Solidification/stabilization of arsenic containing solid wastes using portland cement, fly ash and polymeric materials

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Abstract

Stabilization/solidification (S/S) is used as a pre-landfill waste treatment technology that aims to make hazardous industrial wastes safe for disposal. Cement-based solidification/stabilization technology is widely used because it offer assurance of chemical stabilization of many contaminants and produce a stable form of waste. The leaching behavior of arsenic from a solidified/stabilized waste was studied to obtain information about their potential environmental risk. Activated alumina (AA) contaminated with arsenic was used as a waste, which was stabilized/solidified (S/S) using ordinary portland cement (C), fly ash (FA), calcium hydroxide (CH) and various polymeric materials such as polystyrene and polymethyl methacrylate (PMMA). Toxicity characteristics leaching procedure (TCLP) and semi-dynamic leach tests were conducted to evaluate the leaching behavior of arsenic. Formations of calcite along with precipitate formation of calcium arsenite were found to be responsible for low leaching of arsenic from the stabilized/solidified samples. Effective diffusivity of arsenic ion from the matrix and leachablity index was also estimated. Minimum leaching of the contaminant was observed in matrix having AA + C + FA + CH due to the formation of calcite.
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1. Introduction

Various treatment technologies, such as coagulation, precipitation, ion exchange and adsorption, have been extensively used for the removal of arsenic from water but they pose problems of generation of bulky and toxic solid and liquid arsenic rich waste. Activated alumina is one of the promising technologies, which has shown good potential for removal of arsenic from water [1,2].

The sludge generated with high levels of arsenic cannot be disposed of directly into the landfill due to risk of underground water contamination. The high concentration of arsenic in the leachates requires treatment before disposal to a landfill. The solidification/stabilization (S/S) treatment is employed to reduce the mobility of arsenic so as not to present an environmental hazard. Various techniques for sludge disposal have been used depending upon the type of substance to be disposed and environmental conditions prevailing. These include portland cement processes, lime-based technology, bituminization, emulsified asphalt processes, polyethylene extrusion and vitrification [3–9].

Currently, the US Environmental Protection Agency (USEPA) recognizes cementitious solidification as the "best demonstrated available technology (BDAT)" for land disposal of most toxic elements. Alkaline matrices such as $Ca(OH)_2$ and cement are commonly used in waste conditioning because they are inexpensive, readily incorporate wet wastes and their alkalinity reduces the solubility of many inorganic toxic or hazardous metals. U.S. Department of Energy [10] performed experiments in an attempt to quantitatively define the waste form performance response to the four binding constituents (i.e. portland cement, fly ash, blast furnace slag, clays), as well as to different combinations and concentrations of the phosphate salts. Results of waste form performance testing indicated that the use of blast furnace slag and fly ash increased the allowable salt loading in cement, but the use of clays resulted in forms with unacceptably low compressive strength (<500 psi).

Cement and hydrated lime $(Ca(OH)_2)$ and other pozzolanic materials such as fly ash have been widely and successfully used for the stabilizion of As [11–13]. Diet et al. [14]

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studied the behavior of portland cement during the S/S of synthetic heavy metal sludge. They observed that cement hydration of the metal hydroxide sludge depends on nature and amount of treated sludge. Rouf and Hossain [15] reported that arsenic and iron sludge could be used for brick making. Results also indicated that the sludge proportion and firing temperature were the two key factors determining the quality of brick. Roy and Eaton [16] studied the solidification and stabilization of a synthetic electroplating waste in lime-fly ash (LFA) binder. They observed that the microstructure of LFA is not significantly affected by the addition of sludge; the presence of sludge only affects the degree of hydration. The consistent presence of the heavy metals on fly ash surfaces indicated that fly ash helps in S/S by adsorbing the waste species. Increase in Fe/As mole ratios has shown no benefit in the immobilization of arsenic. The leaching of calcium was linked with the leaching of arsenic for all solidification/stabilization formulations. Increase in the calcium leachate concentration resulted in immediate decreases in the arsenic concentrations [17,18].

The present study aims at developing an appropriate method for the safe disposal of arsenic [As(III)] rich activated alumina waste as As(III) is more toxic and mobile than As(V). This waste was a product of the process where arsenic [As(III)] has been successfully removed by activated alumina [2].

2. Materials and methods

The waste material studied for disposal was originated from the arsenite removal by activated alumina adsorption process, the results of which have been given elsewhere [2]. All the experiments were carried out using double distilled water. Ordinary portland cement (C), fly ash (FA), $Ca(OH)_2$, polystyrene and polymethyl methacrylate (PMMA) have been investigated for stablization/solidification studies. Both polystyrene and PMMA were used in the cylindrical beads form (1 mm diameter and 2.5 mm length). These materials were mixed with arsenic solid waste in the ratio as shown in Table 1.

2.1. Preparation of monolithic matrices

The binder materials (C, FA, CH, PS and PMMA) were mixed with arsenic solid waste in a definite ratio as given in Table 1. Six different solidified waste matrices were prepared by mixing different compositions of binder materials. Water was added in a proportion to make slurry of these binders and arsenic rich waste materials.

For the preparation of solid matrices, PVC pipe of diameter 0.016 m and length 0.064 m were cut into pieces to give a length to diameter ratio of 4.0 as per the ANSI/ANS standards. One of the ends of the empty pipe was blocked with a flat plastic plate and the arsenic waste along with other desired materials (C, FA, CH, PS and PMMA) were added to the pipe from the other end. These PVC moulds were kept at a temperature of 298 ± 2 K for 24 h for curing and setting of the matrices. Initial arsenite loading of 125 mg/g (12.5% w/w arsenic trioxide) of solid matrix materials was kept for all the samples. Ratio of leachant volume to volume of specimen was kept as 1:10 (v/v) as per ANSI standard method [19].

2.2. Toxicity characteristic leaching procedure (TCLP)

Before proceeding for toxicity characteristic leaching procedure (TCLP) leaching tests, the solid matrices was crushed. The particle size of the crushed sample was kept less than 9.5 mm according to the requirement of the TCLP procedure. For semi-dynamic leaching tests, monolithic matrices were used.

The TCLP test used in this study followed the standard procedures described by USEPA. For this test, 10 g of waste samples were weighed and kept in a polypropylene bottle. 0.2 l of the TCLP extracting solution (CH_3COOH or NaOH) was added to each bottle. These samples were tumbled in a shaker at temperature 298 K for 18 h. At the end of the extraction, the leachates were filtered with Whatman glass fiber filter paper. The pH of the filtrate was measured. The leachates were maintained highly acidic by adding nitric acid (pH < 2) to avoid any change in arsenic concentration.

2.3. Semi-dynamic leachability test

Semi-dynamic leach tests are generally used to determine the leachability of contaminants from monolithic solidified waste forms. The term 'semi-dynamic' means that the leachant is replaced periodically after intervals of static leaching. In this study, the stabilized/solidified waste samples were leached in a closed vessel without agitation using distilled water at a leachant to solid mass ratio of 10:1 (Fig. 1). Leachates were collected after a fixed duration of 2, 7, 24, 48, 72, 96, 120, 456, 1128 and 2160 h (90 days) as per ANSI/ANS 16.1 standard [17–20].

Runs were made in triplicate in order to confirm the reproducibility of the data and average values were used for the interpretation of the data. The mean, standard deviation, coefficient of variance and standard error was calculated for the

Table 1
Composition of different S/S waste samples

| Matrices composition	Notations	Ratio (w/w)
Activated alumina + cement	AA + C	3:1
Activated alumina + cement + fly ash	AA + C + FA	3:1:0.5
Activated alumina + cement + fly ash + calcium hydroxide	AA + C + FA + CH	3:1:0.5:0.5
Activated alumina + cement + fly ash + polystyrene	AA + C + FA + PS	3:1:0.5:0.5
Activated alumina + cement + polystyrene	AA + C + PS	3:1:0. 5
Activated alumina + cement + polymetylemethacralate	AA + C + PM	3:1:0.5
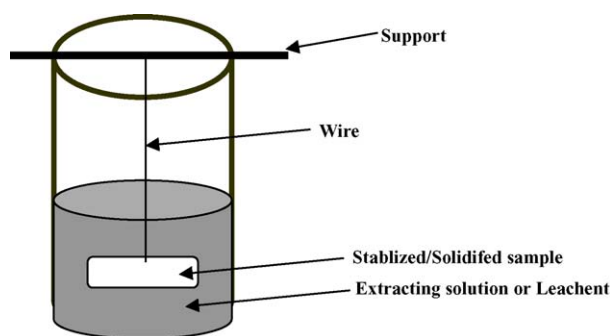
Fig. 1. Experimental set up for leachability studies from S/S samples for safe disposal of arsenic.

analysis of the data. The results were reported with 95% confidence limit. The D_e and L values reported are mean values of 10 leaching interval.

2.4. Diffusion coefficient D_e and leachability index L (ANS method)

Leachability of arsenic from the different materials was evaluated using the ANS 16.1 method. The cumulative fraction of arsenic leached versus time is determined. Usually, a mathematical diffusion model based on Fick's second law is used to evaluate the leaching rate with respect to time [12,19]. ANS has standardized a Fick's law-based mathematical diffusion model as follows:

$$D = \left(\frac{\frac{a_n}{A_0}}{\Delta t_n}\right)^2 \cdot \left(\frac{V}{S}\right) T \tag{1}$$

where D is the effective diffusivity (cm^2/s), V the volume of specimen (cm^3), S the geometric surface area of specimen (cm^2), $(\Delta t)_n$ the duration of nth leaching interval (s) and a_n, A_0 are the concentrations of solidified species at nth leaching time and at the beginning.

2.5. Leachability index

The leachability index is a quality parameter, which is limited to the material for which it was determined. It is meaningful only if the specimen did not undergo significant change during the leach test and if the actual waste form to which it applies does not undergo change in its material characteristics. To evaluate the leachability of a diffusing species, leachability index was calculated as:

$$L = \left(\frac{1}{m}\right) \sum_{n=1}^{m} \left(\log\left(\frac{b}{D_e}\right)\right)_n \tag{2}$$

where $b = 1$ cm^2/s,

$$L = \left(\frac{1}{m}\right) \sum_{n=1}^{m} (-\log(D_e))_n \tag{3}$$

where n is the number of the particular leaching period and m is the total number of individual leaching period. This leachability

index can be used to compare the relative mobility of arsenic species on a uniform scale, which varies from 5 ($D_e = 10^{-5}$ cm^2/s very mobile) to 15 ($D_e = 10^{-15}$ cm^2/s immobile).

2.6. Effective diffusion coefficient (penetration theory)

As most S/S waste forms have relatively low permeability, diffusion is normally the main operating mechanism. The driving force of diffusion is the difference in the chemical potential between solid and fluid leachant fluid. According to the penetration theory, assuming a constant diffusion coefficient, the diffusion flux (J) across the solid/solution interface of a semi-infinite medium has been given by Dutre and Vandecasteele [9,18]:

$$J = \sqrt{\frac{D_e}{\pi t}} \cdot C_0 \tag{4}$$

where C_0 is the initial concentration of the leaching substance in the S/S waste specimen (mg/cm^3), D_e the effective diffusion coefficient (cm^2/s) and t is the leach time (s). The total amount of arsenic leached that has diffused out of the medium surface at time t, M_t can be derived from Eq. (4) by integration over time and surface area, and is given by Dutre and Vandecasteele [17]:

$$M_t = 2M \frac{S}{V} \sqrt{\frac{D_e t}{\pi}} \tag{5}$$

where M, S and V are the total initial amount of arsenic in the S/S waste specimen (mg), surface area of specimen (cm^2) and volume of specimen (cm^3), respectively.

Diffusion is mainly responsible for leaching of arsenic from S/S matrices. Assuming a constant diffusion coefficient for each S/S matrix, cumulative fraction of a substance (CFR) can be defined as a ratio of M_t to M and is given by:

$$\text{CFR} = \frac{2}{\sqrt{\pi}} \frac{S}{V} \sqrt{D_e t} \tag{6}$$

2.7. Mould characterization

To ascertain the mechanism of arsenic leaching and other micro structural properties, XRD and SEM analysis was performed. X-ray diffraction (XRD) patterns of the different matrices were obtained from a Philips Mat. Holland PW 1730/10 diffractometer to determine the arsenic immobilization mechanism. The X-ray was generated with a current of 30 mA and a potential of 40 kV. The scan was performed between 2θ values of 10° and 70° at a scan rate of 0.06° min^{-1}.

For scanning electron microscope (SEM), samples were pasted on a brass bar with adhesive. The sample was then coated by silver in JEOL (JFC-1100E Ion) sputtering device and transferred into the JEOL (Cambridge 360) scanning electron microscope) sample chamber where an accelerating voltage of 15–40 kV was used for scanning.

2.8. Analytical methods for arsenic determination

A Varian Spectra AA 220 atomic absorption spectrophotometer equipped with a graphite furnace was used to analyze arsenic

concentrations in solution. All measurements were based on integrated absorbance and performed at 193.7 nm by using a hollow cathode lamp (Varian, Australia). Pretreatment temperature was 1400 K; atomization temperature was 2500 K. The calibration range taken was 20–100 μgl^{-1}. Solution pH was measured with a CD APX 175 pH meter (Scientific Systems, India). The analysis was conducted in triplicate to check the reproducibility of results. Acid-washed (30% HNO$_3$) HDPE plastic bottles were used for TCLP tests.

3. Results and discussion

3.1. Leaching of waste material

The leachability of arsenic ions from activated alumina waste was examined by submitting the waste to the extraction test. Ten grams of waste material was added to a volume of 100 ml of distilled water and was shaken for a period of 24 h. Arsenic was found to be the most easily leachable element as arsenic concentrations in the leachate was more than 2 g/l in 24 h. These results were compared with USEPA standards, which define when a waste sample should be classified as hazardous. When the untreated waste is leached in distilled water, the pH of the leachates varies between 3 and 3.5, which indicates the acidic nature of the waste.

Arsenic rich exhausted activated alumina was crushed and subjected to TCLP in static leaching test. The results showed that the standard TCLP test was unable to differentiate the leachability of metal contaminants in various cement-based waste matrices. As all cement-based matrices have strong buffering capacity, which neutralizes the extraction solution and limit metal leaching process, semi-dynamic leaching tests were undertaken to study the mechanism of leaching.

3.2. Semi-dynamic leaching

The results of different solidified waste samples subjected to semi-dynamic extraction test are shown Fig. 2 in which the concentration of arsenic in the leachate, at the end of each leaching period, is plotted as a function of time. It is essential for the semi-dynamic leach tests that no equilibrium should be reached during any period of static leaching. Higher leaching of arsenic was observed in case of waste samples having composition of AA + C + PS and AA + C + PM. The initial sharp rise in arsenic concentration leached from monolithic stablized/solidified (S/S) samples could be attributed to mass transfer controlled processes whereas diffusion becomes predominant at the later stages.

Effective diffusion coefficient (D_e) was calculated from the slope of the plot of CFR as a function of square root of time t. This was proven by the linear relationship that was exhibited when plotting the cumulative fraction released value versus the square root of time as can be seen from Fig. 3. Effective diffusion coefficients thus calculated from the slope of these plots were also confirmed with ANSI/ANS 16.1 [19].

Diffusion coefficient calculated for the various matrices have been shown in Table 2. Matrix prepared from AA + C + FA + CH showed lowest diffusion coefficient (8.93×10^{-11} cm^2/s) than
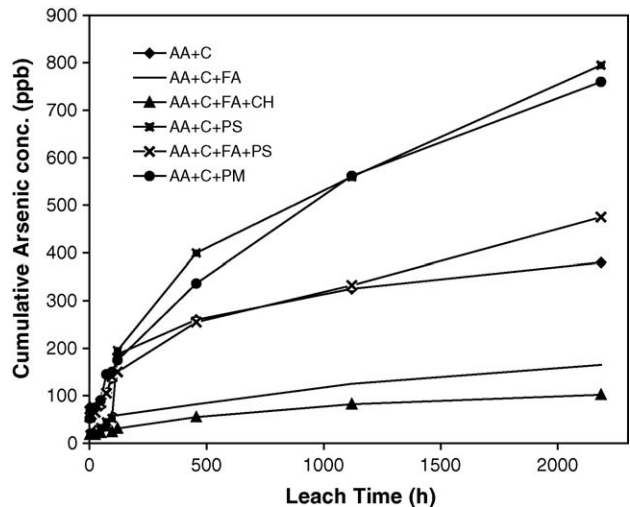

Fig. 2. Cumulative concentration of arsenic in the leachate of S/S waste as a function of leach time for the semi-dynamic leach test.

other solidified samples. All the solidified samples except AA + C + PS and AA + C + PM have shown diffusion coefficients (D_e) of the order of 10^{-10} cm^2/s. PS and PM have been extensively used in the stabilization/solidification of different radioactive compounds and other hazardous materials [21]. On the basis of their chemical interaction, these materials were selected in the present study, but the higher diffusion coefficients for AA + C + PS and AA + C + PM matrices obtained in this study could be due to the wider void space due to the cylindrical shape of the beads used in the experiments. AA + C + FA + PS sample had higher crystallite size and wider pore openings and thus had higher leachability. Addition of polystyrene and PMMA do not interact chemically with the other components (cement and sludge) of the matrix under the ordinary conditions. On the other hand, it enhances pore size of the matrix, which results in more leaching of the contaminant from the solidified sample. Dutre and Vandecasteele [17,18] reported diffusion coefficients in the order of 10^{-10} to 10^{-11} cm^2/s for arsenic solidified sample having cement and lime. Matrix having AA + C + FA have
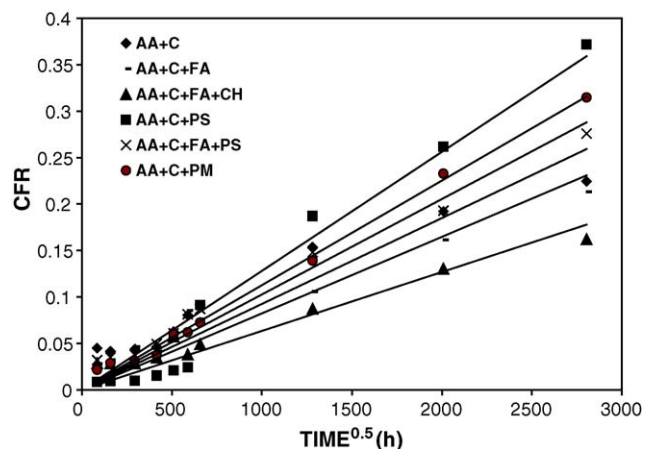

Fig. 3. Cumulative fraction of arsenic released from the S/S waste as a function of square root of leach time for the semi-dynamic leach test.

Table 2
Comparative studies of effective diffusion coefficients and leachablity index

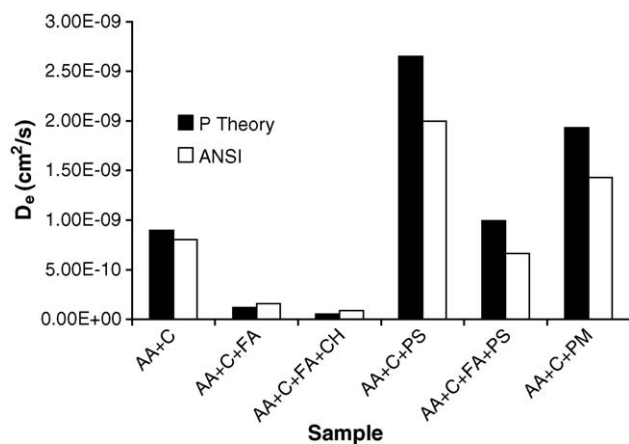| Composition	Effective diffusion coefficient D_e (10^{-10}) (cm²/s)			Leachability index (L)	
	ANSI	Penetration theory	R^2	ANSI	Penetration theory
AA + C	8.95	8.04	0.80	9.0	9.0
AA + C + FA	1.17	1.59	0.88	10.2	9.7
AA + C + FA + CH	0.512	0.893	0.89	10.66	10.0
AA + C + FA + PS	6.64	9.93	0.95	9.4	9.0
AA + C + PS	26.5	19.5	0.94	9.5	8.7
AA + C + PM	8.73	14.3	0.99	9.2	8.8

Fig. 4. Comparison of diffusion coefficient curve for the arsenic [1 = AA + C; 2 = AA + C + FA; 3 = AA + C + FA + CH; 4 = AA + C + FA + PS; 5 = AA + C + PS; 6 = AA + C + PM].

also shown comparable results as the presence of silica enhances binding capacity and reduces permeability of the matrix. This can also be explained on the basis of pozolanic reaction products, which contain silicic acid. This reacts with lime in the presence of water to produce material properties similar to cement. Common sources of pozzolans are fly ash and lime kiln dust. The silicates can react with polyvalent metal ions, resulting in reduced metal solubilities. Pozzolanic cement solidification/stabilization has been successfully demonstrated on certain metals including lead, cadmium, zinc and chromium. A comparison of diffusion coefficients obtained from ANSI and penetration theory is depicted in Figs. 4 and 5.
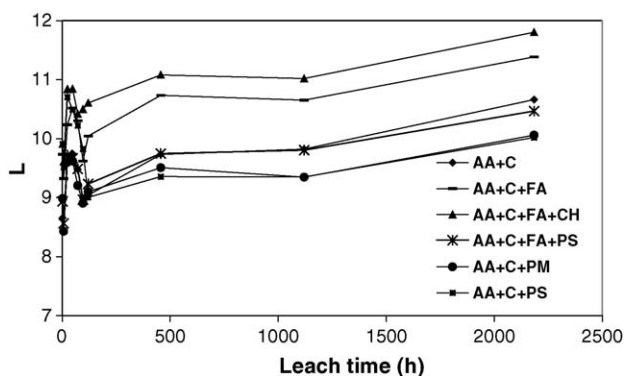

Fig. 5. Variation in leachability index with leach time for different S/S samples (ANSI theory).

In order to estimate the microscopic structures and morphology of surfaces, XRD and SEM of these matrices were done. SEM images of the different matrices are shown in Fig. 6(a–f). In contrast to the results of earlier studies on S/S with cement and fly ash [4,20], no formation of ettringite crystals was observed in matrices having AA + C + FA, AA + C + FA + CH and AA + C + FA + PS (Fig. 6b–d). Ettringite formation as shown in literature was the result of the addition of sulfate either through ferrous sulfate or some other form of sulfate. In the present study, no salt containing appreciable amount of sulfate salt was added, which resulted in lack of sulfate thus no formation of etttringtite was observed. Ettringite is undesirable as it is responsible for swelling and deterioration of the S/S matrices, which increases matrix porosity, and hence leaching of arsenic. Absence of ettringite could be one of the causes for less leachability of arsenic from the AA + C + FA + CH matrix.

As can be seen from Fig. 6(e and f), polymeric matrices (PS and PM) have higher crystal size and pore openings which in turn increased the leaching of arsenic into the solution. These results are in agreement with the experimental observations where lower leachability indices were observed in matrices having AA + C + PS and AA + C + PM.

XRD of the plate-like structure formed on the AA + C + FA + CH surface has been shown in Fig. 7. Calcium arsenite and silicates peaks were observed at 2 theta values of 29°, 26° and 33.80°. XRD pattern indicated the presence of peaks of calcite, Ca(OH)₂ and silicates (Fig. 7).

3.3. Leachability index

A waste contaminant with $L < 6.5$ is considered to be very mobile whereas $6.5 < L < 8.0$, it is moderate and $L > 8.0$ is considered of very low mobility [18]. Experimental leachability index results revealed that the all solidified samples performed well under the given set of experimental conditions as the leachability index values for each sample was higher than 6.0 (Table 2). Minimum amount of leaching ($L = 10.66$) was observed in the case of mould containing sludge, cement, fly ash and Ca(OH)₂ in the weight ratio 3:1:0.5:0.5. Variation in leachablity index as a function of leach time (Fig. 5) shows that in the initial phase of semi-dynamic test, most of the solidified samples have similar leachability indices. Monolithic samples having composition AA + C + FA + CH and AA + C + FA showed low mobility of arsenic ($L > 10.0$). Other samples have shown moderate mobility of arsenic ions.
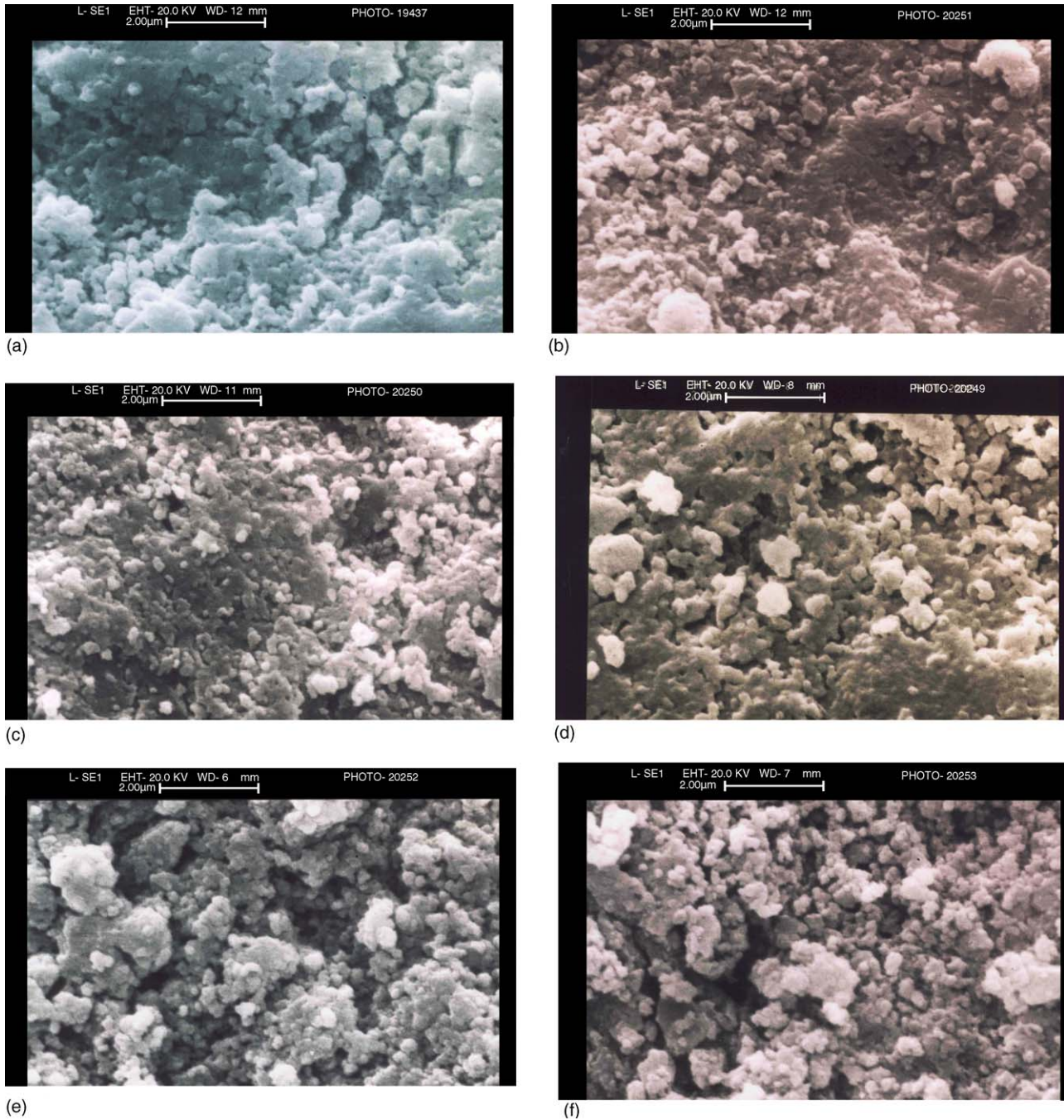
Fig. 6. SEM photographs of different stabilized/solidified matrices (a) AA + C; (b) AA + C + FA; (c) AA + C + FA + CH; (d) AA + C + FA + PS; (e) AA + C + PS; (f) AA + C + PM.

Portland cement used was heterogeneous mixture of (i) 50–70% of tricalcium silicate (Ca_3SiO_2), (ii) 20–30% of dicalcium silicate (Ca_2SiO_4), (iii) 5–12% of tricalcium aluminate ($Ca_3Al_2O_6$) and (iv) 5–12% of calcium aluminoferrite ($Ca_4Al_2Fe_2O_{10}$). When cement was added to an acidic medium, calcium readily dissolves and released into leachate, which results in an increase in pH of the leaching fluid during the leaching procedure. $Ca(OH)_2$, when added to a different mould, along with cement acts as a reservoir of calcium and maintains high pH for a considerable period of time. Calcite formed during carbonation reaction seal the pores in the cement matrix, which

hinder the penetration of contaminant from the matrix as well as passage of atmospheric CO_2 into the interior of the mould. This process may be responsible for the reduction in leaching from the matrix having cement as well as $Ca(OH)_2$. Precipitation and conversion into a non-soluble form (calcium arsenite) in the cement also contribute towards low leaching in the solidified samples [22,23]. Chuanyong et al. [24] also reported significant reduction in arsenic mobility with cement because of the formation of the sparingly soluble calcium arsenate. They also confirmed the conversion of the adsorbed arsenic to calcium arsenate precipitate with a FTIR peak at $860\,cm^{-1}$. Reduction in the concentration

Table 3
Calculation for standard error in S/S studies

Sample	Leachablity index (L)	S.D. (σ_n)	Precision of measurement $s_n = (n/n-1)^{1/2} \cdot \sigma$	Standard error $S_n = s_n/n^{1/2}$
AA + C	9.04	0.72	0.547	0.173
AA + C + FA	10.25	0.576	0.350	0.108
AA + C + FA + CH	10.66	0.586	0.362	0.114
AA + C + FA + PS	9.46	0.432	0.196	0.0622
AA + C + PS	9.57	0.576	0.358	0.110
AA + C + PM	9.27	0.519	0.284	0.0900

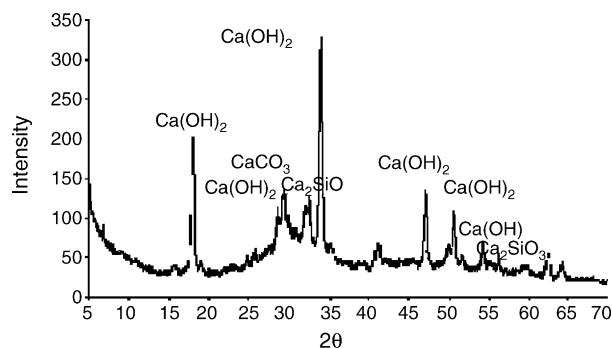n is the number of experimental data.

Fig. 7. XRD pattern of flakes like formation found on the surface of S/S matrix AA + C + FA + CH.

of arsenic in the leachate, mainly of As(III), due to the formation of insoluble $CaHAsO_3$ in the leachate in the presence of $Ca(OH)_2$ has also been reported by Vandecasteele et al. [11]. Another main reason of arsenic immobilization in cement-based S/S matrices could be due to the alkaline nature and buffering capacity provided by calcium hydroxide and C–S–H [25].

The presence of Ca has also affected the leaching of arsenic from the stabilized/solidified matrices as more leaching was observed in case of AA + C than AA + C + FA and AA + C + FA + CH. The addition of calcium and silica improved the diffusion coefficient. Pozolanic reaction products formed with the addition of fly ash also resulted in reduction of arsenic leachability. Similar kind of low leaching with higher Ca/As ratio and fly ash has also been reported in the literature for arsenic stabilization/solidification [4,26].

The statistical error analysis for the calculation of leachability index is given in Table 3. Standard errors were also calculated from the results and the data have been reported with a confidence limit of 95%.

4. Conclusion

Solidification of arsenic solid waste with cement is a suitable technique to reduce the leachablity of arsenic. Semi-dynamic tests have shown reduction in leaching of arsenic from arsenic containing waste. Arsenic waste can be solidified/stabilized with cement, fly ash and $Ca(OH)_2$ This was due to the formation of calcite which seals the pores of the solidified sample and precipitate formation of calcium arsenite whereas cement acts as a binder. XRD and SEM also confirmed the experimental observations as well as formation of calcite. The addition of calcium

and silica improved the diffusion coefficient. Pozolanic reaction products formed with the addition of fly ash also resulted in reduction of arsenic leachability. Precipitation and conversion into a non-soluble form (calcium arsenite) in the cement also add to the mechanism for low leaching in the solidified samples. No physical or chemical interaction was observed between polystyrene and PMMA with the arsenic species as these materials were used in cylindrical bead form. Highest leaching of arsenic was observed for the matrix having cement with PMMA, which shows that this matrix is not fit for arsenic waste stabilization/stabilization. A linear relationship between CFR and square root of leach time suggested that the diffusion is the responsible mechanism for arsenic leaching. A good agreement between diffusion coefficients and leachability indices as obtained from both penetration theory and ANSI was also observed.

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