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Original Research Paper

Compressive Strength Development of Solidified PAHs Contaminated Soil Using Cement and Micro Silica

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ABSTRACT

High organics contaminated soil is a common concern for the solidification and stabilization (S/S) method. A probable problem with using S/S technology for the immobilization of organics is that these compounds are generally non-polar and hydrophobic; thus, they do not react with the inorganic binders and may interfere with solidification by disrupting the gel structure of the pozzolanic mix. In this research, the effect of microsilica (as an additive) in solidification of polycyclic aromatic hydrocarbons (PAHs) contaminated soil samples, collected from Ray Petrochemical Industry, was investigated. Solidification of soil sample was carried out by using cement and microsilica (MS) with different mass ratios. The behaviour of the solidified pastes has been analysed in terms of unconfined compressive strength (UCS). The results of this research indicated that the highest UCS of 1274.5 Kpa was achieved for samples contained 25% of cement and 8% of MS which was greater than the guidance value of 344.7 Kpa (50 psi) proposed by USEPA.

INTRODUCTION

In recent times, petroleum compounds contamination has become a major concern for the environment (Torabian et al. 2010). Leaking underground and above ground storage tanks, improper disposal of petroleum wastes, and accidental spill are major routes of soil and groundwater contamination with petroleum products (Nadim et al. 2000, Gitipour et al. 2011). Polycyclic aromatic hydrocarbons (PAHs) are one of such contaminants, consist of two or more benzene rings joined together comprising a group of ubiquitous environmental contaminants (Lang et al. 2012).

Soil contamination can be treated by varieties of methods like soil washing, soil flushing, solidification/stabilization, etc. Stabilization/solidification (S/S) system has been widely used for immobilization of metals and other contaminants in hazardous wastes, industrial sludges, power plant residues, municipal ashes, nuclear wastes, and contaminated soils and debris before final disposal since 20 years ago (Gong & Bishop 2003). Cement S/S techniques are generally low in cost, utilize straightforward technologies, and possess a potential for long term satisfactory performance (Leonard & Stegemann 2010).

However, a common concern about the S/S method is that it is much more viable for inorganic waste, but not suitable for the organic high content wastes such as petrochemical oily soil. A probable problem with using S/S technology for the immobilization of organics is that these compounds are generally nonpolar and hydrophobic; thus, they do not react with the inorganic binders and may interfere with solidification by disrupting the gel structure of the pozzolanic mix. Several additives like fly ash, modified clay, etc. can be utilized to solve this issue (Della 1989).

Microsilica (MS) is a by-product of the smelting process of silicon metal and ferrosilicon alloy production (Köksal et al. 2008). It is with amorphous structure, high SiO_2 content, and large surface area (about 20 m^2/g) makes it reactive with calcium hydroxide produced by cement hydration (Mark Atkins & Frederik 1989, Mehta 1989).

The aim of this research was to evaluate the effects of using MS in properties of the Portland cement used for solidification of high oily PAHs contaminated soil.

MATERIALS AND METHODS

Sample preparation and analysis methods: Soil samples were collected from a petrochemical industry (Rey Petrochemical Industry) which is located in the south-east of Tehran. To prepare the samples for geotechnical tests and PAHs analysis, the samples were air dried for 24 hours and screened by passing through a 60 mesh sieve. Following that, each of the samples were transferred to the glass jars (1,000 cm^3) and subsequently capped and placed in a refrigerator at 4°C for the tests. The soil properties were determined according to ASTM methods, consisting of gravel (4.3%), sand (69.5 %), and silt and clay (26.2%), and with moisture content of 38 % and pH of 6.21 (ASTM 2005a, 2007a, 2007b) (Table 1).

In the next phase, 5 g of each sample was extracted and weighted by a digital scale to prepare for gas chromatography (GC) analysis. To accomplish the GC analysis on the extract, the fluid was placed in a rotary evaporator for 15 min until its volume reached 1 cm^3. Following concentrating of the extract, 1 μL of it was injected into the GC equipped with a FID (340°C for the PAHs analysis).

The analysis results showed that the concentration of 4 polycyclic aromatic hydrocarbon presented in soil samples in mg/kg were: 328, 935, 607 and 284 for naphthaline, chrysene, Benzo[A]anthracene and benzo[A]pyrene, respectively.

Binders: Binders used for S/S of PAHs contaminated soil were Portland cement (PC) type II and MS. PC is considered as the most common S/S binder due to economical considerations and its availability. It is made by heating a mixture of limestone, clay and other materials, including fly ash and shale. A number of additives, such as activated carbon, fly ash, MS, modified clay and so on are available to improve the cement hydration process (Diet et al. 1998, Cheilas et al. 2007, Garces et al. 2008).

Typical properties of MS could be viewed in Table 2. The properties of MS depend on the type of producing and the process used for its manufacture. It is in form of spherical particle shape. The surface area of MS is about 20,000 m^2/kg and its average diameter is smaller than 1 μm. MS comes in three forms of powder, condensed and slurry. Its colour varies from light to dark grey which depends on the process in the manufacturing (Panjehpour et al. 2011, Wang et al. 2014).

Preparation of specimens: All the pastes were prepared by mixing contaminated soil, PC and MS with the water (Table 3). After thorough mixing, samples were transferred into the molds which have been made as per ASTM D 1633:00 method A with the height to diameter ratio of 1.15 (ASTM 2007).

Analytical methods: ***Unconfined compressive strength (UCS)***: UCS was measured as per ASTM D 2166M-13 method. All the samples were mechanically tested in 7, 14 and 28 days. All the experiments were conducted in duplicate (ASTM 2013).

RESULTS AND DISCUSSION

Unconfined compressive strength: Two groups of concrete mixes were investigated for compressive strength development. The first one was a series of 25% Portland cement mixed with 0, 4, 8 and 16% MS indexed as $C_{25}MS_0$, $C_{25}MS_4$, $C_{25}MS_8$ and $C_{25}MS_{16}$, respectively. On the other hand, the second group was mixed 35% Portland cement with the same ratio of MS indexed as $C_{35}MS_0$, $C_{35}MS_4$, $C_{35}MS_8$ and $C_{35}MS_{16}$, respectively. The average compressive strength results for both of the groups are presented in Table 4. Also, Fig. 1 and Fig. 2 are prepared for a better understanding of UCS results.

According to Table 4, the 28-day UCS of solidified soil blocks varied between 142.8 Kpa and 1274.5 Kpa for addition of 25% and 35% Portland cement, and 0% to 16% MS.

Table 1: Characteristics of soil samples in Ray Petrochemical Industry.

Soil Properties	Value
Gravel	4.30%
Sand	69.50%
Silt & Clay	26.20%
Moisture Content	38%
pH	6.21

Table 2: Typical of microsilica.

Fineness (m^2/kg)	15,000 to 35,000
Bulk density (kg/m^3) main elements	1350 to 1510
Silicon (% as SiO_2)	> 85
Aluminium (% as Al_2O_3)	< 2
Iron (% as Fe_2O_3)	< 1
Calcium (% as CaO)	< 1
Chloride (% as Cl)	< 0.3

Table 3: Mixing ratio.

Mix code	PC /Soil Ratio	MS/ Contaminated soil	Contaminated Soil	Water/PC (Grams)
$C_{25}MS_0$	0.25	0	55	0.35
$C_{25}MS_4$	0.25	0.04	55	0.35
$C_{25}MS_8$	0.25	0.08	55	0.35
$C_{25}MS_{16}$	0.25	0.16	55	0.35
$C_{35}MS_0$	0.35	0	55	0.35
$C_{35}MS_4$	0.35	0.04	55	0.35
$C_{35}MS_8$	0.35	0.08	55	0.35
$C_{35}MS_{16}$	0.35	0.16	55	0.35

Table 4: UCS results (Kpa).

Mix Code	Compressive Strength (Kpa)		
	7 Days	14 Days	28 Days
$C_{25}MS_0$	91	123.7	142.8
$C_{25}MS_4$	425.3	595.4	715.2
$C_{25}MS_8$	659.6	921.62	1274.5
$C_{25}MS_{16}$	170.71	245.75	302.45
$C_{35}MS_0$	132.1	181.8	209.88
$C_{35}MS_4$	509.5	702.2	1033.1
$C_{35}MS_8$	375.24	535.5	879
$C_{35}MS_{16}$	183.15	263.4	304.1

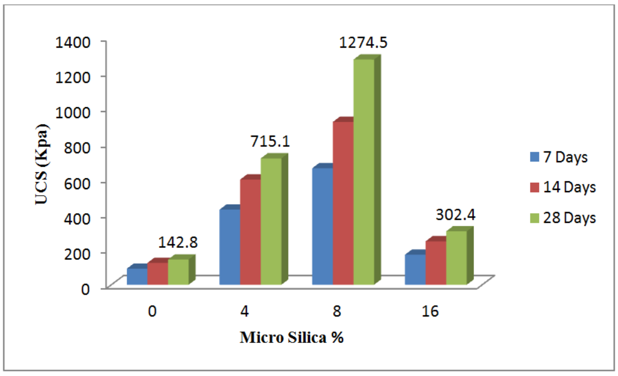

Fig. 1: Unconfined compressive strength for combination of 25% cement and microsilica.

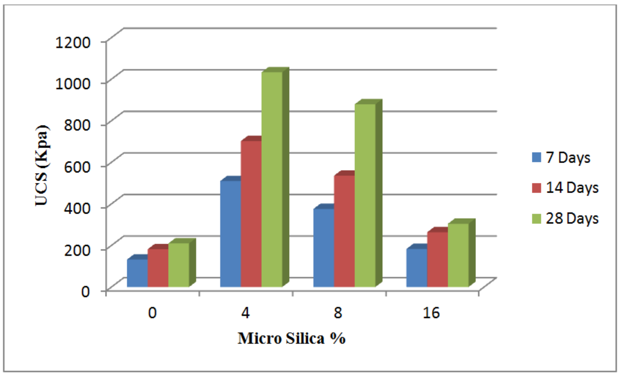

Fig. 2: Unconfined compressive strength for combination of 35% cement and microsilica.

The average strength of samples, with no MS, was 176.34 KPa, which delineates the effect of MS in S/S technique on increasing the strength of solidified samples.

With the combination of 25% PC and 8% MS, the highest strength of 1274.5 Kpa amongst all the 8 samples could be obtained after 28 days. Also the corresponding values were 1033.1, 879 and 715.2 Kpa, respectively, for $C_{35}MS_4$, $C_{35}MS_8$ and $C_{25}MS_4$. These observed values are greater than the guidance value of 344.7 Kpa (50 psi) proposed by EPA for successfully solidifying liquid waste before landfill disposal (USEPA 1999).

Equations 1 and 2 were adjusted for regression analysis of UCS of solidified soil after 28 days. These results indicated that UCS changes obey nonlinear parabolic function. Equation 1 is based on regression of the samples contained 25% of cement, furthermore, equation 2 stranded for the specimens with cement ratio of 35%. In these equations, MS is abbreviation of microsilica and the R^2 are 0.98 for both equations.

Eq. 1: $UCS = -1.1489MS^3 + 10.39MS^2 + 143.85MS + 47$

Eq. 2: $UCS = 1.5475MS^3 - 46.107MS^2 + 341.44MS + 306$

According to Fig. 1, 2 and Eqs. 1, 2, Pastes with no additive showed the value of 142.8 Kpa for $C_{25}MS_0$ and 209.8 Kpa for $C_{35}MS_0$ which are much lower than the ones solidified with addition of MS. Based on the above observations; it can be argued that the use of MS effectively improved the performance of solidified cement pastes.

As mentioned before, two ratios of cement were used to solidify contaminated soil. In solidified pastes with 25 percent of PC (Fig. 1), the lowest strength was related to the sample with no MS which was predictable according to the presence of organic contaminants and their negative effects on cement hydration. By adding MS, the compressive strength of pastes increased more than 5 times for the pastes with 4% MS (715.1 Kpa) and 9 times for specimens contained 8% MS (1274.5). Finally, adding MS more than 8% posed adverse effect on final compressive strength of the $C_{25}MS_{16}$ pastes decreased to 302.4 Kpa.

Furthermore, samples with 35% cement (Fig. 2) showed different behaviour compared to samples containing 25% of PC. Although the sample with no additive has not showed high compressive strength, but with addition of 4% of MS the final strength increased significantly (1033.1.3 Kpa was gained). Then, increasing more MS to the solidified pastes the compressive strength reduced (879 Kpa for C35MS8 and 304.11 Kpa for $C_{35}MS_{16}$). As per Figs. 1, 2 and 3, it can be observed that the ratio of cement content for the solidified samples can be reduce by using MS as an additive.

These observations were contributing to the facts that MS increases the water demand of concrete because of addition to concrete directly, not as a replacement of cement and also its' ultra fineness (Malhotra et al. 1987, Köksal et al. 2008). Also, Fig. 3 illustrates these results in a better way.

CONCLUSION

In the present study, UCS was conducted to evaluate the effect of adding MS on behaviour of solidification/stabilization of PAHs contaminated soil. The results showed that using MS for stabilization/solidification posed a good effect for gaining soil with higher strength. S/S offers technological advantages over the alternative remedial options for contaminated soils and sediments. It reduces the mobility of hazardous substances and contaminants in the environment by physical means, preventing further movement of the contaminant. Conclusions of this study are highlighted below:

1. The best combination of solidified samples studied for treatment of contaminated soil was 25% cement and 8% ($C_{25}MS_8$) MS and the specimen had strength of 1274.5 Kpa.
2. The increased amount of MS produced positive effect

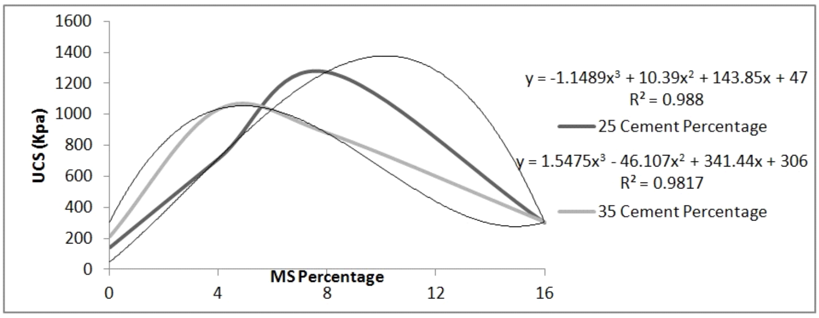

Fig. 3: Relation between UCS and MS percentage for solidified samples after 28 days.

on strength development of the samples with 25% cement until 8% whilst for specimens with cement ratio of 35%, adding more than 4% MS decreased the final strength of solidified samples.

3. Solidified samples with combination of 25% cement and different ratio of MS have better results in compressive strength after 28 days.
4. Increasing MS resulted in declined strength of the solidified samples due to the fact that MS increases the water demand of concrete because of addition to concrete directly, not a replacement of cement and also its ultra fineness.

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