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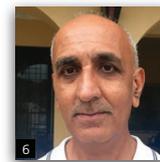
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# Utilisation of carbonating olivine for sustainable soil stabilisation

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This paper describes the first study demonstrating the potential of olivine as a soil stabiliser. Olivine has been shown to provide a reactive source of magnesium oxide capable of sequestering carbon dioxide. The effects of olivine additions on consistency limits, compaction characteristics and unconfined compressive strength (UCS) of soil are described. The effect of carbon dioxide pressure, and carbonation period, on the UCS of olivine-treated soil is of great importance in defining treated properties. Results highlight the benefits of olivine in soft soil stabilisation with reference to the UCS. Use of 20% olivine decreased the plasticity index and optimum moisture content while increasing the maximum dry density of the soil. The greatest strength was developed after carbonation at 200 kPa for 168 h in the soil containing 20% olivine. Structural and compositional analysis using scanning electron microscopy and X-ray diffraction confirmed the benefits of olivine in terms of decreasing the discontinuity of soil. This was attributed to the crystallisation products responsible for strength development after carbonation, respectively. The paper is significant as it presents a more environmentally friendly method of stabilising soils compared with alternative methods using high embodied energy binders such as cement.

## Introduction

Soil stabilisation is one of the most extensively used techniques for ground improvement. Cement, lime and other admixtures are commonly used in construction to enhance bearing capacity, reduce settlement, control shrinking and swelling and reduce permeability. Owing to its high strength, cement is one of the most important and widely used binders in soil stabilisation (Horpibulsuk *et al.*, 2011; Pakbaz and Alipour, 2012; Pourakbar *et al.*, 2015). However, cement production consumes various types of fuel that results in emissions of carbon dioxide (CO<sub>2</sub>) to the atmosphere. While the cement industry only accounts for around 5% of global carbon dioxide emissions, associated releases of carbon dioxide from cement production have continued to grow by 2.5% per year on average over the past decade (Friedlingstein *et al.*, 2014; Worrell *et al.*, 2001).

Recent developments in ground improvement have heightened the need for using sustainable materials (e.g. reactive magnesia, zeolite) or by-products (e.g. fly ash, rice husk ash, cement kiln dust, palm oil fuel ash, recycled glass foam, recycled concrete aggregate and crushed brick, calcium carbide residue and ground-granulated blast-furnace slag) for partial or full replacement of cement as a result of the significant environmental impacts (Arulrajah *et al.*, 2015; Basha *et al.*, 2005; Cai *et al.*, 2015; Disfani *et al.*, 2014; Du *et al.*, 2011, 2015; Jegandan *et al.*, 2010; Pourakbar *et al.*, 2015; Yi *et al.*, 2013a). The search for alternative binders identifies materials that are cost-effective and would affect the surrounding soil to a lesser extent upon treatment.

Governments throughout the world recognise a growing urgency to find safe and cost-effective solutions to counteract climate

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change by reducing the carbon dioxide levels of the atmosphere. Carbon dioxide sequestration through reactive minerals has been widely proposed by several researchers (Andreani *et al.*, 2009; Chen *et al.*, 2006; Haug *et al.*, 2011; Prigiobbe *et al.*, 2009b). Olivine,  $(\text{Mg,Fe})_2\text{SiO}_4$ , as one of the most promising candidates, has been the focus of many studies (Hänchen *et al.*, 2006; Olajire, 2013; Olsson *et al.*, 2012). All the carbon dioxide created by burning 1 litre of oil can be sequestered by less than 1 litre of olivine; in other words, 140 g of olivine will sequester 176 g of carbon dioxide (Schuiling and Praagman, 2011). As a result of carbon dioxide sequestration, a stable mineral carbonate is produced. In addition, in accordance with ASTM D5370, olivine can be considered as a pozzolanic material due to the high levels of silicon dioxide ( $\text{SiO}_2$ ), iron(III) oxide ( $\text{Fe}_2\text{O}_3$ ) and aluminium oxide ( $\text{Al}_2\text{O}_3$ ), which potentially can contribute to the soil improvement (ASTM, 2014). Furthermore, the significant proportion of magnesium oxide ( $\text{MgO}$ ) contained within olivine provides a large potential for hydration, contributing to the soil strength.

Olivine is widely distributed around the world. It has been found everywhere all over the Earth: Egypt, Myanmar, South Africa, Russia, Pakistan, Norway, Sweden, France, Brazil, Germany, Mexico, Australia, China and also the USA (TARGET MAP, 2012). A geological survey of Malaysia shows that there is a large number of volcanic rocks of the andesite–dacite–basalt in the Tawau Mountains in Sabah, which is the main source of olivine (Tahir *et al.*, 2010). According to several studies, the carbonation of olivine breaks the chemical bond between magnesium oxide and silicon dioxide to produce magnesium carbonate ( $\text{MgCO}_3$ ), with quartz being the main product (Daval *et al.*, 2011; Dufaud *et al.*, 2009; Saldi *et al.*, 2013). Recently, many attempts have been made to use reactive magnesia in soil stabilisation. In these studies, magnesium oxide grout was mixed with the soil using an auger and the columns were then carbonated with gaseous carbon dioxide introduced in two different ways: one using auger mixing and the other through a perforated plastic tube system inserted into the treated soil (Cai *et al.*, 2015; Yi *et al.*, 2013b). Their results demonstrated that the reactive magnesia, after carbonation, contributed to an increase in the shear strength of the soil. The studies also revealed that the brucite ( $\text{Mg}(\text{OH})_2$ ) and magnesium carbonate were responsible for soil strength (Yi *et al.*, 2013a, 2013b, 2013c). It was reported by several researchers that in situ carbonation of the reactive magnesium oxide-treated soils can be achieved by resorting to two approaches, including a mass carbonation stabilisation method and a deep mixing carbonation method (Cai *et al.*, 2015; Yi *et al.*, 2013b). However, a major problem with this type of application is that the reactive magnesia is usually produced by heating magnesium carbonate that releases carbon dioxide into the atmosphere. Since olivine is an excellent source of magnesium oxide, it is more prudent to produce the magnesium carbonate from the reaction between olivine as a natural mineral source and carbon dioxide. Equations 1 and 2 describe the chemical reaction of olivine and water to form brucite. According to this reaction, the same number of

moles of serpentine ( $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$ ) and brucite is produced and all elements are preserved in the rocks with the exception of water (Okamoto *et al.*, 2011).



The rate of carbonation of olivine by this reaction is dependent on several factors, such as the presence of water, carbon dioxide pressure, pH and temperature (Kwon *et al.*, 2011; Prigiobbe and Mazzotti, 2011; Prigiobbe *et al.*, 2009a). According to the previous discussion, besides the role of olivine as a sustainable carbon dioxide capture strategy, olivine could potentially be utilised to stabilise soil through magnesite production during olivine carbonation, which may be capable of binding soil particles. Nevertheless, the consumption of minerals as natural resources should also be managed appropriately. This study aims to investigate the role of olivine as an intelligent sustainable soil stabiliser. The effects of incorporating olivine and carbonated olivine on mechanical properties of soil and the underlying mechanisms of stabilisation are investigated.

## Material and methods

### Materials used

#### Soil properties

The soil was collected from a depth of 2 m from Hulu Langat located in the state of Selangor, Malaysia. The selected soil contained 10% sand, 60% silt and 30% clay, with a mean particle size ( $D_{50}$ ) of 11.759  $\mu\text{m}$  and specific surface area of 1.04  $\text{m}^2/\text{g}$ . Table 1 shows the physicochemical properties of the soil, based on the Unified Soil Classification System, the soil is classified as highly plastic (CH). Figure 1 indicates that the particle size distribution of the soil ranges from 1 to 800  $\mu\text{m}$ .

| Properties          | Value                       |
|---------------------|-----------------------------|
| OWC                 | 23.3%                       |
| PL                  | 30%                         |
| LL                  | 54%                         |
| PI                  | 24%                         |
| Specific gravity    | 2.65                        |
| Maximum dry density | 1.58 $\text{g}/\text{cm}^3$ |
| <i>Particles</i>    |                             |
| Sand                | 10%                         |
| Silt                | 60%                         |
| Clay                | 30%                         |

**Table 1.** Physicochemical properties of soil. LL: liquid limit; OWC: optimum water content; PI: plasticity index; PL: plastic limit

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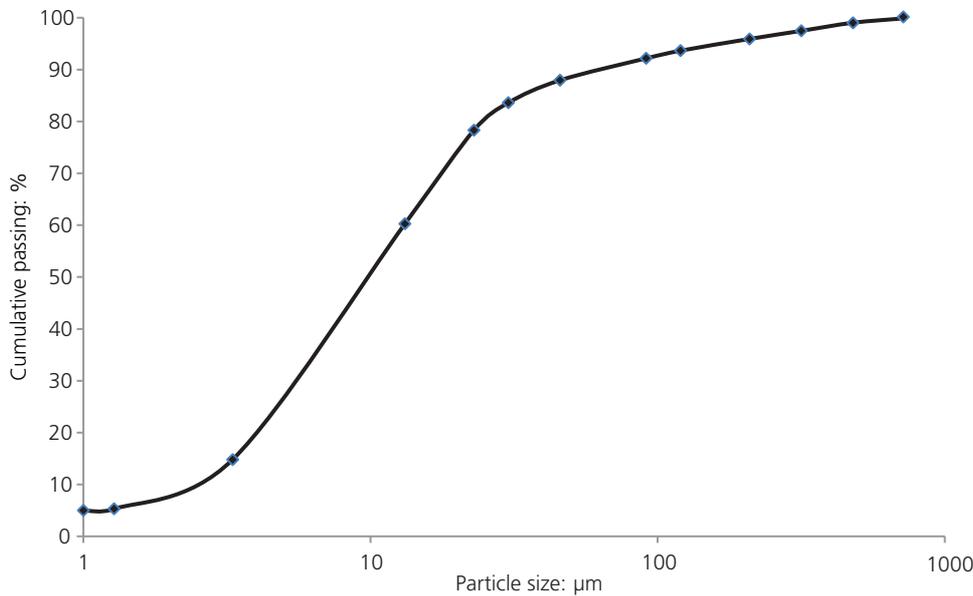


Figure 1. Particle size distribution of soil

**Olivine properties**

The olivine was obtained from MAHA Chemicals Sdn Bhd in Malaysia. The physicochemical properties of the olivine are given in Table 2. According to this table, olivine has a high magnesium oxide content of around 48% and a silicon dioxide content of around 40%. The olivine in its received state had a large particle size, which required ball milling for 24 h at a speed of 60 rpm (around 65% of critical speed) to decrease and homogenise the particle size prior to addition to the soil. Figure 2 shows the particle size distribution of olivine before and after milling. The analysis indicates that the  $D_{50}$  and specific surface area of olivine after milling were changed from 465.68 to 2.24 μm and 0.015 to 6.07 m<sup>2</sup>/g, respectively.

**Laboratory tests**

**Atterberg limit test**

Liquid limit (LL) and plastic limit (PL) tests were performed to assess the effects of the olivine on the physical properties of soil. The ball-milled olivine was applied with the rate of 5, 10, 15, 20,

22.5 and 25% by mass of dry soil. Soil and olivine with defined amounts were mixed and conveyed to the experimental pots. After mixing, the specimens were cured in a plastic cover for a day, air dried, pulverised, and passed through a 425 μm test sieve. The Atterberg limits were then determined in accordance with the British Standard methods (BSI, 2003a).

**Soil compaction test**

The standard Proctor compaction test was conducted to attain the moisture–density relationship of the soil-stabiliser mixtures according to British Standard methods (BSI, 2003b). These tests were performed to measure the maximum dry unit density (MDD) and the optimum water content (OWC) of the untreated soil and olivine-treated soil at different olivine dosages (i.e. SO<sub>5</sub>, SO<sub>10</sub>, SO<sub>15</sub> and SO<sub>20</sub> samples). The host soil was mixed with the previously ball-milled olivine at dosages of 5, 10, 15, and 20%. Water was added as required to facilitate the mixing and compaction processes for each percentage of olivine. The MDD and OWC of the treated soil was then determined. Compaction was performed immediately after mixing to avoid the formation of voids in the specimens, which lowers the unit weight.

**Carbonation set up and method**

Figure 3 shows a schematic diagram of the triaxial cell used to infuse pressurised gaseous carbon dioxide throughout the samples. Samples were subjected to a confining pressure of 400 kPa, followed by upward permeation of the carbon dioxide. The outflow tube was placed underwater in order to detect whether or not the samples were fully saturated with carbon dioxide gas. After a few minutes, the outflow tap was closed while keeping the inlet open. Since the carbon dioxide mineralisation of olivine is significantly influenced by carbon dioxide exposure time and

| Properties                     | Value  |
|--------------------------------|--------|
| Specific gravity               | 3.43   |
| MgO                            | 48.28% |
| SiO <sub>2</sub>               | 40.32% |
| Al <sub>2</sub> O <sub>3</sub> | 1.37%  |
| Fe <sub>2</sub> O <sub>3</sub> | 8.9%   |
| CaO                            | −1.13% |

Table 2. Physicochemical properties of olivine

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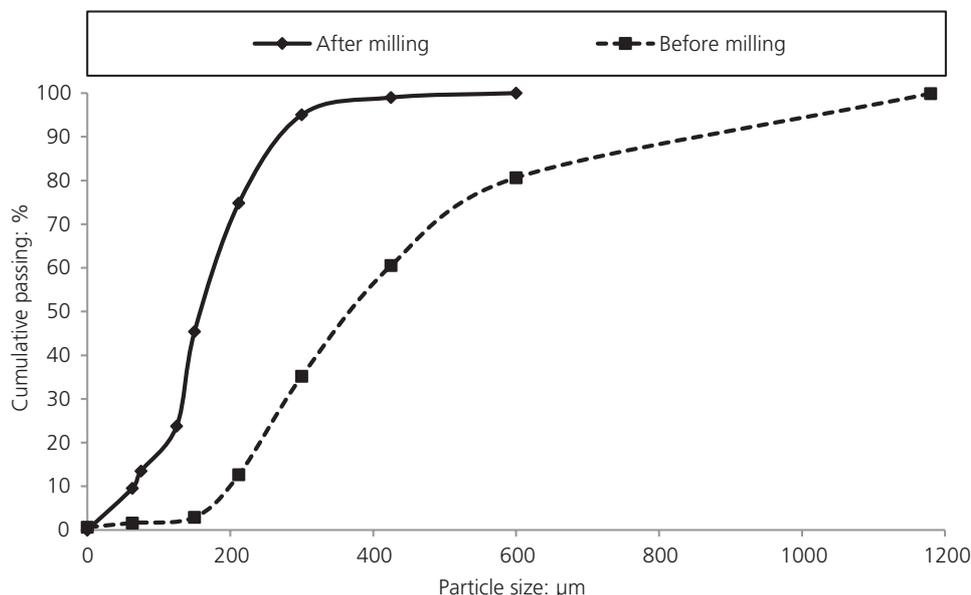


Figure 2. Particle size distribution of olivine before and after milling

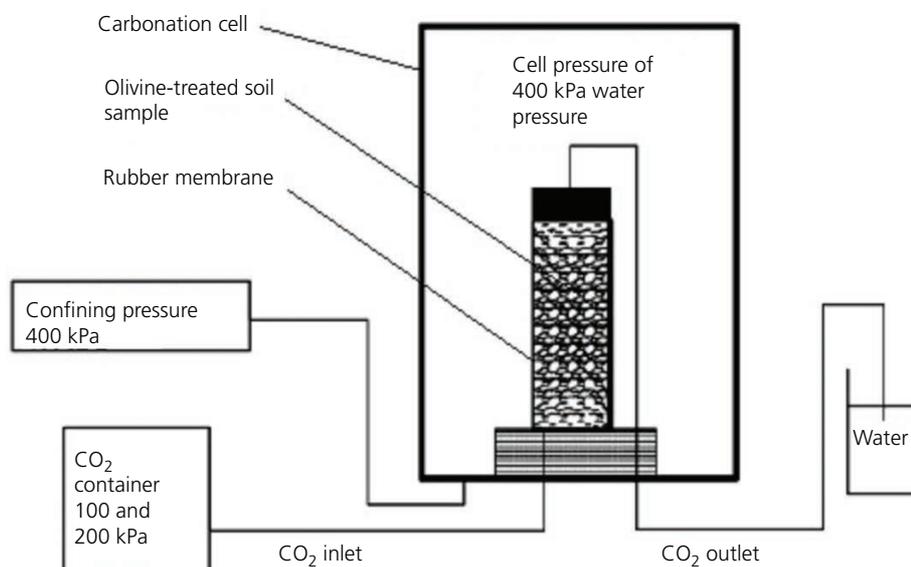


Figure 3. Schematic diagram of triaxial setup for carbonation of olivine-treated soil samples used in this research

pressure to olivine (Kwon *et al.*, 2011), the samples were then carbonated at various carbon dioxide pressures of 100 and 200 kPa at various carbonation periods of 7, 48 and 168 h.

#### Unconfined compressive strength before and after carbonation

In the first series of the unconfined compressive strength (UCS) tests, the ball-milled olivine was applied into the soil at dosages of 5, 10, 15 and 20%. Each specimen was then compacted in a

cylindrical mould with a diameter of 50 mm and a height of 100 mm in three layers, at the MDD and OWC for each percentage of olivine. The unconfined compressive tests were conducted on treated specimens at curing times of 7, 14, 28 and 90 d according to British Standard methods (BSI, 2003c). The UCS defined as the stress corresponds to the peak stress condition. Stress values were measured at specified ages in three specimens for each mixture and all data points deviated less than 5% from the average. Consequently, the most effective percentage

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| Mixing                          | Curing time: d | Olivine dosage: % | Carbonation period: h | CO <sub>2</sub> pressure: kPa |
|---------------------------------|----------------|-------------------|-----------------------|-------------------------------|
| SO <sub>B</sub> T               | 7, 14, 28, 90  | 5, 10, 15, 20     | —                     | —                             |
| C <sub>TP</sub> SO <sub>B</sub> | —              | 20                | 7, 48, 168            | 100, 200                      |

SO<sub>B</sub>T: soil-olivine<sub>(dosage)</sub>-curing time

C<sub>TP</sub>SO<sub>B</sub>: carbonated<sub>(time, pressure)</sub>-soil-olivine<sub>(dosage)</sub>

**Table 3.** Research design of samples in this study

of olivine obtained from the first series of tests was applied to the soil. Figure 3 shows the soil samples carbonation setup used in this investigation. As before each specimen was compacted in the cylindrical mould, in three layers, at MDD and OWC. Finally, the second series of the unconfined compressive tests were conducted on treated specimens according to BSI (2003c).

A summary of all tests is presented in Table 3. Within this table, the following abbreviations are used: SO<sub>B</sub>T as olivine-treated soil based on different percentages of olivine (i.e. 5, 10, 15 and 20%) at different curing times (i.e. 7, 14, 28 and 90 d), and C<sub>TP</sub>SO<sub>B</sub> as carbonated olivine-treated soil at different periods (i.e. 7, 48 and 168 h) and at different pressures (i.e. 100 and 200 kPa).

#### Microstructure analysis

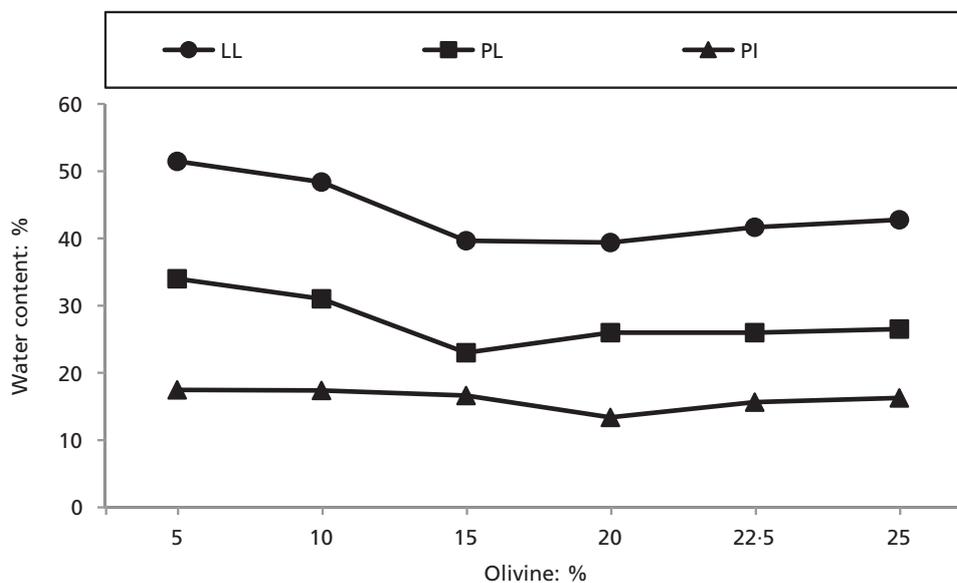
Treated and untreated specimens were examined after 90 d curing time and carbonation using a field emission scanning electron microscope (SEM; JSM 5700) coupled with an energy dispersive X-ray spectrometer. Samples were sputtered and coated with gold for 2 min, utilising Emitech K550X Sputter Coater to improve the electrical conductivity of surface and reduce charging. Crystalline phases were investigated using a Philips X-ray diffraction (XRD)

and X'Pert software over a 2-theta range of 3°–50°. Analyses were conducted on samples SO<sub>20</sub>90, C<sub>(7,200)</sub>SO<sub>20</sub>, C<sub>(48,200)</sub>SO<sub>20</sub> and C<sub>(168,200)</sub>SO<sub>20</sub>.

## Results and discussion

#### Effect of olivine on consistency limits of soil

The effect of different proportions of olivine on the LL, PL and plasticity index (PI) of soil is given in Figure 4. As shown in this figure, ball-milled olivine application generally decreased the LL, PL and PI of the soil. Decreases in LL and PL may be attributed to the fineness of ball-milled olivine particles (Figure 2), which resulted in an increased aggregate stability and lowered absorptive capacity of the mixture for water. This may be due to high specific areas of the ball-milled olivine. The main clay fraction of soil was kaolinite (Table 1). Therefore, the addition of olivine also may decrease the thickness of the double layer due to the higher valence of magnesium, which again may bring down the LL. Further increases in the olivine content (i.e. greater than 20%) were found to slightly increase LL and PL. It may possibly be due to particle flocculation and an increase in the water-holding capacity of the mixture due to a high silica content of the mixture (Table 2).



**Figure 4.** The effect of olivine dosage on the consistency limit of soil

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These results are in agreement with the findings of other studies (Barker *et al.*, 2006; Dash and Hussain, 2012; Sivapullaiah *et al.*, 2000), where the addition of lime releases  $\text{Ca}^{2+}$  ions into the pore fluid. As a result, the electrolyte concentration of the pore water influences the thickness of the diffuse double layer on the soil particles, resulting in a lower LL (Dash and Hussain, 2012; Liu and Yu, 2013; Sivapullaiah *et al.*, 2000). The PI reflects the range of moisture content over which the soil is susceptible to compaction by external forces. The lower the PI value, the lower the range of moisture over which the soil is susceptible to compaction. In general, PI decreased with an increase in the applied olivine; however, the lowest PI values corresponded to an application of 20% olivine to the soil. This may be explained by the fact that when olivine is dissolved in water, it dissociates into  $\text{Mg}^{+2}$  and  $\text{OH}^-$  (Consoli *et al.*, 2011; Harichane *et al.*, 2011; Vandeperre *et al.*, 2008a). In soils, this dissociation may result in an immediate ion exchange of magnesium ions for the existing cations at the negative charge sites on the clay particle surface. The ion exchange may cause aggregation of mineral particles and a reduction of PI (Dash and Hussain, 2012). Therefore, a further improvement in workability occurs. Moreover, brucite has the potential to precipitate on the external surfaces of the soil, forming a more or less complete coating around the clay particles. Some reactions may also occur between magnesium hydroxide ( $\text{Mg}(\text{OH})^+$ ) and the  $\text{Al}^{3+}$  and  $\text{Si}^{4+}$  species within the clay structure, producing Mg–aluminosilicate minerals; these minerals also precipitate on the clay surface. Coverage of the clay particles could reduce the surface properties, involving bonding of the clay particles faces by semiorganised brucite layers developed in the interlayer space; and coating, aggregation and cementation of the clay particles by the external phase of the brucite precipitate of the minerals in soil resulted in a decrease in PI (Xeidakis, 1996).

#### Effect of olivine on the soil compaction test

Compaction curves of the untreated and olivine-treated soil samples are shown in Figure 5. These were developed by plotting the dry unit weight of the compacted samples against the corresponding water content. It is apparent from the figure that increases in the olivine content increase the flatness of the compaction curve. This phenomenon may occur because the olivine takes up the spaces that would have otherwise been occupied by the water. As can be seen from the figure, olivine additions to the soil decreased the OWC and increased the MDD of the soil. However, the lowest OWC and the highest MDD were observed when 20% olivine was incorporated into the soil. The test results indicate that as the percentage of olivine increases, the MDD increases up to 20% olivine, after which MDD continues to decrease, with an increase in olivine content. Correspondingly, the OWC decreases up to 20% of olivine, after which the OWC increases with an increase in olivine content. Reduction in OWC is likely due to the lower affinity of olivine for water compared with that of soil. The increase in OWC of olivine-treated soil beyond the 20% incorporation of olivine could be a result of an increase in the water-holding capacity of the mixture due to the high silica content of the mixture and also aggregation and coagulation of clay particles.

In general, the increase in the MDD can be attributed to both the particle size and specific gravity of the binder and natural soil; due to high specific gravity of the ball-milled olivine, the olivine situated between the host soil constituents increased the MDD of the soil. Since the specific gravity of the olivine is greater than that of the untreated soil, the MDD also increases at the same compaction effort and water content. The increase in MDD is usually an indicator of improvement of soil properties (Basha *et al.*, 2005; Rahman, 1987).

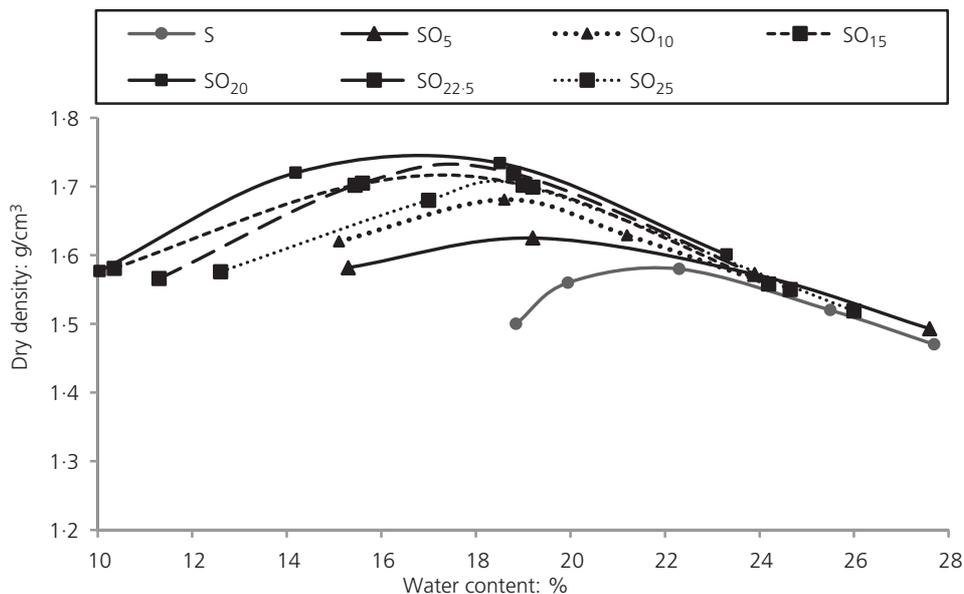


Figure 5. The effect of adding olivine on the MDD and OWC of soil

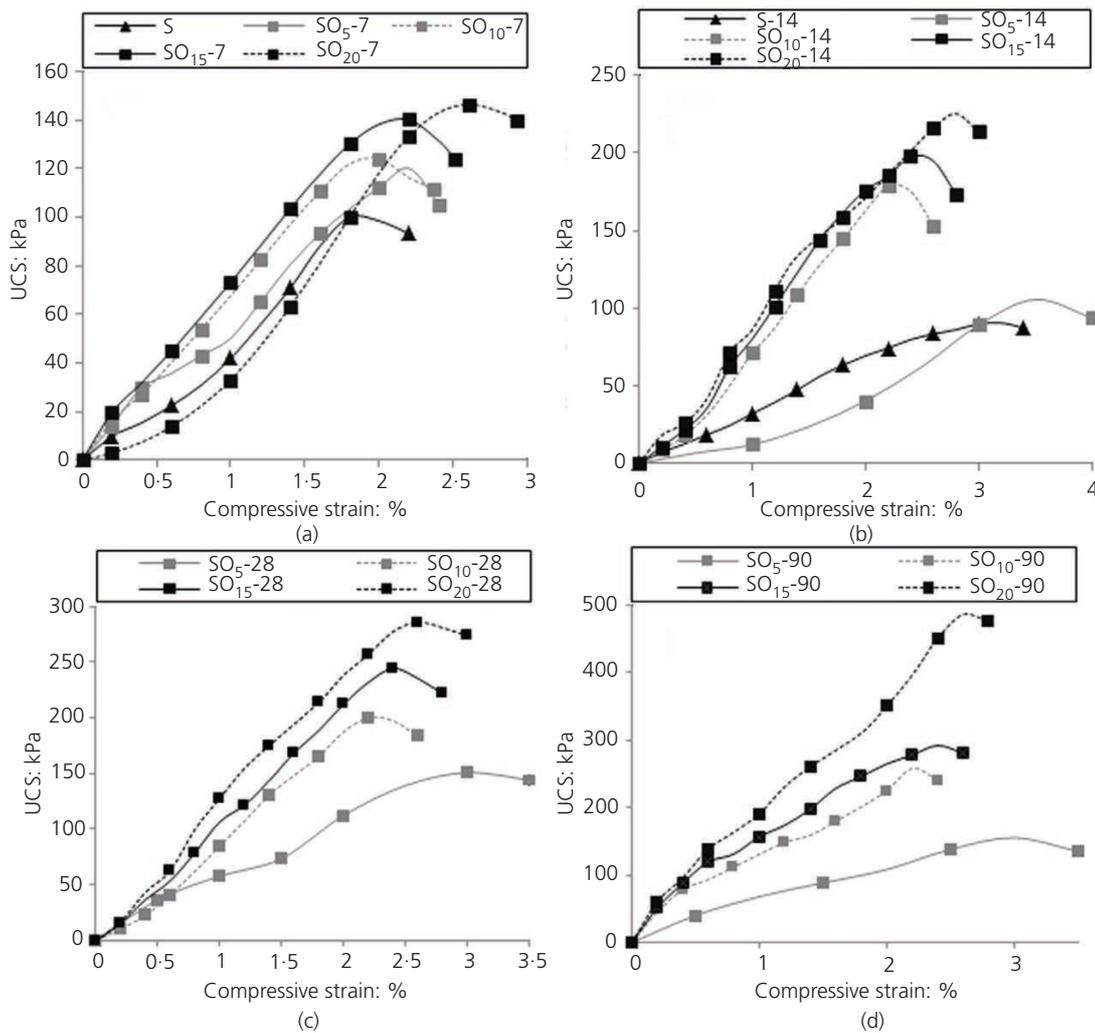
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**UCS**

**UCS of olivine-treated soil**

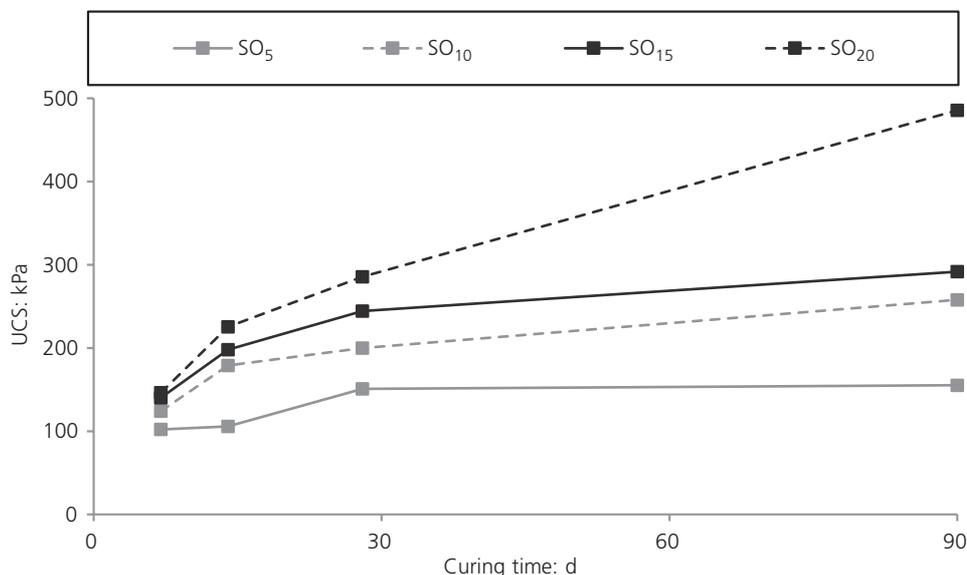
Figure 6 shows the stress–strain behaviour of untreated soil specimens and olivine-treated specimens at curing times of 7, 14, 28 and 90 d. As can be seen from the figure, untreated soil specimens (S) showed a ductile behaviour with a UCS of 100 kPa at a failure strain of 1.8%. In this study, the ball-milled olivine was introduced into the soil at concentrations of 5, 10, 15 and 20%. Since the engineering properties of soil are largely dependent upon MDD or densification (Basha *et al.*, 2005), the highest percentage of olivine incorporated was 20%, which corresponded to the maximum dry density obtained for the compaction tests (Figure 5). It was observed that addition of olivine to the soil increased the UCS at different curing times. According to the graphs, the maximum strengths of stabilised soil were observed at SO<sub>20</sub> samples where 20% olivine was

incorporated into the soil. It can be seen that the addition of 20% olivine increased the slope of the ascending branch of the stress–strain curve and increased the failure strain of the treated specimens depending on curing time. As illustrated in the figure, the area under the curves increased when soil was treated with olivine. The increased area under the curve indicates higher energy absorption per volume of treated specimens. Figure 7 shows the UCS of all specimens at their respective curing period (i.e. after 7, 14, 28 and 90 d curing time). As shown, the compressive strength increased in all samples with curing age. The increasing trend was almost identical for all treated samples with olivine from ages of 7 to 28 d; nonetheless, the strength obtained was higher in samples treated with 20% olivine from 28 to 90 d (i.e. SO<sub>20</sub>90 specimen). For sample SO<sub>20</sub> enhanced pressures of 146.41, 225.51, 285.64 and 485.54 kPa were observed at 7, 14, 28 and 90 d, respectively.



**Figure 6.** The UCS of different dosages of olivine-treated soil at different curing times; (a) 7 d, (b) 14 d, (c) 28 d, (d) 90 d. S: untreated soils specimens

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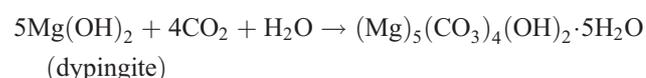
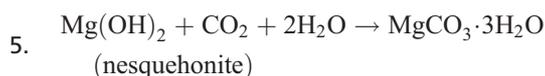
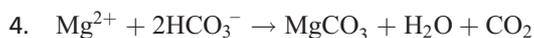
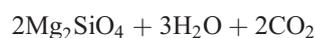
**Figure 7.** UCS of different dosages of olivine-treated soil at different curing times (7, 14, 28 and 90 d)

Higher compressive strength and energy absorption of olivine-treated samples may be attributable to hydration and pozzolanic reaction of olivine with soil. The origin of olivine is from mafic and ultramafic rocks with high magnesium oxide contents. Table 1 highlights the high concentration of natural magnesium oxide within the olivine used in this study (i.e. 48.28%). It is well-documented that magnesium oxide can be successfully utilised for soil stabilisation. According to this study, SO<sub>20</sub> specimens showed insufficient strength development from 7 to 28 d. This finding is in agreement with studies that indicate that magnesium oxide has a low reactivity at earlier ages (i.e. less than 28 d), leading to a delayed hydration. Therefore, since the hydration of magnesium oxide is not fast enough for many applications, researchers in previous studies used secondary additives such as cement and pozzolanic materials, including fly ash and ground granulated blast furnace slag, to provide an enhanced strength in soil stabilisation applications (Jegandan *et al.*, 2010; Yi *et al.*, 2013a, 2014). Interestingly, the chemical constituent of olivine categorises it within the class of defined pozzolans (see Table 2). The iron(III) oxide, aluminium oxide and silicon dioxide of the olivine amount to almost 50% by weight, indicating that olivine is a pozzolanic material (ASTM, 2014). It is likely that this will have an added benefit to the resultant strength of the soil.

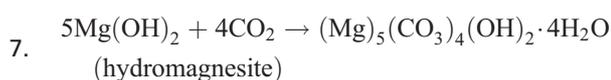
The olivine contributed to the strength development in soil, albeit rather slowly. The UCS of stabilised soil either by cement or lime usually falls between 0.1 and 5.0 MPa (Åhnberg and Johansson, 2003; Horpibulsuk *et al.*, 2010; Obuzor *et al.*, 2012), varying significantly between countries and for different applications. Therefore, in the next step, mechanical behaviour of olivine-treated soil after carbonation was investigated.

#### UCS of olivine-treated soil after carbonation

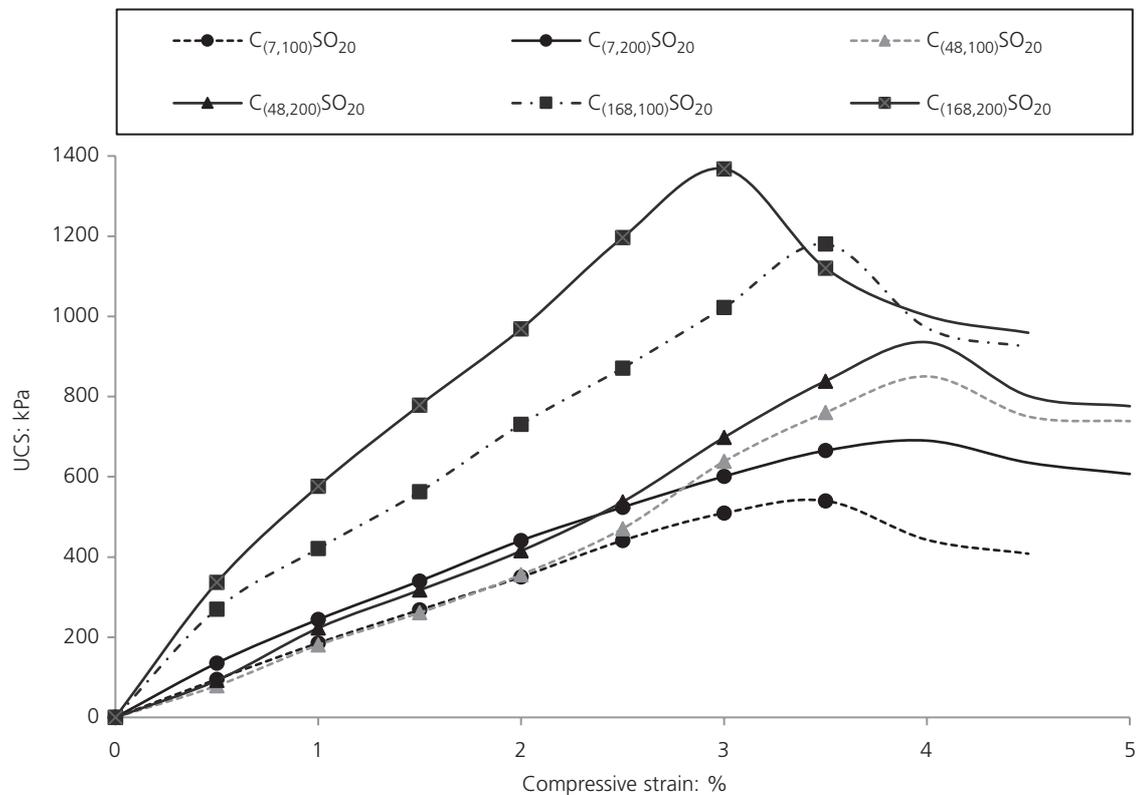
Figure 8 indicates the stress–strain behaviour of 20% olivine-treated soil following carbonation at different pressures and carbonation periods. The figure shows that carbonation of olivine-treated soil modified the sample ductility depending on carbon dioxide pressure and carbonation period. These changes may correspond to the formation of hydrated magnesium carbonates, such as nesquehointe ( $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$ ) and hydromagnesite ( $(\text{Mg})_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$ ), and dypingite ( $(\text{Mg})_5(\text{CO}_3)_4(\text{OH})_2 \cdot 5\text{H}_2\text{O}$ ) Equations 3–7)



6.



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**Figure 8.** The UCS of carbonated 20% of olivine-treated soil, at 100 and 200 kPa carbon dioxide pressure for a periods of 7, 48 and 168 h of carbonation

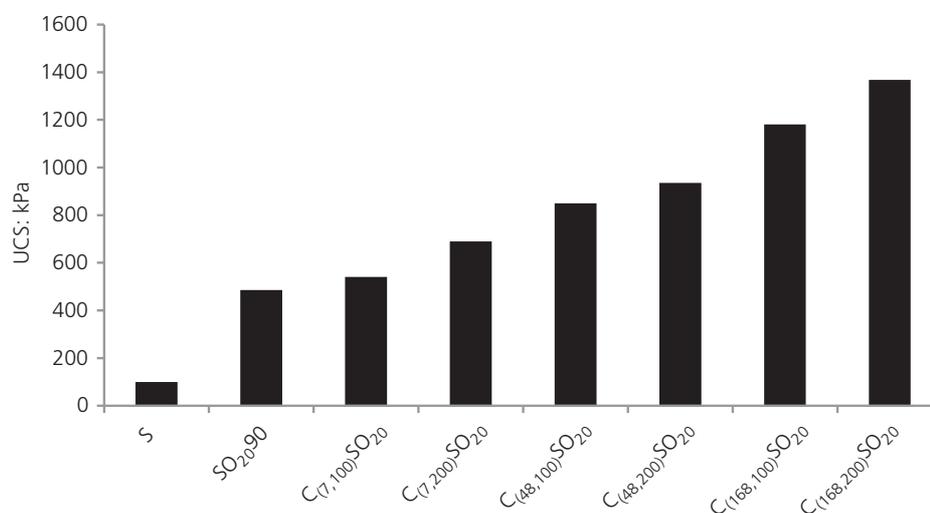
The main reaction products of carbonated magnesia responsible for the strength development are nesquehointe ( $MgCO_3 \cdot 3H_2O$ ), hydromagnesite ( $(Mg)_5(CO_3)_4(OH)_2 \cdot 4H_2O$ ), and dypingite ( $(Mg)_5(CO_3)_4(OH)_2 \cdot 5H_2O$ ). According to Yi *et al.* (2013a), the carbonation of magnesium oxide consumes water as a result of the effect of suction contributing to the strength development. Consequently, the study shows the hydration of magnesium oxide and carbonation of brucite are expansive reactions and the solid volume of magnesium oxide increases during the carbonation; the hydration of magnesium oxide significantly fills available pores. These hydrated magnesium carbonates also form well-ramified networks of massive crystals with a very effective binding ability, resulting in substantial and rapid strength increase.

In light of various recent investigations, it is clear that there is great interest in the use of magnesium oxide for soil stabilisation (Jegandan *et al.*, 2010; Yi *et al.*, 2013a, 2014). An important benefit of utilising magnesium-based compounds from olivine, and derived from the reaction of olivine with soil and carbon dioxide, is that negligible initial energy input is required. In comparison, the magnesium oxide used in previous studies is usually formed by heating magnesium carbonate to release carbon dioxide (decarboxylation), thus consuming substantial

amounts of energy, which offset environmental benefits. The associated release of carbon dioxide into the atmosphere has raised some environmental concerns due to its activity as a greenhouse gas, while the high magnesium oxide content is naturally available in olivine, making it a suitable product as a soil stabiliser.

As can be seen from the figure, olivine modified the behaviour of the soil to that of a more ductile material. The higher slope of the ascending branch in the stress–strain curve of the carbonated specimens indicates an increased ductility of the soil. Moreover, the increased area under the curve indicates higher energy absorption per volume of carbonated specimens. The figure shows that the rate of strength development is proportional to the carbon dioxide pressure and carbonation period. At the same period of carbonation, a rapid strength increase can be seen, when the carbon dioxide pressure increased from 100 to 200 kPa. According to previous research, the degree of strength growth with increasing carbon dioxide pressure depends on the magnesium content of the blend. However, increasing carbon dioxide pressure increased the pore solution acidity, resulting in a greater amount of  $Mg^{2+}$  ions being available for carbonation reactions (De Silva *et al.*, 2009; Mo and Panesar, 2012).

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**Figure 9.** The comparison of UCS of soil, 20% olivine-treated soil and 20% carbonated olivine-treated soil at different periods and pressures

Figure 9 shows that a significant contribution of the carbonation to strength is observed where the unconfined compressive stress increased up to approximately three times that compared with the corresponding 90 d ambient cured olivine-stabilised soils (i.e. 1.38 MPa). Higher compressive strength and energy absorption of carbonated specimens may be attributed to less favourable conditions for olivine to carbonate, forming one or more magnesium carbonate phases that are responsible for soil strength. According to previous studies, the main magnesium carbonate phases that can exist in an MgO–CO<sub>2</sub>–H<sub>2</sub>O system are magnesite, nesquehonite and hydromagnesite. The stability of these carbonates depends on the carbon dioxide pressure and water vapor pressure of the environment. Nesquehonite was found to be stable at higher carbon dioxide pressures (De Silva *et al.*, 2009). The results showed that adequately carbonated olivine-treated soils could, in a few hours, reach a sufficient strength range for ground improvement applications, thus, the study confirms that olivine is promising as a sustainable stabilisation binder through the sequestration of carbon dioxide. It should be noted that carbonation of olivine-treated soil in a real field case may be achieved by resorting to two possible approaches, including a mass carbonation stabilisation method or a deep mixing carbonation method (Cai *et al.*, 2015; Yi *et al.*, 2013b).

#### Microstructure analysis

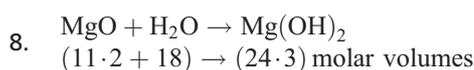
In order to attain an enhanced knowledge on the physical properties and chemical composition of olivine-treated soil and also carbonated olivine-treated soil, SEM and XRD have been undertaken. According to the UCS test results, 20% is the most effective percentage of olivine for soil stabilisation before and after carbonation. SEM and XRD analysis was therefore performed on specimens of this specific percentage.

#### SEM

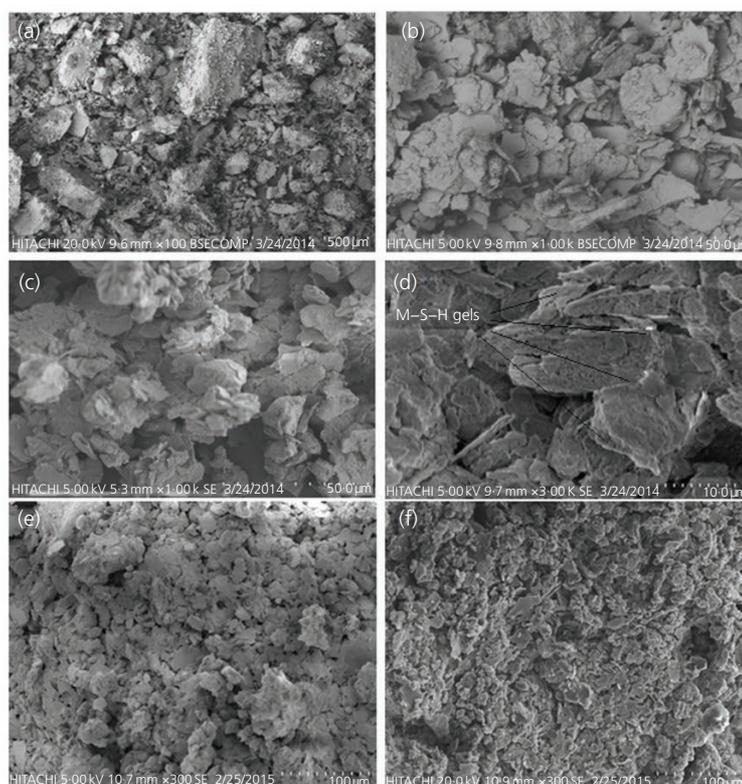
A SEM image of the ground olivine, as received, is shown in Figure 10(a). The image was taken at a relatively low magnification with a scale bar of 500 μm (0.5 mm). The particles shown correspond to the lower sieve aperture sizes as presented in Figure 2. Particles are rounded in nature and have a low aspect ratio. In comparison, the soil particles are shown in Figure 10(b) at a higher magnification with a scale bar of 50 μm. The overall particles sizes shown are smaller than those visible in the olivine. A number of the particles are shown ‘edge on’, which reveals their ‘plate-like’ high aspect ratio morphology. These characteristics are consistent with the clay phases typically found in soil.

Figures 10(c–d) show SEM images of solid treated with 20% olivine following a 90 d curing time at magnifications corresponding to 50 and 10 μm scale bars. Improved structures with higher contrast due to charging are visible when a direct comparison is made between images (Figure 10(b)) of olivine, and (Figure 10(d)) olivine-treated soil, which have the same magnifications. This could be attributed to magnesium hydroxide deposits on the surface. Figure 10(d) shows particles with micro porosity which is believed to be magnesium–silicate–hydrate (M–S–H) phases.

As shown in Figure 10(c–d), microstructures of the stabilised soil were denser than the untreated soil. This could be due to the volume change associated with the hydration of magnesia. The volume changes associated with this reaction are illustrated by Equation 8 below, where brucite is formed (Harrison, 2008; Vandeperre *et al.*, 2008b; Yi *et al.*, 2013a; Zhu *et al.*, 2013)



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**Figure 10.** The SEM of (a) olivine, (b) soil, (c and d)  $SO_{2090}$  with low and high magnification, (e)  $C_{(168,100)}SO_{20}$  (f)  $C_{(168,200)}SO_{20}$

Figure 10(e)–10(f) shows the SEM analysis of carbonated olivine-treated soil at carbon dioxide pressures of 100 and 200 kPa after a carbonation period of 168 h. In the case of the carbonated sample, Figure 10(f), the soil structure is noticeably denser when compared with the uncarbonated olivine-treated soils shown in Figure 10(c) and 10(e). The reduction in discontinuities on the surface structure of carbonated soil is believed to be a result of the olivine that was able to carbonate at the high carbon dioxide pressure of 200 kPa. According to literature, the decrease in total pore volume and increase in apparent density due to the formation of carbonates may also have contributed to the performance of the microstructural network (Mo and Panesar, 2012). This is likely a result of the extensive carbonation observed in the olivine-treated soil treated at a carbon dioxide pressure of 200 kPa.

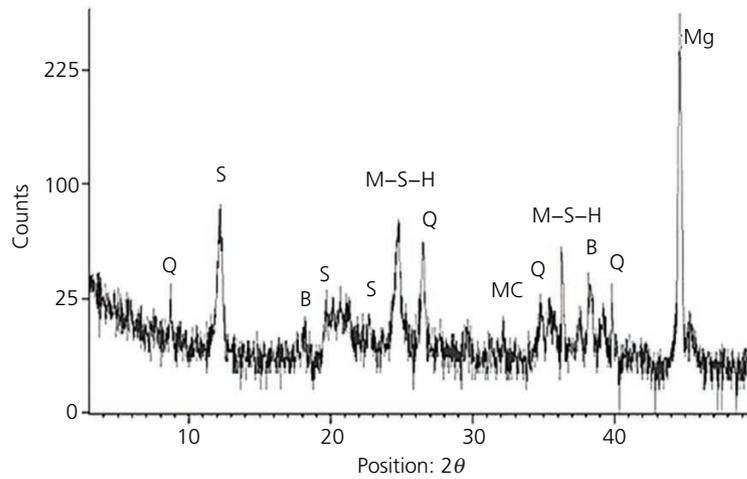
#### X-ray diffraction (XRD)

Figure 11 shows the X-ray diffraction patterns of soil treated with 20% olivine after 90 d curing time ( $SO_{2090}$ ). As shown in this figure, peaks corresponding to brucite at  $2\theta$  values between 18 to 20° and 38 to 40°, quartz (silicon dioxide) at  $2\theta$  values of 8.5°, between 26 to 28° and 39.8°, and serpentine at  $2\theta$  values of 11 to 13°, 19 to 20° and 22 to 23° were observed. The peaks were identified with reference to previous studies (Temujin *et al.*, 1998; Yi *et al.*, 2013a; Zhu *et al.*, 2013). A high intensity peak corresponding to magnesium oxide was observed

at a  $2\theta$  value of 45° (Yi *et al.*, 2013b). Also, as shown in Figure 11, the changes in soil structures are not only due to the addition of olivine to the soil in the presence of water but also as a result of brucite and serpentine production as described in Equation 2. From the literature, brucite was the major hydration product when magnesium oxide hydrates alone or in the presence of fumed silica (Vandeperre *et al.*, 2008a). In addition, the formation of M–S–H is in principle possible due to the silica present. In this regard, M–S–H gels are known to form in aqueous solutions containing  $Mg^{2+}$  and  $SiO_3^{2-}$  ions (Temujin *et al.*, 1998). Also, based on several studies, the two major broad peaks evident at  $2\theta$  values between 25 to 30° and 35 to 39° were assigned to M–S–H gel (d’Azevedo *et al.*, 2006; Brew and Glasser, 2005; Chiang *et al.*, 2014).

Figure 12 shows the XRD patterns of olivine-treated soil carbonated (200 kPa) over different periods. According to this figure, the early disappearance of the magnesium oxide peak, at the  $2\theta$  value of 45, together with weak peaks corresponding to brucite, at  $2\theta$  values of 18 to 20° and 37 to 38°, suggests the rapid carbonation of the magnesium oxide, resulting in the formation of nesquehonite, identified from peaks at 14, 25, 29, 35 and 47° and dypingite hydromagnesite, identified from peaks at 16, 21, 43°. This is associated with expansion attributed to the filling of voids in the soil structure and in agreement with

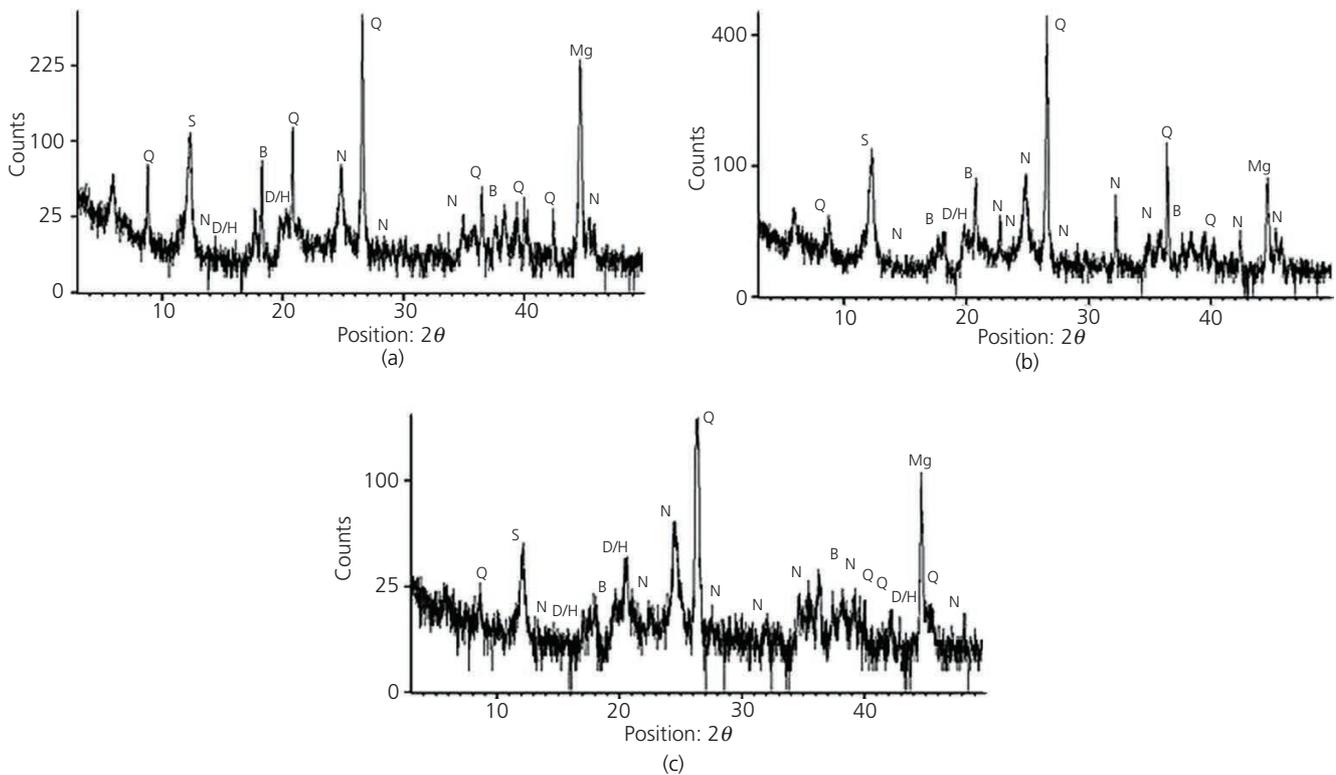
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**Figure 11.** The XRD pattern of  $SO_{20}$  after 90 days curing time. B: brucite; Mg: magnesium oxide; MC: magnesium carbonate; M-S-H: magnesium silicate hydrate; Q: quartz, S: serpentine

previous studies that used carbonating magnesium oxide for soil stabilisation (Temuujin *et al.*, 1998; Yi *et al.*, 2013a, 2013c). Also, according to Equation 1 during carbonation of olivine silicon dioxide was formed in addition to magnesium carbonate.

In addition, the XRD patterns also indicated the presence of quartz. According to the literature, quartz plays an important role as a pozzolanic material in soil stabilisation (Andersson *et al.*, 2002; Walker and Pavia, 2010).



**Figure 12.** XRD pattern of (a)  $C_{(7,200)}SO_{20}$ , (b)  $C_{(48,200)}SO_{20}$ , (c)  $C_{(168,200)}SO_{20}$ . B: brucite; D/H: dypingite–hydromagnesite; Mg: magnesium oxide; N: nesquehonite; Q: quartz; S: serpentine

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## Conclusions

This study demonstrates the utilisation of olivine as a soil stabiliser and highlights increases in strength enabled through sequestration of carbon dioxide, which was identified as an important environmental benefit. The following concluding remarks can be drawn from this study.

- PI decreased with increasing olivine content; however, the lowest PI was obtained from soil that contained 20% olivine.
- Incorporating olivine from 5% to 20% into soil caused an increase in the maximum dry density and a decrease in optimum moisture content and PI.
- Furthermore, the study showed that utilisation of 20% olivine enhanced the UCS of soil; however, the highest strength was achieved when 20% olivine and soil were carbonated at a carbon dioxide pressure of 200 kPa for a curing time of 168 h.
- The SEM microstructures of the stabilised soil verified that the soil matrix was relatively dense with no greater discontinuity than that of untreated soil.
- X-ray diffraction analyses identified as the magnesium-based phases dypingite, hydromagnesite and nesquehonite as being responsible for strength development following carbonation.

## Acknowledgement

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