

## **Technical** Note

# Stabilization of fuel oil contaminated soil—A case study

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Abstract. Fuel oil contamination brings adverse effect on basic geotechnical properties of foundation soil. The present study pertains to one such case, from the petrochemical complex near Vadodara City in Gujarat State, India. Here, the fuel oil contaminated soil samples exhibit drastic changes in their geotechnical parameters. Noteworthy among such deleterious changes are: decrease in maximum dry density (-4%), cohesion (-66%), angle of internal friction (-23%) and unconfined compressive strength (UCS) (-35%) and increase in liquid limit (+11%). An attempt has been made to stabilize the contaminated soil using various additives viz., lime, fly ash and cement independently as well as an admixture of different combinations. It is apparent from the test results that the stabilization agents improved the geotechnical properties of the soil by way of cation exchange, agglomeration, and pozzuolanic actions. The best results were observed when a combination of 10% lime, 5% fly ash and 5% cement was added to the contaminated soil. The improvement in unconfined compressive strength (UCS), cohesion and angle of internal friction can be attributed to neo-formations such as Calcium Silicate Hydrates (CSH, CSH-1) that coats and binds the soil particles. Formation of stable complex between oil and metallic cations, results in reduction of leachable oil.

Key words. additive admixture, fuel oil contamination, neoformation, soil stabilization

## 1. Introduction

Hydrocarbon contaminants from oil exploration, transportation, production and processing affect the safety of civil engineering structures (Preslo et al., 1989; Nicholson and Tsugawa, 1996; Shroff et al., 1998). Some of the common deleterious effects due to oil contaminants are excessive settlement of tanks, breakage of pipelines, etc (Mackenzie, 1970). Several methods have been suggested to improve the oil contaminated areas (Troy, et al., 1994; Nicholson and Tsugawa, 1996; Mulligan, et al., 2001). Horizontal migration of the oil away from the sources of spillage, through the groundwater, can be controlled by construction of impervious barrier dykes adjacent to the tank (Shroff et al., 1998). Cleaning of the groundwater can be done by removal of the floating material (hydrocarbons) from pumped well water. Cleaning up of hydrocarbon-contaminated soil is a complicated task by virtue of high cost and limitations in disposing the excavated soil.



Figure 1. Location map of the study area

The investigated area, i.e. petrochemical industrial area around Undhera Village, Vadodara District, India (Figure 1) offers one such classical case wherein point source contamination resulted in degradation of sub-grade soil to an extent of 200 m<sup>2</sup>. The main source of fuel oil contamination is from leakage of storage tanks of petrochemical industry. Attempts had been made to check further lateral migration of contaminants (Shroff et al., 1998) by way of constructing impervious (bentonitic) dykes. However, treatment of contaminated soil is warranted for the overall reclamation of the land resource. Therefore, in this study, attempt was made to stabilize the soil and restrict the internal migration of the oil in line with the works of Morgan and Novoa, 1984; Pankoski et al., 1988; Preslo et al., 1989.

## 2. Methodology

The adopted methodology includes detailed sampling at a regular interval of 50 m covering the entire contaminated area. Sixteen disturbed soil samples were collected upto a depth of 2 m along with the insitu parameters such as natural moisture content and field density. The soils were subsequently evaluated for the quantity of fuel oil contamination following the methods of Standard Methods for Evaluation of Water and Wastewater (SMEWW, 1989). The weight percentage of oil was found to vary between 7% and 10%. Both contaminated and uncontaminated soils (eight samples) were analysed for index properties and classified as per unified soil classification scheme. As the soil (CL-Class) was exhibiting uniform index properties, strength parameters such as UCS, cohesion, angle of internal friction and permeability (IS 2720, Part 5,7,10,11) were carried out on three samples only.

Additional contaminated samples (four numbers) were generated by adding 10% of fuel oil to the uncontaminated soil at predetermined Optimum Moisture Content (OMC) and Maximum Dry Density ( $\gamma_{dmax}$ ) using mechanical mixer. The mixture was allowed to cure in closed container at ambient temperature for seven days. Subsequently, the mixture samples were evaluated for Atterberg limits and compaction characters. Results of these tests indicate that the laboratory generated samples do not vary significantly with respect to their field counterparts.

Contaminated soil was divided into a number of sets. Each set was then treated with additives of hydrated lime, Portland cement and fly ash individually at different weight percentages (5%, 10% and 20%) and as an admixture of four different combinations (10% lime + 10% fly ash; 10% cement + 10 fly ash; 15% lime + 5% fly ash and 10% lime + 5% fly ash + 5% cement). Maximum amount of additives used did not exceed 20% by weight due to economic considerations. Atterberg limits, and strength parameters of the treated soil samples were evaluated after curing the sample at room temperature for seven days. Three samples were tested from each batch and the results are expressed as a mean value. As the Atterberg limits improved significantly, when the soil was treated with an admixture combination of 10% lime, 5% fly ash, and 5% cement, further detailed studies were restricted to this combination only.

Changes in the cohesion and angle of internal friction are expressed as a ratio between stabilized soil and contaminated soil normalized to uncontaminated soil for quick interpretations.

Scanning Electron Microscopic (JEOL–T300) and X-ray diffractometry (Philips, PW 1720–Cu K $\alpha$  source) techniques were adopted for identifying the new minerals/compounds formed due to the action of stabilization agents.

## 3. Results

#### **3.1. INDEX PROPERTIES**

The soil is basically loamy silt with fine fractions varying between 48% and 52%. The Atterberg limits of the uncontaminated soil were: liquid limit–38%, plastic

limit–17.42%, plasticity index –20.58% and flow index –8.58%. It is apparent from the above details that the soil falls under **CL** class of unified soil classification scheme (**CI**- of IS: 1498–1970). The fuel oil contamination has caused significant changes in the Atterberg limits of the contaminated soil. This has been indicated by conspicuous increase in liquid limit (11%), plastic limit (34%) and decrease in plasticity index (7%). Similarly, fuel oil contamination resulted in lowering the  $\gamma_{\text{dmax}}$  and OMC also (Table 1).

On treating these oil contaminated soil samples with each one of the chemical additives separately at different percentages and as an admixture; the Atterberg limits have improved (Figure 2, Table 2). Addition of fly ash at different percentages i.e. 5%, 10% and 20 %, resulted in decrease of liquid limit (12–18%), plastic limit (20–27%) and plasticity index (4–8%). In the case of cement addition, reductions in both liquid and plastic limits were evidenced. Addition of lime did not yield any definite trend with respect to increase or decrease in Atterberg limits.

Similar results were obtained for different attempted combinations of admixture (Table 4). The best results were obtained (liquid limit 25%, plastic limit 19%, plasticity index 6%), when the contaminated soil was treated with the additive admixture having combination of 10% lime + 5% fly ash + 5% cement. The OMC (15.80%) and  $\gamma_{\rm dmax}$  (1750 kg/m<sup>3</sup>) also improved significantly when the soil was treated with 10% lime + 5% fly ash + 5% cement admixture.

Soil properties	Uncontamin	ated soil	Contaminated soil	
	Mean	S.D.	Mean	S.D.
Natural moisture content (%)	14.00	1.56	10.50	2.43
Insitu density (kg/m <sup>3</sup> )	1360	0.16	1080	1.87
Liquid limit (%)	38.00	0.86	42.00	1.77
			46.71*	1.44*
Plastic limit (%)	17.42	1.95	22.90	2.84
			$26.17^{*}$	$2.96^{*}$
Plasticity index (%)	20.58	1.91	19.10	2.37
•			$20.54^{*}$	3.91*
Flow index (%)	8.58	0.67	10.50	0.84
Maximum dry	1750	1.25	1680	0.11
density (kg/m <sup>3</sup> )			$1450^{*}$	$0.15^{*}$
OMC (%)	16.48	1.53	12.50	1.05
			$15.52^{*}$	$0.89^{*}$
UCS (kPa) 🛛	0.58		0.38	
Cohesion (kPa)	0.63		0.20	
Angle of internal friction $\square$	$18^{\circ}$	—	14°	_

Table 1. Index properties of uncontaminated and contaminated soils

 $\boxtimes$  – Average of three tests;

\* - Values for Laboratory generated specimens.



Figure 2. Observed range of variation in Liquid Limit

## 3.2. UNCONFINED COMPRESSIVE STRENGTH

The unconfined compressive strength of the fuel oil contaminated soil (37.27 kPa) was lower than that of the fresh or uncontaminated soil (56.88 kPa). Addition of these stabilization agents resulted in marked increase in UCS (Table 3, 4). The increase in UCS varied from 207–315% by lime addition; 200–218% by cement addition and 105–205% by fly ash addition. The highest strength of 138.28 kPa (371%)

Table 2. Atterberg limits for different additives in stabilized soil

	Additives								
	Lime		Fly ash		Cement				
Atterberg Limits	5%	10%	20%	5%	10%	20%	5%	10%	20%
Liquid Limit (%) Plastic Limit (%) Plasticity Index (%)	42.22 25.80 16.42	41.80 23.70 19.10	40.64 19.44 21.20	37.30 19.00 18.30	36.80 18.70 18.10	35.50 17.90 17.60	38.50 19.28 19.22	37.90 18.50 19.40	36.60 17.80 18.80

	U C S (kPa)			
Additives	5%	10%	20%	
Lime	77.47 (207%)	105.91 (284%)	117.68 (315%)	
Cement	74.53 (200%)	77.47 (207%)	81.40 (218%)	
Fly Ash	61.78 (165%)	68.65 (184%)	76.49 (205%)	

Table 3. UCS of stabilized soil with different additives

Values in parenthesis are % increase with respect to contaminated soil.

Table 4. Atterberg limits and UCS of soil treated with additive admixtures

Additives	LL	PL	PI	UCS in (kPa)
10% Lime + 10% Fly Ash	42.84	23.11	19.73	109.84 (294%)
10% Cement + 10% Fly Ash	38.46	18.45	19.96	87.28 (234%)
15% Lime + 5% Fly Ash	40.73	22.38	18.35	122.58 (328%)
10% Lime + 5% Fly Ash + 5% Cement	25.59	19.87	5.72	138.28 (371%)

Values in parenthesis are % increase with respect to contaminated soil.

higher than that of contaminated soil) was obtained when an additive admixture of 10% lime, 5% fly ash and 5% cement was added to the contaminated soil.

#### 3.3. COHESION AND ANGLE OF INTERNAL FRICTION

The angle of internal friction and cohesion of the uncontaminated soil were 18° and 58.84 kPa respectively. Contamination of fuel oil in this soil had caused reduction in both cohesion and angle of internal friction (19.69 kPa, 14°). Normalized variations of internal friction angle ( $\phi_{\text{stabilized soil}}/\phi_{\text{original soil}}$ ) and cohesion ( $C_{\text{stabilized soil}}/C_{\text{original soil}}$ ) with different percentage of additives were evaluated (Figure 3A, B).  $\phi_{\text{stabilized soil}}/C_{\text{original soil}}$  of the lime stabilized soil ranged between 0.78 and 0.88;  $C_{\text{stabilized soil}}/C_{\text{original soil}}$  counterpart ranged between 0.33 and 0.89. Similarly, the cement ( $\phi_{\text{stabilized soil}}/\phi_{\text{original soil}} = 0.78-0.83$ ;  $C_{\text{stabilized soil}}/C_{\text{original soil}} = 0.33-0.76$ ) and fly ash ( $\phi_{\text{stabilized soil}}/\phi_{\text{original soil}} = 0.78-0.79$ ;  $C_{\text{stabilized soil}}/C_{\text{original soil}} = 0.33-0.43$ ) stabilization also resulted in increase in angle of internal friction and cohesion.

The contaminated soil treated with an admixture of 10% lime, 5% fly ash, 5% cement resulted in increase in angle of internal friction ( $\phi_{\text{stabilized soil}}/\phi_{\text{original soil}} = 0.78-1.4$ ) and cohesion ( $C_{\text{stabilized soil}}/C_{\text{original soil}} = 0.33-3.4$ ). This quantum of increase in angle of internal friction and cohesion had not been achieved in the other three admixture combinations.

## 3.4. PERMEABILITY AND LEACHATE ANALYSIS

Coefficient of hydraulic conductivity of uncontaminated soil measured at its OMC and  $\gamma_{dmax}$  was of the order of  $6.779 \times 10^{-7}$  cm/sec., whereas in the case of fuel oil



*Figure 3.* Variation in normalized internal friction angle (3A) and normalized cohesion (3B) of contaminated soil with increased percentage of additives

contamianted soil this coefficient was  $4.753 \times 10^{-8}$  cm/sec. The leachates collected from the permeability test of four different samples (untreated soil, 5% cement treated, 10% lime treated and admixture of 10% lime, 5% fly ash, 5% cement treated) were analysed for oil content. These leachates were collected after 24 h of steady state water permeation. The leachate concentrations of oil obtained by this analysis are a measure of freely leachable oil. The oil content in the leachate of the untreated soil was 380 mg/l. When treated with 5% cement, this reduced to 107 mg/l and with 10% lime treatment, the concentration of leachable oil further reduced to 51 mg/l. The lowest leachable oil content (30 mg/l) was found in the soil treated with an admixture of 10% lime, 5% cement, 5% fly ash.

## 3.5. X-RAY DIFFRACTION ANALYSIS

X-ray diffractometry of both contaminated soil and soil treated with an admixture of 10% lime, 5% fly ash, 5% cement, (Figure 4 A and B) was carried out to evaluate the neo-formation of mineral/chemical compounds due to the action of stabilization agents.

The uncontaminated soil comprised mainly quartz, feldspar and clay minerals such as illite, montmorillonite and chlorite.

From the X-ray diffractogram it was evident that several new peaks (d = 3.353, 3.232, 3.219, 3.180, 3.029, 2.488, 1.993) were observed in the case of soil treated with



Figure 4. X-ray Diffractograms of (A) uncontaminated soil and (B) stabilized soil



*Plate 1.* SEM photomicrograph of uncontaminated soil (A), Stabilized soil-neoformed ribs (B), neoformed cement coating (C) and dense matrix (D)

stabilization admixture. Some of the newly developed peaks (d = 3.353 and 3.029) could be attributed to Calcium Silicate Hydrate (CSH). The rest of the new peaks could be attributed to other cementing compounds such as Calcium Aluminate Hydrates (CAH) and Calcium Alumino-Silicate Hydrates (CASH).

## 3.6. SEM ANALYSIS

To understand the changes in soil texture and to identify the neo-formations, both uncontaminated soil (Plate 1A) and admixture treated soil samples (Plate 1B) were subjected to SEM studies. The SEM photomicrograph indicated that the addition of stabilization admixture (10% lime, 5% fly ash, 5% cement) resulted in the formation of non-crystalline chemical compounds. These compounds coated over soil particles (Plate 1B, C) and bridged them. The newly developed coating over the sand-silt grains of soil enhanced the bridging action (ribs of about 3.6  $\mu$ m length) between the soil particles. These coating and bridging actions of neoformations resulted in the development of a strong cellular and nodular mass.

## 4. Discussion

Fuel oil contamination in soil causes adverse effects on the basic geotechnical properties of soil (Sridharan and Sivapulliah, 1987; Yaji, 1995; Tuncan, et al., 1996; Shroff, et al., 1998). In the present case, the fuel oil contamination of CL type of soil depicted increase in Atterberg Limits and decrease in UCS, cohesion, angle of internal friction and permeability. Effect of the fuel oil contamination and increase in Atterberg limits of the contaminated soil can be attributed to increase in double layer thickness of clay minerals such as kaolin, chlorite and illite (Shridharan and Sivapulliah, 1987; Sivapulliah, 1987; Tuncan et al., 1996). It is apparent from Figure 2 that the contaminated soil exhibits a wide range of liquid limits (41–46.5%), a phenomenon that can be attributed to inhomogeneous distribution of non-polar liquid (fuel oil) in soil. Excessive compressibility and reduction in shearing strength are some of the other adverse effects of hydrocarbon contamination in soils (Lancaster et al., 1996; Nicholson and Tsugawa, 1996).

The general procedures involved in treating such fuel oil contaminated soils are either by leaching (Young and Warith, 1989), evaporation (Paul and McLaren, 1975), stabilization (Nicholson and Tsugawa, 1996), or bio-remediation. In the present work, stabilization of soil using lime, cement and fly ash was attempted. The stabilization agents were added to the soil both independently at different weight percentages (5%, 10%, and 20) as well as an admixture of four different combinations. It is clear from Figure 2 and Table 2 that the addition of fly ash and cement resulted in the reduction of liquid and plastic limits. However, in the case of lime addition, liquid and plastic limits increased at 5% addition and subsequently decreased at 15% and 20% additions. These phenomena can be attributed to the **'amelioration effect'** i.e. immediate cation exchange, flocculation and agglomeration process (Akoto and Singh, 1981).

Fuel oil contamination affected the compaction characters of the soil (Table 1). Further, the improvement in atterberg limits contributeed to improvement of OMC and  $\gamma_{dmax}$  and hence the strength parameters (Doshi and Guirguis, 1982). Improvement of both Atterberg limits and OMC,  $\gamma_{dmax}$  similar to that of the uncontaminated soil was achieved when an admixture combination of 5% cement, 5% fly ash and 10% lime was used.

Addition of these stabilization agents at different weight percentages resulted in increase of unconfined compressive strength. The UCS progressively increased with increasing percentage of additives (Table 3). Manifold increase in the UCS was observed, when the contaminated soil was treated with an admixture of two or more of the said additives in different proportions (Table 4). Increase in UCS by 371% was observed when the soil was treated with an admixture of combination of 10% lime, 5% fly ash and 5% cement.

Similarly, the cohesion and angle of internal friction of the contaminated soil also showed a marked increase (Figure 3 A,B) after the addition of stabilization agents. Maximum values of normalized angle of internal friction ( $\phi_{\text{stabilized soil}}/\phi_{\text{original}}$ soil = 1.4) and cohesion (C<sub>stabilized soil</sub>/C<sub>original soil</sub> = 3.4) was observed in the case of above said admixture combination. The increase in UCS, cohesion and angle of internal friction can be attributed to both colloidal reactions (cation exchange, flocculation, agglomeration depression of double layers of clay minerals and cementing reactions (pozzuolana actions) among the additives and soil (De Silva et al., 2001; Mckinley et al., 2001; Prasanth et al., 2001). However, there was major decrease in the values of  $\phi_{\text{stabilized soil}}/\phi_{\text{original soil}}$  at lower percentages (5%, 10%) of lime. Factors such as immediate cation exchange, dispersive action can be attributed to this initial decrease in angle of internal friction (Nicholson and Tsugawa, 1996; Rajasekaran and Rao, 1995, 1997, 1998; Rao and Rajasekaran, 1996). But, subsequent increase in the strength parameters (20% lime addition) was due to both, pozzuolanic reactions (between lime and soil silica, soil alumina) and presence of excess of cation in soil. However, addition of fly ash at different percentages (Figure 3A,B) did not improve the cohesion and angle of internal friction. This phenomenon can be attributed to poor or no pozzuolanic action in the absence of lime.

X-ray diffraction studies (Figure 4B) aimed to identify the new compounds/minerals also indicated appearance of several new peaks in the treated soil. Some of these peaks (d = 3.353, 3.029) corresponded to Calcium Silicate Hydrates (CSH and CSH-1) (Akoto and Singh, 1981; Joshi et al., 1981). However, there were a number of additional peaks, which could not be identified (d = 3.232, 3.219, 2.488, 3.180) probably due to the existence of neoformations in gel form. SEM photomicrographs (Plate 1B, C) also indicated development of neoformations that coated and bridged the soil particles, thereby forming a dense, compact groundmass.

It is noteworthy to point out that with the additives, a progressive decrease in the percentage of leachable fuel oil was observed in contaminated soil (untreated soil: 380 mg/L, 5% cement treated: 107 mg/L, 10% lime treated: 51 mg/L, admixture treated: 30 mg/L). This progressive decrease in the leachable oil concentration can be assigned to the formation of a complex between metal and polycyclic aromatic hydrocarbons. Increase in the concentration of metallic ions due to the additives could have facilitated the formation of stable metal-fuel oil complex (Rao, 1997).

#### Conclusion

In this study, fuel oil contamination caused deleterious effects to the basic geotechnical properties of the CL type of soils. Oil contaminated soil when treated with different stabilization agents like lime, fly ash and cement either independently or as an admixture showed an improvement in the geotechnical properties. This improvement can be attributed to dispersion of oil, cation exchange, agglomeration, and pozzuolanic actions of additives namely lime, fly ash and cement. Best results were observed when soil was treated with a combination of 10% lime, 5% cement and 5% fly ash. In the process of stabilization fuel oil might have formed a stable complex with metals. Increase in the strength of the soil can be attributed to neoformation of compounds like CSH, CSH-1, that coat and bridge soil grains.

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