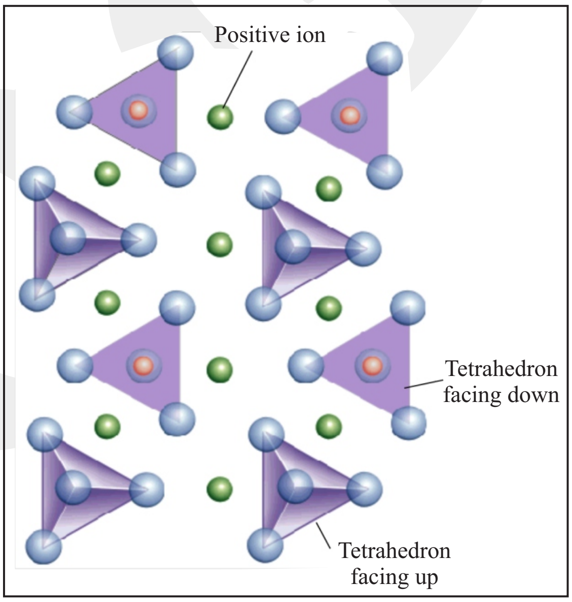

Figure 1. Olivine chemical structure.

Olivine in Jordan is abundant and a low cost raw material found in different areas associated with basaltic rocks belonging to Cenozoic volcanism (Barberi *et al*., 1979). Karak dike in Kharazeh area (31°15'10"N and 35°35'19"E) (Figure 2) is one of the important olivine rich locations in southern Jordan. Another location is Jurf Eldaraweesh (JDO). This research paper aims to characterize the physical and chemical properties of Jordanian olivine and to investigate the potential use of local natural olivine rich rocks in CO_2 capturing. OL was chosen due to its availability and high concentration in Jordanian basaltic rocks.

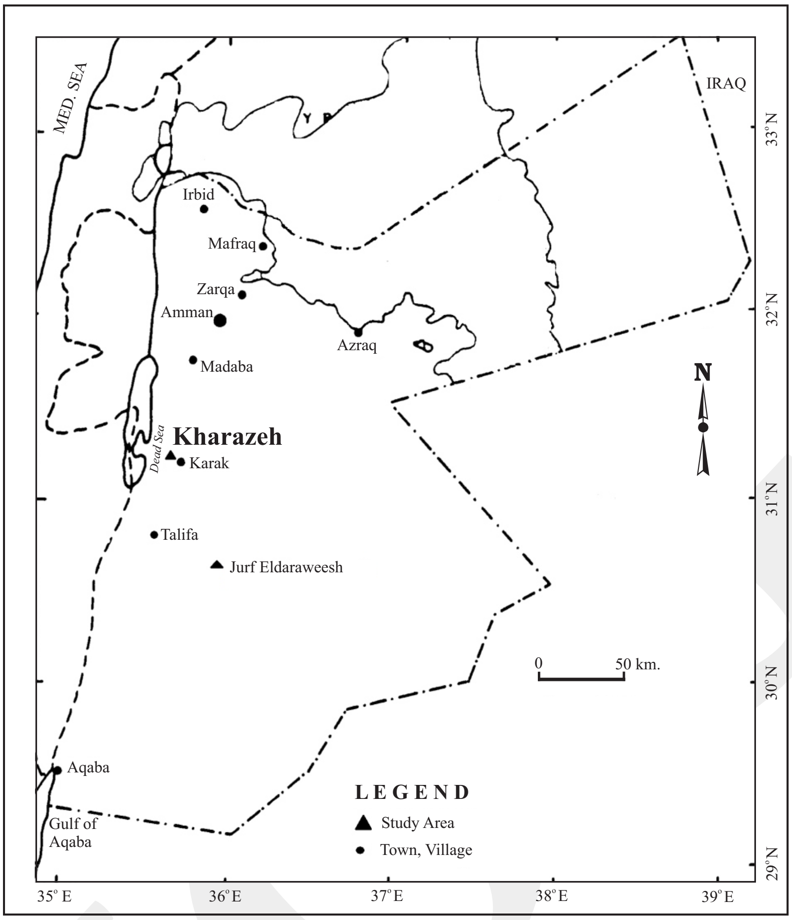

Figure 2. Locality map of Kharazeh olivine (KHO) and Jurf Eldaraweesh olivine (JDO), southern Jordan.

Materials and Methods

Two types of fresh basaltic rocks were chosen to be used in the mineralization experiments. They are Kharazeh olivine rich basaltic rock (KHO) and Jurf Eldaraweesh basaltic rock (JDO), situated in southern Jordan (Figures 2 and 3).

Characterizations of the basaltic samples were analyzed for their mineralogical and chemical compositions. Mineralogical identification was determined using thin section petrography and X-Ray Diffraction (XRD) analyses. The chemical composition was determined using X-Ray fluorescence (XRF) apparatus. Characterization studies were carried out in the Faculty of Engineering Laboratory at Al Housin Ben Talal University in 2016.

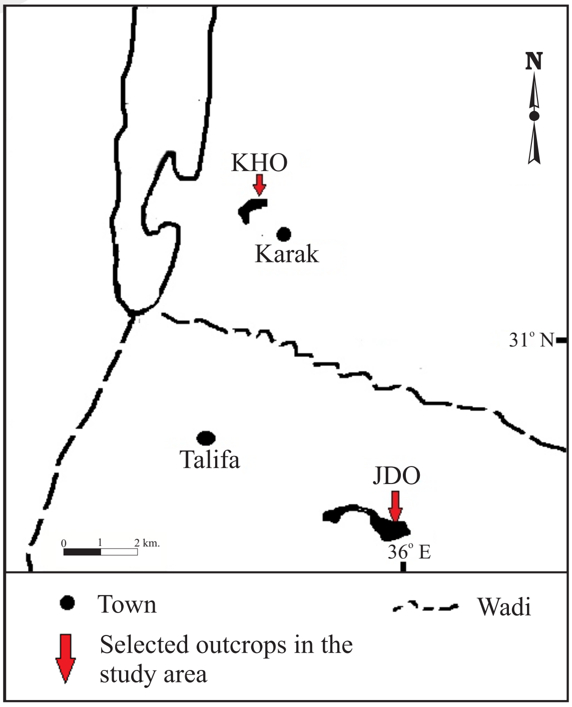

Figure 3. Geological map where the samples were taken.

KHO and JDO samples used in the capture experiments were crushed using jaw crusher with an aperture of 5 cm, and sieved into two sizes;

the first one is -1+0.700 mm, and the second one is -0.700 +0.300 mm (Table 1). Olivine rich samples were used in the experiments without any modification or chemical treatment.

Table 1. Grain Size Fraction for the Used Olivine in CO_2 Capture Experiments

Location	Sample Name	Used Sizes	Grain size (mm)
Kharazeh olivine	KHO	KHO1	-1 +0.700
		KHO2	-0.700 +0.300
Jurf Eldaraweesh olivine	JDO	JDO1	-1+0.700
		JDO2	-0.700 +0.300

A series of direct carbonation experiments were conducted in TTU for CO_2 capturing based on combining CO_2 chemically with olivine rich rocks to form stable carbonate minerals (Figure 4) by using the direct carbonation reaction (gas-solid carbonation) (Huijgen and Comans, 2003) as follow:

$$\text{Fe/Mg-silicate (S)} + CO_2\ (g) \rightarrow \text{(Fe/Mg)}\ CO_3\ (S) + SiO_2\ (S)$$

A simple reactor was developed and fabricated for the purpose of mineralization composed of an Erlenmeyer flask filled with 50 gr of olivine grains and 200 ml of tap water with CO_2 inlet and outlet. The Erlenmeyer flask was put on a shaking table using magnetic stirrer and hot plate (Figure 4). Each olivine type (KHO1, KHO2, JDO1, and KDO2) were filled separately in to the reactor. The time series including tests were conducted for 1, 2, 4, 8, 12, and 24 hours. The used sizes, temperature, and contaminated time are listed in Table 2.

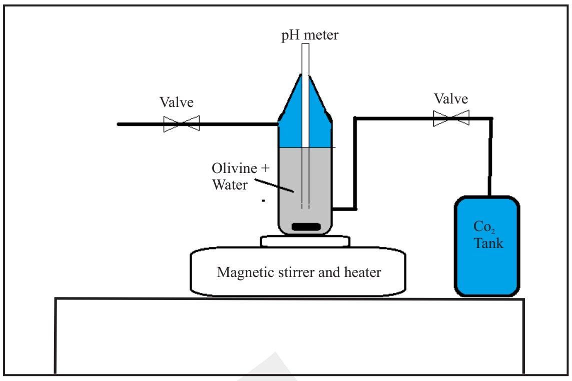

Figure 4. A pilot plant scale reactor for CO_2 capturing.

Results and Discussion

Mineralogy Results

The hand sample of KHO shows large green colour olivine crystals which can be seen by eyes, while JDO hand samples show black to green colour with a medium to small olivine crystals (Figure 5). The microscope analysis indicates the presence of primary rock forming minerals including olivine, iddingsite, pyroxene, plagioclase, feldspathoids, alkali feldspar, and opaques

Figure 5. Hand sample for KHO and JDO showing the presence of olivine crystals.

Table 2. Size Fraction and Time Used in the Batch Experiments Using Olivine Samples

Olivine Sample	Column #	Size fraction (mm)	Time in Hours	Temperature C°
KHO1	1	-1 +0.700	1, 2,4,8,12,and 24	50
KHO2	2	-0.700 +0.300	1, 2,4,8,12,and 24	100
JDO1	3	-1 +0.700	1, 2,4,8,12,and 24	50
JDO2	4	-0.700 +0.300	1, 2,4,8,12,and 24	100

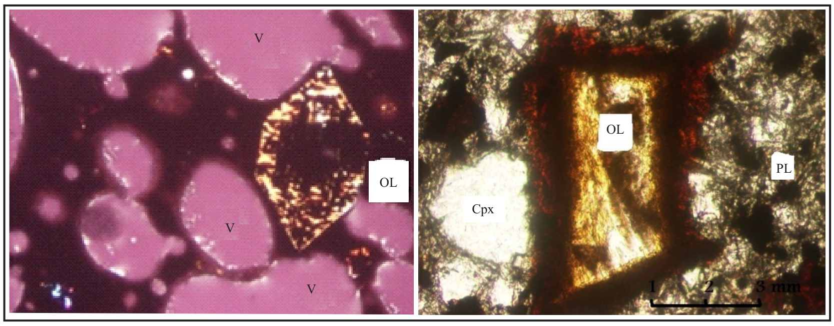

Figure 6. Photomicrographs of Jordanian olivine under XPL, 4x*10x magnification (4x= 2.25 mm). V= vapour; Cpx= clinopyroxene; OL= olivine; PL= plagioclase.

in both KHO and JDO samples (Figure 6). Thin section studies in most of the studied slides show that KHO and JDO are colourless and is highly fractured. The olivine crystal lengths range from 0.3 to 2 mm. Olivine is altered to iddingsite (Id).

Olivine is usually destroyed or replaced during the early stage of weathering. The most common product is iddingsite, which may be optically homogeneous and crystallographically oriented with respect to the parent olivine (Wilson, 2004).

The XRD pattern for KHO and JDO is shown in Figure 7, while the mineral content is listed in Table 3. The results indicate the presence of olivine in all samples.

Chemical Composition Results

The XRF analyses are listed in Table 4. The obtained XRF results show that the SiO_2 concentration ranges between 42.10 and 44.30% for KHO, and between 41.78 - 42.70% for JDO. The MgO% varies from 47.5 to 50.13% for KHO, and from 41.78 to 42.70% for JDO.

CO_2 Mineralization Experiment Results

The results of direct CO_2 capturing experiment using KHO and JDO are listed in Tables 5 and 6, and represented in Figures 8, 9, 10, and 11. The data in Table 5 represent the results of direct CO_2 capturing using KHO1, KHO2, JDO1, and JDO2 within different shaking times and temperatures.

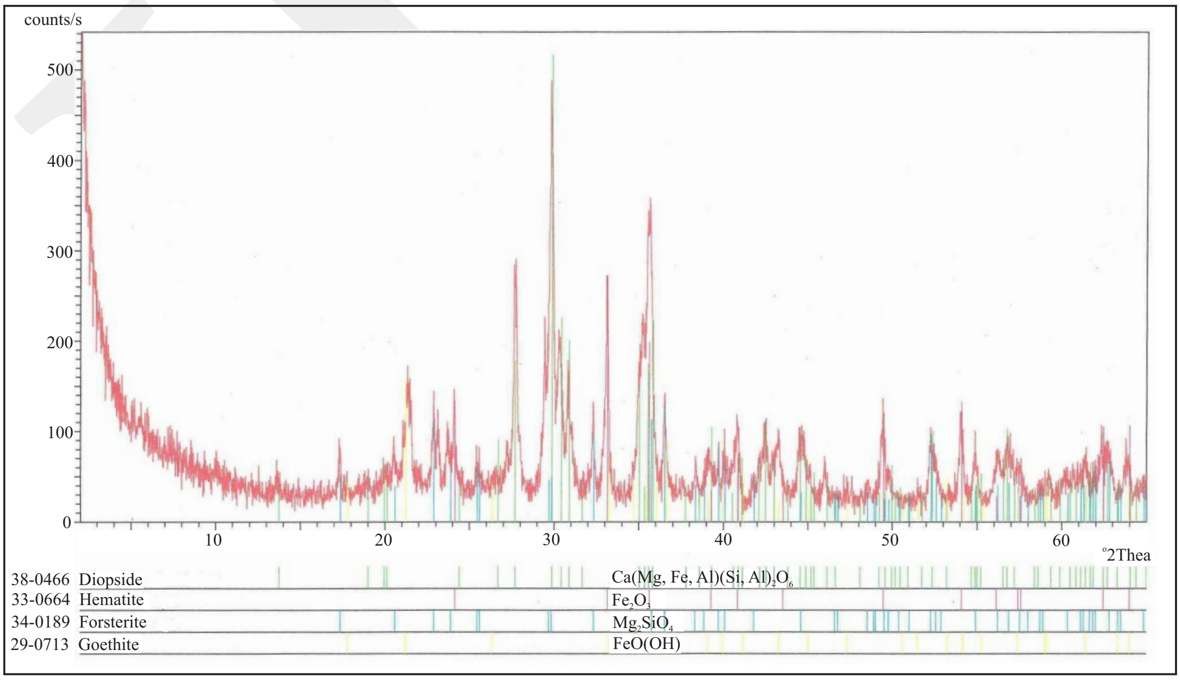

Figure 7: X-ray diffraction pattern of the olivine rock using Cu K α radiation for KHO.

Table 3. Minerals Identified by XRD Method for Jordanian Olivine

Location	**Olivine**	**Plagioclase**	**Clinopyroxene**	**Opaque Minerals**
(KHO)	*	*	*	*
(JDO)	*	*	*	*

Table 4. Chemical Composition for KHO and JDO Olivine

	KHO1	**KHO2**	**JDO1**	**JDO2**
SiO_2%	42.10	44.30	41.78	42.70
Al_2O_3%	0.231	0.211	0.231	0.221
MgO%	47.5	50.13	43.5	44.6
Fe_2O_3%	2.961	2.45	2.57	2.71
FeO%	5.98	6.201	5.32	5.12
K_2O%	0.006	0.007	0.006	0.005
CaO%	0.040	0.032	0.029	0.028
Na_2O%	0.086	0.097	0.091	0.0081

Table 5. Percentage Removal Obtained from Batch Experiments for CO_2

Olivine Samples	**Shaking Time (hours)**	**Percentage Removal % CO_2 at T=50°C**	**Percentage Removal % CO_2 at T=100°C**
KHO1	1	0.21	0.3
	2	0.23	0.35
	4	0.25	0.39
	8	0.27	0.41
	12	0.28	0.43
	24	0.29	0.45
KHO2	1	0.24	0.35
	2	0.26	0.38
	4	0.27	0.41
	8	0.29	0.43
	12	0.30	0.47
	24	0.31	0.48

Table 6. Percentage Removal Obtained from Batch Experiments for CO_2

Olivine Samples	**Shaking Time (hours)**	**Percentage Removal % CO_2 at T=50°C**	**Percentage Removal % CO_2 at T=100°C**
JDO1	1	0.21	0.24
	2	0.25	0.24
	4	0.27	0.25
	8	0.29	0.25
	12	0.3	0.26
	24	0.31	0.27
JDO2	1	0.27	0.27
	2	0.28	0.3
	4	0.28	0.31
	8	0.29	0.33
	12	0.29	0.36
	24	0.30	0.37

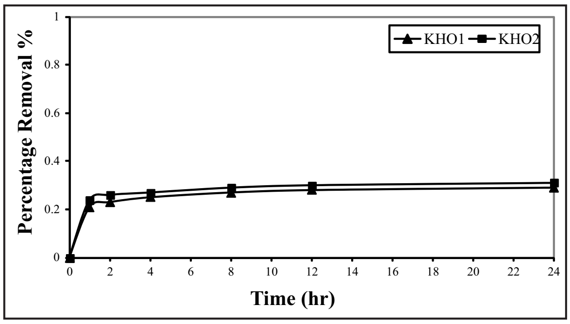

Figure 8. Percentage removal of CO_2 using different grain sizes of KHO and temperature equal 50° C.

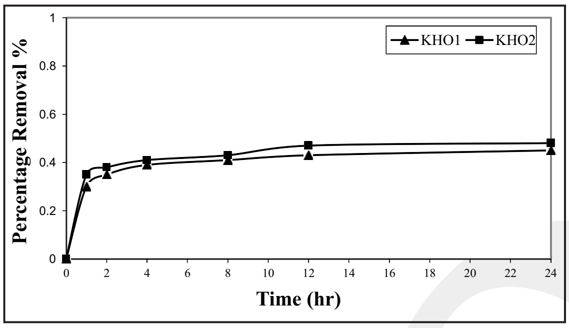

Figure 9. Percentage removal of CO_2 using different grain sizes of KHO and temperature equal 100° C.

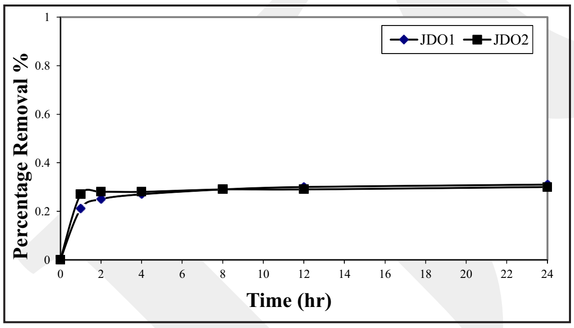

Figure 10. Percentage removal of CO_2 using different grain sizes of JDO and temperature equal 50° C.

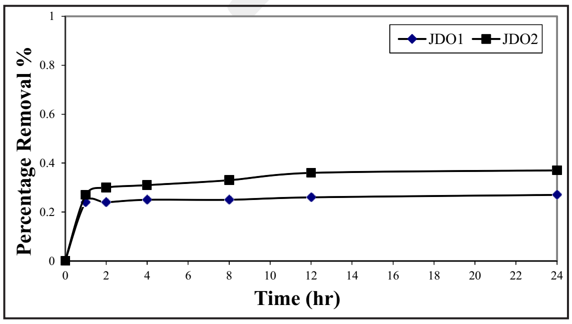

Figure 11. Percentage removal of CO_2 using different grain sizes of JDO and temperature equal 100° C.

KHO and temperature equal 50° C (Table 5) results show that the CO_2 capture percentages range between 0.21% and 0.29%, and from 0.24% to 0.31% by using KHO1 and KHO2, respectively. While by using temperature equal 100°C, the CO_2 capture percentages varies from 0.30% and 0.45%, and from 0.35% to 0.48% by using KHO1 and KHO2, respectively.

JDO and temperature equal 50° C (Table 6) results show that the CO_2 capture percentages range between 0.21% and 0.31%, and from 0.27% to 0.30% by using JDO1 and JDO2 respectively. Furthermore, by using temperature equal 100°C the CO_2 capture percentages range between 0.24% and 0.27%, and from 0.27% to 0.37% by using JDO1 and JDO2, respectively.

CONCLUSION

The use of Jordanian olivine rocks from southern Jordan (KHO and JDO) as the capture material for CO_2 is effective and suitable for the mineralization processes. Utilization of olivine for CO_2 capture can be an alternative and effective technique for capturing CO_2 from the air. KHO sample with size fraction (0.3 - 0.7mm) is suitable for the direct mineralization of CO_2. The increase of shaking time and temperature has increased the reaction in addition to the decreasing of grain size. By using the available equipment and material (simple reactor), the achieved reduction of CO_2 emissions reaches 0.48% in maximum.

Experimental data have revealed that Jordanian natural olivine (KHO and JDO) is effective for capturing CO_2 and controlling CO_2 emission from industry. Olivine fraction KHO2 is the suitable material and more effective than the other sizes in capturing CO_2.

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