



## Simultaneous stabilization of Pb and improvement of soil strength using nZVI

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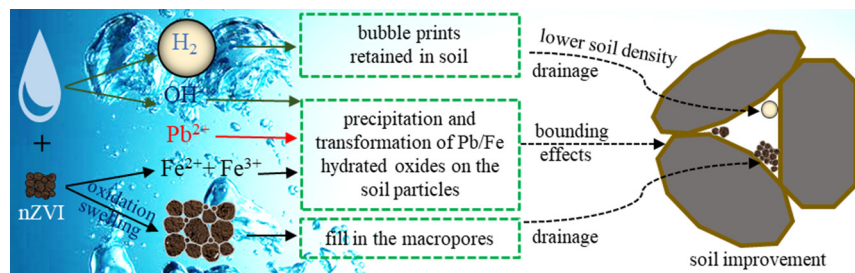
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### HIGHLIGHTS

- The feasibility of nZVI for simultaneous stabilization of Pb and soil improvement is demonstrated.
- Soil improvement is primarily attributed to the precipitation and transformation of nZVI-derived hydrated oxides.
- Microbubbles generated from the oxidation of nZVI also contributed to the soil improvement.
- This study can facilitate the application of nZVI in redevelopment of contaminated soil.

### GRAPHICAL ABSTRACT

Mechanisms for simultaneous stabilization of Pb and improvement of soil strength using NZVI includes: 1) the precipitation and transformation of Pb-/Fe-hydrated oxides on the soil particles and their induced bounding effects; 2) the increased drainage capability of soil as the occupation of NZVI aggregates (volume swelling resulted by oxidation) in the macropores space and 3) lower soil density derived by the increase of microbubbles (hydrogen generated from the redox of NZVI) retained in the soil.



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### ABSTRACT

This study demonstrates the feasibility of nanoscale Zero-Valent Iron (nZVI) for simultaneous stabilization of Pb and improvement of soil strength via batch experiments. The soil samples were prepared using slurry and pre-consolidation method at nZVI doses of 0.2%, 1%, 5%, and 10% (by dry weight). The physicochemical and geotechnical properties of Pb-contaminated soil treated by nZVI were analyzed. The results indicate that the contamination of Pb(II) resulted in a notable reduction in the undrained shear strength of soil from 16.85 kPa to 7.25 kPa. As expected, the Pb in exchangeable and carbonate-bound fractions decreased significantly with the increasing doses of nZVI. Meanwhile, the undrained shear strength of Pb-contaminated soil enhanced substantially as the increase of nZVI, from 25.83 kPa (0.2% nZVI treatment) to 69.33 kPa (10% nZVI treatment). An abundance of bubbles, generated from the oxidation of nZVI, was recorded. The mechanisms for simultaneous stabilization of Pb and soil improvement primarily include: 1) the precipitation and transformation of Pb-/Fe-hydrated oxides on the soil particles and their induced bounding effects; 2) the increased drainage capability of soil as the occupation of nZVI aggregates and bubbles in the macropores space and 3) the lower soil density derived from the increase in microbubbles retained in the soil. This study is provided to facilitate the application of nZVI in the redevelopment of contaminated soil.

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## 1. Introduction

Nanoscale Zero-Valent Iron (nZVI) is a well-established nanoparticle that has been used widely in the remediation of contaminated soil and groundwater (Bartke et al., 2018). The purpose of using nZVI is to remove or to stabilize the pollutants so that to reduce the potential threats caused by the pollutants to the ecology (Chen et al., 2017). Considerable endeavors have been made to improve the efficiency of nZVI in field application, such as degradation of various pollutants, mobility in subsurface, and revealing the fate and impacts of nZVI on ecology (Huang et al., 2016; Vilardi et al., 2018a).

Nevertheless, civil engineers consider both the pollutants and the injected nZVI as non-native components of soil. Their concerns stem mainly from the geotechnical properties of remedied contaminated soil because these properties are important in the follow-up construction on the contaminated site or the reuse of contaminated soil, such as backfill at another site. The introduction of nZVI and its derived removal of contaminants in soil could potentially disturb the microstructures of soil and lead to alteration of its geotechnical properties. This means that the application of nZVI in remediation of contaminated site may pose potential threat to serviceability, and reliability of a facility. Just like the ecological and human health risk posed by contamination, the geoenvironmental risk may break out at some point in the future, considering the high reactivity of nZVI and its long-term effect on the physicochemical properties of soil (Clayton, 2001). Thus, it is significant to investigate the behavior and mechanism of nZVI treatment in changing of geotechnical properties of soil. However, few studies have thoroughly addressed the effects of nZVI in this regard.

Most authorities accept that chemical contaminants are capable of changing properties of soil, either by directly changing the chemical or physical state of the soil, or through indirect changes, such as influencing biological activity and groundwater, which in return, modifies its properties (Ma et al., 2017; Xu et al., 2018). Rogers et al. (2004) found that the contamination of  $Pb^{2+}$  and  $Fe^{3+}$  had a significant effect on the initiation and development of the lime-clay reactions during both short-term modification and longer-term solidification. Dror et al. (2015) summarized that engineered nanomaterials have the potential to inflict irreversible changes on the physical and chemical properties of pristine soils. Gil-Diaz et al. (2014) concluded that an immobilization of  $Pb^{2+}$  and  $Zn^{2+}$  by nZVI posed no negative effects for physicochemical and biological properties of soil. However, their researches did not reveal the changes in its geotechnical properties associated with their physicochemical counterpart, such as the shear strength of the soil which is the most important parameter in the design of civil engineering projects, neither did it reveal the mechanism of the effects. The reader should note that the changes in physicochemical and biological properties of soil do not always result in significant changes in its geotechnical properties. For instance, Cuisinier et al. (2011) reported that no evidence showed any impact of chloride on the geotechnical behavior of their tested soils. Nasehi et al. (2016) performed an investigation of the effects of nZVI and nanoscale hydrated lime on the geotechnical characteristics of a diesel-contaminated soil. They suggested that the introduction of nZVI to the diesel-contaminated soil led to an increase in Unconfined Compressive Strength (UCS), maximum dry density, optimum water content, and shear strength parameters. Nevertheless, the mechanism of nZVI in soil improvement remains ambiguous given that oxidation, adsorption, and precipitation may be involved in the removal of diverse contaminants.

This study is an investigation of the stabilization of lead by nZVI in a clayey soil and its effects on the geotechnical properties of soil. Particularly, lead (Pb), a heavy metal of well-known toxicity, prevalence and responsible for liver and kidney damage, anemia, infertility, and mental retardation among other consequences, is employed as an artificial contaminant (Huang et al., 2018). Different doses of nZVI were added to the Pb-contaminated soil. The mobilization effect on lead ions, physicochemical characterization, and shear strength of the soil were examined

to analyze the effects of nZVI on the physicochemical and geotechnical properties of the Pb-contaminated earth. The results of this study can facilitate the application of nZVI in the remediation of contaminated sites where remedied soil will be used as backfill, such as remediation of brownfield and dredged sediment.

## 2. Methodology

### 2.1. Soil preparation

The researcher collected the soil samples from a construction site in Arealia Preta, Macau (N22°12'35.84", E113°33'27.96"). According to the Unified Soil Classification System (USCS, ASTM D2487-17, 2017), the Atterberg limits of the preconsolidated soil were found to be 61, 32, and 29 for the liquid limit, plastic limit, and plasticity index respectively. The particle size distribution, when analyzed by the Malvern Particle Size Analyzer indicated that the D60, D30, and D10 values were 8.5, 3.08, and 1.01 mm respectively. The soil was classified as MH: silt with high plasticity.

The slurry and pre-consolidation method was employed to prepare the soil samples (Choudhary et al., 2016). Firstly, the soil was first cut into small chunks and immersed in distilled water for two days, after which the soil fractions were passed through a No. 16 sieve (1 mm opening) to achieve a soil slurry. The water content of the obtained slurry was increased up to twice of the liquid limit with vivid agitation for approximately 10 min to obtain a homogeneous state. Subsequently the slurry (approximately 17 kg for each) was subjected to pre-consolidation in a container under an overburden pressure of 200 kPa (Indraratna and Redana, 1998). When the settlement of pre-consolidation was stable (usually took two weeks), soil samples with a height of 84 mm and diameter of 38 mm were trimmed.

For the preparation of Pb-contaminated soil, artificial pollutant of lead nitrate ( $Pb(NO_3)_2$ ) (AR, powder solid state) at the lead mass ratio of 400 mg/kg (dry soil mass) was added to the soil slurry (the weight of the soil slurry was approximately 120 kg). To achieve a homogeneous state, the spiked soil slurry was mechanically stirred for approximately 10 mins. Then, the spiked slurry was matured for two weeks to achieve equilibrium of adsorption and desorption prior to pre-consolidation or further treatment. As for the preparation of nZVI treated soil samples, 0.2%, 1%, 5%, and 10% (mass ratio of nZVI to the spiked dry soil) nZVI were added respectively to the aged Pb-spiked soil slurry and agitated vividly prior to pre-consolidation. The nZVI was supplied by Shanghai Xiangtian Nano Materials Co., LTD. The average diameter of nZVI used in this study was about 50 nm, with a specific area of 30 m<sup>2</sup>/g (provided by the manufacturer).

Fig. 1 summarizes the experimental procedure. For all the natural, Pb-contaminated and nZVI treated soil samples, sequential extraction procedure, shear strength tests, and characterization were carried out to investigate the mechanism of the effect of nZVI treatment on the physicochemical and geotechnical properties of the treated clayey soil.

### 2.2. Shear strength of soil

The Laboratory Vane Shear Test (Wykeham Farrance, 27-WF1730) was conducted to evaluate the undrained shear strength of the soil samples, as per the Standard Test Method for Laboratory Miniature Vane Shear Test for Saturated Fine-Grained Clayey Soil (American Society for Testing Material, ASTM D4648/D4648M-13). Particularly, two kinds of vane blade with the same width of 12.7 mm, yet the different heights of 19 mm and 12.7 mm were employed. The rotation rate of the vane during testing was maintained at approximately 6°/min.

### 2.3. Characterization and testing

The mineralogy of the soil was characterized by X-ray diffraction (XRD) on Rigaku Smartlab (anode Cu, in configuration  $\theta/2\theta$ , with an

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