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Immobilization of Lead Contaminated Low Plastic Clay

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***Abstract:** We Indians are living in an age of industrialism where our soils may become contaminated by the accumulation of heavy metals through emissions from the rapidly expanding industrial areas, land application of fertilizers, wastewater irrigation, disposal of high metal wastes etc. Heavy metals constitute an ill-defined group of inorganic chemical hazards which generate leachate during leaching process causing the soil and ground water contamination during its interaction with the both. This is not only harmful for the subsurface water aquifers but also for structures existing on it. Any changes in engineering properties and behaviour of soil strata may lead to loss of bearing capacity, an increase in settlement of foundation system consequently leading to failure of structures thereby causing loss of property and life. The adequate protection and restoration of soil ecosystems contaminated by heavy metals require their characterization and remediation.*

In this study the soil contaminated is immobilized, which result in the redistribution of contaminants from solution phase to solid phase, thereby reducing their bioavailability and transportation in the environment. Thus leaching is restricted by isolating the waste within a solid cementitious matrix. In the present study low plastic clay was artificially contaminated using Lead Acetate at different concentration and an attempt is made to immobilize it using a mixture of class F fly ash and OPC. The effectiveness of the treatment was evaluated by means of compressibility, permeability, plasticity tests and toxicity characteristic leaching procedure (TCLP).
Keywords: Heavy metal remediation, Stabilization/solidification, Lead contamination, TCLP Test, Soil contamination

I. INTRODUCTION

The insistent and rapid industrialization in India calls for the need to effectively dispose and manage generated wastes. Soil is the dumping ground of most of the waste products. Every year the solid wastes dumped into the soil are increasing at an alarming rate. The pollutants in soil remain in direct contact with soil for relatively longer periods. Different heavy metals vary in their tendency to end up in water held in the soil or underlying groundwater, or bind tightly to the soil. These affect the soil mineralogy and clay content. It also causes alterations in soil properties like pH, swell shrink behaviour, permeability and shear strength. Kerala having the highest literacy rates, have a developed society and have all kind of waste generation including heavy metals, biochemical etc.

With greater public awareness of the implications of heavy metal contamination ,.on soils and human health and due to need for ‘green areas’ there has been increasing interest in the development of technologies to remediate contaminated sites and facilitate sustainable industrial development. Factors to be considered for the selection of reclamation method are its effectiveness, impact on environment, limitations, cost and availability. Amidst the numerous soil remediation methods in use, stated that the immobilization technique appears the most effective due to its ability to entrap the waste within a solid cementitious matrix and cost effectiveness. In the present study low plastic clay is artificially contaminated using different concentration of Lead Acetate and then an attempt is made to immobilize the soil using a mixture of Flyash - OPC. The variation in properties due to contaminant and amendment addition is then studied.

II. MATERIALS AND METHODS

A. Low Plastic Clay

Commercially available Low plastic kaolinite clay was used in this study. It was purchased from English East India Clay Limited Trivandrum.

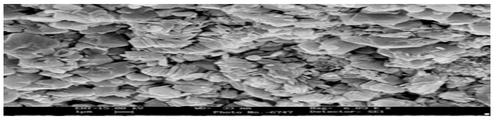

Fig 1. SEM Analysis of Kaolinite Clay

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TABLE I PROPERTIES OF KAOLINITE CLAY

Soil Properties	Values Obtained
Specific Gravity	2.64
Liquid limit (%)	34.3
Plastic limit (%)	22.8
Plasticity index (%)	11.5
IS classification	CL
OMC(%)	22.5
Dry density(g/cc)	1.585
% clay	59.8
% silt	27.7
% sand	12.5
UCC strength (kN/m^2)	53.1
Free swell index	0.52
Coefficient of Permeability (cm/sec)	$4.89*10^{-7}$

The properties of the samples are presented in Table 1.The soil sample was mixed with different molarity of lead acetate solution. The SEM image of Kaolinite is shown in Fig 1.

B. Flyash

Flyash also known pulverized fuel ash is one of the coal combustion products composed of finer particles driven out of the boiler with the flue gases. Fly ash used in this study is class F fly ash which was purchased from thermal power plant in Thoothukudi. The chemical and physical properties of fly ash are presented in Table 2 and 3. In order to immobilize the artificially contaminated soil, optimum dosage of flyash was found out carrying out compaction and compressibility tests. The SEM image of flyash is shown in Fig 2.The optimum flyash – OPC mixture is added only to the artificially lead contaminate soil and the variation in properties are studied.

TABLE II CHEMICAL PROPERTIES OF FLY ASH

Composition	Percentages
SiO_2	57.5
Al_2O_3	33
Fe_2O_3	4.8
TiO_2	1.4
CaO	0.5
MgO	0.2
Na_2O	0.2
K_2O	0.4
Loss on Ignition (% Max)	1.5

TABLE III

PHYSICAL PROPERTIES OF FLYASH

Soil Properties	Flyash
Specific Gravity	2.18
Liquid limit(%)	28.3
Plastic limit(%)	Non Plastic
Plasticity index(%)	Non Plastic
% clay	16.25
% silt	29.75
% sand	46
Uniformity coefficient(Cu)	6.31
Coefficient of curvature(Cc)	1.383
IS classification	SM
OMC(%)	31.3
Dry density(g/cc)	1.16
Free Swell	0.75
pH	8.9
Coefficient of Permeability (cm/sec)	$2.79 * 10^{-5}$
UCC strength (kN/m^2)	92.33
Angle of internal friction (^{o})	34
Class of Flyash (ASTM C618)	F

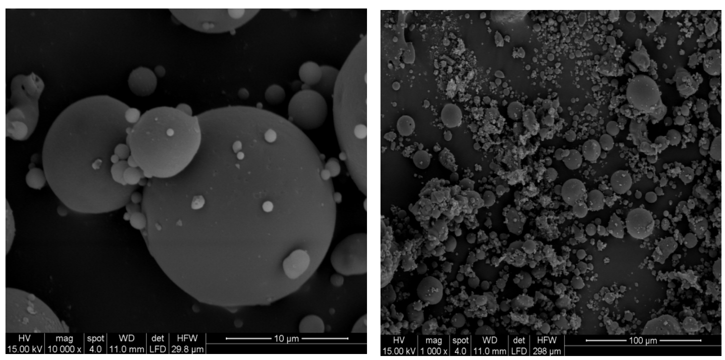

Fig. 2 SEM Analysis of Flyash

C. Cement

Since the fly ash is class F it has pozzolanic properties, but little or no cementitious properties. Therefore small percentage of OPC 43 grade cement is added for soil modification and to provide the fly ash cementitious property. Hence the percentage of OPC is fixed as 3%.

D. Heavy Metal Solution

In this investigation, [$(CH_3COO)_2Pb.3H_2O$] Lead acetate salt of analytical reagent grade was used for preparing different concentrations (molarity) of lead solutions for artificially contaminating the soil. Lead contamination can occur from many sources including mining operations, industrial emissions, waste incineration etc. (Source: Gautam SP, CPCB). Lead ranks fifth behind Fe, Cu, Al and Zn in industrial production of metals. It is highly toxic for human and animals. EPA defines a soil lead hazard as 400 ppm in the residential and 1200 ppm and industrialized areas. In contaminated lands, wide variations in lead levels have been reported ranging from less than 100 ppm to as high as 1100 ppm. Kabata et al., (2001) stated that lead becomes immobile if it goes from soluble form to form complexes with lime, phosphates, organic materials and clay minerals.

E. Immobilization of Contaminated Soil

Soil immobilization or stabilization/solidification (S/S) appears the most effective due to its binding ability. The waste material is

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converted into solid forms via entrapping within a monolithic matrix by adding appropriate reagents. This technique involves a combined application of the solidification and stabilization process and thus ensures the mixing of the contaminated waste materials with the treatment agents, and consequently, both the physical and chemical immobilization of the hazardous components occurs. This technique is a non- destructive approach to eradicate or inhibit the mobility of contaminants in the waste materials (US EPA 1999). It has been evaluated that the average operational time for the S/S projects was shorter (~1.1 months) than many other approaches (e.g. soil vapour extraction, land treatment and composting) and cost effective with an average cost per cubic yard between USD 194 to 264 (US EPA 2000),i.e. approximately 13000 to 17500 rupees. In the present study mixture of Fly ash - OPC are being used to immobilize the contaminated soil. For ensuring the effectiveness of this method USEPA have provided various criteria's to be fulfilled as listed in Table IV.

TABLE IV
TYPICAL SOLIDIFICATION/STABILIZATION CRITERIA FOR SOILS (USEPA)

Parameter	**Units**	**Average Value**
Unconfined Compressive Strength	Pounds per Square	>50
Hydraulic Conductivity	cm/s	$< 10^{-8}$
Hydraulic Conductivity (stabilized waste destined for land	cm/s	$<10^{-5}$
Atterberg Limits (Morgan et al., 1984) Plastic Limit	%	20 – 50
TCLP (US EPA, 1986)	mg/L	<= Regulatory

III. RESULTS AND DISCUSSIONS

A. Optimum Percentage of Amendment on Low Plastic Clay

In the present study a mixture of Fly ash - OPC is used to immobilize the heavy metal contaminated soil. Since the fly ash is class F it has pozzolanic properties and to provide it more effective binding ability OPC is added and the percentage of OPC is fixed as 3%. Compaction and UCC test is carried out on soil mixed with varying percentages of fly ash (2%, 4%, 6%, 8%, 10% and 12%) and the optimum percentage of fly ash is found out from the test results. Variation in dry density, OMC is shown in Table V. The graphical representation of variation in UCC strength with the addition of varying percentage of fly ash is shown in Fig 3.

TABLE V
VARIATION IN DRY DENSITY AND OMC WITH THE ADDITION OF VARYING PERCENTAGE OF FLY ASH

Flyash (%)	0	2	4	6	8
γd (g/cc)	1.585	1.56	1.528	1.472	1.439
OMC	22.5	23.35	24.81	25.52	26.43

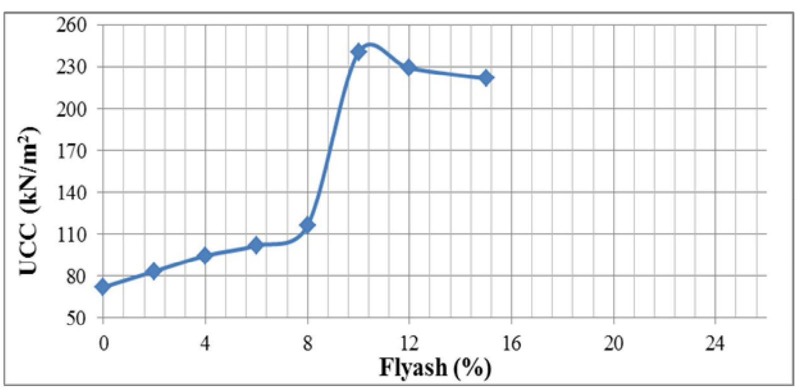

Fig 3. Variation in UCC with the Addition of Varying Percentage of Fly Ash

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The decrease in dry density may be because of the lower dry density of fly ash compared to the kaolinite clay and the higher optimum moisture content maybe due to the poor grain size distribution and the presence of large and hollow cenospheres in flyash (Fig 2) which increases its water holding capacity and increase in UCC may be attributed to the better bonding of clayparticles by fly ash. From the results the optimum percentage of fly ash for CL is 10%.The addition of cement above 5% does not contribute to strength (Ahmet et al., 2005),as the mixture does not contain all the elements for the cementation. Therefore 3% OPC is added to the fly ash to provide it cementitious property. The compaction curve of the CL mixed with 10% flyash- 3% cement is shown in fig 4.

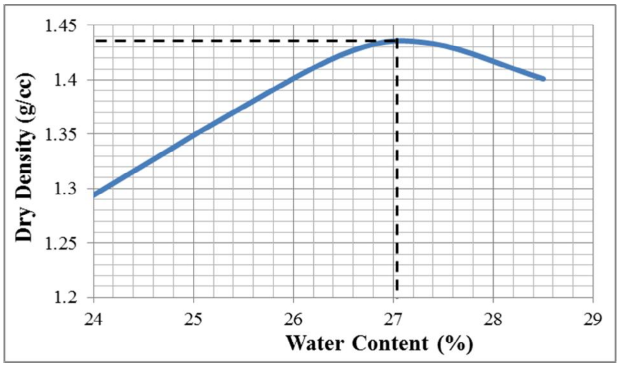

Fig 4. Compaction Curve of Kaolinite + Optimum Mix

B. *Effect of Amendments on Contaminated Clay*

1) *Effect of Amendment on pH*

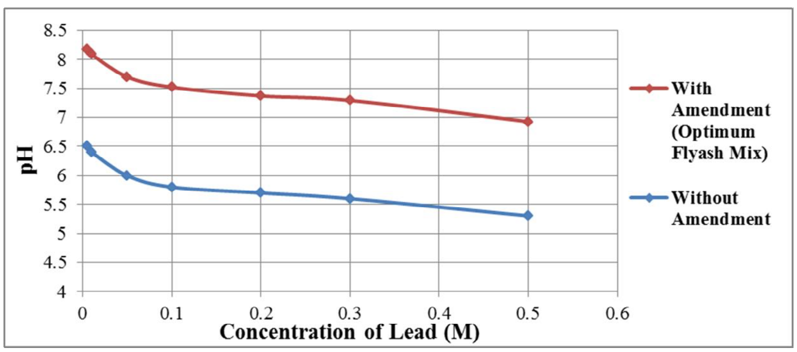

Fig 5. Variation of pH on Contaminated CL Clay Due to Addition of Amendment

Addition of lead decreases the soil pH to relatively acidic nature as shown in Fig 5 and causes damage to soil structure while the addition of fly ash to contaminated soil make the pH almost neutral. This is because; addition of alkaline nature fly ash to the soil with heavy metal of acidic nature tends to neutralize the soil pH.

2) *Effect of Amendment on Atterberg Limit*

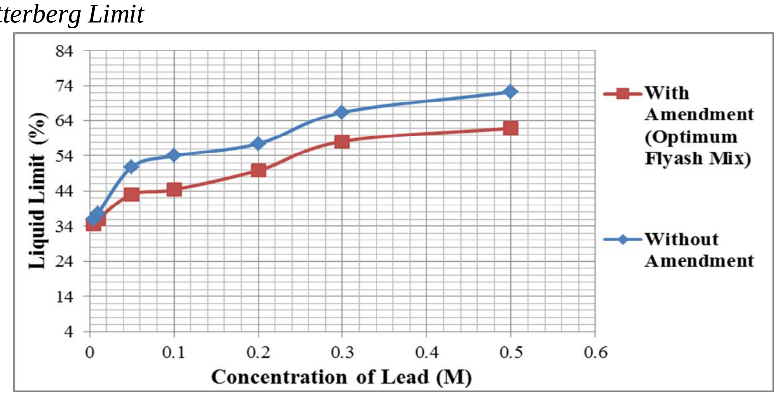

Fig 6. Variation of Liquid Limit on contaminated CL clay due to addition of amendment

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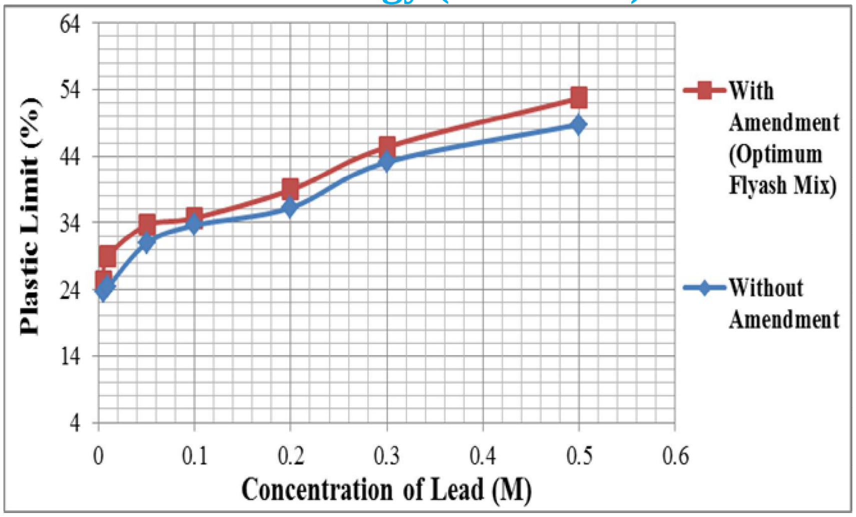

Fig 7. Variation of Plastic Limit on contaminated CL clay due to addition of amendment

As the concentration of lead increased the liquid limit, plastic limit increases which is a negative trend. This may be attributed to the relatively acidic pH obtained by the addition of heavy metal. At low pH the metal ion of the pollutant badly damages the soil structure thereby increasing its plasticity index. By the addition of amendments the liquid limit and plasticity index decreases which is a positive trend. This may be because of the effect of pH, by the addition of fly ash and cement of alkaline nature the pH was almost neutral which made the soil structure stable. Also the Si^{4+} and Al^{3+} in the fly ash might have replaced the metal ions in double layer hence reducing the double layer thickness and thus reducing the liquid limit. The plastic limit of most of the sample satisfies the typical criteria for soils (USEPA) (Table IV) and hence remediation is effective.

3) Effect of Amendment on UCC Strength

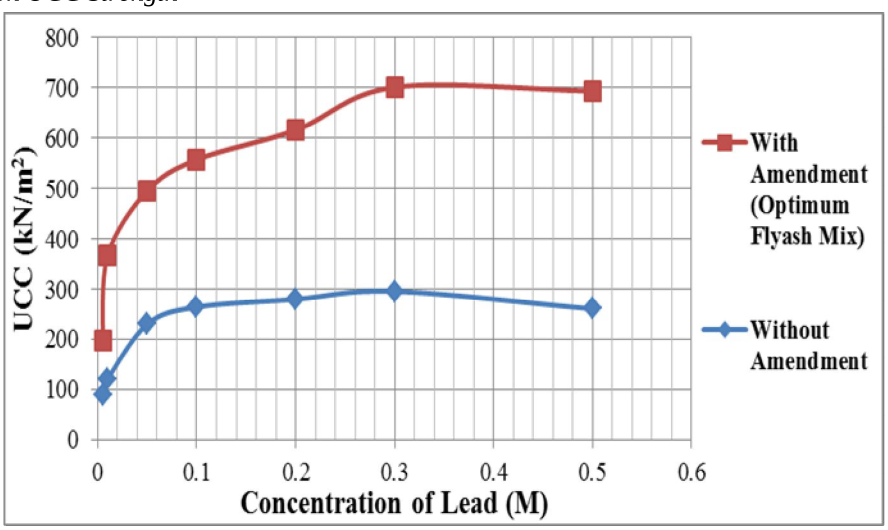

Fig 8. Variation of UCC on contaminated CL clay due to addition of amendment

As the concentration of lead increases the amount of lead adsorbed in clayey soils also increases to certain extent and tends to reduce the thickness of diffused double layer and thus causes flocculation. Thus an increase in strength characteristics of the soil is observed. However as the concentration increases UCC value starts decreasing since at increasing concentration of lead the soil becomes acidic and the structure of soil is damaged. But by the addition of amendment to the contaminated soil UCC value increased. This increase is strength is mainly due to the pozzolanic nature of fly ash and cementitious nature of OPC which caused a better bonding between the clay particles and thus increasing the UCS. The UCS of most of the samples satisfies the typical Solidification/Stabilization Criteria for soils (USEPA) (Table IV) and hence remediation is effective.

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4) Effect of Amendment on Permeability

TABLE V
VARIATION IN DRY DENSITY AND OMC WITH THE ADDITION OF VARYING PERCENTAGE OF FLY ASH

Concentration Of Lead (M)	Coefficient of permeability (cm/s) [Without Amendment]	Coefficient of permeability (cm/s) [With Amendment]
0.005	$4.50*10^{-7}$	$5.54*10^{-9}$
0.01	$5.64*10^{-7}$	$6.02*10^{-9}$
0.05	$6.01*10^{-7}$	$6.68*10^{-9}$
0.1	$6.83*10^{-7}$	$7.89*10^{-9}$
0.2	$7.68*10^{-7}$	$8.72*10^{-9}$
0.3	$9.29*10^{-7}$	$9.35*10^{-9}$
0.4	$1.16*10^{-6}$	$1.27*10^{-8}$
0.5	$1.27*10^{-6}$	$2.39*10^{-8}$

As the concentration of lead increased the permeability of the soil also increased. This may be attributed to the relatively acidic pH obtained by the addition of heavy metal. At low pH the metal ion of the pollutant badly damages the soil structure thereby increasing its permeability. By the addition of amendments the permeability is decreased because of the better bonding of clay particles by fly ash and OPC. Pozzolanic reactions will result in the formation of calcium aluminium and calcium silicate hydrate (CAH and CSH) cementitious products, which in turn provides for the physical behaviour enhancement of the treated matrix. The coefficient of permeability of most of the samples satisfies the typical criteria for soils (USEPA) (Table IV) and hence remediation is effective.

5) TCLP Test Results

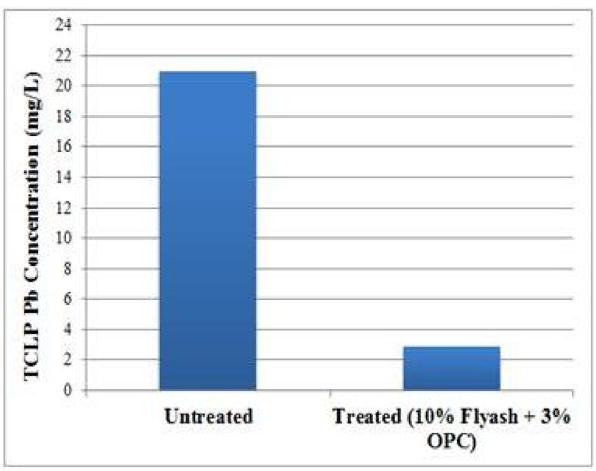

Fig 9. TCLP Pb concentration of untreated (control), and treated samples

Treated soils were tested for their regulatory levels of heavy metal leaching by means of conducting TCLP tests. Pb concentrations in the solution were measured using an ICP atomic emission spectrometer. The leaching solution was deionized water (pH 6.80).After 28 days of specimen curing, for 0.1M Lead acetate contaminated untreated cases, the amount of Pb release was 21mg/L. Whereas, following 10% of flyash and 3% OPC treatment, these values were sharply reduced to 2.9mg/L respectively, which indicated that levels of TCLP Pb release were below the regulatory benchmark of 5ppm when amendments were added to the contaminated soil as shown in fig 9. An efficiency of ~86% was obtained by this immobilization technique.

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IV. CONCLUSIONS

In this study, artificially Pb contaminated low plastic clay was immobilized/remediated using a mixture of Flyash and OPC. The effectiveness of the immobilization process was evaluated using compressibility, permeability, plasticity tests and TCLP. With the increase in salt concentration an increase in liquid limit, plastic limit, coefficient of permeability and UCC strength is observed. However with the addition of amendments liquid limit, plastic limit and coefficient of permeability decreased and UCC strength increased. The combined treatment of 10% Flyash and 3% OPC caused a drastic reduction in Pb (~86%) leachability based on TCLP test. The Plastic limit, coefficient of permeability and UCS of most of the samples and the TCLP test results satisfies the typical Solidification/Stabilization Criteria for soils (USEPA) (Table IV) and hence remediation is effective.

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