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Sustainable Reuse of Heavy Metal Contaminated Dredged Sediment Using Industrial Waste as Stabilizers

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ABSTRACT

Urban areas face soil pollution due to unscientifically disposed industrial effluents and solid waste. The soil and water bodies get contaminated with heavy metals and seriously harm the surrounding ecosystem and groundwater in the long run. Dredging activities undertaken for restoring water bodies require the disposal of dredged sediment. With advanced technologies, new forms of waste are being generated that can cause damage to nature. Conversely, the construction industry faces a shortage of virgin materials. Considering both problems, dredged contaminated sediment is experimented on as a construction material by stabilization/solidification into blocks to utilize as partition blocks. For the experiments, contaminated sediment was collected, which contained heavy metals that can leach out and contaminate groundwater. To understand the efficiency of immobilization, sediment was artificially contaminated by dosing it with Cadmium, Nickel, and Zinc. The effect of stabilization on the leachability of heavy metals. The leachability of the material was tested as per the USEPA1311 and USEPA1312 methods. Industrial waste, Ground Granulated Blast Furnace Slag (GGBS), was used in combination with lime for stabilization in various proportions, and it was found that 30% binder (a combination of Lime and GGBS) gives desirable results in terms of strength and complete reduction in leachability. And hence contaminated sediments can be effectively reused in making partition blocks as a sustainable approach.

Keywords: Reuse of dredged sediment, heavy metals, leachability, lime ggbs stabilization

1 INTRODUCTION

An increase in urbanization is causing waterbodies to get contaminated with toxic heavy metals. The toxic heavy metals get into natural bodies through smelting, tannery and other industrial wastes and settle in the sediment over time, contaminating sediment and the water. Urban areas also face soil pollution due to unscientifically disposed industrial effluents and solid waste. The soil and water bodies get contaminated with heavy metals and pose a danger to the surrounding ecosystem and groundwater in the long run due to the toxic heavy metals. Exposure to toxic heavy metals can have ill effects on health, such as cancer or damage to vital organs. (Algül F., & Beyhan M. 2020) Hence it is necessary to immobilize these metals to avoid their exposure to plants, animals, and humans.

There are several ways to immobilize heavy metals where chemicals are mixed with the contaminated sediment. The immobilization mechanism can be achieved in two different ways; most of the time, this happens simultaneously; first is chemical stabilization, where the heavy metals in the soil are converted to a less soluble form, and the second mechanism is physical encapsulation, where the heavy metals are encapsulated in the binder gel and get isolated from the environment. (Guo B. et al, 2017)

Dredging activities need to be undertaken to restore water bodies, which requires the disposal of dredged sediment that can have high concentration of toxic heavy metals dure to pollution. (Wang L. et al., 2018) With advanced technologies, various new forms of wastes are also being generated that can cause damage to nature. On the other side, the construction industry is facing a shortage of virgin materials and a high carbon footprint due to cement. (Wang L. et al., 2016a). Considering both problems, dredged contaminated sediment is experimented on as a construction material by stabilization/solidification into blocks to utilize as partition blocks. The successful implementation can lead to a reduction in soil pollution, reduced usage of virgin material and, in turn, reduction in carbon footprint.

Reuse of dredged marine sediment as construction material by stabilization/solidification using cement and other additives and its leachability has been explored in the last decades (Todaro F. et al, 2020; Yoobanpot N. et al, 2020). The literature available for dredged contaminated lake sediments to be used as partition blocks without cement is limited and the same was explored in the studies.

Ground Granulated Blast Furnace Slag (GGBS) is a by-product Iron industry. This being rich is CaO, SiO_2 and Al_2O_3, which can lead to strength gain on hydration and has gained popularity as a binder in combination with hydrated lime, MgO. (Wang F. et al., 2021) Addition of GGBS can lead to an increase in the soil and stabilize the soil. UCS of soil has shown an increase when 20% GGBS is added to the soil along with 15% lime. (Islam S. et al., 2016)

2 MATERIALS AND METHODOLOGY

2.1 Materials

Bellandur lake, one of the largest lakes of Bangalore, covering about 3.7 square km area, had been infamous for its pollution and foul stench. As a remedial method, authorities had directed to desilt the lake and dredge silt out of the lake which amounts for almost 1 lakh cubic meter. The samples from a few accessible sites were collected and checked for heavy metals and was found to have excess quantity of Cadmium, Nickel, and Zinc with reference to the safe standards. The soil was tested for physical properties including, specific gravity (D 854 -14), Sieve analysis (IS 2720- part 4), Atterberg’s limits, OMC, MDD by Proctor compaction test (D 698-12) and chemical properties such as pH (ASTM 9045 D), electrical conductivity (EC) (IS 14767: 200), Organic matter and nitrates. The aim of this topic was to study the leachability characteristics and immobilization efficiency of heavy metals. The sediment was spiked using analytical grade salts $CdSO_4.7H_2O$, $Ni(NO_3)_2.6H_2O$ and $Zn(NO_3)_2.6H_2O$ for Cd, Ni and Zn respectively to have the soil contaminated with 50 mg/kg of Cd, 400 mg/kg of Ni and Zn. The sediment was kept in contact with the metal salt solution for 4 weeks and later was allowed to dry. The air-dried sediment was crushed using wooden hammer and sieved through 2 mm sieve. The density of soil compacts was fixed at 1.8 g/cc. GGBS was introduced as the pozzolanic material in combination with lime. The binders were increased from 15% to 30%. GGBS was introduced in three proportions, 10%, 15% and 20% and hydrated lime in 5% and 10% was introduced for each GGBS content, which would help in interpreting role of the binders in arresting leachability. The content of lime and GGBS was varied as given in the Table 1.

Table 1. *Mix proportions used for experiments.*

Mix	M1	M2	M3	M4	M5	M6
Binder %	15	20	20	25	25	30
GGBS	10	10	15	15	20	20
Lime	5	10	5	10	5	10

2.2 Methodologies

The dried sediment was mixed with the binders and three different moisture content, 10%, 15% and 20%. Out of which 15% was found to give desirable results and hence fixed. More than 15% moulding moisture content was leading to separation of water from mix during static compaction process.

2.2.1 Strength Test

The soil was moulded into compacts of a diameter of 38 mm and height of 76 mm. All the samples were cured at 97% humidity and tested for compressive strength at 3 days, 7 days and 28 days (ASTM D 1633). A strength test was conducted at a loading rate of 0.24mm/min. The crushed sample was then collected, powdered, and kept for air drying for 1 hour.

2.2.2 Leachability Tests

The pH of the air-dried powdered samples was checked. The pH was more than five even after the addition of HCl, Toxicity characteristic leaching procedure (TCLP) as per USEPA 1311, and extraction fluid #2 with 2.88 pH was used to check the leachability of heavy metals. This fluid was prepared with glacial acetic acid (CH_3COOH) diluted in reagent water to adjust the pH to 2.88. Synthetic precipitate leaching procedure (SPLP) is recommended by USEPA 1312 for industrial waste and hence used where the extractant fluid Extraction fluid # 1 is a 60/40 weight percent mixture of Sulfuric acid (H_2SO_4) and Nitric acid (HNO_3) with required dilution to obtain pH of 4.2. USEPA1312 also recommends reagent water as an extraction fluid. Hence, reagent water with pH 6.8 was used as extractant fluid 3. The details of the leachate (extractant fluid) are tabulated as given in Table 2. Analytical-grade reagents were used for all the experiments.

Table 2. *Details of leachate (Extractant Fluid)*

Fluid no	Reference	Constituents	pH
1	USEPA 1311 Extractant Fluid #2	CH_3COOH in 1 Litre Reagent water	2.88
2	USEPA 1312 Extractant Fluid #1	60/40 weight percent mixture of H_2SO_4/HNO_3	4.2
3	USEPA 1312 Reagent water	Demineralised water	6.8

2.2.3 Analysis of heavy metals

After strength tests, the dried samples were powdered, placed in conical flasks, agitated with the three above fluids for 18 hours, centrifuged, filtered through 0.45-micron PVDF filters, and checked for eluate pH. These filtered samples were tested in Thermo Fisher Scientific (ICE 3000) Atomic absorption spectrometer (AAS) for Cd, Ni, and Zn. Cd was analysed using its primary wavelength of 228.8. Nickel was analysed using a wavelength of 337.8 nm, Zn was analysed using a 213.8 nm wavelength, and deuterium lamp correction was applied during the analysis. The energy level of the lamp was decided based on the experiments conducted previously for known concentration. The limit of detection for the metals were in the range of 1-5 µg/L, 2-10 µg/L, and 5-20 µg/L, respectively.

X-Ray Diffraction (XRD) was carried out on the powdered samples in the Brukers Germany machine in the central X-Ray facility IISc to identify the crystallinity of the materials. The same procedure was conducted for 3,7 and 28 days. SEM analysis was performed on the powered samples of 28-day age using Jeol SEM Japan from Advance Facility for Microscopy and Microanalysis (AFMM) IISc.

3 RESULTS AND DISCUSSION

3.1 Physical Properties

The dredged sediment collected was oven-dried, and experiments were conducted for physical properties using standard protocols. The results obtained are tabulated in Table 3.

Table 3. *Physical Properties*

Soil Classification	Specific Gravity	Liquid Limit	Plastic Limit	OMC (%)	MDD (kg/m^3)
ML	2.56	27.82	Non-Plastic	9.36	1.85

3.2 Chemical Properties

Stabilization is a chemical process, so the chemical properties of the materials used play an important role in strength and leachability. In case of contaminated soil or sediment to be used as a construction material, the knowledge of contaminants becomes necessary to decide the mix proportion and predict the properties of the final product. Chemical properties were analysed using respective standard protocols, and the results are given in Table 4.

Table 4. *Chemical Properties*

Parameter	**Values**
EC (milli siemens /cm)	0.83
pH	7.24
Organic Matter (%)	12%
Total nitrogen (N)	0.15%
Zinc (Zn)	80 mg/kg
Nickel (Ni)	30 mg/kg
Cadmium (Cd)	28 mg/kg
Nitrate (NO_3)	0.22%
Nitrite (NO_2)	< 0.01%
Ammoniacal nitrogen (N)	< 0.01%
Phosphate (PO_4)	0.008%
Calcium (Ca)	0.70%
Potassium (K)	0.26%
Organic carbon	4. 30%
Sulphur (S)	< 0.01%

3.3 Strength Properties

To understand variation in strength and leachability with respect to the age of stabilized soil, the samples were tested at 3 days, 7 days, and 28 days of 97% humidity curing. The strength of samples obtained in Mpa is given in Fig 1. The samples showed a brittle failure irrespective of binder percentage or GGBS to lime ratio. A gradual increase in strength from 3 days to 28 days was observed for all the samples. M1 showed a 31% increase in UCS from 3 to 7 days and a 26.9% increase from 7 to 28 days. M2 showed a 31.4% and 20.4% increase in strength from 3 to 7 days and 7 to 28 days, respectively. For M3, the increase was 16.8% and 15.5% in 7 and 28 days UCS. M4 showed a 27.9% increase from 3 to 7 days; however, from 7 to 28 days, only a 9% increase was observed. M5 and M6 showed an 18.25% and 23% increase in 7 days strength and a 14.7% and 23% rise in 28 days strength, respectively. Overall, the strength increased as the binder content increased. The highest strength, 8.13 MPa, was obtained for 30% binder. It was observed that the binder percentage and the GGBS to lime ratio played a role in strength gain. The rate of strength gain showed variation with a change in the ratio. The maximum ratio of 0.4 had given the highest strength, and the trend showed that the strength increased slightly with the increase in the ratio. (Yaolin Yi 2016). In the present studies, the ratio varies from 0.25 to 1, and variation in strength is observed.

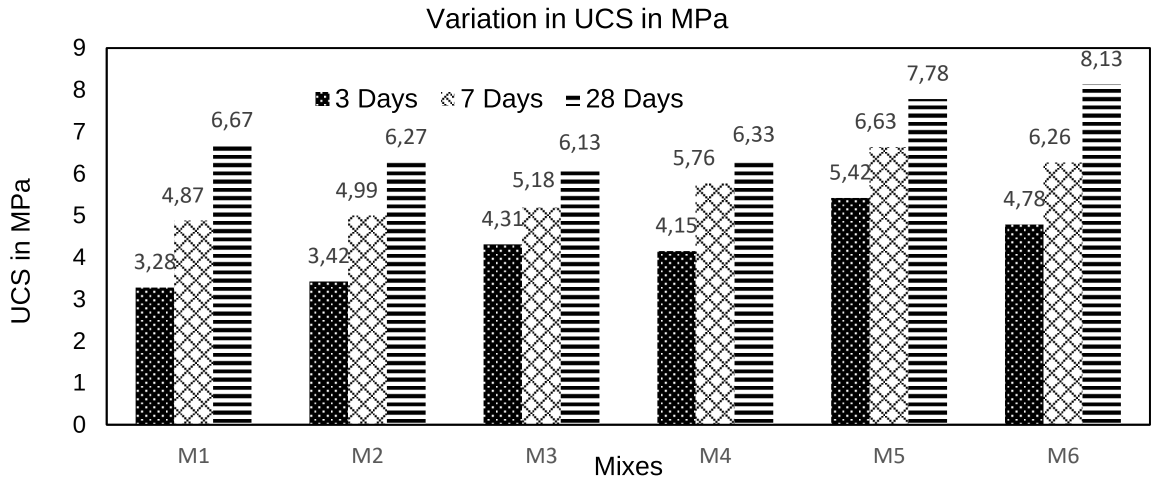

***Figure 1.** Variation in UCS*

3.4 Leachability Test Results

Leachability of Cd was found to reduce from 12 mg/kg to less than the blank value for all three extraction fluids indicating the complete immobilization of Cd within 3 days of curing and showed no leaching even after 7 or 28 days. Ni and Zn, even though showed a reduction in leachability to less than blank for fluid 2 with 4.2 pH and fluid 3, reagent water with pH 6.8, and for fluid 1 with pH 2.88, both the metals leached out. The results are graphically represented in 2. For the mixes M2, M4, and M6, where 10% lime was added, both Ni and Zn did not leach out from the samples, which can be attributed to the high pH or high alkaline nature of the eluate. The samples M1, M3 and M5 with 5% lime, demonstrated leaching of Ni and Zn after 3 days of curing. This can be attributed to the leaching behaviour of cations, which shows maximum activity at lower pH, particularly at around pH 3 (Anna Król, 2020) from the industrial slag, this might have happened due to 5% lime addition did not increase the pH of eluate beyond the acidic level and the heavy metals could dissolve in the extraction fluid. GGBS and 10% lime could stabilize the both the cations from soil as well as the slag at high pH of 12 resulting in reduced leachability. Ni leachability reduced as the curing time increased which could have been due to the mechanism of encapsulation of metals in matrix due to CSH formation reduces leachability of Ni. (Li W. et al, 2019) and thus the leaching reduced gradually from as the time of curing increased from 3 days to 28 days. Leachability of Zn marginally showed in increase which could be due to the CSH gel contributing to increasing leachability for lower alkaline eluates. (Wei M. et al, 2022) Although Zn was found to be leaching after 28 days of curing, the concentration was less than permissible limits. Overall immobilization efficiency is graphically represented in Fig. 3. The efficiency is calculated as the ratio of change in concentration of heavy metals to the initial concentration of heavy metals that leached out from the samples.

The pH used for leachate or extractant fluids as already discussed is given in Table 2. The fluid that is extracted after the contact time of leaching procedure (18 hours) is called eluate. The pH of eluate for all the samples was measured and graphically represented in Figure 4. It was observed that eluate pH in soil stabilization can be used to gauge the extent of contamination of the surrounding environment. (Wan-lu Zhang, 2020). The pH of the eluate was found to be lower, between 5 and 7, for the mixes containing 5% lime compared to the mixes containing 10% lime where the pH of eluate lay between 8 and 10. The low pH of eluate can justify the leachability of Ni and Zn for these mixes at 2.88 pH of extractant fluid. However, the concentration of both Ni and Zn was reduced to safe limits and because naturally, the pH going less than 3 can be a rare phenomenon hence from practical point of view, it can be stated that the heavy metals were brought down to less than permissible limits and the soil with heavy metal contamination can be safely used for construction purpose.

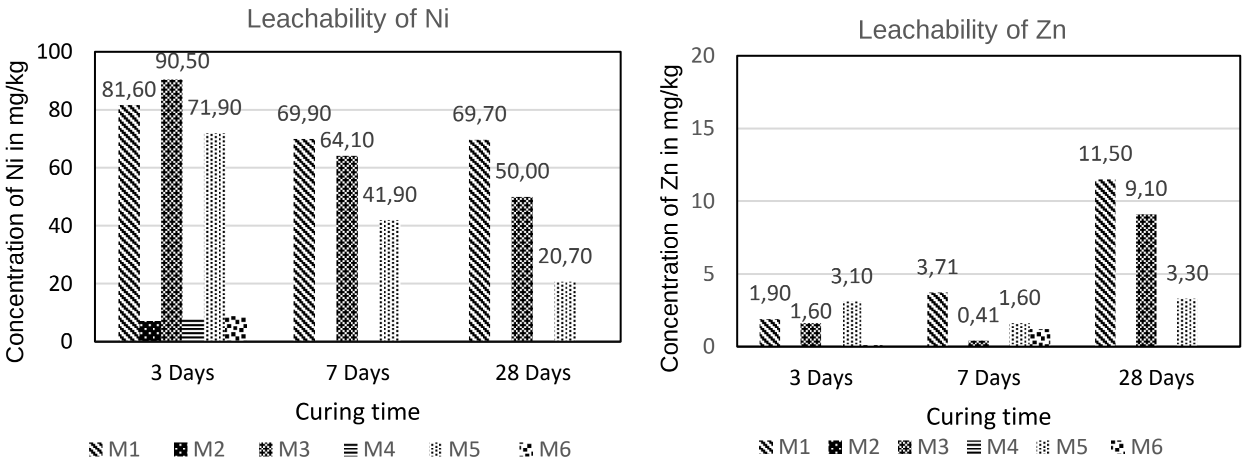

Figure 2. *Leachability variations with respect to curing time for Ni and Zn*

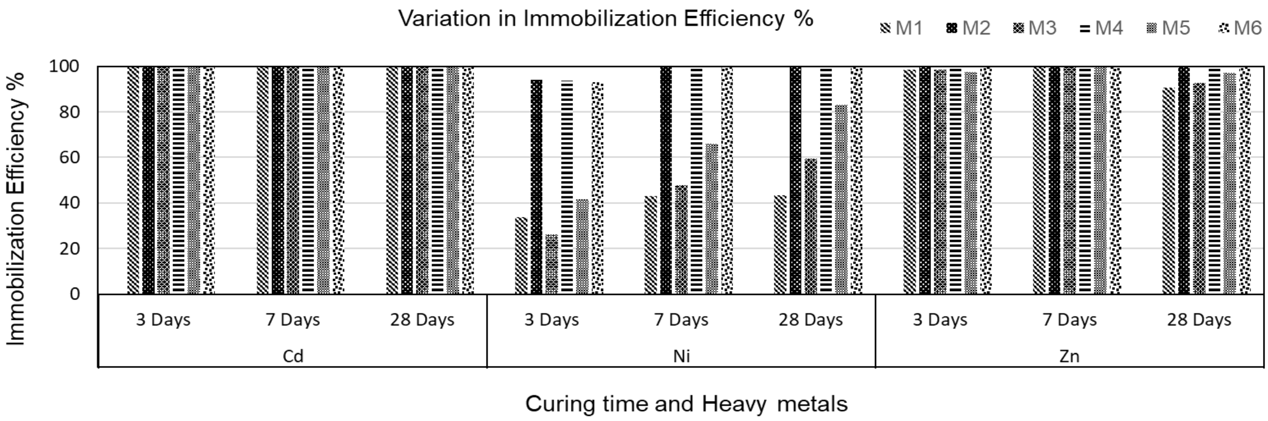

Figure 3. *Variation in immobilization efficiency*

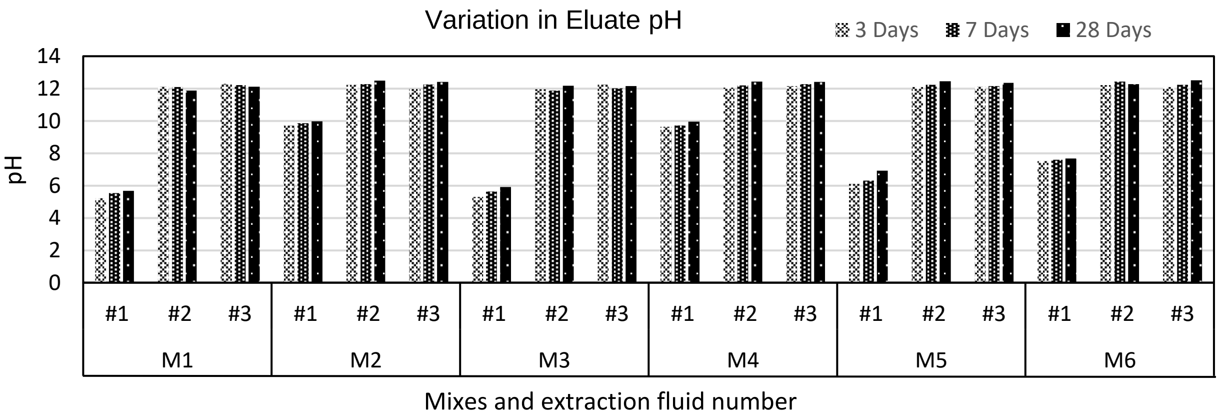

Figure 4. *Variation in Eluate pH with respect to days, pH of leachate and mix*

4 CONCLUSIONS

Soil with heavy metal contamination was stabilized using industrial waste, GGBS, and lime to check its feasibility as a construction material, particularly for use as partition blocks. Dredged sediment from Bellandur Lake was spiked with heavy metals and then stabilized using six different mixes with variations in binder percentage and the ratio of GGBS to lime. The UCS and leachability in three different pH leachates were studied, and the following conclusions are drawn from the results.

- The highest strength obtained was 8.13 MPa for 30% binder at the end of 28 days. The strength also varied with binder percentage and GGBS to lime ratio. The rate of strength gain was observed to vary with the percentage of binder and ratio of the mixes.
- The Immobilization efficiency of Cd was 100% even with 3 days of curing, and Cd did not leach out at any pH, implying complete immobilization.
- Ni showed leaching behaviour only for the extractant fluid of 2.88 pH, which gradually decreased with an increase in the days of curing. Other leachates i. e. the extractant fluids of pH 4.2 and 6.8, did not show any Ni leachability giving 100% immobilization efficiency after 3 days of curing.
- Zn showed similar leachability trends except that at 28 days, where the heavy metal leached out. However, the leachability was lesser than the permissible values.
- Overall, 30% binder showed the most desirable results and can be considered for practical application because pH less than 3 is a sporadic phenomenon. The heavy metals were within safe limits after three days of curing.
- Even with a 5% lime addition, the compressive strength obtained was above 6MPa after 28 days of curing.

Pre-treatment of the dredged sediment can help reduce the leachability of heavy metals. Thus, further reduction of leachability with minimum addition of lime which can lead to a sustainable partition block production with the least carbon footprint can be the future scope of the studies.

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