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Leaching Characteristics of Fly Ash Generated from Suratgarh Thermal Power Plant

RAHUL DANDAUTIYA, PRANAV MAYEKAR, MADHUKAR TOSHNIWAL, AJIT PRATAP SINGH AND SANGHAMITRA KUNDU

Birla Institute of Technology & Science Pilani, Rajasthan, INDIA

***Email:** rahuldandautiya@gmail.com, skundu@pilani.bits-pilani.ac.in*

Abstract: This paper presents the results of short-term leach tests conducted on slightly alkaline fly ash collected from Suratgarh Super Thermal Power Plant (SSTPP), Sri Ganganagar, Rajasthan, India. ASTM D3987-85 (ASTM) and toxicity characteristic leaching procedure (TCLP) test methods were applied on fresh fly ash (FA-1) and weathered fly ash (FA-2), to understand the leaching characteristics of the elements Cu, Zn, Mn, Fe, Ba, Ca, Co, Cr, Ni, Si, Pb, Sr, Cd, Al, Mg, V, Ti, and As inherent in the SSTPP fly ash. Morphological, quantitative and qualitative elemental as well as mineralogical analyses of fly ash samples were done using XRD, FEG-SEM and ICP-AES techniques. The results showed that both the fly ashes comprise mainly of Bavenite, Silicon Oxide Quartz and Berlinite minerals. The concentration of V (in both FA-1 and FA-2) and Cr (only in FA-1) exceeded the acceptable limit prescribed by New-Zealand waste acceptance criteria for class-A landfill. The TCLP test rendered more mobility to almost all the elements. The concentrations of As (in FA-1), Fe, Cr (not in ASTM FA-2), Mn, Pd and Se (not in ASTM FA-2) in the leachates exceeded the WHO drinking water standards for both TCLP and ASTM tests whereas Ni and Ba surpassed only in TCLP test. The high elemental concentrations of various elements in the leachates thereby suggest a fair amount of pollution potential of SSTPP fly ash.

***Keywords:** Leaching, Fly Ash, TCLP, XRD, ICP-AES*

1. Introduction

Electricity, the principal source of energy, has increasingly become an essential requirement in the constant pursuit of urbanization, technical innovations and sustainable development over the years. This demand for electricity is majorly fulfilled by combustion of coal in thermal power plants (TPPs), which are the backbone of India. As of today, about 172.87 million tonnes of coal fly ash is generated by 143 coal/lignite based thermal power stations in India [1]. With a present utilization rate of 57.63%, in concrete and construction, in road sub-bases, in the construction of cement, as mineral fillers in asphaltic mixes, as a low-cost adsorbent for the removal of organic compounds, flue gas and metals, as light weight aggregate, in mine back fill, zeolite synthesis, as landfill liner material, as mineral resource, as an abrasive agent, as soil amendment material in agriculture, and also as an additive in polypropylene polymer [2-8], a major portion of it still remains unused and face problems of disposal as it not only require large areas of precious land for its disposal but also pose a grave environmental concern as a source of pollution of both air and water. Significant quantities of various trace elements are present in coal and during combustion of coal, enrichment of these trace elements occur in the fly ash as a result of carbon loss due to carbon dioxide [9]. These trace elements, with concentrations often 4-10 times higher than their original concentrations in the coal samples, are emitted largely in the fly ash or are associated with the surface of the fly ash particles due to evaporation and condensation [9, 10]. Under favourable conditions, the elements may be susceptible to easy mobilization and leaching by interaction with rainwater or other aqueous solutions in ponds or landfills. Numerous studies provide proof of leaching of toxic elements from fly ash leading to pollution of the surrounding environment [11-24]. In fact, various studies [10, 25-54], Are being done throughout the world to understand the leaching behavior of the fly ash and to understand the nature and magnitude of pollutants present in the leachate, so as to develop proper sustainable environmental control technologies.

The most often used approaches to assess this elemental mobility are the laboratory batch extraction tests and column leaching experiments. Short term leach tests (batch tests) are a simple means to compare the dissolution behavior of various components in considerably shorter interaction periods thus providing critical information [26, 28, & 38]. The column studies using synthetic water, can simulate the surface leaching from fly ash dumps to predict the natural weathering actions on the fly ashes to a considerable extent [55, 56] and thus give us the idea of the extent of environmental pollution hazard these ponds/landfills pose. The experimental conditions under which the leaching studies are conducted in the laboratory can vary reasonably from the actual environment on field; however, the generated data provides a relatively fair idea of the dissolution characteristics of different materials in the waste. There are 143 coal based [1] thermal power plants in India but with respect to this large number very few studies [31-33, 37, 40, 42, 43, & 50-52] have been done.

This study focuses on short term leaching behavior of Zn, As, Cu, Co, Ni, Cd, Fe, Cr, Mn, Pb, Se, Sr, V, Ti, Al, Ba, Si, Ca and Mg from Suratgarh fly ash using both batch and column tests. The mobility of these elements was investigated using TCLP-1311 and ASTM-3987-85 tests for batch experiments. The concentrations in the final leachates were compared with the national and international standards prescribed for drinking water and also with related limit values of landfill regulations. Elemental analysis of ground water was also done to find out the impact of leaching.

2. Material and Methods

2.1. Fly Ash Sample and Properties

Suratgarh thermal power (1500 MW) plant has consumed 6.386870 million tonne of coal in year 2014-15 and it results generation of 2.015803 million tonne of fly ash [1]. Two samples of fly ash, having particle size 0.002 - 0.075 mm [57], fresh fly ash (FA-1) directly taken from plant and pre weathered fly ash sample (FA-2), were collected from disposal site in a dry state and kept in air tight containers. The samples were initially dried and homogenized before performing experiment. The pH of the samples were measured by pH meter after making L/S ratios 20 and shaking vigorously for five minutes. The mineralogical composition of both fresh and weathered samples was determined by X-ray diffraction (RigakuMiniFlex II) with Cu-Kα (1.54 Å) radiation was done at BITS. The elemental composition and analysis of structure of fly ash samples done by inductively coupled plasma atomic emission (ICP-AES) SAIF, IIT Bombay, the particle morphology was examined using Field Emission Gun-Scanning Electron Microscopes (JSM-7600F) analyses from SAIF, IIT Bombay.

Figure 1 *Elevation of Suratgarh Super Thermal Power Plant, dumping area and residential area (Source: Google earth)*

2.2. Leaching Test

Leaching characteristics of fly ash were find out by the two standard test procedures

2.2.1. Toxicity Characteristic Leaching Procedure (TCLP; EPA method 1311)

TCLP test is performed to simulate most adverse case when the leachant is acidic and there is a possibility of presence of hazardous waste, determined under US EPA Resource Conservation and Recovery Act (RCRA), in solid waste [58]. The Extraction fluid, as described in TCLP protocol, used in TCLP was glacial acetic acid (pH 2.88) as both of the fly ash samples were alkaline in nature. The extraction fluid with the solid waste was put in a 300 ml nitric acid cleaned bottles. The liquid to solid (L/S) ratio used was 20 and horizontal shaking was done at 180 rpm for 18 hours at 25°C temperature.

2.2.2. ASTM D39 87-85

ASTM D3987-85 works as a quick way to find out the inorganic extract of a solid waste [59]. Distilled water was used as a leachant in this method. Which was placed with L/S ratio of 20 in nitric acid cleaned 300 ml bottles. The horizontal shaking test was run for 18 hours for at 180 rpm and the temperature was 20°C.

3. Results and Discussions

3.1. Characterization of Fly Ash

The elements found in ICP-AES are Cu, Zn, Mn, Fe, Ba, Ca, Co, Cr, Ni, Si, Pb, Sr, Cd, Al, Mg, V, Ti and As. It revealed that major elements available in both FA-1 and FA-2 are Si, Al, Mg, Ti, Ca and Fe. Concentration of Si and Fe increase from fresh to weathered sample. The concentration of Ca, Mg Al and Ti is decrease as the time passes, signifies they become constituent of leachate. The results are detailed in the Table 1.

The mineralogical composition of the fly ash samples was determined by XRD analysis. XRD results presented in Figure 2 (a) of FA-1 shows predominance of Si, Al, Ti, Fe and Mg based minerals viz. Bavenite ($Al_2Be_2Ca_4H_2O_{28}Si_9$), Silicon Oxide Quartz (SiO_2), Berlinite (AlO_4P), Magnesium Chromate - $-beta ($CrMgO_4$) and minor occurrence of Rodalquilarite ($ClFe_2H_3O_{12}Te_4$), Milarite ($Al_{0.45}Be_{2.55}Ca_4H_{2.72}K_{1.184}$ $Na_{0.24}O_{30.68}Si_{12}$) and Titanium(III) Nitride Osbornite (N Ti).

In Figure 2 (b) for FA-2 shows predominance of Si, Al, Fe, Mg and Ti containing minerals as Bavenite (Al_2 $Be_2Ca_4H_2O_{28}Si_9$), Silicon Oxide Quartz (SiO_2) and Berlinite (AlO_4P) and trace amounts of Rodalquilarite ($ClFe_2O_{12}Te_4$), Nickel Titanium Oxide (NiO_3Ti), Milarite ($Al_{0.81}Be_{2.19}Ca_2H_{3.52}$ $KNa_{0.188}O_{30.668}Si_{12}$) and Magnesium Chromate - $-beta ($CrMgO_4$).

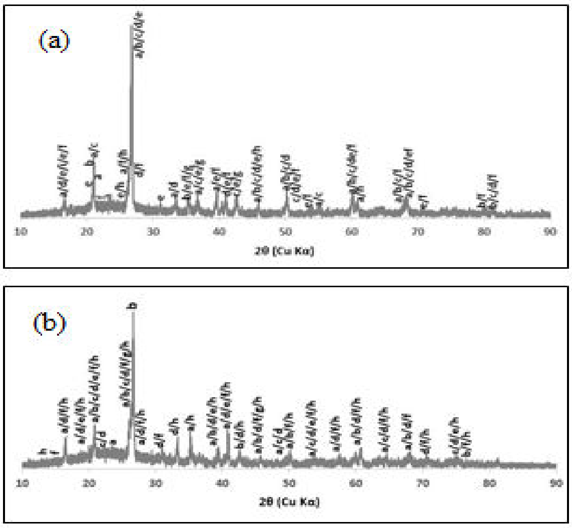

Figure 2 *(a) XRD of FA-1, a: Bavenite ($Al_2Be_2Ca_4H_2O_{28}Si_9$); b: Silicon Oxide Quartz (SiO_2); c: Berlinite (AlO_4P); d: Magnesium Chromate - \$-beta ($CrMgO_4$); e: Rodalquilarite ($ClFe_2H_3O_{12}Te_4$); f: Milarite ($Al_{0.45}Be_{2.55}Ca_4H_{2.72}K_{1.184}Na_{0.24}O_{30.68}Si_{12}$); g: Titanium (III) nitride Osbornite (NTi); h: Strontium (Sr)*
(b) XRD of FA-2, a: Bavenite ($Al_2Be_2Ca_4H_2O_{28}Si_9$); b: Silicon Oxide Quartz (SiO_2); c: Berlinite (AlO_4P); d: Rodalquilarite ($ClFe_2O_{12}Te_4$); e: Nickel titanium oxide (NiO_3Ti); f: Milarite ($Al_{0.81}Be_{2.19}Ca_2H_{3.52}KNa_{0.188}O_{30.668}Si_{12}$); g: Strontium (Sr); h: Magnesium chromate - \$-beta ($CrMgO_4$).

The porosity of the FA-1 and FA-2 comes out to be 41% and 32% respectively which shows considerable compaction of samples and increase in fineness due to weathering action. The presence of calcium oxide transforms the pH of the samples towards basic region. FA-1 pH comes around 9 while because of decrease in calcium content pH of FA-2 was 8.21.

Table 1: *Concentration of elements in FA-1 & FA-2 (conc. mg/kg)*

Element	FA-1	FA-2	Screening criteria	
			Class A landfills	Class B landfills
Cu	40	20	100	10
Zn	80	10	-	-
Mn	10	20	-	-
Fe	4440	6570	-	-
Ba	70	640	2000	200
Ca	8400	5100	-	-
Co	20	9	-	-
Cr	150	90	100	10
Ni	50	20	200	20
Si	296200	334600	-	-
Pb	0	0	-	-
Sr	16	90	-	-
Cd	0	0	-	-
Al	589800	230900	800	80
Mg	13600	8200	-	-
V	200	100	40	4
Ti	297400	211500	-	-
As	0	30	100	10

3.2. Morphology

The morphology of fly ash particle may affect the mobility of the elements in it. Denser packing with nonporous outer surface may resist leaching of heavy metals [60, 61]. Morphological studies were done with the help of Field Emission Gun-Scanning Electron Microscopes (JSM-7600F) analyses from SAIF, IIT Bombay. The results of FEG- SEM (Figure 3 (a) and 3 (b)) give an idea of structure, shape and pattern of samples. However, both the samples (FA-1 and FA-2) of fly ash are having solid and hollow spherical structures, FA-1 results look very smooth edges structures as compared to FA-2. After the weathering action the structural elements of FA-2 are more clustered and seem soft lumps opposed to the smooth surfaces of fresh sample. In the FA-2 the fineness is relatively low than the FA-1 and that is the cause of low porosity of FA-2. The specific surface area of FA-1 is also more than FA-2. That results, because of leaching, decrease in fine particle than fresh FA. In FA-2 some other shape, bundles of wool like patterns, are visible, indicates the availability of some tile shape crystals [62].

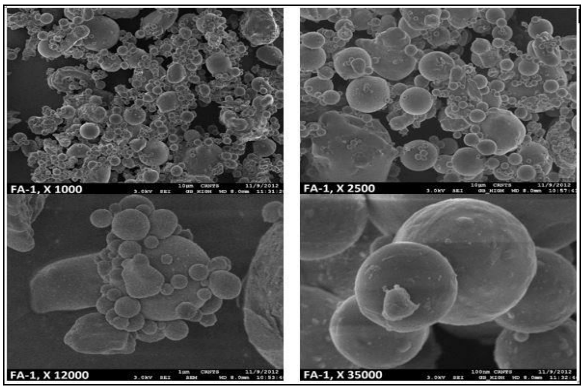

Figure 3 (a) *Images of FEG-SEM FA-1*

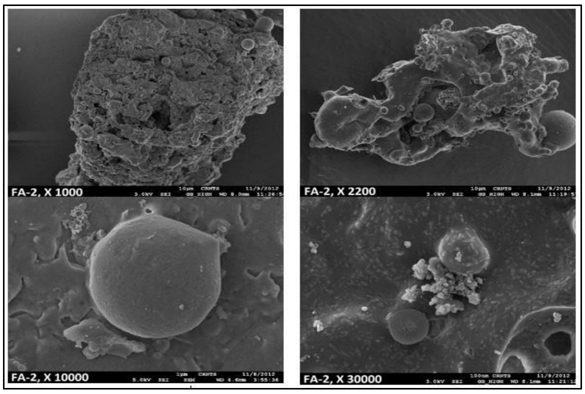

Figure 3 (b) *Images of FEG-SEM FA-2*

3.3. Batch Experiment

The results of elemental concentration in leached samples from TCLP and ASTM test are detailed in Table 2 and compared with the prescribed regulatory value of TCLP and drinking water limits setup by WHO. The concentration of all the elements is higher in TCLP leachate than ASTM leachate for both fresh and weathered fly ash. Which shows a significant dependency of leaching on pH of leachant. All the results are very much under the TCLP regulatory limits. The concentration of Zn, Cu, Ca and Mg in leachate is well below the WHO drinking water standard from both TCLP and ASTM test results. The value of Cd for FA-1 in TCLP is resembles the WHO limit. Pb concentrations are exceeding slightly in both TCLP and ASTM results which is a little alarming. The Concentrations of As, Se and Ba in FA-2 results are under permissible limit for both test except ASTM results of Ba, which are little over the WHO permissible value, but in FA-1 results As concentration increases about 20 folds for both test results. Se and Ba surpass slightly the WHO limits for both tests except ASTM results of FA-1. The Ni concentration exceeds from WHO limits only in TCLP results shows its higher affinity with the acidic leachant. Fe concentration is about 44 times and 6 times higher in FA-2 and FA-1 results of TCLP test than the WHO guidelines. In ASTM test Fe concentration is 3 times higher in FA-1 result and nearly equal in FA-2 result than the WHO guideline. The concentration of Mn is more than the permissible values in all the results and it's about 23 and 30 times higher in TCLP and 10 and 4 times higher in ASTM results of FA-1 and FA-2. Cr Concentration is also exceeding the WHO guideline for FA-1 results in both tests but for FA-2 results its concentration is nearly equal.

The concentration of all the elements is well below the hazardous limit of waste acceptance criteria. Only As and Se concentrations exceed the non-hazardous waste acceptance limit in the results of FA-1 for both TCLP and ASTM tests. The waste acceptance concentration limit in inert waste is crossed by many elements. Zn and Cu concentration in TCLP results for both FA-1 and FA-2 surpass by up to 3 times the waste acceptance concentration limit in inert waste. Concentration of Cd in FA-1 and Pb in FA-2 of TCLP test exceed with a very little amount from the inert waste acceptance limit. Ni and Cr both exceed the inert waste acceptance limit. Ni surpasses by about 4 times in TCLP test for both FA-1 and FA-2 results and twice in ASTM FA-1 result than the inert waste acceptance limit. The concentration of As in FA-1 for both TCLP and ASTM results exceed at a considerable amount of about 11 and 8 times by the waste acceptance concentration limit of inert waste. Value of Cr is increase by 10 and 5 times in the results of FA-1 for both tests and about 3 times in FA-2 of ASTM test. Ba concentration exceeds very little only in FA-1 of TCLP test. The concentration of Se is little higher for FA-2 in both the tests but its notably higher (about 21 and 12 times) than the inert waste acceptance criteria for FA-1 in both TCLP and ASTM tests.

Table 2: *Elemental Concentration in FA-1 and FA-2*

Elements	TCLP (mg/l)		ASTM (mg/l)		Desirable drinking water limit as per WHO (mg/l)	Regulatory limits for TCLP (mg/l)	TCLP (mg/kg)		ASTM (mg/kg)		Waste Acceptance Criteria (mg/kg)		
	FA 1	FA 2	FA 1	FA 2			FA 1	FA 2	FA 1	FA 2	Inert	Non-Hazaardous	Hazardous
Zn	0.382	0.315	0.161	0.0815	4		7.64	6.3	3.22	1.63	4	50	200
As	0.2845	0.0025	0.202	0.0037	0.01	5	5.69	0.05	4.04	0.075	0.5	2	25
Cu	0.159	0.2795	0.0695	0.007	2	-	3.18	5.59	1.39	0.14	2	50	100
Co	0.036	0.0195	0.015	0.004	-	-	0.72	0.39	0.3	0.08	-	-	-
Ni	0.089	0.0825	0.042	0.013	0.07	-	1.78	1.65	0.84	0.26	0.4	10	40
Cd	0.003	0.0005	0.001	0.0005	0.003	1	0.06	0.01	0.02	0.01	0.04	1	5
Fe	2.45	17.59	1.2655	0.4985	0.4	-	49	351.8	25.31	9.97	-	-	-
Cr	0.2535	0.0725	0.1385	0.0095	0.05	5	5.07	1.45	2.77	0.19	0.5	10	70
Mn	0.943	1.234	0.403	0.164	0.04	-	18.86	24.68	8.06	3.28	-	-	-
Pb	0.019	0.0285	0.013	0.0175	0.01	5	0.38	0.57	0.26	0.35	0.5	10	50
Se	0.1055	0.0055	0.0635	0.006	0.04	1	2.11	0.11	1.27	0.12	0.1	0.5	7
Sr	0.9965	0.6605	0.575	0.515	-	-	19.93	13.21	11.5	10.3	-	-	-
V	0.87	0.0125	0.613	0.009	-	-	17.4	0.25	12.26	0.18	-	-	-
Ti	0.282	0.1905	0.1785	0.048	-	-	5.64	3.81	3.57	0.96	-	-	-
Al	50.13	23.1695	22.0445	0.346	-	-	1002.	463.3	440.8	6.92	-	-	-
Ba	1.1015	0.7175	0.485	0.1775	0.7	100	22.03	14.35	9.7	3.55	20	100	300

Si	49.65	33.85	25.1	7.15	-	-	993	677	502	143	-	-	-
Ca	153.35	46.05	91.5	39.2	150-300	-	3067	921	1830	784	-	-	-
Mg	50.5	26.9	33.45	17.45	150-300	-	1010	538	669	349	-	-	-

3. Conclusion

The results of ICP-AES indicated that concentration of all the elements except As in FA-1 exceeded the Class B New Zealand landfill acceptance criteria. Al and Cu in FA-1 concentration surpassed the Class A landfill guidelines. The ASTM and TCLP batch leaching tests were performed to understand the acidic and alkaline leachant behavior. Zn, Cu, Ca and Mg are well below the WHO standards for both leaching tests, while the concentration of As and Se are above the WHO guidelines for both test in FA-1. Ba concentration is slightly above but only in TCLP test leachate. Fe and Mn concentration are also found higher for TCLP test leachate. The results of FEG-SEM clearly differentiate the structural difference between the FA-1 and FA-2 and show the reason of decrease of porosity and different mineral composition in FA-1 and FA-2. The results also indicate that the value of all the elements in TCLP leachate for FA-1 is higher than permissible limit for inert solid waste acceptance criteria. As, Ni, Cr and Se concentrations are exceeded for FA-1 extracts of ASTM test. Values of As and Se for FA-1 in both ASTM and TCLP leachate are greater than the non-hazardous waste acceptance criteria also. For FA-2 in TCLP test, concentration of Zn, Cu, Ni, Cr, Pb and Se are above the guidelines for inert waste. The results of this study show that for all elements leaching is much higher for the acidic leachant.

The results of this study give an idea about how much the disposal of fly ash impact on groundwater, as there is no availability of any earlier specific research that give any picture regarding the ground water characteristics of disposal site. The Suratgarh region is considered as nearly drought condition (annual rainfall of 200-400mm), this may lead to restrict the mobilization. This study is bounded to give an idea about the leaching for a short period. To get more detailed information about the leaching, a better long duration leaching study will more feasible to understand elemental mobility.

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