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IN-SITU REMEDIATION OF HEAVY METAL CONTAMINATED SITES THROUGH MECHANICAL STABILIZATION USING INDUSTRIAL WASTE PRODUCTS

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Highlights

- The indiscriminate and unscientific disposal practices of solid wastes in and around the dumping sites in developing as well as developed countries are causing severe lithospheric (soil and water) pollution in these areas.
- The present study aimed to examine the efficacy of a balanced mixture of three locally available additives viz. fly ash, blast furnace slag and quick lime to immobilize the heavy metals as well as to improve the geotechnical properties of the dumping site soil.
- The concentrations of heavy metals in the leachate of the multimetal-contaminated soil were decreased by almost 100% after curing for a period of 3 and 7 days.
- Improvement of the physico chemical properties achieved in stabilized soil in comparison to the virgin contaminated soil.
- Numerical modeling was performed using PLAXIS 3D software to evaluate the safety factors as well as vertical displacement values of an embankment founded on contaminated as well as stabilized soil.

Abstract. The present study aimed to assess the stabilization performance of fly ash, blast furnace slag and quick lime for heavy metals in contaminated soil at a landfill site at Kolkata, West Bengal, India. The physical properties and strength parameters of the contaminated soil substantially increased after additives application. Moreover, the heavy metal concentrations in the leachate of the polluted soil were found almost nil after optimum blending of the additives mechanically with the soil and post-curing for 7 days. The numerical modeling studies were also carried out using PLAXIS^{TM} 3D software to ascertain the improvement of safety factor and deformation caused at the foundation level of an embankment constructed on such stabilized soil. The vertical displacement of the embankment founded on stabilized soil reduced from 194.3 to 136.3 mm and the safety factor of the embankment slope (1 V:1.5 H) increased from 2.5 to 3.2 under drained condition.

Keywords: contaminated soil, heavy metals, remediation, additives, mechanical stabilization, embankment, numerical modeling.

Introduction

The indiscriminate and unscientific disposal practices of solid wastes in and around the dumping sites in developing as well as developed countries are causing severe lithospheric (soil and water) pollution in these areas (Raja & Pal, 2019). The sub-surface migration of the pollutants (organic as well as metals) from these sites causing severe diminution of the qualities of the underground aquifers as well near-surface soils, which limits the exploitation of these sites for future infrastructural developments (Xia et al., 2018). However, restoration of these polluted sites is not only time consuming but also requires enormous capital investment and most of the cases, effective in-situ treatment technologies are lacking (Tica et al., 2011). The problems become more complex to deal with sites contaminated with heavy metals due to its non-degradability nature (Hu & Cheng, 2013) and high advective mobility in soil and water (Ahmad et al., 2012). The people residing nearby these sites suffer from colossal health issues due to prolonged exposure to these toxic contaminants (Maheshwari et al., 2015).

Though the remediation technologies have come a long way over the past decade, the technologies for the

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restoration of metals contaminated sites are still in the nascent stage in the geo-environmental engineering field (Tica et al., 2011; Xu & Lu, 2012). The various in-situ/off-situ land decontamination techniques such as soil washing, phytoremediation, vitrification, excavation and disposal, etc. are readily available, however, these are either time consuming or expansive or both (Gong et al., 2018). Solidification/Stabilization (S/S) technology has emerged as an efficient method for reclamation of metals contaminated sites because of its inexpensive, time-saving technique and most importantly, highly efficient than other conventional technologies (Du et al., 2014; Zhang et al., 2015). Solidification forms a solid matrix through the physical encapsulation of contaminants, whereas stabilization ease contaminants mobility in the process of a chemical reaction (Mulligan et al., 2001). The S/S processes involve the mechanical mixing of specific additives with the contaminated soil and thereby lead to metals precipitation in the form of hydroxyls and also lessen their mobility through cement hydration (United States Environmental Protection Agency [USEPA], 2002). Researchers studied the potential of various additives on the solidification/stabilization of metal contaminated soil. Xi et al. (2014) found that the proportional mixture of additives, namely fly ash, lime, cement in a mass ratio of 1:1:2 are highly efficient in reclaiming land which is significantly contaminated with Pb (concentration of 10000 mg/kg) through S/S techniques. Rachman et al. (2018) studied the efficacy of Portland cement in attenuating gold mine tailings enriched with Hg through mechanical mixing in the ratio of 90:10 (tailings: Portland cement) on a mass basis. They observed substantial reduction (95% and above) of the Hg concentration level in the leaching of the solidified/stabilized soil and also found an encouraging UCS value of 2570 kN/m^2 of the stabilized mass.

The present study aimed to examine the efficacy of a balanced mixture of three locally available additives viz. FA, BFS and QL to solidify/stabilize As, Cr, Cd, Pb, Zn, Hg, Cu, Ni polluted site as stated above through mechanical blending. Moreover, attempts were also made to lessen the metals leachability from the contaminated site through stabilization and to improve the strength parameters of the stabilized soil through solidification so that the reclaimed land can be used for habitation as well as infrastructural developments. Moreover, numerical model was also carried out considering a geotechnical engineering practical problem to ascertain the degree of strength improvement achieved of the solidified/stabilized multi-metal contaminated soil. So, immobilization of the heavy metals in the contaminated land as well as improvement of the geotechnical properties indicate mechanical stabilization as a good solution in contaminated soil remediation. Thus, the metal-contaminated sites can be rejuvenated for various infrastructural activities as well as waste minimization can also be achieved.

1. Material and methods

The soil samples were collected using handheld boring auger equipment from a depth of 1 m below the existing soil surface at the partially closed landfill site (22° 32" 33" N, 88° 24' 23" E) at Kolkata, West Bengal, India, as shown in Figure 1. The engineering properties of the virgin soil samples were evaluated following the procedures illustrated in Bureau of Indian Standard ([BIS] 2720) and presented in Table 1. The optimum amounts of fly ash (FA), quick lime (QL) and blast furnace slag (BFS) to be used as additives in the soil were estimated using Box Behnken Design (BBD) model in Response Surface Methodology (RSM) tools described by researchers elsewhere in the literature (Bingöl et al., 2012; Turan et al., 2013). The additives blended soil was tested in the laboratory to determine it's specific gravity, soaked CBR value, as well as MDD & OMC according to the guidelines described in the (BIS 2720). The UCS value of the amended soil sample was determined following the below-mentioned test procedure. The soil sample of the predetermined amount was first mixed with water at OMC to form a uniform mixture, which was then placed in a cylindrical mould in three layers, and each layer was compacted with standard rammer until MDD was achieved. Three nos. of cylindrical samples (Height: 7.6 cm and Dia.: 3.8 cm) were extracted from the compacted soil in the mould and cured in water for 7 days for UCS testing.

The toxic characteristics of the leachate generated through the polluted soil, as well as additives, amended soil were examined following the procedure depicted in the USEPA (1992). The soil samples, screened through 600 μ IS sieve, were mixed with double distilled water in the ratio of 10: 1 (water: soil). The pH of the soil-water suspension was kept close to 5.0 by mixing 0.5 N acetic acid. The suspension was placed for 24 h in a rotary shaker at 100 RPM speed and after that, centrifuged for 20 minutes under a speed of 10,000 RPM to separate liquid and solid phase. The liquid part was then filtered through 0.45 μm Whatman filter paper, and then the concentrations of the metals in the aliquot were determined by using the atomic absorption spectrophotometer (AAS).

Numerical modeling was performed using PLAXIS 3D software to evaluate the safety factors as well as vertical displacement values of an embankment founded on contaminated as well as stabilized soil. A comparative study was performed on the outcome of the modeling to assess the degree of strength improvement achieved of the stabilized soil post addition of additives. The numerical analysis of the stability of the embankment slope was performed using finite element modeling (FEM) with a strength reduction approach through PLAXIX 3D software (Dyson & Tolooiyan, 2018). The soil material properties for the foundation and slope were described using the Mohr-Coulomb constitutive model, which relates the shear strength of the material to shear parameters (cohesion & angle of internal friction) and normal stress, as illustrated by Equation (1). The failure of the slopes occurs

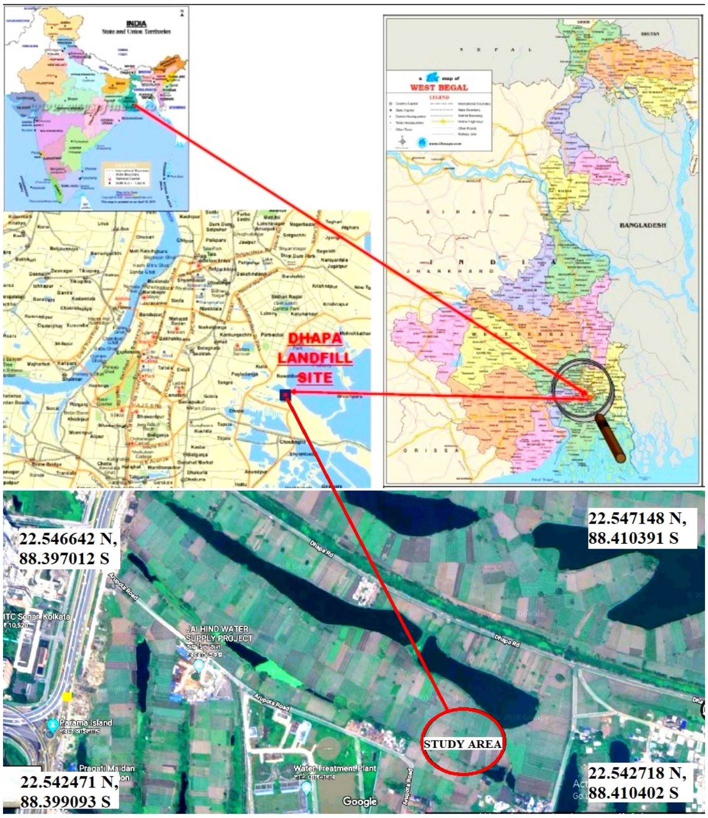

Figure 1. Location map of the study area (Courtesy Google Map)

when the shear strength of the soil on the sliding surface is inadequate to resist the shear stresses. The stability of slopes is examined by the Factor of safety (FOS) value.

$$\tau = c + \sigma \tan\varnothing, \quad (1)$$

where: τ = shear strength at failure; c = unit cohesion; $\varnothing$ = angle of internal friction; σ = normal stress.

Table 1. Engineering properties of the soil sample collected from the landfill site

Engineering properties	Parameter values
Sand (%)	36
Silt (%)	40
Clay (%)	24
Specific Gravity, G	2.06
Liquid limit, LL (%)	32.54
Plastic limit, PL (%)	18.26
MDD (kN/m^3)	12.94
OMC (%)	23
UCS (kN/m^2)	50
Soaked CBR (%)	4.82

2. Result and discussions

The background heavy metal concentrations of the soil sample collected from the landfill site are depicted in Table 2. The soil was found highly contaminated with As, Cr, Cd, Pb, Zn, Hg, Cu, Ni, and well above the threshold values as prescribed in the Canadian Soil Quality Guidelines (Canadian Council of Ministers of the Environment [CCME], 2007). Moreover, the surface, as well as sub-surface water in and around the study area, are also contaminated due to prolong lateral and vertical advective dispersion of the pollutants as reported by Maiti et al. (2016). The quality of virgin soil was found to be poor (Table 1), as reflected by MDD (12.94 kN/m^3), UCS (50 kN/m^2), and soaked CBR values (4.82%). The occurrence of such poor geotechnical properties was perhaps due to prolonged exposure of pollutants in the soil (Estabragh et al., 2017). Table 2 also exhibits that concentrations of heavy metals in the leachate of the multi-metal contaminated soil were decreased by almost 100% after blending with FA, QL & BFS additives at an optimized amount of 5, 20, and 20% by weight of soil mass after the curing for a period of 3 and 7 days. The substantial decline of the concentrations of leachable metals in the polluted soil by mechanical blending with FA, QL, and BFS seemed to be happened due to the immobilization of the pollutants as well as metal precipitates formations, which transformed the physical state of the contaminated soil (Estabragh et al., 2017). The increment of the quantities of the additives in the soil also helps in the decrement of the percentage of leachable metals due to the increment of the amount of immobile as well as insoluble metal precipitations (Wang et al., 2014). Moreover, it may be inferred that the additives stabilized metal contaminated soil owns a minimum or almost nil amount of exchangeable contents and a significant amount

Table 2. Concentrations of heavy metals in the leachate of the untreated and treated soil

Heavy metals	Metal concentration of untreated soil (mg/kg)	Metal concentration of treated soil (After 3 days of curing) (mg/kg)	Metal concentration of treated soil (After 7 days of curing) (mg/kg)	Permissible values (mg/kg) as per CCME (2007) guidelines
Arsenic (As)	6.20	<0.05	<0.05	12
Chromium (Cr)	28.93	<0.05	<0.05	64
Cadmium (Cd)	1.37	<0.05	<0.05	1.4
Lead (Pb)	335.04	<0.05	<0.05	70
Zinc (Zn)	564.28	0.33	<0.05	200
Mercury (Hg)	2.98	<0.05	<0.05	6.6
Copper (Cu)	193.96	0.70	0.52	63
Nickel (Ni)	16.86	<0.05	<0.05	50

of residual content than the untreated soil (Xia et al., 2018). The solidification/stabilization reduced the metals exchangeable fraction and further lowered the metals leachability from the multi-contaminated soil.

The optimum quantities (percentage by weight of the soil mass) of blending agents (FA, QL, and BFS) were estimated by using the BBD model under the RSM tool. The maximum improvement of the geotechnical properties of the metalcontaminated soil was achieved at FA: 5%, QL: 20%, and BFS: 20% with the desirability of 0.942 by using desirability function with a prefixed goal and weight for each factor (Derringer & Suich, 1980). The desirability ramp plot (Figure 2) shows the optimal experimental conditions for the improvement of MDD and soaked CBR values at maximum desirability of 0.942. An additional confirmatory experiment was conducted in the laboratory by mixing optimal values of FA, QL & BFS with the soil. The test was conducted in three replicate samples, and average values were considered. The average MDD and soaked CBR values were obtained as 14.23 kN/m^3 and 28.60%, respectively.

The model predicted results were observed in close agreement with the experimental results. The BBD model, in combination with the desirability function, was found an effective tool in optimizing the additive contents for the reclamation of contaminated land as well as enhancement of its geotechnical properties.

It was observed from Figure 3 that an increment of 13% of the MDD value of the stabilized soil was achieved in comparison to the virgin soil, when the FA, QL, and BFS content increased to 5, 20, and 20%. The additives

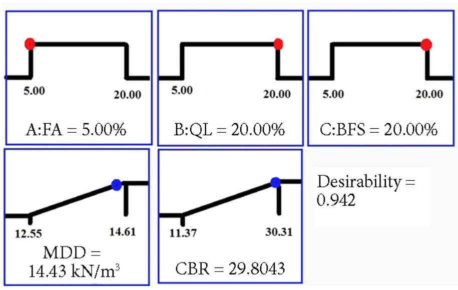

Figure 2. Desirability ramp plot for numerical optimisation

assist in metal precipitates formation, which in turn fill up the soil pore spaces leading to a substantial reduction of the pore volume and thereby causing the formation of denser soil mass (Horpibulsuk et al., 2009; Xia et al., 2018) and thus increase the UCS and MDD values of the stabilized soil mass (Ismail et al., 2002).

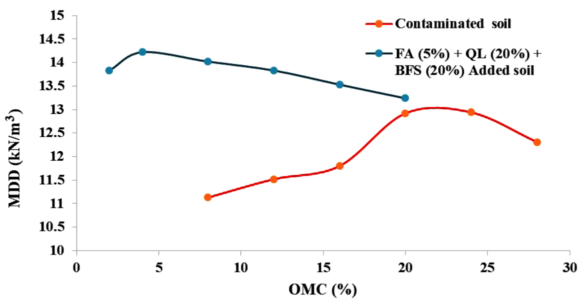

Figure 3. Comparison of compaction characteristics of contaminated and additives stabilized soil

The improvement of G, UCS, and soaked CBR values of the stabilized soil with the addition of optimized additive contents are displayed in Figures 4, 5 and 6. It was observed that G, UCS, and soaked CBR values were increased to 2.48 and 65 kN/m^2, 28.60% respectively after curing for 7 days. The formation of the metal precipitation in the soil with the addition of additives contents caused denser soil mass (Du et al., 2014) and which in turn

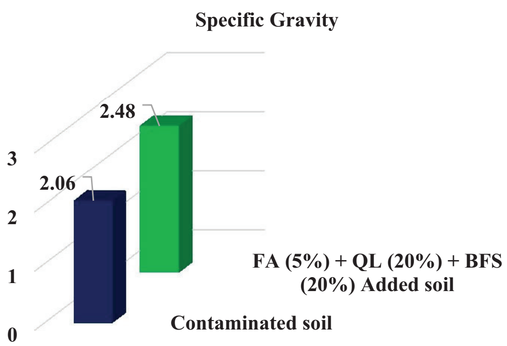

Figure 4. Comparison of Specific Gravity values of contaminated and additives stabilized soil

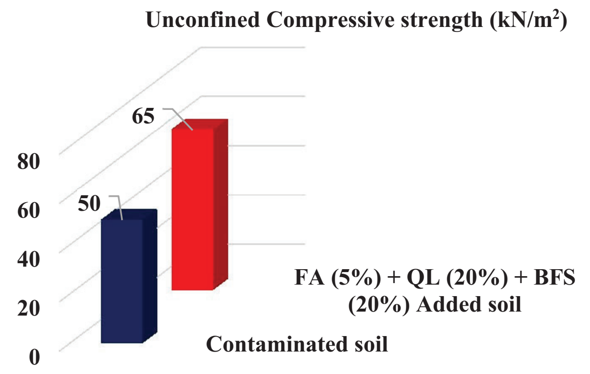

Figure 5. Comparison of UCS values of contaminated and additives stabilized soil

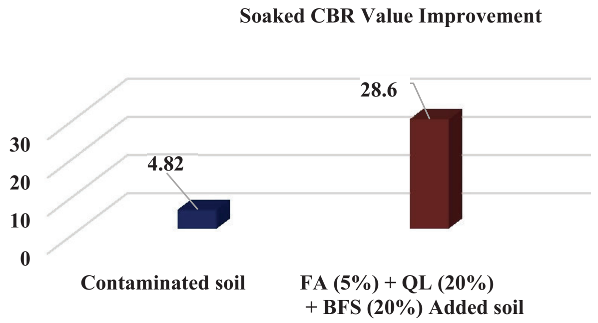

Figure 6. Comparison of soaked CBR values of contaminated and additives stabilized soil

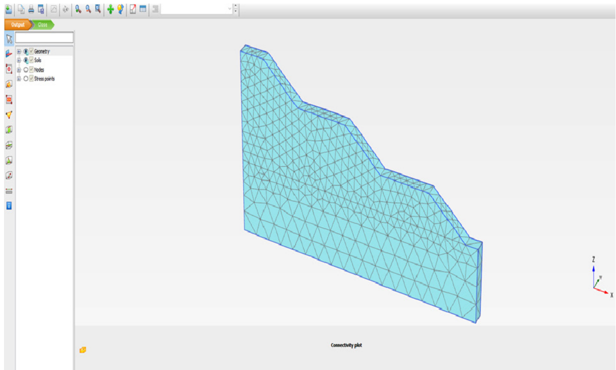

Figure 7. PLAXIS^{TM} 3D model generated mesh diagram of the embankment (1 V: 1.5 H)

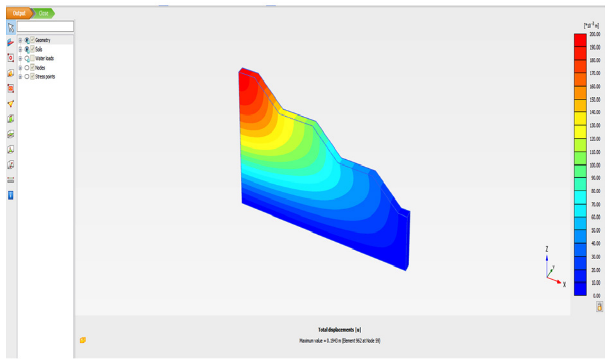

Figure 8. The vertical displacement of the embankment founded on contaminated soil (drained condition)

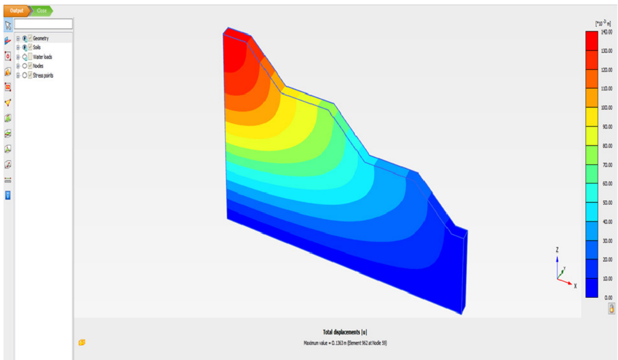

Figure 9. The vertical displacement of the embankment founded on stabilized soil (drained condition)

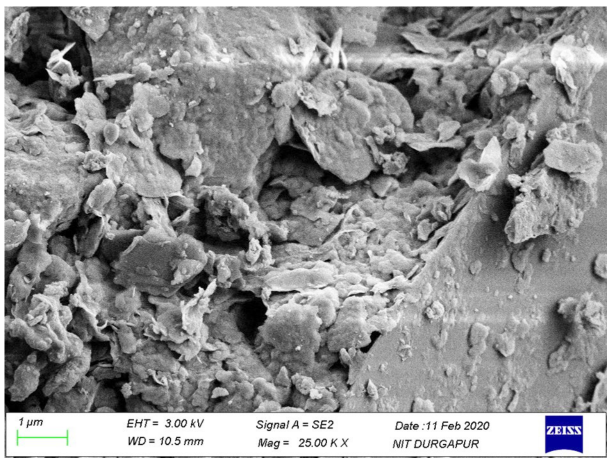

Figure 10. SEM micrograph of heavy metal contaminated soil

increased soil compressive resistance (Xia et al., 2018).

PLAXIS^{TM} 3D software generated mesh diagram of the embankment slope (1 V: 1.5 H) is shown in Figure 7. The vertical displacement of the embankment founded on contaminated soil was estimated as 194.3 mm under drained condition (Figure 8). Whereas, the vertical displacement of the same embankment founded on stabilized soil was estimated as 136.3 mm under drained condition (Figure 9). The factor of safety value was also found to be increased from 2.5 to 3.2 under the drained condition of the model embankment slope founded on additives stabilized soil.

The scanning electron microscope (SEM) micrographs of the heavy metal contaminated and additive stabilized soil are shown in Figures 10 and 11. The insufficient spaces and flaky shapes were observed for the micrograph of the contaminated soil. Soils with flakey shapes are likely to pose low strength. The micrograph of the contaminated soil was compared with the stabilized soil. The aggregation seen in the stabilized soil is absent in the contaminated soil.

The SEM micrographs of the stabilized soil indicated the crystalline format with visible edges. However, few pores are also observed for the micrograph of stabilized soil. Overall, a much denser matrix is observed, showing the formation of fibers and the continuous mass of the stabilized soil, which increased the strength of the contaminated soil.

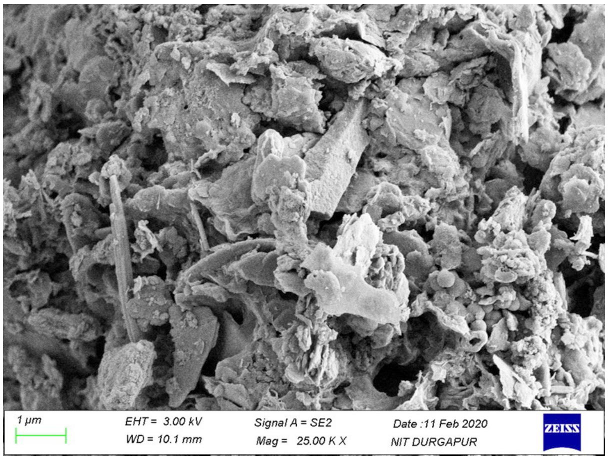

Figure 11. SEM micrograph of additive stabilized soil

Conclusions

- In the present study, the efficacy of three additives namely FA, QL, and BFS, was studied to solidify/stabilize the multi-metal contaminated site in Kolkata, West Bengal, India, through mechanical mixing. The optimization studies were also performed to estimate the optimum quantities of the additives to be blended with the soil to achieve the maximum improvement of the soil qualities. It is observed through laboratoryscale experimental results that the optimum quantities (FA: 5%, QL: 20% and BFS: 20%) of additives addition (by weight of the soil amount) increased the MDD, UCS, soaked CBR values of the soil at a substantial margin after a curing period of 7 days. The significant improvement of the contaminated soil qualities was happened as the addition of the additives assist in metal precipitates formation, which in turn fill up the soil pore spaces leading to a substantial reduction of the pore volume and thereby causing the formation of a denser soil mass.
- Moreover, the mechanical blending of the additives in the contaminated soil and after that water curing caused a major reduction of the quantities of leachable metals contents, which was happened due to the immobilization of the pollutants as well as the formation of metal precipitates. Thus, it can be inferred that the additives stabilized metal contaminated soil owns a minimum or almost nil amount of exchangeable contents. Thereby, the chances of subsurface pollution are almost restricted at the metal-contaminated sites.
- Furthermore, the numerical modeling study results indicate the reduction of the total vertical displacement and increment of the stability factor of the embankment founded on stabilized soil, which also reinforces the efficacy of the FA, QL, and BFS additives as potent binders in solidifying/stabilizing the multi-metal contaminated soil.
- Finally, the present study results provide a novel idea for in-situ improvement of contaminated sites through mechanical mixing with low-cost binders, and thus reclamation of the polluted sites can be achieved. Future infrastructural activities can also be initiated without replacing the contaminated soil. However, further field studies are to be undertaken to reinforce the novel idea.

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Author contributions

RR and SP conceived this research and designed experiments. AK performed numerical analysis. RR and SP performed experiments and analysis. RR and SP wrote the paper. All authors participated in the revisions of it. All authors read and approved the final manuscript.

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