Remediation of Lead-Contaminated Sand through Cerussite Formation and its Effects on Sand Shear Strength Properties

Hamed Abdeh Keykha [1], Maria Mavroulidou* [1], Jonathan Bush[1], Hadi Mohamadzadeh Romiani[2]

[1] College of Technology and the Environment, London South Bank University, UK
[2] Dept. of Civil Engineering, Imam Khomeini International University-Buein Zahra, Qazvin, Iran

*corresponding author: Maria Mavroulidou
e-mail: mavroum@lsbu.ac.uk

Abstract This study addresses the effect of CO_2 syntesised carbonate minerals use in situ to remediate lead (Pb)-contaminated soil and their effect on the soil mechanical properties. A spiked sandy soil sample contaminated with 3,000 ppm of lead nitrate ($Pb(NO_3)_2$) was subjected to injection of hydroxide (NaOH) solution and CO_2 gas, which resulted in the formation of lead carbonate ($PbCO_3$) minerals, identified as cerussite by scanning electron microscope (SEM) and X-ray diffraction (XRD) investigations. After 8 hours, more than 99% of the lead was converted to lead carbonate and successfully separated from the soil, showing the high promise of using the proposed in situ synthesis method for lead-contaminated soil remediation. On the other hand, direct shear tests of the soil showed some small reduction in the peak angle of friction at 15% cerussite contents, presumably due to the increased fines content; however the peak angle of friction was still high. This finding is important as depending on the soil use after decontamination, good mechanical properties of the soil may be a primary requirement.

Keywords: Lead contaminated soil, carbonate mineral synthesis, CO_2 sequestration, cerussite, shear strength.

1. Introduction

Soil contamination by lead (Pb) due to industrial activities is an environmental liability worldwide, as Pb is known to have adverse effects on ecosystems and human health. It has therefore been extensively investigated, and a wide range of remediation techniques have been developed to mitigate it. Many of these rely on the mechanism of lead immobilisation, through the use of reagents added to the lead-contaminated soil to prevent Pb mobility and reduce its solubility. Such reagents include iron or phosphate compounds, aluminum oxides, natural polymers (e.g., cellulose, lignin), and natural mineral compounds (i.e., zeolite tuff) amongst others (Keykha et al, 2022). Recently, studies showed that elevated carbon dioxide (CO_2), in plant rhizosphere which is leading to an increase in organic compounds, microbial populations, and enzyme activity, could significantly enhance the removal ratio of cadmium (Cd) and Pb (Huang et al, 2017). However, this indirect method of heavy-metal removal by supplying CO_2 in the soil rhizosphere is time-consuming. Following Keykha et al (2022) this paper presents an alternative approach using CO_2 to immobilise Pb in soil, with the dual potential advantage of sequestering waste CO_2 while remediating soil. The paper verifies the previous findings, using different Pb concentrations and, going beyond the previous study, it assesses the effect of the treatment on the mechanical soil properties, which is often overlooked in the soil remediation literature, yet is of primary importance for the use of the remediated land in construction projects.

2. Materials and Methods

A clean dry silica Firoozkuh sand of the particle size distribution curve is shown in Figure 1 was placed in a plexiglass box of 300 mm × 30mm × 20mm (Fig 2) and injected with 3,000 ppm lead nitrate ($Pb(NO_3)_2$). The Pb content of the soil was analysed using flame atomic absorption spectrometry.

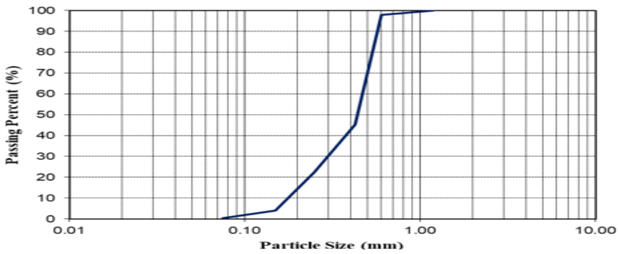

Figure 1. Particle size distribution of sand used in this study

To remediate the Pb-contaminated soil, 2M sodium hydroxide (NaOH) solution (2 M) was injected into the soil, leading to the formation of sodium tetrahydroxyplumbate (II) ($Na_2[Pb(OH)_4]$). Next, CO_2 gas was introduced into the contaminated soil from the top tube into the soil at atmospheric pressure, to induce a reaction resulting into the formation of lead carbonate ($PbCO_3$) minerals (see Equations 1-4).

$$Pb(NO_3)_{2(aq)}+2NaOH_{(aq)}\rightarrow Pb(OH)_{2(s)}+2NaNO_{3\,(aq)} \quad \text{(Eq 1)}$$

$$Pb(OH)_{2(aq)}+2NaOH_{(aq)}\rightarrow Na_2[Pb(OH)_4]_{(aq)} \quad \text{(Eq 2)}$$

$$2Na_2[Pb(OH)_4]_{(aq)}+CO_2\rightarrow 2Na[PbHCO_3]_{(aq)} +2H_2O + 2NaOH \quad \text{(Eq 3)}$$

$2Na[PbHCO_3]_{(aq)} + CO_2 \rightarrow 2PbCO_{3(s)} + 2NaOH$ (Eq 4)

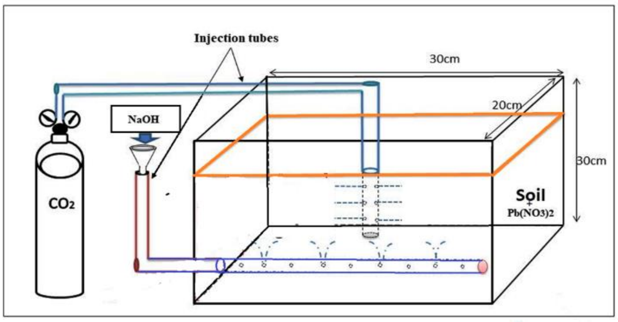

Figure 2. Experimental set up used in this study

After injection, scanning electron microscope (SEM) and X-ray diffraction (XRD) tests were performed to show respectively the morphology and mineralogy of precipitated $PbCO_3$. $PbCO_3$ concentration after treatment was determined by acid washing of soil samples collected from different distances and depths in the box. Finally, direct shear tests on dry sand before and after treatment were conducted under normal stresses of 50, 100, and 200 kPa, at samples of different cerussite content (5%,10%,15%).

3. Results and Discussion

The study showed that as $PbCO_3$ was continuously deposited between soil particles during CO_2 injection (see Fig 3), the Pb concentration in the contaminated soil decreased by >99% after 8 hours of injection. The XRF results analysis results in Figure 4 matched against the JCPDS standard identified the $PbCO_3$ mineral as cerussite with the crystal morphology shown in Fig. 5.

Shear box results analysed in terms of peak angles of friction showed that samples with a low percentage of cerussite (5%-10%) had similar peak angles of friction of ca, 40˚-41˚ (there was an apparent small increase of about 1˚ in the case of 5% cerussite, an increase which is however within the margins of experimental error and result interpretation), while ca. 1-1.5˚ reduction in peak angle of friction compared to the untreated soil was noted for 15% cerussite, possibly due to the gradually increasing fines content.

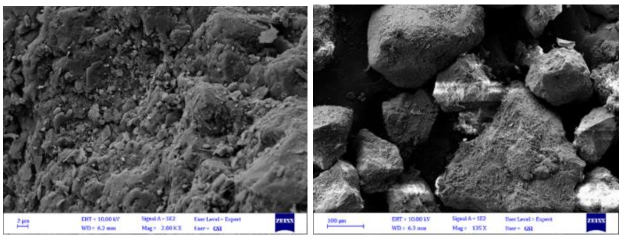

Figure 3. Precipitated crystals between soil particles

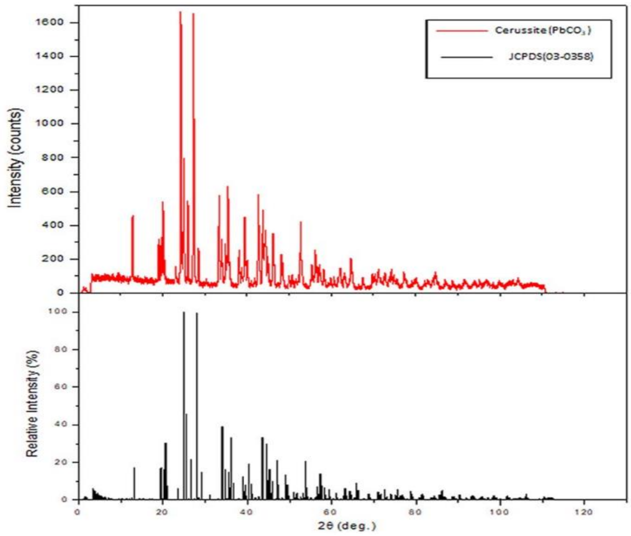

Figure 4. XRD of precipitated $PbCO_3$ crystals

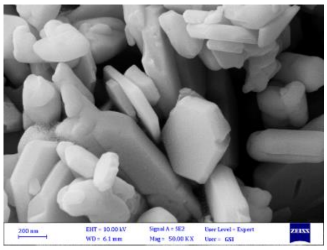

Figure 5. Morphology of precipitated $PbCO_3$ crystals

4. Conclusion

The paper confirmed the ability of a newly proposed method creating carbonate minerals in situ, as an appropriate method of lead-contaminated soil remediation through the formation of cerussite mineral and assessed the resulting effects on the soil frictional properties. The results showed that the proposed remediation method led to a conversion of > 99% of the lead into cerussite mineral after 8 hours of injection due to the absorption of lead nitrate by the supplied CO_2. Cerussite was seen to be deposited in the voids between sand particles. Cerussite, deposition caused a small fluctuation in the friction angle of the soil, however the effect was small and within the margin of experimental error. For the highest cerussite contents of 15% achieved, a reduction in the angle of friction compared to the untreated soil was also small (ca. 1-1.5˚). Therefore, the remediated soil would be adequate for construction applications, if it were to be used as a foundation soil or as a fill material.

Acknowledgement
The research on this topic currently carried out by Dr Keykha at London South Bank University is linked to the UKRI-funded project COSMIC (Grant EP/Y029607/1).

References

Huang, S., Jia X., Zhao Y., Bai B., and Chang Y. (2017). Elevated CO_2 benefits the soil microenvironment in the rhizosphere of Robinia pseudoacacia L. seedlings in Cd- and Pb-contaminated soils. *Chemosphere* 168: 606–616. https://doi.org/10.1016/j.chemosphere

Keykha H.A. et al (2022) Green Remediation for Lead-Contaminated Soil Using Carbon Dioxide Injection *Journal of Hazardous, Toxic, and Radioactive Waste* doi: 10.1061/(ASCE)HZ.2153-5515.0000712