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ORIGINAL PAPER

Evaluation of solidification/stabilization in arsenic-contaminated soils using lime dust and cement kiln dust

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Abstract

Contamination of soil and underground waters is a common environmental problem. Arsenic is a toxic chemical used widely in chemical industrial applications. New methods such as solidification/stabilization developed as requiring much space and propagation of materials in primary treatment methods caused interim methods for disposal of contamination to become inappropriate. The aim of this study was to investigate the use of cement kiln dust (CKD) and lime dust in solidification/stabilization process for arsenic-contaminated soils. Laboratory-prepared samples spiked with arsenic were made and treated with CKD and lime dust ranging from 15 to 30 wt% and 5–15 wt%, respectively. The effectiveness of treatment was evaluated at 28 days of curing based on the solidification/stabilization tests including unconfined compression test, toxicity characteristic leaching procedures (TCLP), and scanning electron microscopy (SEM). The results show that, though lime dust has a minor effect on samples strength, it has a significant effect on arsenic sorption. On the other hand, CKD causes immobilization of arsenic in the soil, as well as a major effect on soil strength. Based on the results, samples with 20 wt% CKD and 5 wt% lime dust have the optimized correlation between strength and acceptable level of Arsenic leakage in the TCLP test.

Keywords Soil solidification/stabilization · Arsenic contamination · Cement kiln dust · Toxicity characteristic leaching procedure

Introduction

Arsenic (As) is the 53rd most abundant element and constitutes about 1.5 ppm of the Earth's crust. Arsenic naturally occurs in the environment through weathering and volcanism in the form of many diverse minerals, usually in combination with sulfur and metals, as well as a pure elemental crystal. Arsenic primary use is in alloys of lead (for instance, in car batteries and ammunition). It has been widely used in industrial applications such as tanning and wood preservation, and for pesticides and herbicides, as well as mining and smelting (Sarkar et al. 2007; Yuan and Chiang 2007). The major sources of arsenic contamination to soils are desiccants, pesticides, and fertilizers (Smith et al. 1998). Arsenic is classified as a group A carcinogen by the Environmental Protection Agency (EPA) with a lethal dose of approximately 1–4 mg/kg for an adult (Southworth 1995). Generally, the heavy metals tend to remain more in the soil (solid environment) than water (aqueous environment), so they gradually release in water resources. Arsenic poses a threat to the ecosystem and human health when incorporated into groundwater or food chain (Tahmasebi et al. 2019). Chronic exposure to arsenic can cause skin lesions, cancer of the organs, impair nerve functions, and inflict liver and kidney damage (Tchounwou et al. 2004; Yoshida et al. 2004). Based on the leaching characteristics, arsenic is a serious risk to human health if the contaminated soil is excavated and reused on sites without suitable treatment. Accordingly, physical encapsulation should be employed in soils to diminish the routes of exposure to arsenic, while chemical destabilization should be avoided when mitigating the environmental risk (Li et al. 2017a).

The construction and infrastructure sector is becoming more complex than ever. Stakeholders have moved from only caring about cost- and time-related objectives to satisfying sustainability constraints (Piryonesi et al. 2018; Piryonesi and Tavakolan 2017; Sahoo et al. 2017). The environmental facet of sustainability has gained more traction over the past few decades especially when it comes to treating contaminants

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(Sözen et al. 2017). One of the widely used practices to help in this regard is known as solidification/stabilization.

Solidification/stabilization (S/S) is a process that entails the mixing of waste with a binder in order to diminish contaminant leachability by both chemical and physical means. It permutes the hazardous waste into an environmentally acceptable waste form suited to store in a landfill or exploited in the construction (Spence and Shi 2004). S/S has been widely used to discard and dispose low-level radioactive and hazardous wastes as well as contaminated site remediation. Stabilization is defined as techniques in which the peril capability of waste is reduced chemically converting the contaminants into less mobile, soluble or toxic forms. The inherent physical nature and treatment characteristics of the waste are not altered by stabilization necessarily. On the other hand, solidification is defined as techniques that enclose the waste and create a solid material and does not ineluctably entail a chemical interaction between the solidifying additives and the contaminants. The result of the solidification process often known as the waste form may be a granular particulate, clay-like material, monolithic block, or other physical forms typically considered solid (Zhou et al. 2006).

Stabilization and solidification processes have different goals. Stabilization tries to diminish the solubility or chemical reactivity of waste by altering its chemical state or by physical entrapment (microencapsulation). On the other hand, solidification systems try to transform the waste into an easily handled solid with lower hazards from volatilization, leaching, or spillage. These two processes are often discussed together since they have the common purpose of enhancing the containment of potential pollutants in treated wastes. Combined processes are often known as "waste fixation" or "encapsulation." According to the United States Environmental Protection Agency (USEPA), S/S is the best demonstrated available technology (BDAT) for the land disposal of most toxic elements and one of the most common treatment processes applied at Superfund sites in the USA (approximately 24% of the sites used S/S between 1982 and 2002) (Agency 1993; Singh and Pant 2006).

S/S is known as an effective treatment for immobilizing arsenic and has been often used to immobilize and restrain arsenic in solid wastes (Leist et al. 2003; Liu et al. 2018; Wang et al. 2015). Cement-based S/S has been used as one of the most effective methods to transform the toxic phases into less hazardous ones (Vinter et al. 2016). In this treatment, an interlocking framework of minerals encapsulates the arsenic present in the soil as cement hydration proceeds, and then the release of arsenic is reduced as the solidified soil has a low surface area and lower permeability (Clancy et al. 2015; Kim et al. 2016). The mobility of arsenic in the soil may be further reduced by adsorption and co-precipitation through employing immobilizing agents (Kogbara 2013). The feasibility of S/S for immobilizing arsenic-contaminated soils requires the proper selection of various binders based on the mechanisms governing arsenic leaching (Li et al. 2017a, b; Tsang et al. 2014). Researchers have evaluated the S/S process to immobilize arsenic by means of diverse S/S agents such as quicklime, cement, fly ash, hydrated lime, and cement kiln dust (Moon and Dermatas 2007; Moon et al. 2008; Tsang et al. 2014; Yoon et al. 2010).

The main objective of S/S is to develop a recipe (binder mixture ratio) that produces a sustainable and stable product posing the minimum threat to the environment (van der Sloot et al. 2007). We specifically chose cement kiln dust (CKD) as the S/S agent because of its certain specification and insignificant cost. In a previous study, the effect of CKD on S/S treatment of arsenic-contaminated soils was investigated (Moon et al. 2008). Laboratory-prepared soil (kaolinite and montmorillonite as surrogate soils) samples spiked with either 0.1 wt% As^{3+} or As^{5+} were treated with up to 25 wt% CKD for curing time of 1 and 7 days. The treatments were evaluated for their leaching behavior using the toxicity characteristic leaching procedure (TCLP). The results show that upon 25 wt% CKD treatment, the As^{5+} concentrations in the kaolinite soils and all arsenic-spiked montmorillonite soils treated with CKD failed to meet the TCLP criteria after 7 days of curing. It was demonstrated that 25 wt% CKD is not enough for the stabilization of As^{3+}, and samples need a longer curing period.

In order to improve the result of the treatment, another S/S agent is needed to cover CKDs shortcoming. Accordingly, this study aims to assess the effectiveness of Ca-containing agents, namely lime dust and cement kiln dust (CKD), as a potential stabilizing agent for immobilizing arsenic in soils using laboratory-prepared arsenic-contaminated soil samples. In this research, the main goals are to evaluate the effectiveness of S/S treatments using lime dust and CKD, to obtain optimum lime dust and CKD doses, and to determine if the minerals formed are consistent with the traditional S/S end products.

Table 1 Typical chemical composition of CKD, Portland cement, and lime

Parameter	Portland cement (%)	Lime (%)	CKD (%)
SiO_2	20.50	0.74	12.96
CaO	63.90	80.15	43.96
SO_3	3.00	–	1.62
MgO	2.10	0.37	0.81
Na_2O	< 1	0.03	0.15
K_2O	< 1	0.01	0.59
Al_2O_3	5.40	0.06	3.99
Fe_2O_3	2.60	0.15	0.29
P_2O_5	–	1.325	–

Table 2 Physicochemical properties of soil sample

Test	Value
Average moisture	4.54%
Atterberg limits	LL = 23.2, PL = 17.7, and PI = 5.5
Specific gravity	2.74
pH	7.58

Materials and methods

A binder is often used to stabilize the contaminants in the waste or contaminated sites and to remove the free liquid. Portland cement is most commonly used because of its availability and low cost. Supplementary cementing materials such as coal fly ash and ground blast furnace slag are often used to partially replace Portland cement, to improve the performance of the treated wastes and to reduce the cost of the binder. Of all the binders, cementitious materials are the most widely used for S/S. Compared with other procedures, cement-based S/S has many advantages, such as (Malviya and Chaudhary 2006; Shi and Spence 2004) relatively low costs, good long-term stability, both physically and chemically documented use, and compatibility with a variety of wastes over decades, material and technology well known, the widespread availability of the chemical ingredients, nontoxicity of the chemical ingredients, and ease of use in processing.

Cement kiln dust (CKD) is a by-product of cement manufacturing. It is a fine powdery material analogous to Portland cement collected from electrostatic precipitators during the production of cement clinker. The generation of CKD is approximately 30 million tons worldwide per year (Siddique and Rajor 2012; van Oss and Kraft 2014). The cost related to CKD disposal is high, and developing beneficial applications for it is interesting for the cement industry. As the price of CKD can be negligible against the cost of Portland cement depending on market conditions and regional availability (for instance, in Iran CKD has no cost and cement industry would be happy to shed it if possible), it becomes an attractive alternative to conventional pozzolans as long as CKDs efficacy in metals immobilization can be demonstrated.

CKD has a chemical composition similar to Portland cement and the major constituents are compounds of lime, iron, silica, and alumina. Table 1 gives the typical chemical composition of CKD against Portland cement and lime used in this study. The pH of CKD-water mixtures is generally around 12. It contains significant alkalis and is considered to be caustic. Trace constituents in CKD comprising specific trace metals such as cadmium, selenium, lead, and radionuclides are typically found in concentrations less than 0.05% by weight.

CKD is used for stabilization and solidification of waste since the absorptive quality of the dust and alkaline inherent nature make it a very efficacious waste treatment. Moreover, CKD is a cheap but plausible alternative to other conventional waste treatment agents such as cement and lime.

In the present study, laboratory-prepared samples were made and treated with CKD and lime dust ranging from 15 to 30 wt% and 5–15 wt%, respectively, up to 28 days in order to achieve an acceptable level of arsenic treatment as well as maximum strength. The toxicity characteristic leaching procedure for total

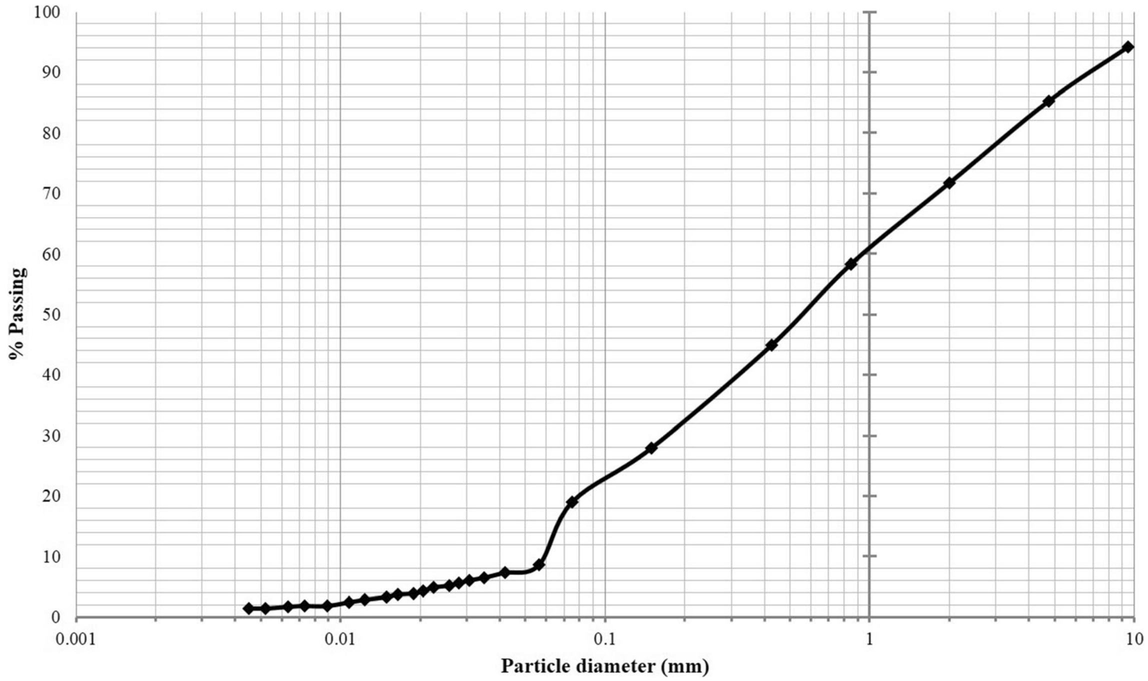


Fig. 1 Particle-size distribution curve of soil sample

Table 3 Test matrix for laboratory-prepared soil samples

Sample ID	CKD (%)	Lime dust (%)	Water content (gr)	Total dry weight (gr)
C15L5	15	5	30	240
C15L10	15	10	30	250
C15L15	15	15	30	260
C20L5	20	5	40	250
C20L10	20	10	40	260
C20L15	20	15	40	270
C25L5	25	5	50	260
C25L10	25	10	50	270
C25L15	25	15	50	280
C30L5	30	5	60	270
C30L10	30	10	60	280
C30L15	30	15	60	290
Control	–	–	20	200

arsenic was implemented to evaluate the effectiveness of that treatment from leaching perspective. On the other hand, the compressive strength of samples using uniaxial compressive test was investigated, and scanning electron microscopy (SEM) was utilized to inquire the crystalline mineral formations.

Soil samples were gathered from a nontoxic area. After the identification of sample characteristics and contaminating procedure, S/S of samples with lime dust and CKD and curing were performed. First, soil samples were contaminated with a specific amount of arsenic and then, adding a various percentage of lime dust and CKD, 12 solidified and stabilized samples and one control sample were prepared. In the end, certain tests including unconfined compressive strength, TCLP, and SEM were performed to evaluate the effect of pozzolanic materials on structure, leachability, and stabilization procedure of contaminated soil.

Classic soil tests and results

In order to determine physicochemical characteristics of the soil sample, classic laboratory soil tests were performed and the results are demonstrated in Table 2.

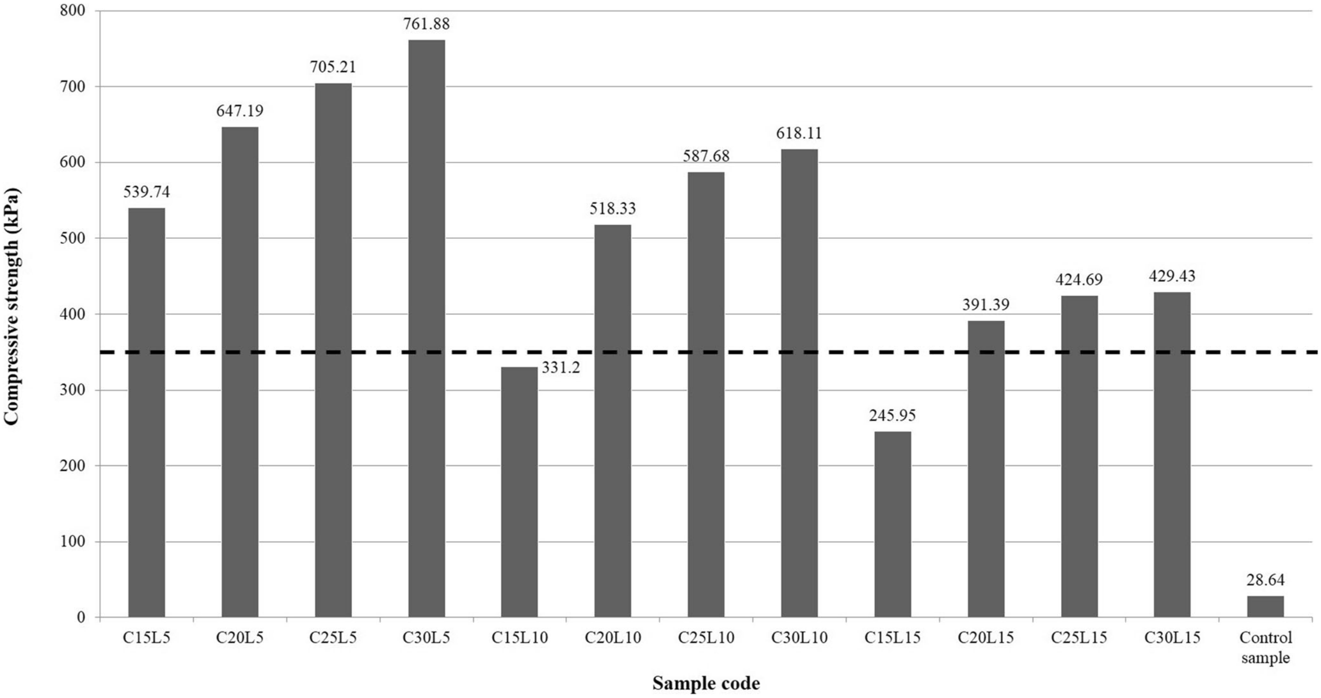


Fig. 2 The unconfined compressive strength for all samples

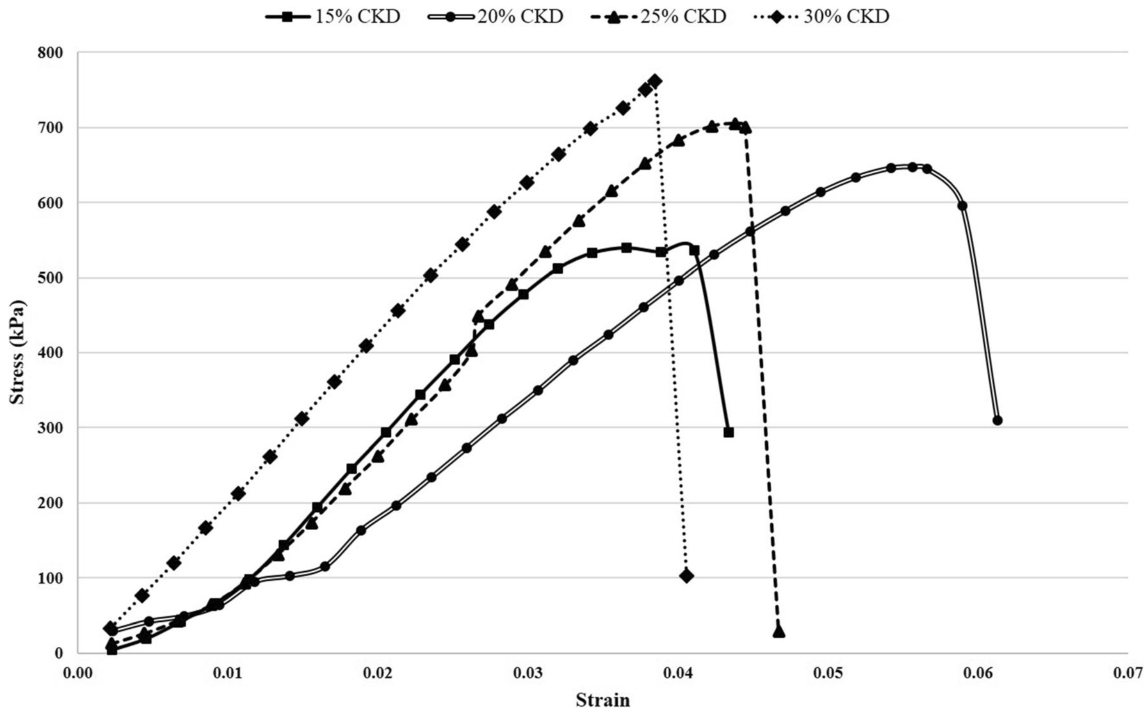


Fig. 3 Stress-strain curves of samples with a constant amount of lime dust (5%)

The particle-size distribution curve of the soil sample is depicted in Fig. 1. From this curve and Atterberg limits, based on the unified soil classification system, the soil sample is in the SC-SM (silty clayey sand) class.

Laboratory-prepared contaminated samples

Sodium arsenite ($NaAsO_2$) was employed as the As^{3+} source. In order to obtain 100 ppm arsenic concentration in soil

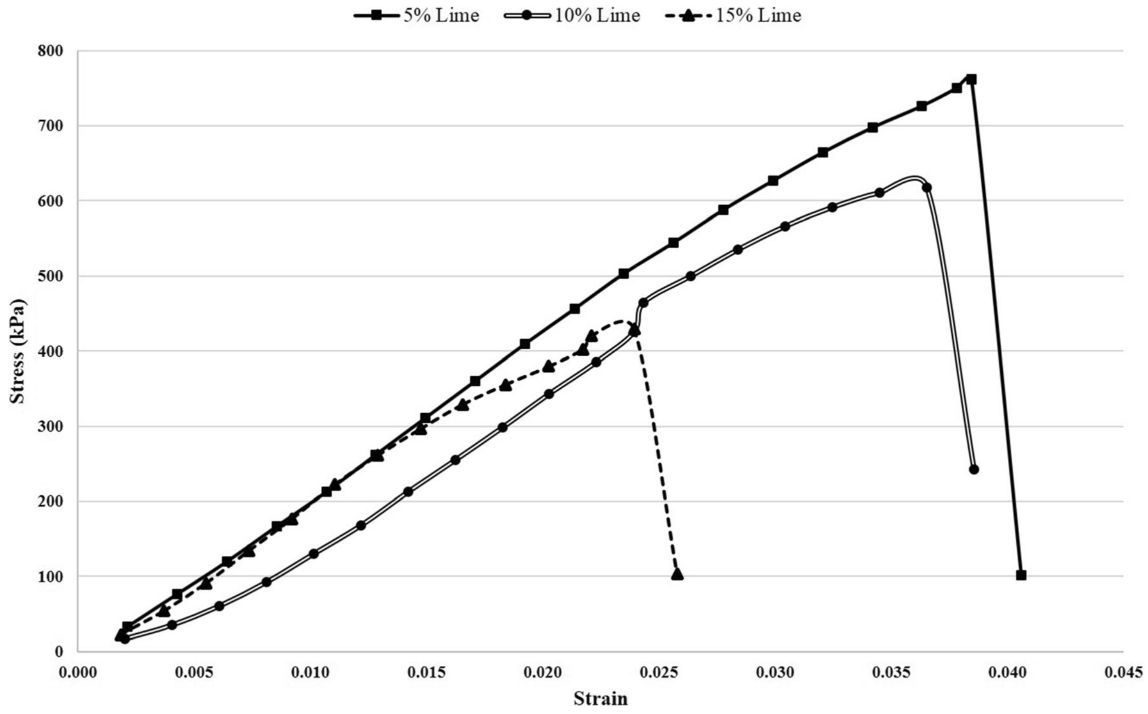


Fig. 4 Stress-strain curves of samples with a constant amount of CKD (30%)

Table 4 TCLP arsenic concentration

Sample ID	Arsenic concentration (ppm)	Arsenic concentration reduction ration (%)
C15L5	0.999	84.14
C15L10	0.186	97.04
C15L15	0.100	98.41
C20L5	0.857	86.39
C20L10	0.119	98.11
C20L15	0.0699	98.89
C25L5	0.113	98.20
C25L10	0.0537	99.14
C25L15	0.0259	99.58
C30L5	0.0554	99.12
C30L10	0.0359	99.43
C30L15	0.0222	99.64
Control	6.300	0

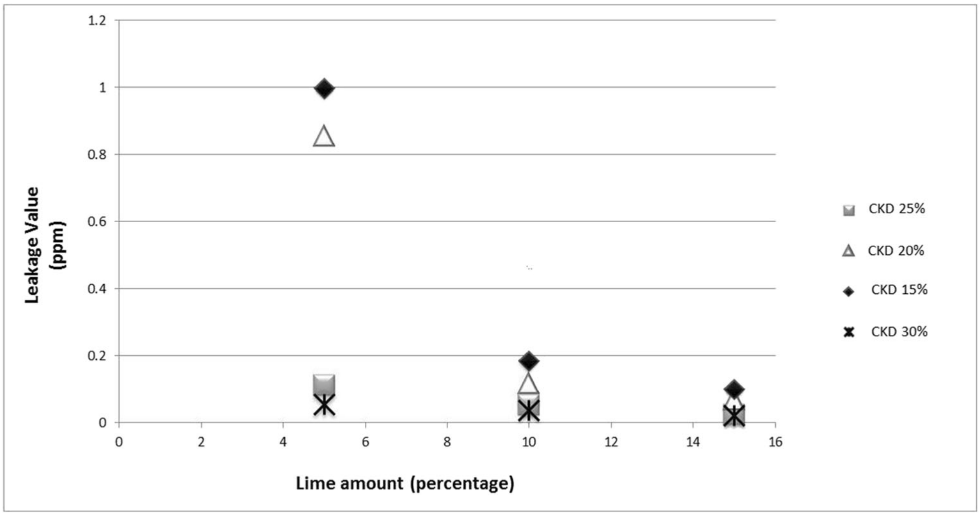
(100 mg toxic agent in 1 kg soil), 173.4 mg sodium arsenite was first dissolved in 100 cc deionized water and then arsenic-spiked soil mixture was prepared. That mixture was used to simulate representative arsenic leaching corresponding to a highly As-contaminated site. To absorb the contamination gradually and distribute uniformly, the mixture has been kept in low temperature and mixed three times a day for a week. Having been prepared, the As-spiked soil was mixed with CKD and lime dust at doses 15–30 wt% and 5–15 wt%, respectively. Two hundred grams As-spiked soil was mixed with those agents until they were visually homogeneous. A control sample without S/S agents was also prepared. The samples were mixed with water (water/CKD ratio equal to 1) and have been cured for 28 days. In order to perform compressive strength tests, samples dimensions were selected in accordance with ASTM D1633 (ASTM 2017).

The specific test matrix of the treatment for laboratory-prepared samples is presented in Table 3.

S/S tests

To evaluate the effect of the S/S process, physicochemical and mechanical properties of output materials were identified employing four test procedures. First, unconfined compressive strength tests were performed to obtain the strength of the solidified samples against mechanical stress. It is a key variable in S/S continuance and hydration reactions. This test offers fruitful information such as the capability of waste against loading, the optimized ratio of water/pozzolanic material, the optimized curing time for CKD, and strength improvement from ordinary waste to stabilized waste.

Fig. 5 Effect of lime dust on samples performance against leakage



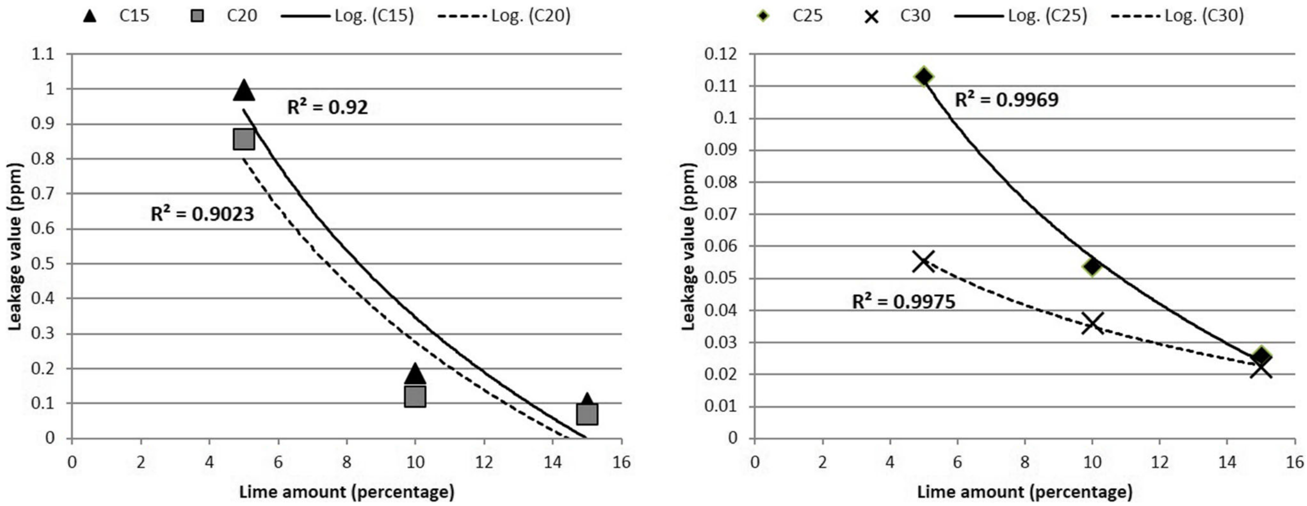


Fig. 6 Logarithmic patterns of lime dust performance

In most cases, the reduction of contamination transmission rate to the environment is the first and most important factor in choosing S/S method. When rain-induced surface water permeates through stabilized material, contamination agents are transferred from stabilized mass and transported to underground water sources. Leakage susceptibility depends on physicochemical features of stabilized material and leaking liquid. The factors that affected leakage rate in the S/S method are alkalinity of stabilized material, the surface to volume rate of mass, and porosity. In this research, toxicity characteristic leaching procedure (TCLP) was selected in accordance with EPA method 1311 (Laboratory and Information 1989).

In order to evaluate the efficiency of the S/S process in the reduction of leakage rate, the soluble arsenic concentrations were identified. For this purpose, output liquid was prepared in accordance with EPA method 7470A and injected to an emission spectrometer using an inductively coupled plasma (ICP) as the excitation source. To observe the deformed soil structure during the reaction between contaminated soil and CKD/lime dust, S/S samples and control sample were evaluated by scanning electron microscopy (SEM) analyses.

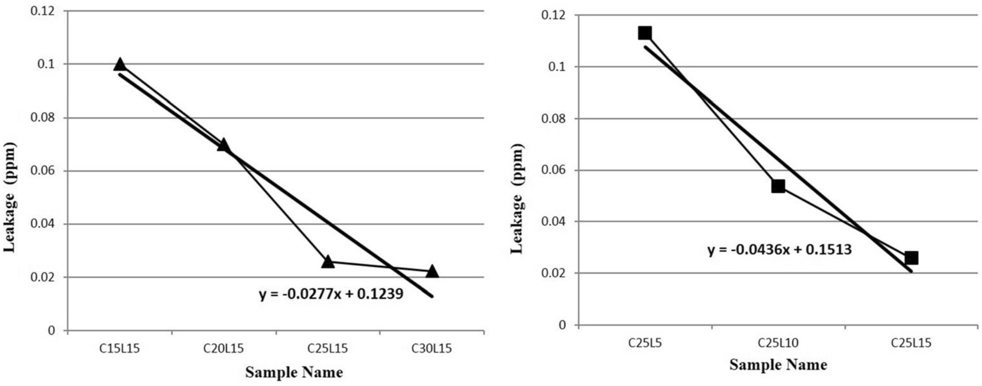
Results

Unconfined compressive strength test results

The unconfined compressive strength results obtained from 13 samples are demonstrated in Fig. 2. Environmental Protection Agency (EPA) specifies the minimum of 350 kPa for 28-days strength of solidified waste.

The compressive strength of S/S samples is far more than the control sample. This is one of the advantages of the S/S method. All of the S/S samples meet the EPA criteria except C15L10 and C15L15 samples. In contrast with the result of Shafiqu and Abass (2018) in which the increase in CKD and lime percentage and curing period has improved the unconfined compressive strength, in this research, in the samples with an equal amount of lime dust, strength increased by adding CKD. Stress-strain curves of samples with 5% lime dust are depicted in Fig. 3. However, increasing the amount of lime dust had a negative impact on strength so that in the samples with 15% lime dust, the negative effect almost neutralized the positive effect of CKD. Figure 4 illustrates the

Fig. 7 Comparison of CKD and lime dust performance against leakage



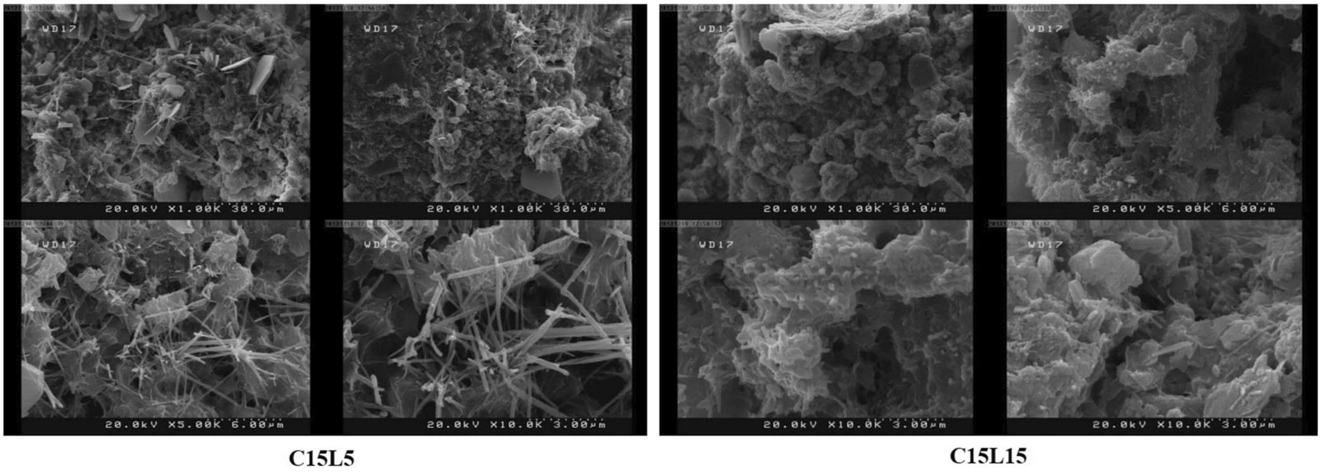


Fig. 8 Microscopic structure of samples C15L5 and C15L15

stress-strain curve of samples with 30% CKD. C30L5, the sample with the maximum amount of CKD and the minimum amount of lime dust had the maximum strength. The range of unconfined compressive strength in this research is 0.3–0.8 MPa sufficient to prevail many geotechnical problems of structures located on this material (Antemir et al. 2010).

As depicted in Fig. 3, with increasing the amount of CKD, samples showed more brittle behavior. CKD generates further adhesion resulting in more strength and brittle constitutive behavior. Conversely, based on Fig. 4, sample strength was decreased by adding lime dust, yet same as CKD, failure strain was reduced leading to brittle behavior.

TCLP results

The TCLP results obtained from inductively coupled plasma optical emission spectrometer are presented in Table 4. The control sample is considered the leakage benchmark (100% leakage).

To evaluate the efficiency of lime dust in stabilization, sample performance is demonstrated in Fig. 5.

Forming insoluble calcareous-arsenic precipitates is the main mechanism of arsenic immobilization in the presence of CaO compounds (Dutré and Vandecasteele 1998; Moon et al. 2008). Same result in this research indicates the effective contribution of lime (containing 80% CaO) in the stabilization of arsenic contamination. Figure 6 presents logarithmic patterns of arsenic leakage, with a high coefficient of correlation, stated that adding only a little CaO to arsenic-spiked soil decreases an impressive amount of leakage from the mass matrix.

Based on Table 1, lime dust has a double amount of CaO in comparison to CKD. Therefore, lime dust has a more stabilizing effect and reduces leakage more effectively. In fact, the effect of lime dust in arsenic concentration reduction is double of CKD effect, as illustrated in Fig. 7.

Among the samples, C30L15 had the least arsenic leakage and the worst result belongs to C15L5 in which S/S method

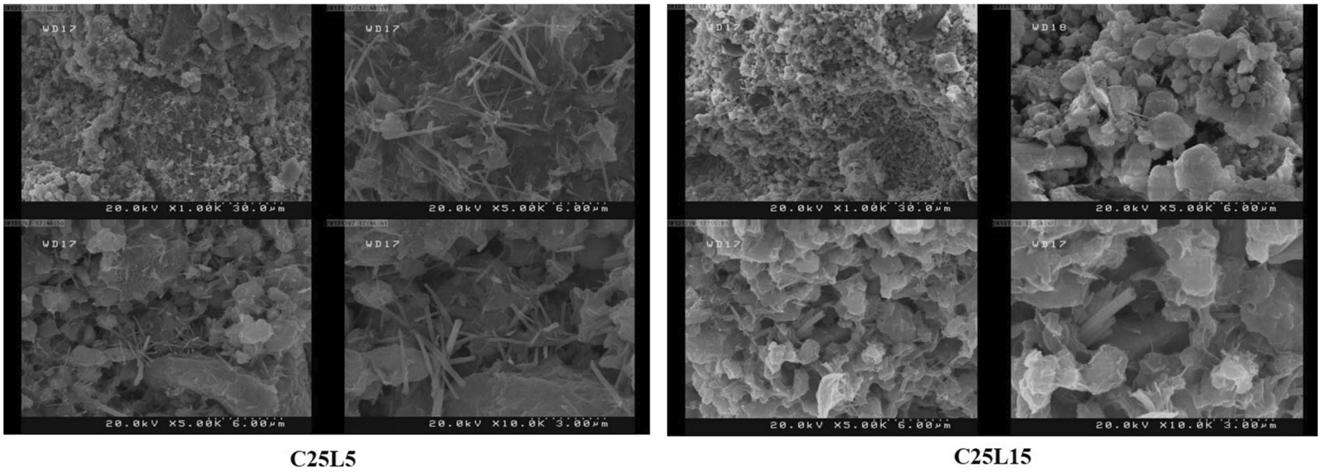


Fig. 9 Microscopic structure of samples C25L5 and C25L15

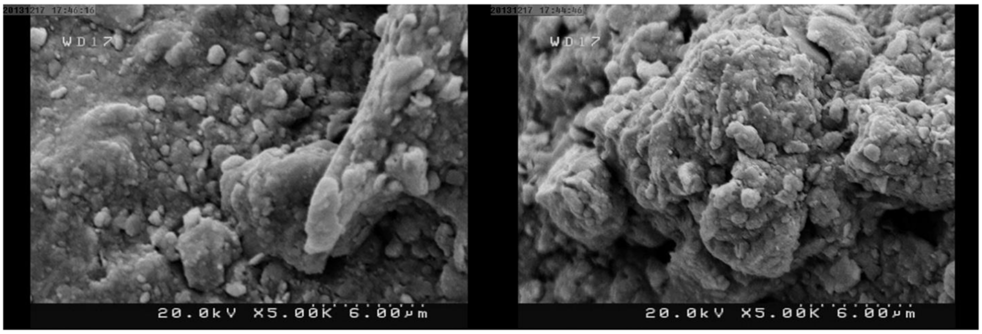


Fig. 10 Microscopic structure of control sample

prevented 84.14% of leakage. As a result, the leachable arsenic concentration decreased with increasing lime dust and CKD content. Though arsenic concentration leakage in control sample exceeded the TCLP-As regulatory limit of 5 mg/L, all treated samples satisfied that criteria well.

SEM analysis results

To evaluate the effect of lime dust and CKD, microscopic structure of five samples (including C15L5, C15L15, C25L5, C25L15, and control sample) was observed before and after the S/S procedure. Observations are depicted in Figs. 8, 9, and 10 at the scale of 30, 6, and 3 μm.

Based on Fig. 8, an increase in the lime dust content has a negative impact on the development of ettringite in samples causing strength reduction. Conversely, comparing two samples with an equal amount of lime dust but different amount of CKD, reveals that adding CKD leads to unified distribution and slaking of ettringite. Structure of sample C25L5 is denser and has less porosity against sample C15L5 causing permeability reduction and more strength. Increase of lime dust content prevented ettringite structure development in soil and somehow made it premature.

Conclusion

In this research, we used lime dust and cement kiln dust (CKD) as the solidification/stabilization (S/S) treatment agents against arsenic-contaminated soil. Laboratory-prepared samples spiked with arsenic were treated with a variety range of CKD and lime dust mixtures and cured for 28 days. The treated samples were evaluated for compressive strength and leakage behavior employing unconfined compressive strength test and toxicity characteristic leaching procedures (TCLP). Lime dust had a key role in contamination absorption while CKD was the operative of strength increment. The S/S samples had far more compressive strength against the control sample. Though lime dust had a considerable effect on reducing arsenic leakage, it had a negative impact on compressive strength. However, that negative effect would not cause unacceptable reduction if a suitable amount of CKD were used. Correlations between leakage and lime dust content, against the constant amount of CKD, with univariate regression, were studied and logarithmic patterns indicate that adding a small amount of lime dust has an impressive effect on leakage prevention. The effect of CKD on reducing leakage was half of the lime dust effect. Scanning electron microscopy (SEM) analyses show that lime dust prevents ettringite development while CKD causes development and unified distribution of that needle-shaped structure in the samples. A designed mixture of CKD and lime dust could treat arsenic-spiked soil and enhance compressive strength effectively.

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