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Bangladesh J. Sci. Ind. Res. **55(2)**, 131-138, 2020

BANGLADESH JOURNAL OF SCIENTIFIC AND INDUSTRIAL RESEARCH

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Stablization of medical waste incineration fly ash in cement mortar matrix

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Abstract

Laboratory experiments were performed to assess the suitability of using medical waste incineration fly ash in cement as a construction material based on the engineering properties of fly ash-cement matrix and the leaching potential of toxic heavy metals from the stabilized mix. Fly ash-cement samples were prepared with different proportions of fly ash (0%, 5%, 10%, 15% and 20% by weight) in the laboratory under controlled conditions. The solidified matrix exhibited a compressive strength from 3950 to 4980 psi when fly ash is mixed in varying proportions. The 28-day compressive strength has been found to decrease with the increase in fly ash content but it meets the minimum requirement of compressive strength for cement-mortar. Soundness test exhibited acceptable results for cement-mortar mixes having up to 15% fly ash. Final and initial setting times of cement have been found to generally increase with fly ash content. Water requirement (for normal consistency) also increased with the increase in fly ash content in cement. Based on physical properties of the cement-mortar matrix it is recommended that up to 10% (by weight) medical waste incineration fly ash can be incorporated for producing cement-mortar of optimum quality. Leaching behaviors of several targeted heavy metals (As, Cu, Ni, Cd, Pb, Hg and Zn) were analyzed using Toxicity Characteristics Leaching Procedure (TCLP) of fly ash and solidified fly ash-cement matrix which shows that the leached concentrations of As, Cu, Cd, Pb and Zn were reduced by 80.13%, 89.47%, 33.33%, 23.9% and 100% respectively for 10% fly ash incorporated cement-mortar matrix compared to that of original fly ash. The leached concentrations of heavy metals from the matrix were far below the EPA land disposal limits. These results suggest that the solidified fly ash incorporated cement-mortar matrix can effectively confine and immobilize the heavy metals contained in the fly ash without significantly diminishing the engineering properties of cement-mortar.

Received: 20 March 2019

Revised: 10 July 2019

Accepted: 11 November 2019

DOI: https://doi.org/10.3329/bjsir.v55i2.47633

Keywords: Medical waste; Incineration; Fly ash; Leaching; Heavy metal

Introduction

Bangladesh, being a developing country, has a rapidly growing urban population. Hospitals, clinics, private individual practitioners, dental clinics, diagnostic centers are growing in urban centres in order to meet the demand for health care services. In Dhaka city the number of health care facilities is almost 1200 and they contribute to the generation of 200 tons medical waste daily on an average, a significant portion of which is toxic (PRISM, 2013). Incineration is a commonly adopted method to neutralize or eliminate medical waste which generally results in decrease in waste volume. However, heavy metals present in the waste can be incorporated in the fly ash at elevated concentrations and inappropriate disposal of fly ash can pose environmental hazards (Tang *et al*., 2016). Release of heavy metals under certain conditions may lead to contamination of aquifers, surface water or drinking water system (Agamuthu and

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Chitra, 2009). If proper care is not taken during generation, fly ash can also spread out to greater distances by wind and contaminate the air environment. In Bangladesh, there is no specific legislation in the disposal, handling and dumping of medical waste under the Environmental Protection Act, 1995 (Akter, 2000). Although incinerators are operated by medical waste service providers (e.g. PRISM Bangladesh) and some major hospitals in Bangladesh, the incinerated fly and bottom ashes are usually dumped into the landfills without taking into account their potential toxicity and possibility or release of heavy metals into the environment. One technique to contain contamination from toxic substances is to incorporate it in construction materials. Here in this paper, we examine the leachability of heavy metals from medical incineration fly ash and fly ash-stabilized cement mortar mixes. Cement mortars were prepared varying the fly ash proportions and their suitability was assessed based on normal consistency, setting time, compressive strength and soundness. Leaching test of the fly ash- incorporated cement was also carried out to check the effectiveness of stabilization against the leaching of heavy metals in the environment by comparing them with land disposal limit based on established leaching protocols.

Materials and methods

Collection of fly ash sample

Fly ash sample was collected from the incinerator of the medical waste treatment facility of Dhaka City Corporation at Matuail which is operated by PRISM Bangladesh. In this facility the medical waste from different health care facilities from Dhaka are received and after separation, chemical disinfection, shredding, autoclaving they are taken into the incinerators. Our fly ash sample was collected from the trash bins at landfill site where incinerated fly ash was dumped.

Preparation of the fly ash sample

We adopted the methodology proposed by Tang *et al*. (2016) to process the sample. Fly ash samples were dried in oven for 24 hours to ensure the accurate quality during the experiment. Subsequently they are cooled to room temperature. After cooling the samples were passed through the series of BS standard test sieves (#4, #8, #16, #30, #50, #100, #200) with the use of mechanical sieve shaker. The portion of the sample retained on #200 sieve (75 micron) and received on pan (<75 micron) was used for preparing cement mortar.

Characterization of cement mortar-fly ash mixture

Table I shows the tests that were conducted to characterize the cement and cement-mortar mix with various compositions of fly ash.

Preparation of cement paste

For normal consistency test, cement paste of 650 grams was mixed with 0%, 5%, 10%, 15% and 20% fly ash as a replacement for cement. The required amount of cement was weighed and mixed with water and kept for a duration of 30 secs for absorption. Mixing was done at (140±5 rpm) for 30 secs with mixer. The mixer kept stopped for 15 secs and then mixer was run at medium speed (285±10 rpm) and mixed for 1 min. For setting time test, the sample was prepared as described for normal consistency test. The amount of water added to cement was the amount of water determined from normal consistency test. The cement paste sample was kept undisturbed for 30 min in a moist room.

Preparation of cement mortar mix

One part of cement was mixed with 2.75 gm of graded standard silica sand conforming to the specification of C778-02. A water-cement ratio of 0.485 was used. 700 gm cement paste (with 0%, 5%, 10%, 15%, 20% fly ash as a replacement of cement) was mixed with 1925 gm of sand for preparation of the sample. Immediately upon completion of molding, the test specimens were placed in a moist room for 20 to 24 hours with the upper surfaces exposed to the moist air but protected from getting wet through water. Then the specimens were immersed in saturated lime water in storage tanks constructed of non-corroding materials. For compressive strength test, three specimens from each batch of mortar were prepared for 3 days, 7 days and 28 days test. So total 9 mortar specimens were made with each of the proportions of fly ash (0%, 5%, 10%, 15%, 20%).

Preparation of specimens for soundness test

For soundness test, the compositions for different samples were as shown in Table II. One single molded cement bar was made for each variation of the fly ash composition. So, total 5 specimens were made for the test.

Heavy metal determination

Sample preparation for heavy metal analysis was carried out according to EPA 3050B and the concentration of heavy metals was determined using Atomic Absorption Spectrophotometer (AAS). After collection of incinerated fly ash sample, some portion of fly ash samples were dried in a vacuum oven at 105°C until constant weight, lightly ground for homogenization and passed through 2-mm sieve. For heavy metal analysis, sample preparation was carried out according to EPA 3050B with a slight variation. 5 gm. of dried sample was digested with acid (HNO_3: HCl =1:3 volume ratio) for 24 hours. After adding 350-400ml distilled

Table I. Test names and test method for cement and cement-mortar mix

Name of the Tests	Test Method	Test for
Determination of normal consistency	ASTM C187	Cement
Determination of setting time	ASTM C191-08	Cement
Compressive strength	ASTM C150-12	Cement-mortar
Soundness	ASTM C1038-01	Cement-mortar
Total Heavy Metal	EPA 3050B	Fly ash/cement-mortar
Toxicity Characteristics Leaching Procedure (TCLP)	USEPA 1311	Cement-mortar

Table II. Compositions for different samples for soundness test

Fly ash percentage	Cement (gm)	Fly ash (gm)	Water(gm)	Sand(gm)
0%	300	0	145.5	825
5%	285	15	145.5	825
10%	270	30	145.5	825
15%	255	45	145.5	825
20%	240	60	145.5	825

water, sample was boiled for 2.5 hour and a 500ml solution was prepared. Then, solution was filtered through a 0.45 µm filter paper and filtrate was collected to determine the concentration of seven heavy metals (As, Pb, Cd, Cr, Ni, Cu, and Zn) by using Atomic Absorption Spectrophotometer (AAS) (Shimadzu AA 6800)

Toxicity characteristics leaching procedure (TCLP)

According to Tsiridis *et al*. (2006), a commonly used test for the determination of the leaching characteristics of fly ash is the Toxicity Characteristic Leaching Procedure (TCLP Method 1311) established by the US Environmental Protection Agency (USEPA, 1992). This procedure provides a uniform method to compare the tendency of inorganic elements to leach out from fly ash samples into moderate-to-highly acidic aqueous environments. During the TCLP test, constituents are extracted from the waste to simulate leaching actions that occur in landfills. If the concentration of the toxic constituents exceeds the regulatory limit, the waste is classified as hazardous. TCLP tests were performed following the USEPA 1311 for two types of samples: (1) original fly ash sample (2) Cube specimen containing 10% fly ash.

Results and discussion

Normal consistency

Figure 4 shows that normal consistency increases with the increase of percentage of fly ash. Up to 15% of fly ash the normal consistency value remains close to the ASTM C150 standards range of 20% to 30% but at 20% of fly ash the normal consistency value goes beyond the limit. Kumar *et al*. (2016) studied the effect of cement properties with the partial replacement of biomedical waste fly ash and showed that water requirement increased with the increase in fly ash percentage and workability of concrete decreased with the increase in fly ash content. Since the fly ash is lighter than cement it occupies more volume than cement on equal weight basis. So more water is needed for lubrication and therefore the normal consistency increases. Our test results agree with the behavior of cement with fly ash as previously showed by Kumar *et al*. (2016).

Setting time

In this study the change in the setting time with the increase in fly ash percentage was observed. The limit of the standard values of the initial and final setting time is shown in table III. The variation in the setting time with the increased percentage of fly ash is shown in figure 5. From the graph it is evident that there is a decrease in initial setting time when 5% fly ash is added and then initial setting time increases with increase of fly ash. It can also be seen that the final setting time increases significantly with the increase in fly ash up to 5%. From 5% to 20%, there is no appreciable change of final setting time. With 5% addition of fly ash, final setting time goes beyond the maximum limit of ASTM C150-12 specification standard of 375 min. Augustine (2016) observed that the initial and final setting times for 20% ash content were 300 and 985 min respectively. In this study, the

Fig. 1. Incinerator of PRISM Bangladesh at Matuail landfill site

Fig. 2. Separation of large particles from incineration fly ash manually and storage of the prepared sample in zip-lock bags

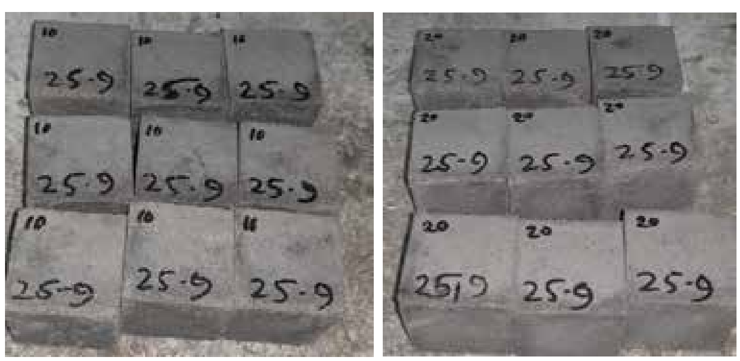

Fig. 3. Cement-mortar cube specimens consisting of 10% and 20% fly ash

initial and final setting time of our experiment for 20% fly ash are 250 and 545 min respectively. It is evident from the study that the introduction of fly ash to a concrete mix design could lengthen the overall hydration period. The presence of Cu, Pb and Zn compounds are set-inhibiting and could have contributed to the increase in setting time (Zain *et al*., 2004). For highway construction, the delays of setting time may be considered advantageous especially for hot weather concreting to keep the concrete workable for a long time.

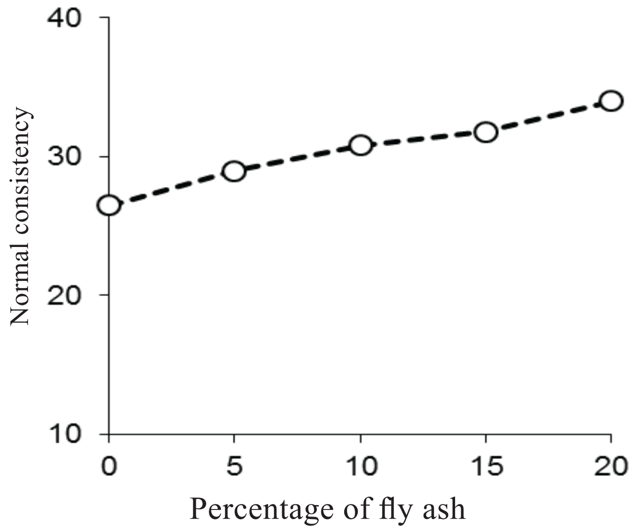

Fig. 4. Relationship between normal consistency and percentage of fly ash

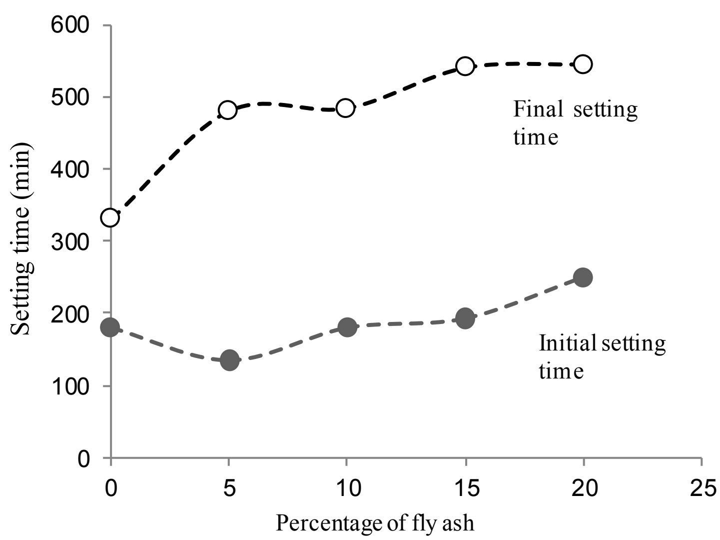

Fig. 5. Relationship between setting time and fly ash content

Compressive strength

Table IV shows the variation of compressive strength with the increase in fly ash proportions. Compressive strength (average of three samples) vs. no. of days in Figure 6 shows that up to 15% addition of fly ash the strength values of 3,7 and 28 days fulfill the minimum ASTM C150-12 requirements for compressive strength for the corresponding time periods. But for 20% addition of fly ash, the strength value of 28 days is 3950 psi which fails to meet the minimum requirement of 4060 psi. In general, the 28-day strength has been found to decrease with increased fly ash content. Although the 3-day and 7-day strengths for cement mortars having 5% and 10% fly ash are very much comparable with the control specimen (0% fly ash), the addition of 15 % and 20% fly ash has reduced the compressive strength significantly. The reason for the decrease in compressive strength with the increase in fly ash beyond a certain limit is that the heavy metals present in the ash such as Pb, Cu, and Cr could have inhibited the solidification of the matrix (Agamuthu and Chitra, 2009). Singh *et al*. (2016) conducted similar tests with fly ash-incorporated concrete and observed that compressive strength of concrete for 7 and 28 days attains a maximum value for 5% to 10% replacement by fly

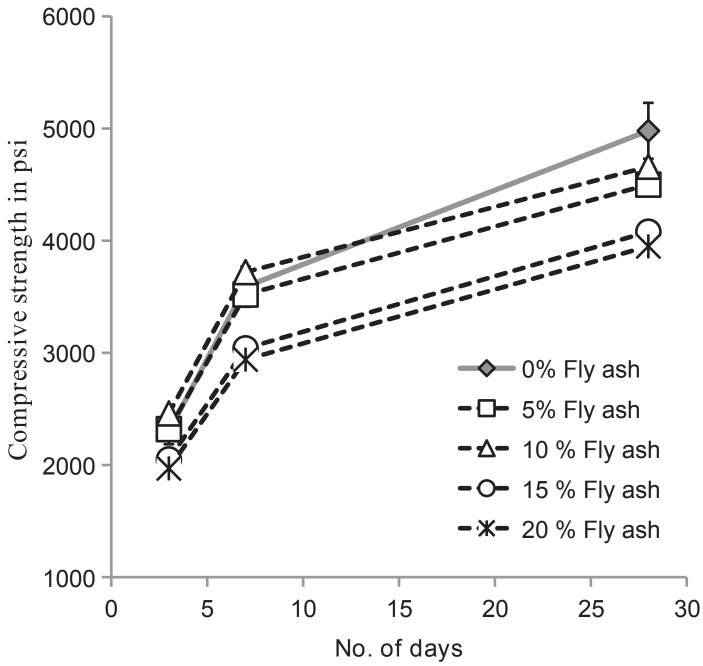

Fig. 6. Change in compressive strength with the increase in fly ash percentages for 3, 7 and 28 days

ash. Agamuthu and Chitra (2009) showed a decrease in compressive strength with the increase in fly ash proportions which follows the similar trend found in these experiments particularly for fly ash content in excess of 10%.

Soundness test

Soundness is an important parameter which signifies that cement should not undergo any significant volume change after setting. According to ASTM C150, a maximum mortar bar expansion of 0.02 percent is specified for all types of Portland cement. From our results shown in Table V, mortar bars having 0% and 10% of fly ash showed volume shrinkage while mortar bar having 5% fly ash did not exhibit any volume change. Mortar bar of 15% fly ash has expanded within the maximum allowable limit but mortar bar of 20% fly ash has shown an expansion exceeding the maximum ASTM C150 limit. The amount of mortar-bar expansion that was obtained according to ASTM C1038 method is related to the amount of sulfate in the cement and excessive expansion was observed because it contained too much sulfate. According to test results, the amount of sulfate increases with increasing of fly ash content and excessive expansion occurs for 20% fly ash due to high amount of sulfate. Yılmaz and Olgun (2008) investigated the effects of low-calcium fly ash on cement and mortar and the replacement of Portland cement by low calcium fly ash ranging from 5% to 40% reduced expansion compared to control cement paste without fly ash. This indicates that unless the sulfate content is controlled, addition of excessive fly ash will generally cause an expansion. In our studies the suitable proportion of medical waste incineration fly ash has been found to be 5% to 15% from the soundness test criteria. The soundness test results obtained from this experiment are given in Table V.

Heavy metal content in fly ash

Heavy metal determination test was conducted on raw fly ash sample in order to understand their relative abundance. The selected heavy metal concentrations for the sample found in the fly ash in this study and their comparison with several international regulatory standards are shown in Table VI. It allows the arrangement of the heavy metals from higher to lower concentrations as: Zn>Cu>Pb>Cr>Ni>As>Cd. The present study found that the concentration of As, Cr, Pb, Ni were well below the permissible limit of Bangladesh, China

Table III. Initial and final setting time test result for various percentages of fly ash

% of fly ash	0	5	10	15	20	ASTM C150-12	IS 4031 (part 5)
Initial setting time (min)	180	135	180	193	250	45 (min)	30 (min)
Final setting time (min)	330	480	484	540	545	375 (max)	600 (max)

Table IV. Average values of the compressive strength for different fly ash proportions

Percentage of fly ash		0	5	10	15	20	ASTM C150-12 requirement
Average Compressive Strength (psi)	3 days	2300	2320	2460	2050	1970	1740
	7 days	3590	3520	3720	3040	2940	2760
	28 days	4980	4500	4660	4080	3950	4060

Table V. Soundness test results of cement-mortar cubes having different proportions of fly ash

Amount of fly ash	Percentage of change	Comment
0%	0.004	Shrinkage
5%	0.000	No change
10%	0.008	Shrinkage
15%	0.004	Expansion
20%	0.052	Expansion

and USA Land Disposal Restriction Limits. The concentration of Cd and Cu are well below the Bangladesh and USEPA limit but exceeds the SEPAC (State Environmental Protection Administration of China) by 2 times. The concentration of Cu in the fly ash is about two times higher than the SEPAC limit. The concentration of Zn satisfies with the USEPA standard but it is more than 2 times higher than the permissible limit in Bangladesh and SEPAC limit.

Toxicity characteristics leaching procedure (TCLP)

TCLP test was carried out on the raw fly ash sample and on the cement-mortar cube mixed with 10% fly ash which was cured over 28 days. Since, the engineering properties of cement mortar with 10% utilization of fly ash provides

Table-VI. Concentration of heavy metals in the fly ash sample comparing with standards limit

Parameters	Heavy metals in the sample (mg/kg)						
	As	Cr	Pb	Cd	Ni	Cu	Zn
Sample	6.72	16.4	42.4	1	13.5	181.2	610.7
Permissible limit in Bangladesh	40	100	100	1.5	50	60	200
SEPAC limit in China	_	250	350	0.6	26600	100	300
USEPA limit	75	3000	840	85	420	4300	7500

Table VII. Result of concentration of in standard TCLP leaching test of leachates from fly ash sample and cube sample mixed with 10% fly ash and comparison with Land Disposal Restrictions Limits (LDR)

Heavy metal	As	Cu	Ni	Cd	Pb	Hg	Zn
Raw fly ash value (mg/l)	0.0297	0.057	0	0.006	0.364	0	0.653
10% Cube mortar (controlled sample) value (mg/l)	0.0059	0.006	0.015	0.004	0.277	0	0
% of reduction	80.13	89.47	-	33.33	23.90	0	100
EPA Land Disposal Limit (LDR) (mg/l)	5	-	11	0.11	0.75	0.25	4.3

optimum values for the normal consistency, initial and final setting time, compressive strength and soundness test, 10% fly ash mixed mortar was considered for this test. Table VII shows that the heavy metal concentration in raw fly ash and mortar cube having 10% fly ash are found to be under the EPA Land Disposal Limit (LDR). This indicates that there would be low risk of ground water contamination by leaching action. In this case, the stabilization of fly ash with the cement mortar reduces the leaching tendency of heavy metal compared to the raw fly ash. Arsenic content reduces by 80.13% ; Copper reduces by 89.47% ; Cadmium reduces by 33.33% ; Pb reduces by 23.90% and Zinc reduces by 100%.

Conclusions

We have shown that heavy metals present in fly ash can be successfully immobilized when it is incorporated in cement-mortar matrix albeit having slightly detrimental effects on its engineering properties such as workability, setting times, soundness and compressive strength. But cement with 10% fly ash can still have desirable engineering properties as per ASTM specifications of cement and cement-mortar matrix. Therefore, medical waste incineration fly ash incorporation into cement appears to be a promising avenue for waste management and successful recycling of waste in the construction material.

Acknowledgement

Authors would like to render their special gratitude to the officials of PRISM medical waste management foundation who assisted in the process of sample and other data collection. In this regard, the authors are grateful to Khondkar Anisur Rahman, Executive Director of PRISM foundation. The authors also acknowledge the heartiest co-operation and assistance of all the staffs of Concrete Laboratory, Environmental Engineering and Geotechnical Engineering Laboratory of BUET during different stages of experiments.

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