JETIR.ORG ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

Leaching Behavior of Elements From Coal Fly Ash Using Continuous and Batch Extraction Process

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ABSTRACT

An attempt has been made to evaluate the leaching behaviors of toxic metals of coal fly ash collected from different thermal power plants. Leaching of the heavy metals namely Aluminum (Al) chromium (Cr) Copper (Cu) Cobalt (CO) Mercury (Hg) Manganese (Mn) Lead (Pb) Iron (Fe) and Zinc (Zn) were investigated using two different leaching methods namely, Toxicity Characterization Leaching Procedure (TCLP) and Water Leaching Test (WLT) to check the possibility of groundwater contamination. This study is performed to evaluate factors affecting the leaching of heavy metals like liquid to solid ratio (L/S) and reaction time. The higher concentration of heavy metals in TCLP test indicates that the pH value of the leaching medium significantly affects the transfer of these elements to the liquid medium. The results from WLT show that the toxic metals from fly ash are not easily leached in neutral water but, reaction time and L/S ratio have an impact on the leaching behaviors of coal fly ash. However, in the entire leaching test for all the samples, leaching is within the regularity level of the USEPA-RCRA-D list. Toxicity Characteristic Leaching Procedure (TCLP) indicates that the toxic metals and elements contained in the coal fly ash could be potentially transferred to the liquid phase depending upon the initial pH of the leaching medium.

Keywords: Coal Fly Ash, Metals Leaching, TCLP, Water extraction

1. INTRODUCTION

Energy generation is a key ingredient in the development of a country. India's sustainable development and rapid economic growth are demanding more energy day by day. To fulfill the energy, demand the Government is utilizing conventional and non-conventional energy sources. Though the world is moving faster towards non-conventional energy sources, in India Coal's contribution towards total power production is more than 50% and it will be consumed up to next 30 years. Indian thermal power plants consume 759.02 million tons of coal which produces 270.82 million tons of ash, which indicates that the coal used in power production is of low-grade quality with ash content 30-45% (CEA, 2022). Coal combustion burns carbonaceous material to produce energy and generates coal ash in the form of fly ash and bottom ash. Electro-static precipitators (ESP) collect fine particles known as fly ash and bottom ash collected at the bottom of hopper. Most of the power plants are facing the problem of handling this coal ash and stored in pond ash nearby power generating units. Transportation of dry ash from unit to storage/disposal location is costly so the most economical option is transportation in slurry form. Coal ash: water ratio for slurry depends upon the ash type, and distance of the pipeline and varies from 1:4 to 1:20 (Roy R et al 2021). The slurry is transported through the pipelines and disposed off in ponds or open pits where ash particles get settled over a period and water flows towards the lower gradient in open space. Researchers (Singh et al., 2010) have stated that fly ash contains toxic heavy metals like Zinc as Zn, Nickel as Ni, Lead as Pb, Manganese as Mn, Aluminum as Al, Cadmium as Cd, Chromium as Cr, Selenium as Se, Boron as B, and Arsenic as As.

Heavy metal present in fly ash can reach to aquatic life and can impact on ecosystem hence, systematic study of the leaching behaviors of fly ash is important. In the last few decades, extensive research (Ivanova et al. 2011, Lau et al. 2001) has been conducted to estimate the toxicity of fly ash. Many researchers (Mahajan et. al. 2022, Leelarungroj et al 2018, Zhao et al. 2020, Davide et al. 2022) have observed that the leaching behavior of elements present in fly ash depends upon various factors like parent coal type, combustion techniques, fly ash particle size, the number of elements present in fly ash, geographical location of the disposal site, and mainly pH of the aqueous medium. The mobility of different elements depends upon the pH of the slurry medium. Laboratory-based leaching studies have proven that elements like Ca, Ni, Fe show greater leachability in acidic conditions. Elements like Se, Cd, and Ni leach out less aggressive conditions while As, Cr, Pb, and Ar leach under more aggressive conditions (Choi et Al. 2002, Shivpuri et al. 2011).

This study aims to investigate the leaching behavior of fly ash in various mediums and the potential impact of fly ash on groundwater and surface water quality. In this study, the leaching potential of heavy metals like Al, Fe, Zn, Cu, Cd, Cr, Mn, Co etc. from fly ash was investigated to predict potential environmental threats.

MATERIAL AND METHODS

Samples (fly ash) were collected directly from the collection point located at dust hoppers of electrostatic precipitators (ESP) in the thermal power plants at Talcher, Odisha. Samples are collected using the zip lock bags and labelled properly according to their location. Collected samples were transferred to the laboratory for further analysis. All the samples are oven dried at 105º before the analysis to remove the moisture content. All glassware was washed regularly using chromic acid, neutralized with diluted dilute alkali, washed with tap water, deionized water and finally oven dried before starting each experiment to avoid possible contamination.

Toxicity Characteristics and Leaching Procedure (TCLP)

A standard leaching approach has been devised to evaluate the environmental impact of hazardous chemicals and to determine their mobility into an aqueous phase. Following US EPA SW-846 technique, 1311, the toxicity characteristic leaching procedure (TCLP) was carried out. Ten grams of ash samples were obtained, and extraction fluid was added in a 1:20 (m/v) ratio. For eighteen hours, the extraction assembly was firmly sealed at room temperature and rotated at 30±2 rpm in an orbital shaker. ICP-MS was used to evaluate the filtrates for heavy metals after the suspension had been filtered. The TCLP extraction fluid was created by combining 500 milliliters of deionized water, 5.7 milliliters of glacial acetic acid, and 64.3 milliliters of 1N NaOH until the mixture reached one liter. The extraction fluid's pH was kept constant at 4.9.

Water Leaching Test (WLT)

Using a modified version of ASTM D 3987, the test is run to find the leachable elements concentration in water. The solid to liquid ratio varies during the water leaching test, and the leaching periods are determined by the contact time. In order to determine the solid to liquid ratio in the range of1:5,1:10, and 1:20, 5 grams of fly ash is combined with 25 milliliters, 50 milliliters, and 100 milliliters of deionized water. After mixing, they are stirred at 30 RPM for eighteen hours. When it comes to contact duration, the normal protocol calls for a constant solid to liquid ratio of 1:20, while the reaction time is allowed to vary, ranging from 6 to 24 hours. After mixing, they are stirred at 30 RPM for six, twelve, and twenty-four hours. To get rid of the ash particles, all of the samples are next filtered through regular filter sheets. By passing the water through a 0.2µm syringe filter, the fine ash particles that remain in it are eliminated. After that, the sample is acidified with HNO3 and preserved by being stored in a refrigerator. To prevent mistakes, each sample is handled three times. Thermo Scientific's iCAP 6300 DUO model is an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) that is used to measure leachable metal concentrations. The detection limit of elements in ICP-OES as Fe (0.0003 ppm), Mn (0.018 ppm), Zn (0.0002 ppm), Pb (0.05 ppm), Cd (0.009 ppm), Cr (0.0006 ppm, and Cu (0.0004 ppm) while detection limit for ICP-Ms is as Mn (0.0001 ppm), Ni (0.005 ppm), Pb (0.009 ppm), Cr (0.01 ppm) , Fe (0.0003 ppm), Zn (0.001 ppm), Cd (0.0001 ppm), Co (0.0004 ppm), Cu (0.0004 ppm) and Hg (0.000075 ppm). Flame Photometer (Model: CL361) was used to analyze the parameters, Na and K. as well as the ICP-MS, or inductively coupled plasma mass spectrometer.

RESULT AND DISCUSSION

Fly ash leaching studies for heavy metals and elements are very important to predict the possible impact on the environment associated with its disposal into the open pit, abandoned mines and reclamation of land and low-laying area.

Toxicity Characteristic and Leaching Procedure (TCLP)

Table 1 displays the heavy metal concentration following TCLP testing. A solution of the sodium hydroxide and glacial acetic acid with a constant pH of 4.9 is employed as the extraction fluid in this process. In comparison to other elements, aluminum (Al) exhibits great solubility in acidic media (figure 10) In the leaching media, copper (Cu), manganese (Mn), and zinc (Zn) were found to be slightly soluble, but Iron (Fe), chromium (Cr), nickel (Ni), and mercury (Hg) were found to be considerably less soluble. Samples are devoid of cadmium (Cd), cobalt (Co), and lead (Pb), with the extraction of cobalt samples S2 and S5. The metal content in TCLP is found to be higher than the metal concentration found in the water leaching test. Since metal solubility typically declines with rising pH, the higher concentration of heavy metals found in the TCLP test can be attributed to the leaching medium acidic pH (Goulds JP et al 1989). While the water leaching test was conducted in deionized water with an initial pH of 7, the extraction of fluid used in the TCLP had a pH of 4.9. Every leaching metal concentration in the test falls within the USEPA-RCRA-D Lists regularity threshold.

Table 1: Heavy metals concentration as a result of TCLP test (Concentration in mg/l)

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
RL	**--**	**1**	**--**	**--**	**5**	**5**	**--**	**--**	**--**	**--**	**5**
S1	15.2	*S	0.003	1.63	*S	0.49	0.0098	0.05	0.433	0.30	*S
S2	12.3	0.011	0.001	1.78	*S	0.38	0.0099	0.03	0.428	0.38	*S
S3	11.7	*S	0.013	1.20	*S	0.30	0.0087	0.03	0.004	0.19	*S
S4	9.7	*S	0.004	0.32	*S	0.40	0.0086	0.04	0.005	0.77	*S
S5	15.2	*S	0.003	1.63	*S	0.49	0.0098	0.05	0.004	0.30	*S
S6	3.1	0.003	0.015	1.14	*S	0.38	0.0086	0.02	0.009	0.11	*S

*S: below detection limit *RL: Regulatory level of USEPA-RCRA-D List.

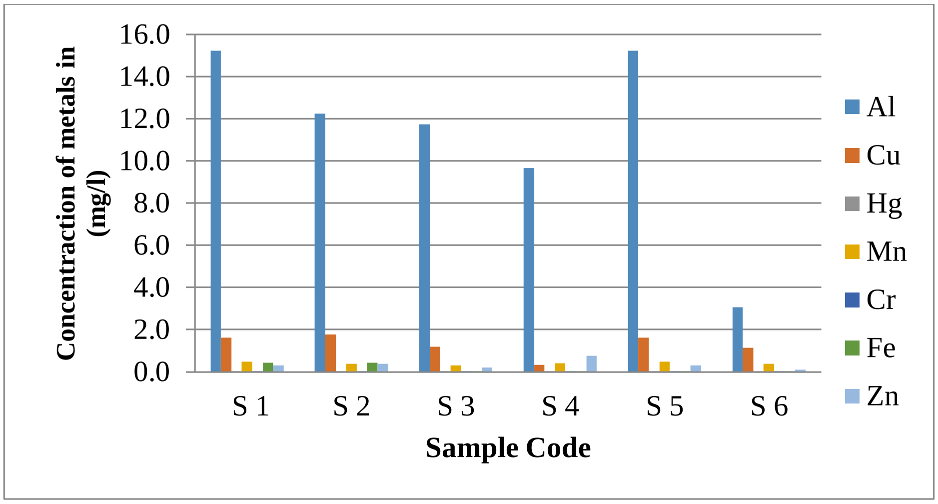

Figure 1: Comparison of heavy metals observed during TCLP test.

Water Leaching Test (WLT)

a) Solid to Liquid ration (S/L)

The concentration of heavy metals leached out with deionized water as result of water leaching test for solid to liquid ratio is shown (Table. 2). Three sets have been run at different solid to liquid ratio i.e. 1:5, 1:10 and 1:20 respectively to observe the leaching behaviors. Same contact time (18hrs) is provided during the experiment for all the three sets. The highest concentration is observed for first set at solid to liquid ratio 1:5 and the lowest concentration observed for 1:20 ratio. It is observed that Al, Mn, and Fe concentration is observed more than the other elements during water leaching tests for all sets (ASTM 199a). While concentration at set 1 (1:5) is found more than the other two sets. The concentration of Al is found more for all samples while Mn and Fe concentration is varying sample to sample Figure 2 and Figure 3.

Table 2: Heavy metals concentration as a result of Water leaching test at solid to liquid ratio 1:5. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	85	0.6	*S	0.005	0.10	11	*S	3.4	69	8	0.04
S2	62	*S	*S	*S	*S	1	*S	0.06	59	*S	*S
S3	64	1.0	*S	*S	0.00	43	*S	2.6	10	*S	*S
S4	88	0.3	*S	*S	0.04	26	*S	0.7	18	*S	*S
S5	89	0.7	*S	*S	0.07	42	*S	1.8	28	*S	0.006
S6	20	*S	*S	*S	0.03	*S	0.05	*S	201	*S	*S

*S: below detection limit

Table 3: Heavy metals concentration as a result of Water leaching test at solid to liquid ratio 1:10. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	58	0.3	*S	0.001	0.07	6	*S	1.7	41	0.5	0.14
S2	20	*S	*S	*S	*S	0.6	*S	*S	44	0.3	*S
S3	29	0.6	*S	*S	*S	25	*S	1.2	9	0.6	*S
S4	9	0.2	*S	*S	0.03	15	0.4	0.4	16	*S	0.02
S5	7	0.3	*S	*S	0.03	23	*S	0.7	21	*S	0.01
S6	11	*S	*S	*S	0.00	0.1	27	*S	45	*S	0.01

*S: below detection limit

Table 4: Heavy metals concentration as a result of Water leaching test at solid to liquid ratio 1:20. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	10	0.1	*S	*S	0.01	2	*S	0.5	9	0.005	*S
S2	7	*S	*S	*S	*S	*S	*S	*S	0.2	0.044	*S
S3	6	0.3	*S	*S	*S	3	*S	0.4	5	*S	*S
S4	0.1	0.1	*S	*S	*S	6	*S	*S	0.1	*S	*S
S5	3	0.2	*S	*S	*S	1	*S	0.2	5	*S	*S
S6	9	*S	*S	*S	*S	*S	*S	*S	2	*S	*S

*S: below detection limit

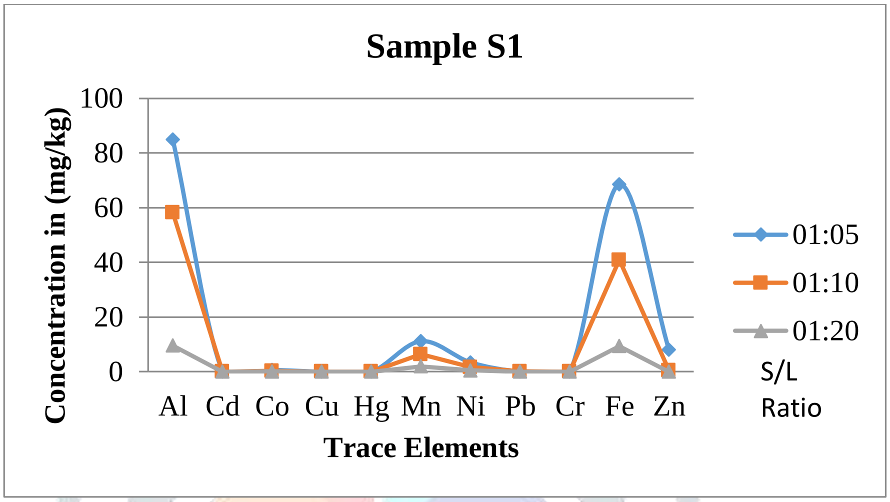

Figure 2: Comparison of heavy metals observed in sample S1during Water Leaching w.r.t solid to liquid ratio.

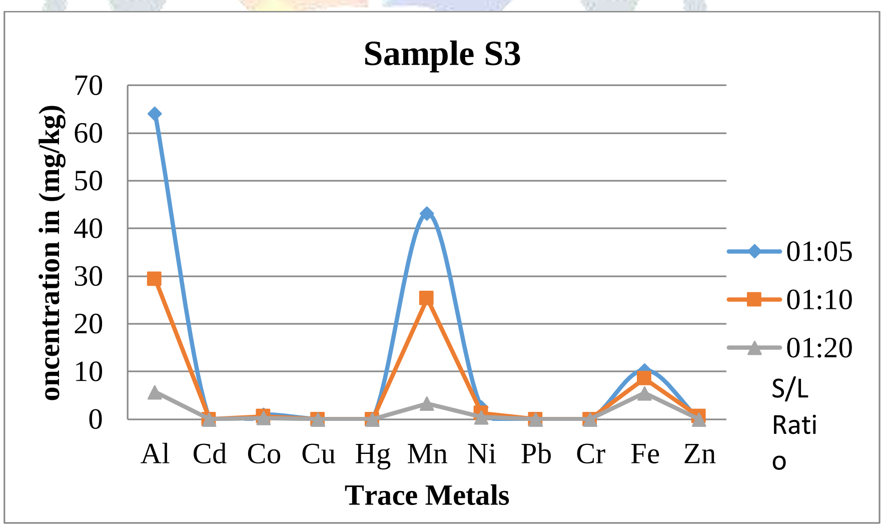

Figure 3: Comparison of heavy metals observed in sample S3 during Water Leaching w.r.t solid to liquid ratio.

b) Contact Time

In order to examine how fly ash leaches in relation to contact time, three batches of the material were run for varying lengths of time (6 hours, 12 hours, and 24 hours) while maintaining a constant solid to liquid ratio of 1:20. The concentration of heavy metals is shown in Tables 5, 6, and 7 in relation to the specified contact times of 6 hours, 12 hours, and 24 hours, respectively. The concentration of iron (Fe) and aluminum (Al) in each sample of each batch is noted (Figures 4 and 5). These two elements have higher leaching percentages than the other elements. Lead (Pb), Copper (Cu). While cobalt (Co), Zinc (Zn), manganese (Mn), and Nickel (Ni) are not detected in the first set (6 hours), low concentrations were noted in sets 2 and 3 (12 and 24 hours), respectively. Mercury

(Hg) and Chromium (Cr) are not discovered in any sample for any set. As contact time increases, the concentration of leached heavy metals rises.

Table 5: Heavy metals concentration as a result of Water leaching test w.r.t. contact time 6hrs. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	4.6	0.02	*S	*S	*S	*S	*S	0.27	5.8	*S	*S
S2	3.3	*S	*S	*S	*S	*S	*S	*S	0.8	*S	*S
S3	0.9	*S	*S	*S	*S	*S	*S	*S	2.7	*S	*S
S4	0.4	0.22	*S	*S	*S	*S	*S	*S	0.9	*S	*S
S5	1.0	*S	*S	*S	*S	*S	*S	*S	3.7	*S	*S
S6	3.7	*S	*S	*S	*S	*S	*S	*S	1.0	*S	*S

*S: below Not Detected

Table 6: Heavy metals concentration as a result of Water leaching test w.r.t. contact time 12hrs. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	7.6	0.13	*S	*S	*S	0.3	*S	0.3	9	*S	*S
S2	4.6	*S	*S	*S	*S	*S	*S	*S	1	*S	*S
S3	1.1	0.67	*S	*S	*S	0.2	*S	0.2	4	*S	*S
S4	0.9	0.13	*S	*S	*S	*S	*S	*S	1	*S	*S
S5	1.7	0.07	*S	*S	*S	0.1	*S	0.1	5	*S	*S
S6	8.6	*S	*S	*S	*S	*S	*S	*S	2	*S	*S

*S: below detection limit

Table 7: Heavy metals concentration as a result of Water leaching test w.r.t. contact time 24hrs. (Concentration in mg/kg).

Sample Code	Al	Co	Hg	Cu	Cd	Mn	Cr	Ni	Fe	Zn	Pb
S1	11	0.2	*S	*S	0.02	0.5	*S	0.8	10	0.005	*S
S2	7	*S	*S	*S	0.02	*S	*S	*S	1	0.003	*S
S3	2	0.7	*S	*S	*S	0.4	*S	0.4	5	*S	*S
S4	1	0.5	*S	*S	*S	*S	*S	*S	2	*S	*S
S5	4	0.2	*S	*S	0	0.2	*S	0.3	5	*S	*S
S6	10	*S	*S	*S	*S	*S	*S	*S	2	*S	*S

*S: below detection limit

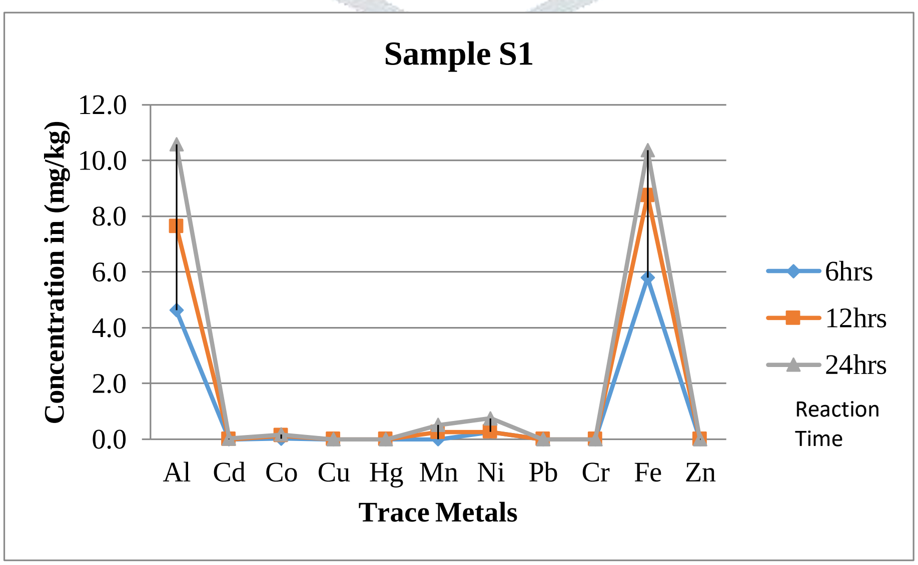

Figure 4: Comparison of heavy metals observed in sample S1 during Water Leaching w.r.t contact time.

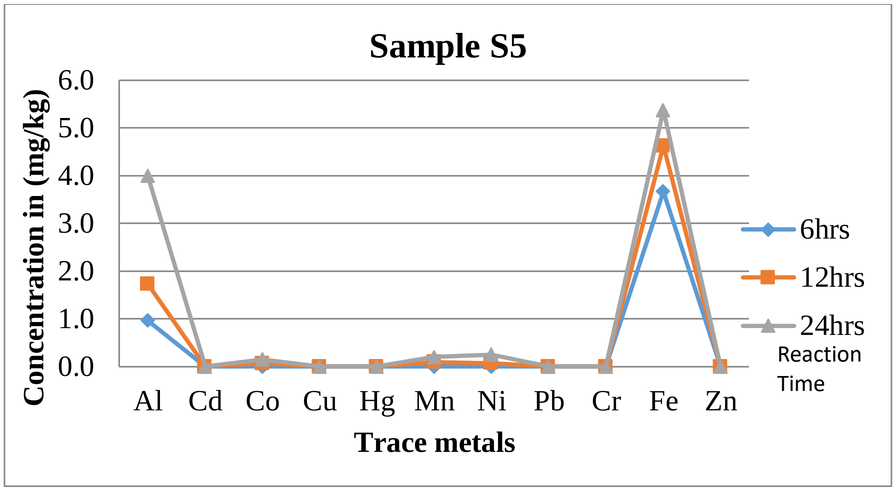

Figure 5: Comparison of heavy metals observed in sample S5 during Water Leaching w.r.t contact time.

The lowest solubility was found in the water leaching test at 1:20 solid to liquid ratio and 24 hrs contact time. Trace metal concentrations in leaching medium were observed within the Indian standards for disposal of effluents. A similar comparison of TCLP and water leach test indicates that all metals were well within specified limits.

CONCLUSION

Based on the study of the leaching of heavy metals from ash, the following conclusion can be drawn.

- There appeared to be significant differences in leaching characteristics with respect to fly ash origin, pH of the leaching medium, liquid-to-solid ratio and contact time provided during an experiment
- Aluminum shows a relatively higher leachability than any other than any other element in fly ash.
- The concentration of leached heavy metals after extraction is directly related to the initial pH of the leaching medium.
- In the TCLP test all the leaching metals concentrations are within the regularity level of USEPA-RCRA-D.

Acknowledgment

The authors are grateful to the Director, CSIR-National Environmental Engineering Research Institute, Nagpur and HOD, WTMD for providing the facilities for this project. The authors are thankful to the environmental management group of the NTPC and BSL for providing fly ash samples.

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