The durability of stabilized sandy soil contaminated with $Pb(NO_3)_2$

S. R. Zeedan[1] A. M. Hassan[2] & M. M. Hassan[2]
[1]*Raw Materials and Processing Department,*
National Center for Housing and Building Research, Giza, Egypt
[2]*Sanitary and Environmental Engineering Department,*
National Center for Housing and Building Research, Giza, Egypt

Abstract

The purpose of this study was to investigate the effectiveness of cement kiln dust (CKD) and silica fume (SF) for the safe disposal of hazardous waste contaminated sand soil with toxic heavy metal as lead nitrate (Pb $(NO_3)_2$. The curing time and the leaching test through immersion in water are test methods for studying the durability of the stabilized/solidified (S/S) hazardous wastes under the soaking condition that leachate through the compacted samples. These methods simulate the leaching by immersion process; S/S hazardous waste under particular landfill conditions, when the S/S contaminated soil is more permeable than its surrounding materials or when the deterioration of solidified waste has reached a statue that ground water can flow – through the compacted soil via the porosity system of S/S waste matrix. The sandy soil was mixed with mixture binder (10% CKD and 2% SF) and mixed by dissolving three different ratios of $Pb(NO_3)_2$ (O.5, 1, and 3%) in tape water and compacted. UCS was determined for 1 and 3 month curing (100% moisture humidity) and for one year soaking in tap water. The concentration of (Pb^{+2} and NO^{--3}) which leachate from the compacted samples to the soaking water was recorded along a time of one year soaking. The UCS was decreased for the compacted samples have high levels of lead nitrate. The leaching of Pb was decreased during the soaking time for the two curing times (30 and 90 days) for the three ratio of $Pb(NO_3)_2$.
Keywords: solidification/stabilization, leaching, CKD, SF, heavy metal.

WIT Transactions on Biomedicine and Health, Vol 10, © 2006 WIT Press
www.witpress.com, ISSN 1743-3525 (on-line)
doi:10.2495/ETOX060271

1 Introduction

The type of waste, cost, legislation and technology limit disposal options for industrial waste. Studies have shown that solidification/stabilization (S/S) processes are viable for most metallic waste streams (Vipulanandan [1]), S/S of hydration waste involves mixing the waste with binder material to enhance the physical properties of the waste and to immobilize any contaminants that may be detrimental to the environmental conditions. Several binder systems are currently available and widely used for S/S (Mayers *et al* [2], Jones *et al* [3]). Binding reagents commonly used include Portland cement (OPC), cement kiln dust (CKD) lime and a number of proprietary reagents. Solidification and stabilization S/S immobilization technologies are the most commonly selected treatment options for metal-contaminated sites (Coner *et al* [4]). Solidification involves the formation of a solidified matrix that physically binds the contaminated material. In case of remediation projects, S/S is often the only reasonably available technology for treating the large volumes of heavy metals – contaminated soil, sludge, or sediments resulting from these operations cement or cementing material is uniquely suited for use as a S/S reagent for metal contaminants because it reduces the mobility of inorganic compounds by 1) formation of insoluble hydroxides, carbonates or silicates; 2) substitution of the metal into mineral structure and 3) physical encapsulation (Adaska *et al*. [5] and Bhatty *et al*. [6]). Alkalies in clinker kiln dust accelerated the setting and hydration of cement and quick setting time (QSA) influenced quick setting and increased the compressive strength of cement. The CKD and QSA modified by adding 2% SF accelerated the QSA. The least amount of heavy metals leached and the highest compressive strength was due to a large proportion of the formation of hydrates and the most effective stabilization of hazardous waste containing multiple heavy metals.

Cement stabilization is one of the most widely used waste stabilization methods for metal contaminants (Cheng *et al*. [7], Cocke [8], Li [9], and Poon and Chen [10]). This was due to the high buffering capacity and high pH value, where most metals are insoluble. Stabilization, also referred to as fixation, usually uses chemical reactions to convert the waste to a less mobile form. The general approach for solidification/stabilization treatment processes involves mixing or injecting treatment agents into the contaminated soils. Inorganic binders, such as cement, fly ash or blast furnace slag, and organic binders such bitumen are used to form crystalline glassy or polymeric frameworks around the waste. Abdel-Ghani *et al*. [11], studies the 10% CKD with 2% SF when compacted with sandy soil gives the best result and high resistance to environmental impact. The aim from reused the CKD waste as a binder material to solidified the contaminated sand soil with Pb $(NO_3)_2$ as economic process. Many studies show the effect of the flow rate of water on the solidification processes but this paper studies the effect of brackish water on the solidification process. By studying the durability effect on the compacted contaminated samples and on the effect of the brackish water by using a tap water as soaking media due to seepage or a rise in the groundwater level in landfill constructions as a leaching filled system. The UCS was determined as mechanical properties to study the ability of this stabilized soil to carry external loads and the maximum strength will be calculated to classify this stabilized soil and determine the kind of structure that will be established on it.

2 Methodology and material

2.1 The soil

Sandy soil as sample of a desert in Egypt from 6th October city.

2.2 Binders

The binding material was a mixture of 10% CKD and 2% SF of the total weight of the batch. It was used to solidify a synthetic contaminated sandy soil samples. The chemical compositions of both binders are given in Table 1.

Cement kiln dust (CKD) was obtained from Helwan Portland Cement Company as a waste material. Silica fume (SF) is a by-product of the smelting process for silicon metal and ferrosilicon alloy production. It was taken from the Egyptian company for Ferrosilicon Alloys situated in Edfo, Aswan.

Table 1: Chemical analysis of the soil and binders.

Symbol	Soil	CKD	SiF
SiO2 (%)	94.32	14.12	94.64
Al2O3 (%)	1.68	4.75	.97
Fe2O3 (%)	1.36	2.13	.93
CaO (%)	0.42	55.35	.55
MgO (%)	0.09	2.48	.35
SO3 (%)	0.47	5.80	0.10
Cl- (%)	.25	3.96	-
Na2O (%)	0.58	0.58	0.2
K2O (%)	0.12	3.53	2.5
L.O.I. (%)	1.22	12.60	2.01
PH	7.33	12.2	8.5
TSS	2.74	-	-
OM	--	-	-
TSS	2.74	-	-

2.3 The contaminants

Three different ratios (0.5, 1, 3%) of toxic lead nitrate were used as a synthetic source of pollution with heavy metal, and were separately added to the marked sandy soil samples.

 © 2006 WIT Press
www.witpress.com, ISSN 1743-3525 (on-line)

The research aimed to:

(a) use the waste CKD as a factor that chemically binds the polluted soil elements together, protecting the environment from the possible potential leak of the harmful content. CKD is waste material, so it is very economic to use as it serves double purposes: to get rid of it in a useful application instead of searching for an alternative method of disposal, and to limit the negative impact of the polluted sandy soil on the surrounding environment. CKD was deliberately selected because of its high alkalinity which helps to increase the efficiency of solidification of the soil, by chemically reacting with the contents, forming non-soluble salts of heavy metals resulting in immobilization of the toxic material. This action leads to the "imprisoning" of the dangerous content in the solidified soil in the shape of a non-soluble deposit.

(b) use the silica fume, which is also a waste material, with the cement kiln dust to enhance the soil strength and accelerate the setting of the hydration of CKD with the sand soil elements and accelerate the solidification process.

3 Solidified samples preparation

1) The sand soil groups of samples were polluted with 3 different ratios of $Pb(NO_3)_2$ then they were mixed with the binder mixture (10% CKD, 2% SF) with the optimum water content.
2) Samples were compacted employing the "modified effort" according to ASTM D [12].
3) There were typically three groups of samples, each of five sub-groups of samples to be cured for 1 and 3 months in a moisture ratio of 100%, soaked in tap water for one of the selected soaking times (30, 90, 180, 360 days), in enough quantities to afford the leaching pollution mobilized concentration and strength tests. They were kept in covered polyvinyl bottles at room temperature (25-30C°).

4 Testing procedure

The research program was carried out as follows:

- The strength of the curing stabilized soil specimens after soaking was determined using the unconfined compression test accordance ASTMD1633 [13].
- pH values were determined according to BS [14].
- the concentrations of Pb in the soaking water were determined after acidified with concentrated H_2NO_3 for each soaking according to AWWA [15].

 © 2006 WIT Press
www.witpress.com, ISSN 1743-3525 (on-line)

5 Results and desiccation

5.1 Unconfined compressive strength (UCS)

UCS tests were initially performed on samples that had undergone two curing conditions at 1 and 3 months as reference values to compare the results that will be obtained for soaked samples in tap water as soaking media for one year was illustrated in table [2].

5.1.1 Effect of soaking time on UCS

The continuous decrease of strength (UCS) of the compacted solidified samples contained 1% and 3% $Pb(NO_3)_2$ after one month curing for the compacted samples, contained 3% $Pb(NO_3)_2$ after 3 months curing and soaking for one year is due to the acidic effect resulting from the presence of 5.4% of SO4 in CKD and the presence of the NO_2 in high percentage in the $Pb(NO_3)_2$, in addition to the presence of CO_2 in the air. All of them, having been dissolved in water, are responsible for the acidification of the soaking water (by the H_2NO_3, H_2 SO_4 and H_2CO_3 respectively). These acidic waters in contact with the cementing materials give rise to the formation of calcium nitrates, sulfates, bicarbonates, etc. Some of these salts are highly soluble and easily leached, which causes pores that negatively affect the strength of the solidified samples. However, it has been recently proven that a strong synergy effect is produced by SO_2 and NO_2 when they exist together.

Table 2: Unconfined compression strength of solidified polluted sandy soil samples.

	0.5% Lead Nitrate				1.0 % Lead Nitrate				3.0% Lead Nitrate			
Time	1	3	6	12	1	3	6	12	1	3	6	12
	UCS Values				UCS Values				UCS Values			
Curing	3416	2949			3198	2797			1143	1099		
Soaking after 1Month curing	1732	2004	2320	3299	1594	1223	1059	900	546	472.3	176	5.4
Decrease of Strength %	49.30	41.33	32.08	3.43	50.16	61.76	66.89	71.86	52.23	58.68	84.60	99.53
Soaking after 3 months curing	2594	2656	3240	4076	2275	1598	2942	3324	862	762	689	548
Decrease in Strength %	12.04	9.94	-9.87	-38.22	18.66	42.87	-5.18	-18.84	21.57	30.66	37.31	50.14

According to the standard guide of the ASTM D4609 6 [16], the UCS of the chemically stabilized soil must be increased by 345(kN/m^2). These values can be attained after one and three months of curing for all solidified samples. And according to U.S.EPA standards [17] the minimum values of UCS are 3.5(kN/m^2) for the disposal of solidified hazardous wastes in landfills. UCS values for 0.5% of $Pb(NO_3)_2$ for one year soaking for both curing time also the UCS for 1% and 3% $Pb(NO_3)_2$ after curing or soaking.

5.2 Leaching of Pb by soaking (mobilized concentration)

The initial concentration of Pb for the three different ratios of contamination by Pb $(NO_3)_2$, (0.5, 1 and 3%) were (1220 mg/L, 2240 mg/L and 7320 mg /L) in the contaminated soil/cement samples respectively. The mobilized concentrations of Pb pollution leached from the compacted specimens (cured for 1 and 3 months, soaked in tap water for the scheduled times: 1, 3, 6 and 12 months) are given in Tables 3 and 4. The same results are graphically represented in Figs. 1 to 6. It is obvious from the results that the mobilized (leaching) concentration of lead has continuously decreased with the increase of the soaking time throughout the year's duration of the experiment with the exception of the 3% polluted samples which shown an increase in the mobilized concentration. However, the decrease in concentration of the leaching pollution was more than 99% in all cases compared to the initial concentration. These are extremely positive results. The initial concentration was cut down by more than 99% during the 1st month of soaking irrespective of the initial pollution status (0.5, 1, 3%) Curing the samples for 3 months showed better results (decreasing in the mobilized concentration) than curing them for one month. The compacted samples of 3 months curing emitted less leaching pollution than the samples of 1 month curing did.

Table 3: Mobilized concentration of Pb leached from solidified sandy soil samples polluted with Pb $(NO_3)_2$ different ratios (0.5,1,3)% by weight, soaked for 30, 90, 180 and 365 days, after 1 month curing.

Efficiency of Solidification ξ %									
Time of Soaking	Initial Concentration of Lead								
	% weight = mg/l								
	0.50%			1%			3%		
	(1220 mg/l)			(2440 mg/l)			(7320 mg/l)		
	Mobilized Concentr mg/l	Wt. of Mob. Concentr mg	ξ %	Mobilized Concentr mg/l	Wt. of Mob. Concentr Mg	ξ %	Mobilized Concentr mg/l	Wt. of Mob. Concentr mg	ξ %
30 Days	6.28	2.6	99.79	12.6	5.2	99.79	97	40.9	99.44
90 Days	3.2	1.35	99.89	21.8	8.99	99.63	115	47.4	99.35
180 Days	1.93	0.8	99.93	15.7	6.48	99.73	153	63.1	99.14
365 Days	0.78	0.32	99.97	6.28	2.59	99.89	187	77.1	98.95

Table 4: Mobilized concentration of Pb leached from solidified sandy soil samples polluted with Pb $(NO_3)_2$ different ratios (0.5,1,3)% by weight, soaked for 30, 90, 180 and 365 days, after 3months curing.

Efficiency of Solidification ξ %									
Time of Soaking	Initial Concentration of Lead								
	% weight = mg/l								
	0.50% (1220 mg/l)			1% (2440 mg/l)			3% (7320 mg/l)		
	Mobilized Concentr. mg/l	Wt. of Mob. mg Concent	ξ %	Mobilized Concentr. mg/l	Wt. of Mob. mg Concent.	ξ %	Mobilized Concentr. mg/l	Wt. of Mob. mg Concent.	ξ %
30 Days	4.15	2.6	99.79	10.8	4.5	99.82	73	30.1	99.59
90 Days	2.05	1.9	99.84	7.7	3.15	99.87	87	35.9	99.51
180 Days	0.6	0.53	99.96	2.3	0.95	99.96	113	46.6	99.36
365 Days	0.22	0.1	99.99	0.75	0.31	99.99	141	58.2	99.20

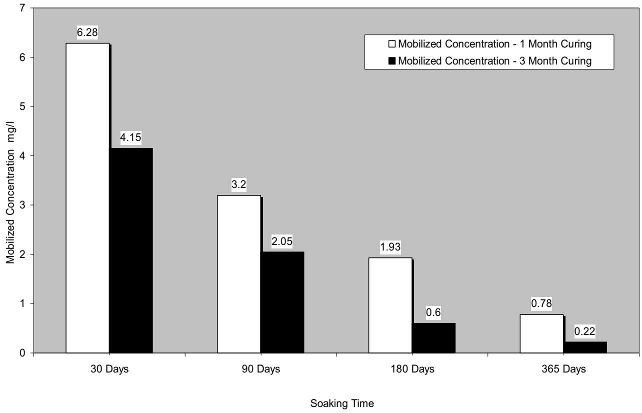

Figure 1: Mobilized concentration of Pb leached from solidified sandy soil samples polluted with Pb $(NO_3)_2$ 0.5% by weight, soaked for 30, 90, 180 and 365 days, cured for 1 and 3months.

The decrease in concentration of lead might be due the increase in the high pH value of the compacted samples (about 10-11) and the brackish soaking water (about 8.5-9.6) in the early time of soaking but this increased gradually to 10.5 to 11 in up to one year so that the solubility of lead decreased to its lowest solubility and precipitate in the form of insoluble salts (M. D. Largrega *et al.* [18]), the forming of insoluble precipitates in different samples producing higher hydraulic resistance and due to the hydration of a cementitous matrix and the formation of CSH gel which increased with time, so that when the water entered the samples by soaking, the porosity of the compacted samples were consequently reduced.

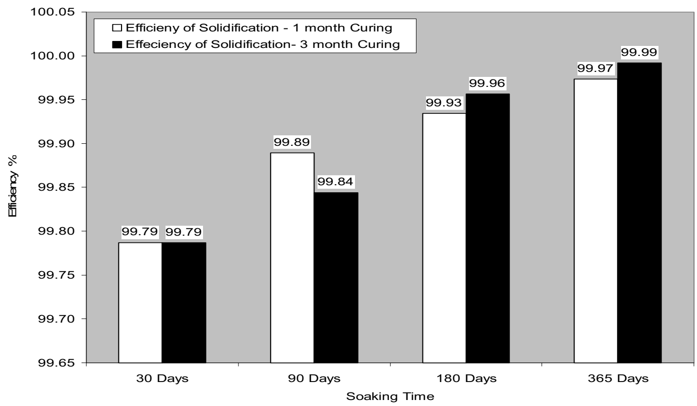

Figure 2: Efficiency of solidification effect on reduction of leachant pollution from samples polluted with Pb $(NO_3)_2$, 0.5% by weight = 1220 mg, soaked for 30, 90, 180 and 365 days, cured for 1 and 3 months.

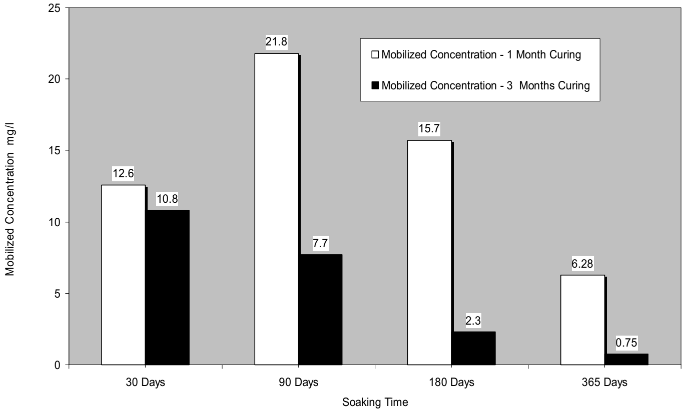

Figure 3: Mobilized concentration of Pb leached from solidified sandy soil samples polluted with $Pb(NO_3)_2$, 1.0% by weight, soaked for 30, 90, 180 and 365 days, cured for 1 and 3 months.

6 Conclusions

The results proved the feasibility of reducing the concentration of the leaching pollution (mobilized concentration) coming out of the polluted soil that had undergone solidification/stabilization, by curing (for 1 and 3 months), and soaking for up to 1 year.

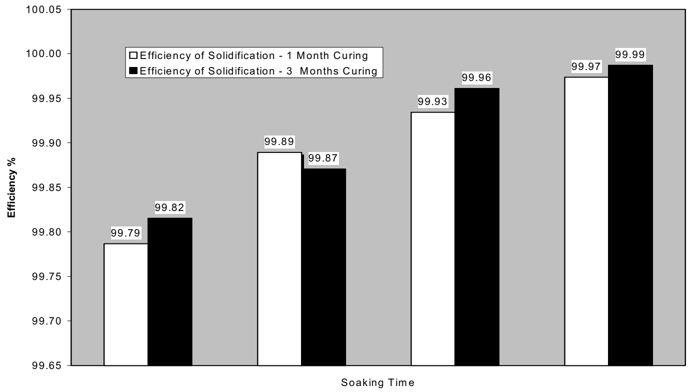

Figure 4: Efficiency of solidification effect on reduction of leachant pollution by lead after 1 and 3 months curing. Soaking time 30, 90, 180 and 365 days. Values for samples with 1% initial pollution.

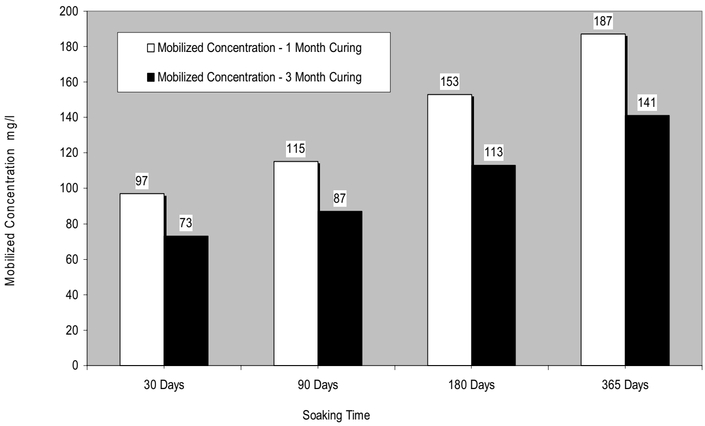

Figure 5: Mobilized concentration of Pb leached from solidified sandy soil samples polluted with Pb $(NO_3)_2$, 3.0% by weight=7320mg. Soaked for 30, 90, 180 and 365 days, cured for 1 and 3 months.

1. The level of pollution (by lead) of the water was reduced to less than the maximum allowable in the applicable Environment Laws. There would be no harmful effect if the water were to be used in agriculture, or if the water leaked into the underground.
2. Lead mobilized concentration was reduced to 0.22 mg/l and 0.75 mg/l for the samples with 0.5% and 1% initial pollution respectively, when cured for 3 months and soaked for 1 year.
3. Lead mobilized concentration was reduced to 0.78 mg/l and 6.28 mg/l for the samples with 0.5% and 1% initial pollution respectively, when cured for 1 month and soaked for 1 year.

4. Lead mobilized concentration was reduced to 187mg/l and 141mg/l for the samples with 3% $Pb(NO_3)_2$ initial pollution, when for 1 month, 3 months respectively and soaked for 1 year.
5. The strength (UCS) of samples with 0.5% and 1% $Pb(NO_3)_2$ initial pollution, was found to have decreased in the first six months of soaking, then increased in the 2nd half of the soaking time (1 year).
6. The strength (UCE) of samples with 3% $Pb(NO_3)_2$ initial pollution experienced a continuous gradual decrease throughout the 1 year of soaking. However, the value of UCS was found to be within the accepted limits, despite the continuous decrease.
7. The cement kiln dust (CKD) and silica fume mixture (10% CKD and 2% SF) can be used as solidified materials for $Pb(NO_3)_2$ contaminated sand soil in small ratios (0.5% and 1%).
8. All compressive strength results can be used in landfill structures.
9. The curing time plays a good role in the solidification/stabilization process.
10. With a high ratio of $Pb(NO_3)_2$, there must be a high ratio of CKD.

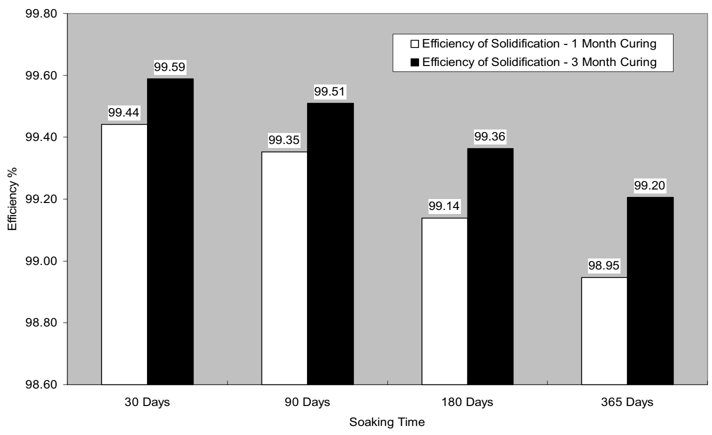

Figure 6: Efficiency of solidification effect on reduction of leachant pollution from samples polluted with Pb $(NO_3)_2$, 3% by weight = 7320 mg, soaked for 30, 90, 180 and 365 days, cured for 1 and 3 months.

References

[1] Vipulanandan, S. Krishnan, XRD analysis and leachability of solidified phenol -Cement mixtures, Cem, Conc. Res. 23(4) (1993) 792-802.

[2] T.E. Myers, A Simple procedure for acceptance testing of freshly prepared solidified waste, in: J.K.Peros Jr., W.J Lacy, R.A. Conway (Eds.), Hazardous and Industrial Solid Waste Testing and Materials, Philadelphia, PA, 1986 .

[3] L. W. Jones, Interference Mechanisms in waste solidification / stabilization processes, EPA-600/s-89/067(NTIS No. PB90-

156209/AS),USEPA. Risk Reduction Engineering Laboratory, Cincinnati, OH,1989 .

[4] Coner, J.R. Chemical Fixation and solidification of hazardous wastes; Van Nostrand Reinhold: New York, 1990.

[5] Adaska, W.S. et al. Solidification and stabilization of wastes using Portland cement, 2nd Edition ;EBO71; Portland Cement Association: Skokle, IL,1198

[6] Bhatty, J.L. et al. stabilization of heavy metals in Portland cement, Silica fume / Portland cement and masonry cement matrices; RP348; Portland Cement Association : Skokle, IL., 1999.

[7] Cheng, K.Y. (1991). Controlling Mechanisms of Metals Release from Cement –Based waste from in acetic acid solution, PhD. Thesis, University of Cincinnati, Cincinnati.

[8] Cocke, D.L. (1990) Journal of Hazardous Materials, 24,-231-253.

[9] Li, X.d,. Poon, C.S., Sun, H., Lo, I.M.C. and Kirk, D.W. (2001) Journal of Hazardous Materials,A82,215-230.

[10] Poon, C.S., Chen, Z.Q. and Wai, O.W.H. (2001) Journal of Hazardous, B81, 179-192.

[11] Abdel-Ghani, Kh., I., "Strength Durability of Sandy Soil Stabilized by Preheater Fines", Journal of the Egyptian Geotechnical Society, Cairo, Vol. 13, Part 1, June, 2002.

[12] ASTM D698-91, "Standard Test Method for Laboratory Compaction Characteristics of Soil Using Standard Effort", 1998.

[13] ASTM1633 compressive strength of molded soil cement cylinders

[14] British Standard Institution Methods of test for soils for civil engineering, BS1377, BS.1975

[15] Standard methods for the examination of water and wastewater .18th edition. Am. Works Assn. (AWWA), Washington, D.C, (1992).

[16] STM D 4609 "Standard guide for screening chemicals for soil stabilization (2002).

[17] U.S. EPA.(1996). Test. Methods for Evaluation solid wastes, Physical /chemical methods, SW-846, 3rd Edition. Office of solid waste and emergency response, Washington, D.C.

[18] Largrega.M.D.Buckingham P.L, and Evans J.C. Hazardous Waste Management, McGraw Hill Inc., USA, 640-696, 1994.