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Encapsulation of Contaminated Soil in Concrete Mortar

by Sreedevi Dawadi, M. R. Hansen, and Bruce W. Berdanier

A mixture of arsenic-contaminated soil and mortar was developed to study the effect of arsenic-contaminated soil on the strength of mortar and the effectiveness of the mortar in containing the arsenic. Preliminary laboratory analyses were conducted with spiked soil samples to verify percent recovery of the toxicity characteristic leaching procedure (TCLP). Five parameters were chosen for variation during the investigation: amount of contaminated soil (as a weight percentage of sand content), concentration of arsenic in the contaminated soil, water-cement ratio (w/c), amount of fly ash, and amount of silica fume. Compressive strength tests and TCLP tests were conducted on mortar samples. The test results showed that with the changes in the w/c, silica fume, and fly ash contents in the presence of arsenic, the compressive strength slightly increased from 7 to 28 to 90 days but did not show any major effect on the TCLP tests. The TCLP results were at the level (3 to 20 ppb) predicted to be established as the Drinking Water Equivalent Level (DWEL) by the United States Environmental Protection Agency (USEPA), indicating that mortar containing arsenic and contaminated soil may be a valid disposal solution.

Keywords: compressive strength; mortar; soil; water-cement ratio.

INTRODUCTION

Studies have been performed to investigate the technical feasibility, cost effectiveness, and implementation of stabilization/solidification (S/S) technology for the remediation of contaminated soils. Stabilization processes rely on additives that reduce the hazardous nature of a waste by converting the hazardous components of the waste into a form that minimizes the contaminant migration, reduces toxicity, or both. In contrast, solidification processes rely on the addition of reagents to a hazardous material, resulting in a solidified mass that typically has less compressibility and permeability. These processes have been successful in decreasing the potential for contaminant loss from a stabilized mass of materials including metals, volatiles, waste oils, and solvents. Disposing of the solidified/stabilized-contaminated soil in sanitary or secure landfills has been economical in the past,¹ but landfill space in secure landfills is at a premium, and incineration processes are becoming cost prohibitive and socially unacceptable. Therefore, the incorporation of contaminated soils into usable structural concrete may be a valid disposal solution. Properly designed concrete could be used at contaminated sites to build the structures needed for parking lots, access roads, wash-down areas, control structures, and pads for retention ponds.^{1,2}

The main objective of this project was to develop a mixture of contaminated soil and mortar (sand, cement, and water) with a low leaching potential through solidification and to evaluate the strength of the final monolith. The effects of contaminant level and contaminated soil proportion on the resultant strength were observed. The results of this preliminary study will be used to design a more intensive study to optimize the inclusion of the arsenic-contaminated soil in the mixture proportioning of structural concrete.

A homogenized soil sample was spiked with arsenic and mixed into mortar. The compressive strength and standard leaching potential were analyzed. A standard mortar mixture was incrementally modified by replacing the sand with contaminated soil. Standard cubes were made following the procedures of ASTM C 305³ by varying five parameters as follows: amount of contaminated soil, concentration of arsenic added to the soil as a contaminant, water-cement ratio (w/c), amount of silica fume, and amount of fly ash. The compressive strength was determined according to ASTM C 109 at 7, 28, and 90 days.⁴ The TCLP tests were performed following the USEPA Standards⁵ and analyzed to determine the maximum contaminated soil and concentration of arsenic that adversely affects the strength of concrete mortar. Three samples for both compressive strength and TCLP were evaluated for each mixture at each time period. The results presented in the graphs in this paper for compressive strength and TCLP represent the mean values for each mixture at each time.

Study of arsenic-contaminated soils

Inorganic forms of arsenic are much more toxic than organic forms.⁶ The principal valence states of arsenic are +3, +5, and -3.⁷ Arsenical pesticides, natural geothermal sources, and mine tailings increase arsenic concentrations in soils. The adsorption of arsenicals in soil depends on soil pH, texture, Fe, Al, and organic matter. The amount of arsenic adsorbed on soil increases as clay, Fe, and Al content increases. Toxic amounts of arsenic in soils will limit the germination of seeds and reduce the viability of seedlings having a concentration that is greater than 10 ppm. Organic arsenic is used in catalysts, glass manufacturing, alloys, electronics, and weed killer. Inorganic forms of arsenic are used to kill insects or rodents, to preserve wood, and as a medicine for asthma and psoriasis. Arsenic levels in municipal sewage are variable from 1 to 18 ppm. An upper limit of 0.2 ppm is recommended for arsenic in livestock drinking water and an upper limit of 0.05 ppm for water intended for human consumption. In soils, the total arsenic concentration normally ranges from 1 to 40 ppm.⁸ Currently, United States Environmental Protection Agency (USEPA) is considering amendment to the safe drinking water act to set the Drinking Water Equivalent Level (DWEL) at 5 ppb.

RESEARCH SIGNIFICANCE

Arsenic contamination resulting from natural or xenobiotic sources in ground and surface waters is a major health concern for waters designated for agricultural or human

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consumption uses. A great deal of time and money is being expended to conduct research and development of processes for removal of arsenic from such waters to concentrations as low as 5 ug/L. This research project is significant in developing recommendations for encapsulation of arsenic-contaminated soils in concrete mortar while limiting leaching potential and maintaining sufficient strength. This study provides operational boundaries for the initiation of a more detailed study of arsenic encapsulation in structural concrete. Additionally, this study can also form the basis for development of a methodology for the concrete industry to use in the protection of ground and surface waters from contamination due to the leaching of arsenic from contaminated soils.

Table 1—Proportions for soil samples

Mixture no.	Soil, g	Arsenic, mg/kg of soil	Arsenic, g
S1	25	0	0.000
S2	25	5	0.001
S3	25	50	0.005
S4	25	100	0.010
S5	25	200	0.021
S6	25	2000	0.208
S7	25	20,000	2.083

Table 2—Mortar proportions for 20 mixtures

Mixture no.	Cement, g	Sand, g	w/c	Soil, % of aggregate	Soil, g	Arsenic, mg/kg of soil	Arsenic, g	Fly ash, % of cement	Fly ash, g	Silica fume, % of cement	Silica fume, g
A1	740	2035.0	0.485	0	0.0	0	0.0000	0	0.0	0	0
A2	740	1933.3	0.485	5	101.8	0	0.000	0	0.0	0	0
A3	740	1831.5	0.485	10	203.5	0	0.000	0	0.0	0	0
A4	740	1729.8	0.485	15	305.3	0	0.000	0	0.0	0	0
A5	740	1628.0	0.485	20	407.0	0	0.000	0	0.0	0	0
A6	740	1831.5	0.485	10	203.5	0	0.000	0	0.0	0	0
A7	740	1831.5	0.485	10	203.5	5	0.0042	0	0.0	0	0
A8	740	1831.5	0.485	10	203.5	50	0.0424	0	0.0	0	0
A9	740	1831.5	0.485	10	203.5	100	0.0848	0	0.0	0	0
A10	740	1831.5	0.485	10	203.5	200	0.1696	0	0.0	0	0
A11	740	1831.5	0.450	10	203.5	50	0.0424	0	0.0	0	0
A12	740	1831.5	0.485	10	203.5	50	0.0424	0	0.0	0	0
A13	740	1831.5	0.520	10	203.5	50	0.0424	0	0.0	0	0
A14	740	1831.5	0.485	10	203.5	50	0.0424	0	0.0	0	0
A15	592	1831.5	0.485	10	203.5	50	0.0424	20	148	0	0
A16	444	1831.5	0.485	10	203.5	50	0.0424	40	296	0	0
A17	740	1831.5	0.485	10	203.5	50	0.0424	0	0.0	0	0
A18	703	1831.5	0.485	10	203.5	50	0.0424	0	0.0	5	37
A19	666	1831.5	0.485	10	203.5	50	0.0424	0	0.0	10	74
A20	629	1831.5	0.485	10	203.5	50	0.0424	0	0.0	15	111

MATERIALS

Commercially prepared topsoil was purchased for this study and dosed with the arsenic concentrations to have a consistent base material for addition of arsenic. Moisture content of the contaminated soil was determined using ASTM C 128.⁹

The cement used was Type I/II manufactured by GCC Dakota, Rapid City, S.D. Sand was commercial concrete sand obtained from a local concrete plant. Water was municipal tap water. Fly ash was Type F commonly used in the area. Silica fume was from Master Builders, Inc., and commonly used in the area.

EXPERIMENTAL DESIGN

Seven soil samples (S1 to S7), as shown in Table 1, were prepared and TCLP tests were performed to evaluate the percent recovery obtained. This preliminary testing was an evaluation of the experimental methodology for mixing the soil and the arsenic, performing the TCLP, and the calibration of the inductively coupled plasma mass spectrometry (ICPMS) unit. Arsenic was added to the soil in the form of sodium arsenate as shown in the Appendix.

Twenty mixtures (A1 to A20), as shown in Table 2, were designed for the study by varying the five parameters as described previously. Five mortar mixtures (A1 to A5) varied the percent of contaminated soil (0, 5, 10, 15 and 20%) to evaluate the adverse effects this range of percentages had on the compressive strength. Mixtures A6 to A10 maintained constant 10% contaminated soil and varied arsenic concentrations at 0, 5, 50, 100, 200 mg/kg of soil. Mixtures A11 to A20 held arsenic concentration constant at 50 mg/kg of soil, and contaminated soil was maintained at 10% of aggregate. The final ten mixtures were varied as follows: three mixtures (A11 to A13) with w/c of 0.45, 0.485 and 0.52; three mixtures (A14 to A16) with fly ash of 0, 20, and 40% by weight of cement; and four mixtures (A17 to A20) with silica fume of 0, 5, 10, and 15% by weight of cement were also prepared to determine their effect on mortar's

strength and on TCLP results while still maintaining the arsenic-contaminated soil as an aggregate.

After completion of the initial twenty mixtures, five additional mixtures (M1 to M5) were made by keeping the amount of arsenic constant at 10,000 mg/kg of soil and varying the amount of soil by 0, 10, 20, 30, and 40% of sand and constant *w/c* of 0.485 as shown in Table 3. An arsenic concentration of 10,000 mg/kg would be characteristic of hazardous waste sludge or severely contaminated soil. This concentration is within the range of treatability claimed by some commercial solidification products.^{10,11} This procedure will provide an indication of a high upper bound for the use of arsenic-contaminated soil in cement mortar.

Specimens

Specimens of 2 x 2 x 2 in. were made following the ASTM C 305-94 standard³ and water-cured for 7, 28, and 90 days, respectively. The cubes were tested for compressive strength following the standard test method for compressive strength of mortars given by ASTM C 109.⁴ The TCLP test samples were prepared following the USEPA standards.⁵ Leached arsenic concentrations were measured on a inductively coupled mass spectrometer (ICPMS). The ICPMS was calibrated to detection limits of part per billion for arsenic in the TCLP leachate and has capabilities into the part-per-trillion range.

RESULTS AND DISCUSSION

TCLP results on soil samples

Arsenic concentrations of 0, 5, 50, 100, 200, 2000, and 20,000 mg/kg of soil in 25 mg of soil were dosed for TCLP analysis. Figure 1 shows that the leaching of arsenic increased with the amount dosed. TCLP tests recovered close to 100% of the dosed concentration for all the soil samples.

Compressive strength and TCLP results for 20 mixtures

Effect of soil content—The compressive strengths for Mixtures A1 to A5 were determined and the mean values are shown in Table 4 after 7, 28, and 90 days of curing. From Fig. 2 it was observed that the strength appeared to decrease with the increase in the soil content. No TCLP tests were performed for these samples because they were not dosed with arsenic, and the purpose of Mixtures A1 to A5 is to settle on a level of contaminated soil to use in the remaining tests. Ten percent soil content was chosen for the remaining tests as sufficient for this initial analysis of a mortar that could be used in structural concrete mixtures. The 90-day compressive strength still had a mean value close to 5000 psi (30 MPa).

Effect of arsenic content—Mixtures A6 to A10 contained a constant 10% soil and *w/c* of 0.485. Varying concentrations of arsenic dosed as follows: 0, 5, 50, 100, 200 mg/kg of soil. Figure 3 shows that there appears to be no particular trend in the effect of this range of arsenic concentrations on the observed compressive strength.

The 7-, 28-, and 90-day TCLP results in Fig. 4 showed that the amount of arsenic leaching out in all the mixture samples was consistent at low arsenic concentrations over time. The 28-day leached concentrations, however, were considerably higher at initial arsenic concentrations over 50 mg/kg.

Effect of w/c—Mixtures A11, A12, and A13 were made with 10% soil and arsenic of 50 mg/kg of soil constant and *w/c* of 0.45, 0.485, and 0.52, respectively. Figure 5 showed that at 7 and 28 days of testing, there was a gradual decrease

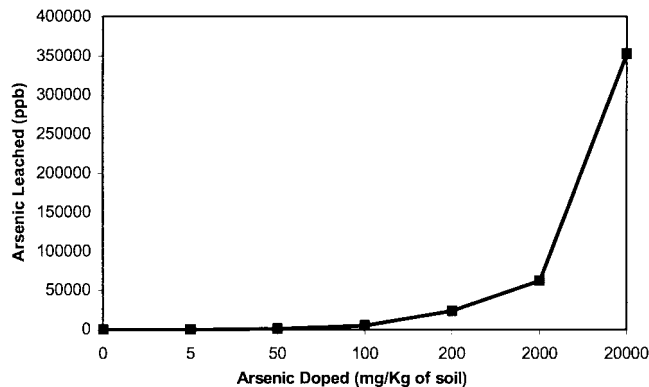


Fig. 1—TCLP results for arsenic content in soil samples.

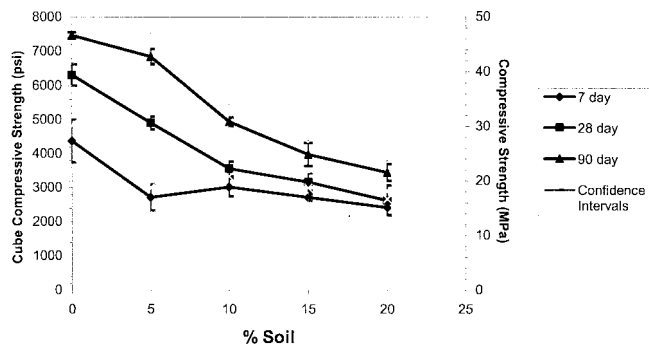


Fig. 2—Compressive strength as affected by soil cement.

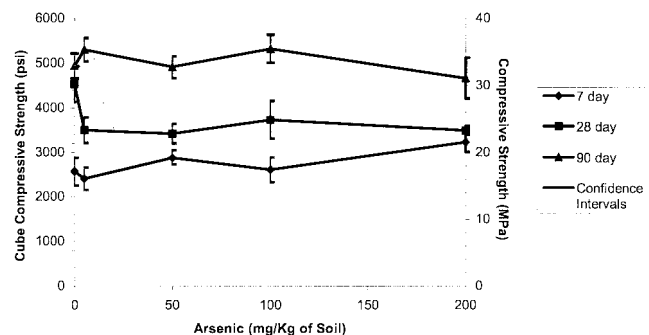


Fig. 3—Compressive strength as affected by arsenic content.

Table 3—Mortar proportions for final mixtures

Mix ture no.	Cement, g	Sand, g	<i>w/c</i>	Soil, % of aggre- -gates	Soil, g	Arsenic, mg/kg of soil	Sodium arsenate, g	Fly ash, % of cement	Silica fume, % of cement
M1	740.0	2035.0	0.485	0	0.0	10,000	0.0000	0	0
M2	740.0	1831.5	0.485	10	203.5	10,000	8.4792	0	0
M3	740.0	1628.0	0.485	20	407.0	10,000	16.9583	0	0
M4	740.0	1424.5	0.485	30	610.5	10,000	25.4375	0	0
M5	740.0	1221.0	0.485	40	814.0	10,000	33.9167	0	0

in the compressive strength, which indicated that the *w/c* of 0.45 was the best proportion to use for getting higher compressive strength.

The 7- and 90-day TCLP results in Fig. 6 showed that the amount of arsenic leaching out in all the three mixture samples appears to be approximately equal with no effect from the *w/c*. The 28-day TCLP results indicated higher levels of arsenic leaching on the order of two to four times the 7- and 90-day values.

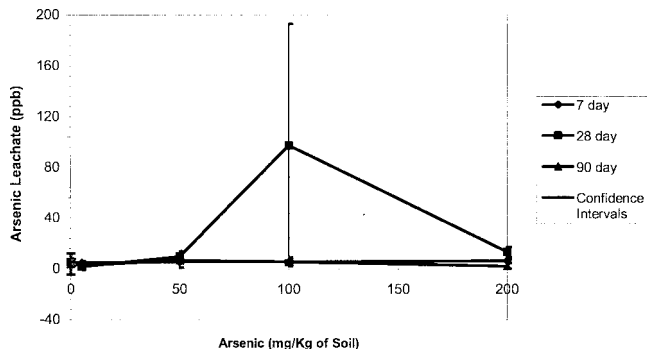


Fig. 4—TCLP as affected by arsenic content.

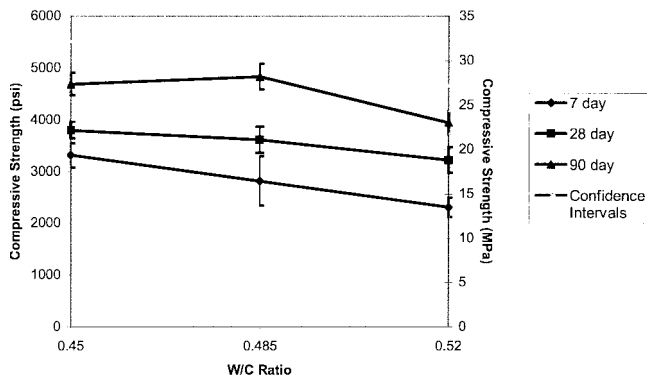


Fig. 5—Compressive strength as affected by w/c.

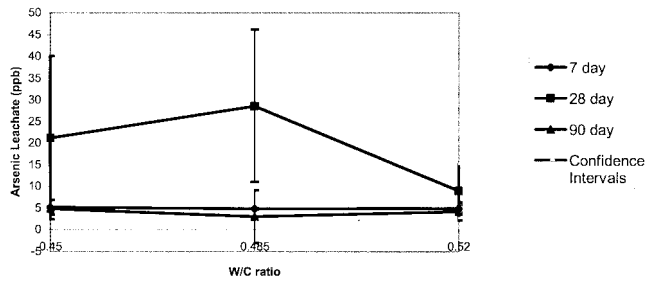


Fig. 6—TCLP as affected by w/c.

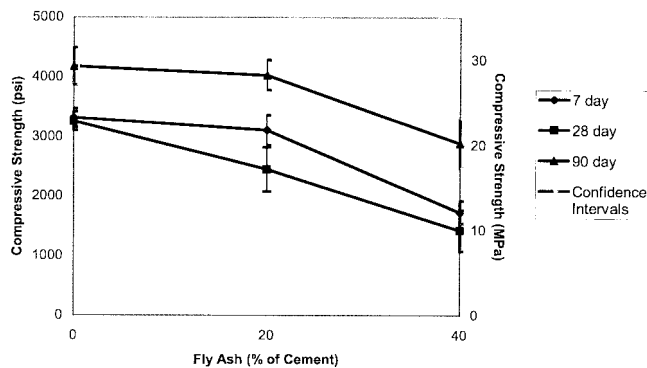


Fig. 7—Compressive strength as affected by fly ash content (initial mixture).

Effect of fly ash content—With fly ash as the variable, Mixtures A14, A15, and A16 were made with 10% of soil, arsenic of 50 mg/kg of soil, w/c of 0.485 constant, and fly ash of 0, 20, and 40% of cementitious material as shown in Table 2. Figure 7 showed that at 7- and 28-day tests, there appears to be a gradual decrease in the compressive strength

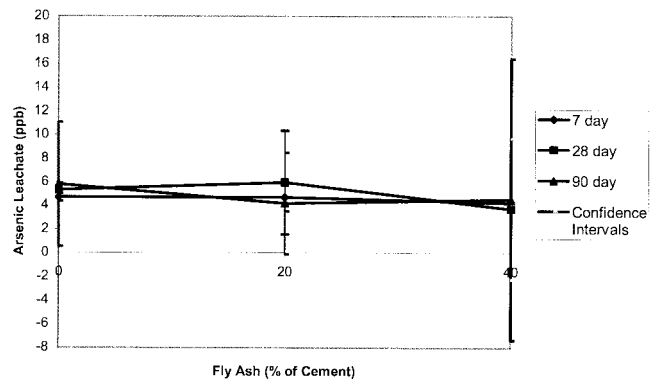


Fig. 8—TCLP as affected by fly ash content (initial mixture).

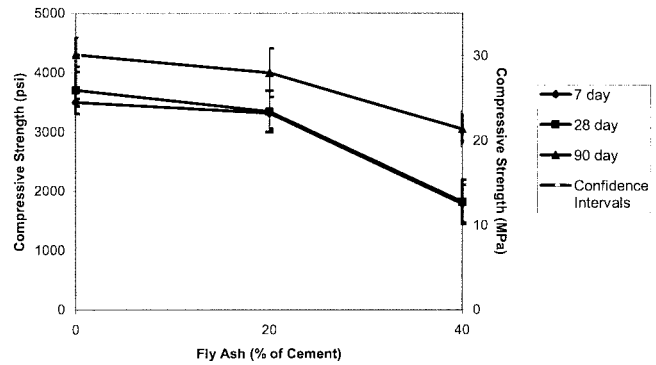


Fig. 9—Compressive strength as affected by fly ash content (repeated mixture).

Table 4—Compressive strength as affected by soil cement

Mixture no.	% soil	7-day compressive strength		28-day compressive strength		90-day compressive strength	
		psi	MPa	psi	MPa	psi	MPa
A1	0	4380	30.1	6320	43.5	7470	51.4
A2	5	2720	18.7	4910	33.8	6850	47.1
A3	10	3020	20.8	3560	24.5	4950	34.0
A4	15	2720	18.7	3170	21.8	3980	27.4
A5	20	2420	16.6	2630	18.1	3450	23.7

with increasing fly ash content, but at 90 days, all strengths are higher, as is normal with mixtures containing fly ash.

The 7-day TCLP results for Mixtures A14, A15, and A16 in Fig. 8 showed that the amount of arsenic leaching out was approximately the same with increasing fly ash percentage and over time. The results are all within a 2 ppb range.

Because the strength decreased from 7 to 28 days, three new mixtures with the same proportions were completed to verify the results. The results shown in Fig. 9 indicate that there was decreasing strength at 7 and 28 days, and the strength at 28 days of all the cubes was slightly higher than that at 7 days. The 90-day compressive strength results were all higher but decrease in strength with the increase in the fly ash content.

The 7- and 28-day TCLP results for the repeated mixtures indicated lower concentrations of arsenic in the leachate compared to the initial mixtures. The concentration of arsenic leachate appeared to consistently increase over the 90-day testing period for all the mixtures, as shown in Fig. 10. However, the results still appear to be around 5 ppb or less. The percent average retained was 97.2%, which indicates that

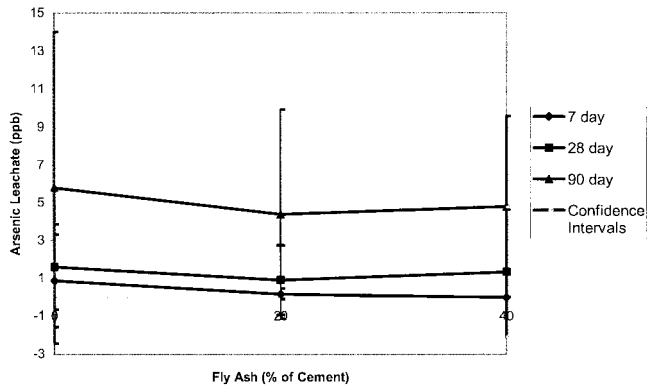


Fig. 10—TCLP as affected by fly ash content (repeated mixture).

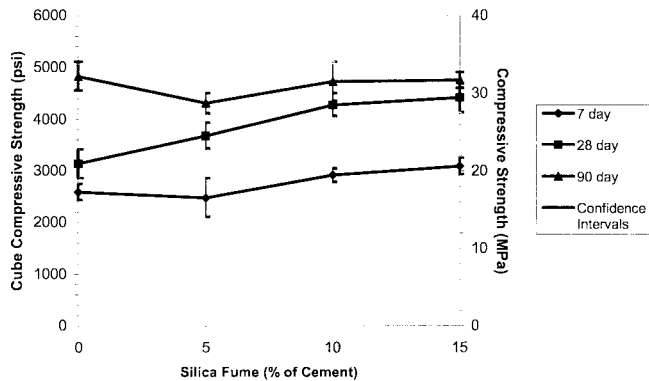


Fig. 11—Compressive strength as affected by silica fume content.

fly ash mortar is a good solidifying material in binding wastes. Further studies may be warranted to investigate the effects of fly ash on strength and arsenic leachate concentration at very low arsenic concentrations and low fly ash percentages.

Effect of silica fume content—Mixtures A17, A18, A19, and A20 were made with 10% soil, arsenic of 50 mg/kg of soil, w/c of 0.485 and silica fume of 0, 20, and 40% of cementitious material as shown in Table 2.

The results in Fig. 11 indicate that 7- and 28-day strength slightly increased with percent silica fume. At 90-day tests, the strength decreased at 5% of silica fume and there was a gradual increase at 10 and 15% silica fume content. There is no significant strength benefit from silica fume.

The TCLP results did not indicate a consistent relationship between the presence of silica fume in the mortar mixture and arsenic concentration in the leachate as shown in Fig. 12. The resultant leachate concentrations were all at the 6 ppb level or less.

Compressive strength and TCLP results for final mixtures

Mixtures M1 to M5 were made with arsenic of 10,000 mg/kg of soil; 0, 10, 20, 30, and 40% of aggregates; and w/c of 0.485 as shown in Table 3.

The compressive strength tests and TCLP tests were performed to study the effect of very high concentration of arsenic and increasing proportions of soil content on mortar strength. Mixture M1 is the control mixture and has the highest compressive strength. Figure 13 indicates a decrease in the compressive strength for the mortar as soil content is increased up to 20%. The 7- and 28-day compressive strengths for the last two mixtures containing 30 and 40% of soil could not be

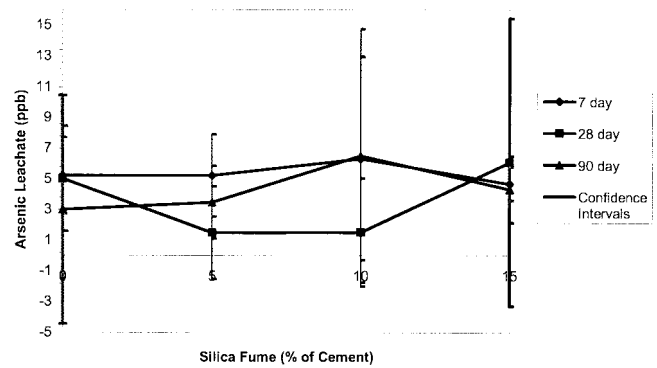


Fig. 12—TCLP as affected by silica fume content.

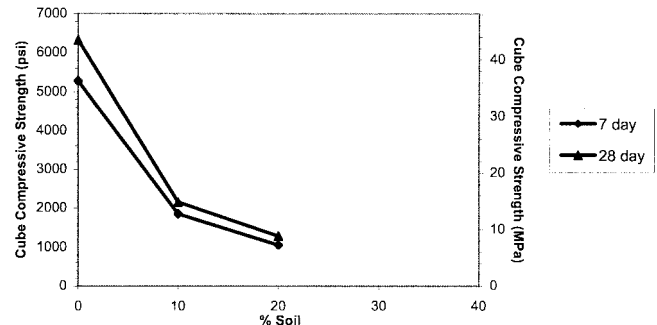


Fig. 13—Compressive strength as affected by soil cement.

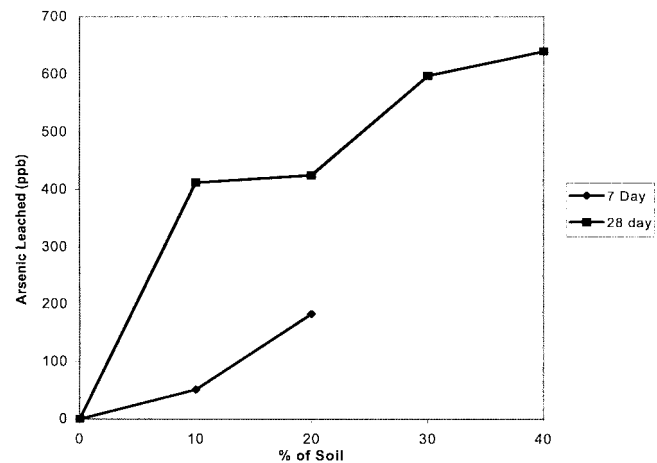


Fig. 14—TCLP results as affected by soil content.

obtained, as the setting time for these mixtures was more than 28 days.

From these results, it appears that the addition of high-arsenic-concentration soils (on the order of 10,000 mg/kg) beyond 10% soil to the mortar mixture would greatly reduce the final compressive strength for structural concrete.

Figure 14 shows the results of the TCLP tests for the high-arsenic-concentration mixtures. As expected, the concentration of arsenic in the leachate increased with percentage of soil. The leachate concentrations are higher than would be accepted if the methodology were used for protection of groundwater. The 7-day TCLP tests were not performed on the last two mixtures, as their set time was more than 7 days. Figure 14 also indicates that the leaching potential increases with time, as the results for the TCLP at 28 days are considerably higher than those at 7 days. It should be noted that although the mortar did not retain the arsenic to the low ppb levels

required for drinking water protection, the method still shows promise as the leached concentration is on the order of less than 1% of the initial dosage.

STATISTICAL ANALYSIS

Linear regression analysis was performed on all of the data for compressive strength and TCLP in relation to the varying of arsenic concentration, w/c, fly ash, and silica fume. Trends observed were found to have low slope values have low correlation coefficients.

Statistical analysis was performed on compressive strength results for mixtures (A1 to A20) and on TCLP results for mixtures (A6 to A20) to evaluate the statistical significance of the results in relation to curing time. One-way analysis of variance (ANOVA) using statistical software was performed for each mixture. Resultant *p* values were considered significant if they indicated that the hypothesis—that the means were equivalent—could be rejected at a level lower than 0.05.

The experimental results showed that the compressive strength appeared to increase from 7 to 90 days for all of the mixtures. The resultant *p* values from the ANOVA confirmed that the compressive strength increase is statistically significant from 7 to 90 days for all of the mixtures.

The TCLP test was performed for the mixtures (A6 to A20) containing arsenic. The ANOVA results showed that apparent increases in the TCLP concentrations from 7 to 28 days were significant for all of the mixtures except for the mixtures with varying percent fly ash and for the 15% silica fume mixture. Apparent decreases in TCLP concentrations from 7 to 28 days were significant only for the mixtures with arsenic at 5 mg/kg and with silica fume at 5%. Apparent decreases in TCLP from 28 to 90 days were significant only in the mixtures with arsenic at 100 and 200 mg/kg and in the mixtures with w/c at 0.45 and 0.485. Apparent increases in TCLP from 28 to 90 days were significant in fly ash at 40%, and in silica fume at 5 and 10%.

CONCLUSIONS AND RECOMMENDATIONS

Based on the results of this study, the following conclusions can be made:

1. The strength of the mortar mixtures decreased with the increase in the soil content, fly ash percentage, and w/c;
2. The concentration of arsenic in the leachate increased with time for the mixtures containing 10,000 mg of arsenic/kg of soil and soil content of 10, 20, 30, and 40%. The evaluated method reduced the leachate concentration to approximately 1% of the dosage concentration but would not meet currently proposed groundwater protection levels for this high of concentration;
3. Testing at 7, 28, and 90 days indicated that the TCLP concentration of arsenic in the leachate and the compressive strength was approximately the same for all dosage concentrations up to 200 mg arsenic/kg soil; and
4. Testing of mortar encapsulation of arsenic-contaminated soil (up to 200 mg arsenic/kg soil) indicates that the method was successful in reducing the arsenic concentration in leachate to approximately 5 ppb.

Recommendations for future research

Future research should be focused on studying the effects of arsenic-contaminated soil on structural concrete.

Additional investigation should be conducted into the interfacial chemistry reaction(s) that determine the ability of the mortar to encapsulate the arsenic-contaminated soil.

Future research should identify the concentration of arsenic contaminated soil in the 1000 to 10,000 mg arsenic/kg soil range that can be successfully encapsulated to meet Environmental Protection Agency standards based on TCLP.

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APPENDIX—CALCULATIONS TO DETERMINE AMOUNT OF ARSENIC REQUIRED

The calculations used to determine the amount of arsenic required for dosing in the soil are explained as follows:

The compound taken was sodium arsenate, Na₂HAsO₄ · 7H₂O. The atomic weight of Na₂HAsO₄ · 7H₂O is

$$= 2(22.989) + 1(1) + 1(74.9216) + 4(15.994) + 7[2(1) + 15.994]$$

$$\frac{311.8946 \text{ g}}{\text{mole}} \text{ Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$$

The amount of arsenic in Na₂HAsO₄ · 7H₂O is calculated as follows:

$$\frac{\text{mole Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}}{311.8946 \text{ g Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}} \times \frac{74.9216 \text{ g As}}{\text{mole Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}}$$

$$\frac{100 \text{ mg As}}{\text{g As}} = \frac{240 \text{ mg As}}{\text{g Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}}$$

Therefore, 1 mg As/kg of soil =

$$\frac{\text{g Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}}{\text{g soil}} \times \frac{1000 \text{ g soil}}{\text{kg soil}} \times \frac{240 \text{ mg As}}{\text{g Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}}$$