



Leaching Behaviour of Fly Ash: A Review

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ABSTRACT

Disposal of fly ash produced in a large quantity is a major problem in coal fired thermal power plants. The coal quality depends upon its rank and grade. Fly ash content in Indian coal ranges from 35% to 50% consisting of a variety of toxic elements including heavy metals that damage the surrounding environment. During transportation, disposal and storage phases, the residues from coal combustion may pollute both ground and surface waters through leaching. The fly ash enriched in Ca, Ni and Fe showed greater leachability in acidic or ion exchangeable conditions. Principal mechanisms of metal leachability are the dissolution of primary solids under aggressive environment and/or precipitation sorption reactions, which are primarily dependent on pH. Fe is found to be tightly bound to the ash and does not leach easily while Ca is highly soluble and leaches out almost all the mediums. The leachability of trace elements in fly ash is time dependent and pH values of the leachant.

INTRODUCTION

The growth of an economy is closely related to growth in its energy consumption, particularly in the case of developing country like India. Electricity, by virtue of being a relatively clean, efficient and convenient form of energy has a vital role to play in the socioeconomic development of the country (Mathur et al. 2003). Fossil fuel (coal, oil and gas) is going to remain the primary source of energy in India in spite of attempts to harness hydel, nuclear and renewable sources of energy. In the sphere of fossil fuels, India is not very well placed with reference to resources of oil and import of oil is very expensive. However, the situation with respect to coal reserves is comparatively better, as it contributes 84 percent of thermal power generation while, gas and oil contribute 13 and 3 percent, respectively for the same (Mudgal 1998, Singh 1998, Lookman & Rubin 1998, Chandra & Chandra 2004).

India has about 211 billion tons of coal reserves, which is known to be the largest resource of energy and presently 240 MT of coal is being used annually to meet the nation's electricity demand (Ahmad et al. 2012). Coal based thermal power plants have been a major source of power generation in India where about 57% of the total power is obtained from coal based thermal power plants (Nawaz 2013). Coal is a combustible sedimentary rock, which consists of organic and inorganic materials and its formation and composition depend on the earth's crust composition, climate and flora of its location (Clark & Sloss 1992, Finkelman 2004, Walker

1993, Pandey et al. 2011). Coal quality depends upon its rank and grade. Ascending order of coal rank on the basis of carbon content is lignite, sub bituminous coal, bituminous coal and anthracite. Indian coal is largely of sub bituminous rank, followed by bituminous and lignite. Ash content in Indian coal ranges from 35% to 50% (Bhattacharjee & Kandpal 2002, HVFAC 2005, Pandey et al. 2011).

High ash coal means more wear and tear of the plant machinery, low thermal efficiency of the boiler, slogging, choking and scaling of the furnace and most serious of them all, generation of a large amount of fly ash. Fly ash is fine glass powder, the particles of which are generally spherical in shape and ranges in size from 0.5 to 100 μm (Senapati 2011). Fly ash quality depends on coal type, coal particle, fineness, percentage of ash in coal, combustion technique, air/fuel ratio and boiler type (Dhadse 2008). Fly ash is classified into two types according to the type of coal used. Anthracite and bituminous coal produces fly ash classified as class F. Class C fly ash is produced by burning lignite or sub bituminous coal (Senapati 2011). Disposal of fly ash produced in a large quantity is a major problem in coal fired thermal power plant.

Fly ash contains a variety of toxic elements that damage the living and non living constituents of the environment with 100 tons of coal burnt produces about 100 kg Pb, 40 kg Zn, 15 kg Cu, 8 kg Cd, 40 kg Cr, 3 kg As, which ultimately find their way in surrounding environment (Sahu 1990). Different environmental issues of concern due to

disposal of such huge quantity of ash on land are the effects on air, soil, vegetation, ground and surface water. The major constraint in block disposal of fly ash is the land requirement as for every 20-25 years (Mishra 2011).

At thermal power plants, fly ash is presently collected or disposed of by using either dry or wet systems. In dry disposal system, electrostatic precipitation (ESP) is the most popular and widely used method of emission control today, which enables the collection of dry fly ash. After collecting the fly ash in ESP, it is then transported by trucks or conveyors at the sight and disposed of by constructing a dry embankment. In wet disposal system, the fly ash is mixed with water and transported as slurry through pipe and disposed of in ash ponds or dumping areas near the plants. Being cheaper than other methods of fly ash removal, it is a widely used method at present in India (Nawaz 2013).

During transport, disposal and storage phases, the residues from coal combustion are subjected to leaching effects of rain and part of the undesirable components in the ash may pollute both ground and surface waters (Benito et al. 2001). These solid residues (fly ash) can be leached in higher concentration than drinking water sources. Fly ash contains trace amounts of toxic metals that may have negative effects on human health and on plants (Mehara et al. 1998).

CHEMICAL COMPOSITION OF FLY ASH

Fly ash is considered as amorphous and mixture of ferroalluminosilicate minerals. However, the composition of fly ash depends mainly upon the geographical factors related to coal deposit, combustion conditions and the type of ash collection devices. The major constituents of fly ash are primarily oxides of Si, Al, Fe, Ca and Mg which constitutes about 95-99% of total constituent. Minor constituents of fly ash are Ti, Na, K and S comprising 0.5-35% (Xiuping & Clark 2011, Ahmad & Prakash 2010, Nawaz 2013). Typical chemical composition and physical properties of fly ash are given in Table 1.

From the X-ray diffraction pattern it is found that the major crystalline phases present in fly ash are quartz (SiO_2) and mullite ($\text{Al}_6\text{Si}_2\text{O}_{13}$) and minor phases are iron oxides such as magnetite (Fe_3O_4) and haematite (Fe_2O_3) (Moreno et al. 2005, Metcalf et al. 2006, White & Case 1990, Kourkouzas et al. 2007, Lee et al. 1999, Khanra et al. 1998, Monte & Sabbioni 1984, Mishra & Das 2010). During the burning of coal, minerals undergo thermal decomposition, fusion, disintegration and agglomeration. Several elements present in a volatile form may be vaporized. A portion of non combustible (ash forming) elements in the boiler enter into slag or bottom ash and rest of the inorganic residue present in the flue gas fly ash (FA) left after the complete combustion of

coal, containing, however, also small amounts of carbon and condensed carbonaceous compounds. One of the most prominent features of fly ash is the gradation of effects of particle size on elemental concentration (Pandey et al. 2011). Davison et al. (1974), Klein et al. (1975), Kaakinen et al. (1975) observed that As, Cd, Cu, Ca, Mo, Pb, S, Sb, Se, Ti and Zn tend to increase in concentration with decreasing fly ash particle size.

Khan et al. (2013) analysed the chemical composition of fly ash from Barapukuria Thermal Power Plant (BTPP) by using x-ray fluorescence technique. The major component of fly ash of BTPP was alumina (Al_2O_3) and silica (SiO_2) with 40.12% and 50.20% respectively. The results obtained from XRF analysis are given in Table 1, which indicate that fly ash (FA) is a useful component for alumina and aluminium recovery. It is also useful to produce aluminium based agrochemicals, construction materials, carbothermal reduction process, etc. Iron oxide ($\text{Fe}_2\text{O}_3/\text{FeO}$) and titanium dioxide (TiO_2) concentrations are little but higher, i.e., 3.32% and 2.38% respectively than other elements. Alkali metals such as sodium oxide (Na_2O), potassium oxide (K_2O), calcium oxide (CaO), magnesium oxide (MgO) content in fly ash is less and found to be 0.06%, 0.93%, 1.29% and 0.20% respectively. On the basis of chemical composition, total amount of alkali oxide (Na_2O , K_2O , CaO , MgO , etc.) was lesser than 10% and the sum of Al_2O_3 , SiO_2 and Fe_2O_3 presented in fly ash was greater than 70% indicating that the analysed fly ash was class F-type. Various trace elements were also found in the fly ash as manganese oxide (MnO) 0.05%, cupric oxide (CuO) 0.015%, nickel oxide (NiO) 0.010%, lead oxide (PbO) 0.007%, zinc oxide (ZnO) 0.006%, phosphorus pentoxide (P_2O_5) 0.66%, and sulphur trioxide (SO_3) 0.45%.

Wang (2014) had assessed the mineralogical and chemical composition of magnetic fly ash fraction. The fly ash samples were collected from the respective electrostatic precipitators of three power plants (denoted as XD, XH and HD) in Xuzhou, China. In order to identify the Fe bearing minerals present in coal ash, x-ray powder diffraction (XRD), scanning electron microscope with energy dispersive x-ray (SEM EDS) and Mossbauer spectroscopy were performed on samples. Major elements including Na, Mg, Al, Si, K, Ca, Ti and Fe in bulk fly ash samples were determined by x-ray fluorescence spectrometry (XRF). Trace metals including Co, Cu, Zn, Pb, Mo, Cd and Ni concentrations were measured by inductive coupled plasma mass spectrometry (ICP-MS). The XRD analysis of nonmagnetic fractions of three fly ash samples showed the presence of quartz (SiO_2) and mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$). The elements revealed by the EDX spectra were Fe, Al, Ti, K, Ca, Si, Na, Mg and C in the magnetic fractions of fly ash. The major elemental concentra-

tions of three bulk fly ashes determined by XRF are summarized in Table 1. The concentration of trace metals in magnetic and nonmagnetic fractions is also listed in Table 1. The results showed that the concentrations of Co, Ni, Cu, Zn, Mo and Cd were high in magnetic fraction than in non-magnetic fraction.

Sushil & Batra (2006) investigated the heavy metal content of fly ash and bottom ash from three major coal fired thermal power plant in North India using flame atomic absorption spectrometry. The collected fly ash samples were tested for chromium, lead, zinc, nickel, cobalt, copper and manganese. Results for the heavy metal analysis of fly ash samples are presented in Table 1.

Choi et al. (2002) compared the chemical composition of feed coal and fly ash from five different power plants in Korea; Boryong (BR), Samcheonpo (SCP), Seocheon (SC), Yongwol (YW) and Yongdong (YD). BR and SCP were burning bituminous coals whereas SC, YD and YW use anthracite coals. Chemical analysis of coals is given in Table 2.

Anthracite coals were found to be higher in SiO₂, Al₂O₃, CaO and K₂O concentrations. The higher K concentration in anthracite coals appeared because it was derived from illite. Other major elements including Fe, Na, Ca and Mg showed similar concentrations. The mineral matter contents in the anthracite coals were 3-5 times higher than in the bituminous coals and such high mineral matter contents were considered to have influenced the higher concentrations for the Si, Al and K in the anthracite coal. As for trace elements, the anthracite coals also showed higher concentrations compared with the bituminous coals, especially for Cr, Cu and V. Contrary to the coal chemistry, the fly ashes from bituminous coals were higher in SiO₂, Fe₂O₃, CaO and MgO concentrations. However, K₂O concentrations were still higher in the anthracite fly ash and fly ash chemistry seems to reflect higher mineral matter contents in the feed coal. It was observed that the higher concentrations of Si, Al, Fe, Ca and Mg in the bituminous coal fly ash appeared due to lower content of mineral matter.

Table 1: Chemical composition of fly ash.

	Nawaz (2013)	Khan et al. (2013)		Wang et al. (2014)		Sushil and Batra (2006)					
	Fly Ash	Fly Ash (Barapukuria TPP)	Fly Ash (XD TPP)	Fly Ash (HD TPP)	Fly Ash (XH TPP)	Fly Ash (FA1)	Fly Ash (FA 2)	Fly Ash (FA 3)			
MgO	0.42-8.79%	0.20%	0.38%	0.61%	0.91%	-	-	-			
Al ₂ O ₃	5.23-33.99%	40.10%	28.8%	28.1%	24.5%	-	-	-			
SiO ₂	27.88-59.40%	50.20%	39.8%	41.8%	39.9%	-	-	-			
P ₂ O ₅	-	0.66%	-	-	-	-	-	-			
Na ₂ O	0.20-6.90%	0.06%	0.11%	0.39%	0.43%	-	-	-			
Cl	-	-	-	-	-	-	-	-			
K ₂ O	0.64-6.68%	0.93%	0.71%	1.12%	1.29%	-	-	-			
CaO	0.37-27.68%	1.29%	5.84%	2.88%	3.89%	-	-	-			
TiO ₂	0.24-1.73%	2.38%	1.05%	0.97%	0.84%	-	-	-			
SO ₃	0.04-4.71%	0.45%	-	-	-	-	-	-			
Cr	-	-	-	-	-	-	-	-			
MnO	-	0.05%	-	-	-	-	-	-			
Fe ₂ O ₃	1.21-29.63%	3.32%	5.22%	3.84%	4.41%	-	-	-			
NiO	-	0.01%	-	-	-	-	-	-			
CuO	-	0.0158%	-	-	-	-	-	-			
ZnO	-	0.0068%	-	-	-	-	-	-			
PbO	-	0.0073%	-	-	-	-	-	-			
LOI	0.21-28.37%	2.0%	-	-	-	-	-	-			
			Mag- netic	Non Mag- netic	Mag- netic	Non Mag- netic	Mag- netic	Non Mag- netic			
Co	-	-	20.07%	7.1%	36.8%	13.2%	28.4%	14.2%	18 mg/kg	13 mg/kg	8 mg/kg
Ni	-	-	41.2%	22.4%	101.6%	41.9%	105.4%	65.3%	63 mg/kg	39 mg/kg	28 mg/kg
Cu	-	-	78.6%	38.5%	88.4%	67.3%	87.3%	64.3%	83 mg/kg	56 mg/kg	58 mg/kg
Zn	-	-	79.6%	65.2%	289%	202%	657.3%	243.3%	124 mg/kg	60 mg/kg	64 mg/kg
Mo	-	-	4.9%	3.2%	8.7%	6.7%	13.2%	7%	-	-	-
Cd	-	-	1.7%	1.8%	0.8%	0.6%	0.4%	0.13%	-	-	-
Pb	-	-	36.2%	36.8%	50.2%	67.3%	84%	67.3%	56 mg/kg	20 mg/kg	34 mg/kg
Cr	-	-	-	-	-	-	-	-	103 mg/kg	90 mg/kg	87 mg/kg
Mn	-	-	-	-	-	-	-	-	62 mg/kg	47 mg/kg	139 mg/kg

Table 2: Chemical composition of the coal and fly ash from five different coal fired power stations in Korea (Choi et al. 2002).

	Coal					Fly ash				
	Sub Bituminous		SC	Anthracite		Sub bituminous		Anthracite		
	BR	SCP		YW	YD	BR	SCP	SC	YW	YD
SiO ₂ (wt %)	8.5	8.0	16.3	21.6	15.2	55.2	52.2	43.5	44.6	47.3
Al ₂ O ₃ (wt %)	3.5	3.6	10.0	13.1	11.0	19.9	23.1	27.5	25.1	29.2
Fe ₂ O ₃ (wt %)	1.5	0.6	1.4	1.8	1.5	9.7	6.1	4.7	3.8	4.4
MgO (wt %)	0.2	0.1	0.3	0.4	0.2	1.2	1.0	0.9	0.8	0.7
CaO (wt %)	0.6	0.4	0.4	0.3	0.2	3.7	3.8	0.9	0.5	0.5
Na ₂ O (wt %)	0.1	0.1	0.1	0.1	0.1	0.3	0.3	0.3	0.2	0.2
K ₂ O (wt %)	0.1	0.1	1.2	1.6	1.4	1.0	1.0	3.8	3.3	3.9
TiO ₂ (wt %)	0.2	0.2	0.4	0.6	0.5	1.2	1.1	1.5	1.5	1.6
P ₂ O ₅ (wt %)	0.1	0.1	0.1	0.1	0.1	0.3	0.5	0.2	0.2	0.2
MnO (wt %)	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.0	0.0
S (wt %)	0.3	0.4	0.5	0.5	1.0	0.1	0.1	0.1	0.1	0.1
Co (ppm)	11	2	4	4	5	53	18	12	8	8
Cr (ppm)	19	8	19	22	39	95	69	86	80	498
Ni (ppm)	41	6	14	14	22	174	74	66	41	72
Cu (ppm)	13	11	19	34	39	66	63	109	77	109
V (ppm)	38	30	46	45	62	147	210	175	130	157
Pb (ppm)	6	14	16	22	28	29	32	103	36	68
Zn (ppm)	21	14	220	19	34	93	93	167	43	97

Drozhzhin et al. (2008) studied the mineral substances in coal combusted at thermal power stations in Russia and of fly ash trapped in the electrostatic precipitators of thermal power stations, as well as the chemical composition and properties of microsphere present in fly ash which is given in Table 3. The chemical compositions of mineral substances in the coals and fly ash trapped in combustion of the coals were observed very similar but the composition of microsphere was somewhat different. As compared with the mineral matter of coals and their fly ash, the alluminosilicate microsphere were characterized by increased concentrations (on an oxide basis) of aluminium, potassium (sodium), and (in most cases) silicon, whereas the amounts of iron and calcium were lower than those in the fly ash of parent fuel.

Malik et al. (2016) reported that the ash composition analysis could be helpful in developing a pollution abatement approach for various applications of fly ash. Their study was dealt with the characterization of fly ash samples collected from different NTPC units in India for eleven major oxides (Na₂O, MnO, SO₃, P₂O₅, MgO, K₂O, TiO₂, CaO, Fe₂O₃, Al₂O₃ and SiO₂) and eleven trace elements (As, Co, Pb, Ni, La, Cu, Zn, Cr, V, Sr and Ba) by using ED-XRF technique. Among the major elements, the concentration of SiO₂ is found to be the highest in the range of 51.36-58.5% and that of Na₂O is found to be the lowest in the range of 0.02-0.17%. Among the trace analyst, Ba content is found to be the highest in the range of 126.6-1393.5 ppm and As is found to be the lowest concentration in the range of 24.8-37.4 ppm, respectively. The study revealed that all the fly

ash samples were of siliceous type as per IS: 3812.

LEACHING PROPERTIES OF FLY ASH

Leaching properties of fly ash under different leaching tests are presented in Table 4. The surface of fly ash particle is only microns in thickness and can contain leachable heavy metals which have condensed on to the surface. Inorganic salts make the particle surface more reactive than the glassy matrix. Elements which are leachable from fly ash such as As, B, Ca, Cr, Mg and Sr, are likely to be dominantly partitioned into the exterior glass surface following condensation reactions during combustion; while elements such as Al, Si, K, Pb and many other trace elements are distributed throughout the particle and are not preferentially concentrated. When fly ash comes into contact with an aqueous environment some constituents will dissolve to a greater or lesser extent and become mobile. The product of this contact is known as leachate (Zandi & Russel 2007). To assess the degree of mobilization and dissolution of constituents in the leachate, leaching tests are carried out. Extraction leaching test and batch leaching test are the two major categories of leaching tests. In extraction or batch leaching test, a specific amount of solid sample and extraction solution are combined for a set period of time. The product of this combination is leachate. The leachate is then filtered and the extract solution is stored for further analysis. The main assumption made by batch leaching tests is that at the end of the test period a steady state of constituent has been achieved (Bushell 1997, Zandi & Russel 2007). In contrast, in dynamic or column tests the leachate flows across the

Table 3: Chemical composition of fly ash (FA) matter and aluminosilicate microsphere (ASM) present in fly ash.

Combusted Coal; Power plant	Combustion Product	SiO ₂	Al ₂ O ₃	MgO	Na ₂ O	TiO ₂	CaO	K ₂ O	Fe ₂ O ₃	Cr ₂ O ₃	MnO
Ekibastuz basin SS; Omskara CHP-5	FA	65	30	0.18	0.33	1.34	1.68	0.44	3.5	-	0.36
	ASM	55	40	0.3	0.14	1	1	1.7	1.2	-	0.02
Donets basin, A; No Vocherkass TPP	FA	54	24	2.9	0.9	1.6	2.4	3.2	10	-	0.2
	ASM	58	28	1.5	1	0.7	1	5	4	0.01	0.04
Kuzhetsk basin,T; Tom- Usinkaya TPP	FA	61	21	2.21	1.6	0.93	4.34	2.41	6.5	-	-
	ASM	68	19	1.5	0.9	0.9	2.2	3.8	3.6	-	0.05
Pechora basin (Vorkuta deposit) Zh; Arkhangelsk ppm CHP-1	FA	61	21	1.9	0.6	0.9	1.5	1.2	10	-	0.2
	ASM	65	21	3.8	0.8	0.8	0.7	2.8	5.5	-	0.05
Pechora basin (Inta deposit) Zh;Cherepo- vetskaya TPP	FA	59	19	3	1.5	1	2.8	1.2	10	-	0.2
	ASM	60	21	3	0.8	0.9	5.2	2.6	5.8	0.03	0.05
Kansk-Achinsk basin, B; Barnaulskara CHP-3	FA	31	12	6	0.7		4	0.8	10	-	-
	ASM	62	25	1	0.9	0.5	4.1	2.1	1.8	-	0.03
Near Moscow basin coal B; Ryazanskaya TPP	FA	55	27	0.18	0.33	1.34	1.68	0.44	3.5	-	0.36
	ASM	56	33	1.7	0.5	1	1.6	1.9	4.1	0.03	-
Bikin deposit, coal B; Primorskaya TPP	FA	58.8	25.9	2	0.9	0.9	2.7	2.1	7.6	-	-
	ASM	60.1	28.2	1.6	1.1	0.8	2.4	2.7	6.1	-	-
Neryungri deposit, coal Gzh; Artemovskaya TPP	FA	59.8	23.6	1.7	1.1	0.7	2.2	2.6	4.1	-	-
	ASM	57.6	27.2	1.4	1.4	0.7	2.2	3.1	3.3	-	-

solid sample continuously (Zandi & Russel 2007).

Many researchers have conducted extensive studies on the leachabilities of heavy metals from different fly ashes (Sivapullaiah & Baig 2010, Baba & Kaya 2004a, Dutta et al. 2009). The leachability of trace elements present in the ash increases with decreasing pH (Sivapullaiah & Baig 2010). It has been suggested that principal mechanisms of metal leachability are the dissolution of primary solids under aggressive environment and/or precipitation sorption reactions, which are primarily dependent on pH (Kim et al. 2003). On the other hand, relatively low leachability of cationic metals from fly ashes is due to their alkaline nature (Kim 2006). For sub bituminous coal ashes from India, it was observed that the concentration of trace elements leached was more at lower L/S ratios than at higher ones (Praharaj et al. 2002).

Praharaj et al. (2002) had collected fly ash samples from four electrostatic precipitators of a power plant combusting sub bituminous coal in Orissa, India. Separate ash samples were subjected to serial batch leaching in polypropylene bottles using Milli-Q water as leachant. The test was carried out for ash samples in four steps at L/S ratios of 4, 8, 12 and 16. In the first step, 125 g of ash was added to 500 mL of Milli-Q water in 1000 mL polypropylene bottles. The bottles were then subjected to horizontal shaking at 60 rpm in an oscillating shaker for 24 hours. The leachates were immediately filtered, and acidified with nitric acid (35%). For each subsequent step, additional 500 mL of Milli-Q water was added to the ash in the bottle along with the filter paper

from the previous step. Each leaching step was carried out for 24 hours. Praharaj et al. (2002) had observed in their study that there was a decrease in the concentration of elements in leachates from L/S 4 to 16 for As, Ca, Mg, Na, S, Zn, Mo, Al, and Si. Potassium showed highest leachability but without any trend in any of the leachates with varying L/S ratios, while manganese showed lowest in all the samples. Finally it was concluded that most of the elements (As, Ca, Mg, Mn, Na, S and Zn) have maximum concentrations in the leachate at low L/S ratio, and decreased at higher L/S ratio.

Shivpuri et al. (2011) carried out sequential extraction leaching procedure to detect leachable metal concentration in fly ash. Fly ash samples were collected from the dust hoppers of electrostatic precipitators of the six thermal power plants in Maharashtra, India. The fly ash samples were subjected to leaching procedure using leaching solutions sequentially in order of increasing aggressive nature. 0.5 g of dried ash samples were mixed with 50 mL of the corresponding leaching solution in a centrifuge tube and set in end to end agitator, specifically fabricated for SEP, at 30 rpm for a specified time period. Thereafter, the samples were centrifuged at 10000 rpm for 10 minutes. The supernatant was decanted, filtered with a 0.2 µm PTFE filter and then acidified to 2% using concentrated nitric acid and preserved for analysis by ICP-AES. The solids remaining after the centrifugation were then contacted with the next leaching solution in the sequence. Following sequential leaching procedure, the fly ash samples showed different leaching trends

for the different leaching solutions. As, Cr, Mn, Zn, Cd, Ba, Pb and Co showed maximum leaching percentage in acid soluble. Se, Cd, Ni and V showed maximum leaching percentage in ion exchangeable phase while Fe leached maximum in residual solids phase. The elements in fly ash also exhibited varying behaviour for different leaching conditions like leaching medium and pH. The fly ash enriched in Ca, Ni and Fe showed greater leachability in acidic or ion exchangeable conditions. Fe was found to be tightly bound to the ash and does not leach easily while Ca was highly soluble and leaches out almost in all the mediums. Se, Cd and Ni were found to be leached at less aggressively in ion exchangeable conditions, while As, Cr, Cd, Pb and Zn leached under more aggressive conditions.

Yuan (2009) had assessed the leaching characteristics of metals in fly ash from coal fired power plants. A modified sequential extraction procedure was developed and applied to characterize the species of metals in fly ash. The two separate fly ash samples were collected from two different types of combustor in a coal fired thermal power plant in the north of China; a pulverized coal boiler (PC) and circular fluidized bed (CFB) boiler. Five-step extraction procedure was adopted in this experiment. The chemical fractions were divided into water extractable fraction (F1), ion exchangeable and carbonate fraction (F2), Fe-Mn oxide fraction (F3), Organics and sulphides binding fraction (F4), and residual fraction (F5). The obtained results in the study of Yuan (2009) revealed that the elements V, Cu, Zn and Cd were the most active metals in the fly ash. For the other elements in fly ash, more than 80% of the total amount was in the most stable fraction, i.e. the residual fraction, which does not pose a serious environmental risk.

Baba & Kaya (2004b) identified leaching characteristics of solid waste (fly ash) generated from thermal power plants; Yenikoy thermal power plant, Kemerkoy thermal power plant and Yatgan thermal power plant in western Turkey and compared the three different procedure of extraction (extraction procedure, toxicity characteristic leaching procedure, ASTM extraction procedure). Fly ash and bottom ash samples were collected from thermal power plants. The extractions were made following the EP, TCLP and ASTM procedure for the extraction of toxic elements from bottom and fly ash. In ASTM extraction procedure, the measured Cd and Pb concentrations were significantly below the specified level of 1.0 and 5.0 ppm, respectively, for all fly and bottom ashes. However, in other extraction procedures (TCLP and EP) the measured Cd and Pb concentrations were considerably above the EP toxicity levels. In the EP extraction procedure the measured Cd and Pb concentrations were found to be considerably above the EP toxicity levels. But according to the TCLP extraction procedure, Pb concen-

tration was considerably above the EP toxicity level, whereas the measured Cd concentrations were significantly below the specified toxicity level of 1.0 ppm in all the power plants. The comparison of results revealed that the ASTM procedure results in much lower dissolved metal concentrations than the EP and TCLP procedures. The results of the study indicated that pH is an important parameter affecting the leaching rate of metals from ash deposits. The lower pH values increase the leaching rate of inorganic constituents of fly and bottom ash.

Singh et al. (2007) conducted a standard open column experiment for studying behaviour and characteristics of leachate. The setup consisted of PVC columns of 10 cm diameter and 75 cm height, open at the top and fitted with an outlet at the bottom to collect the leachates. The column was filled up into layers with the fly ash collected from ash ponds of different thermal power plants. Elemental analyses of leachates were carried out using atomic absorption spectrophotometer. In general, nine elements, i.e. Na, K, Ca, Mg, Mn, Cu, Fe, Zn and Pb were observed in the leachates obtained from the pond ash of different thermal power plant in the study. However, their concentration level was differed from plant to plant. Mn and Cu were found to be below the detection limit (<0.001 mg/L) in leachates of Durgapur thermal power plant, whereas the elements Na, K, Ca, and Mg were found in the leachates of all the our thermal power plants. Pb was found to be nil in leachates of Durgapur thermal power plant and Fertilizer Corporation of India. It was concluded that the characteristics of leachates of the ash pond varied from time to time as well as plant to plant which can be attributed to inherent properties of the coal used in individual thermal power plant and changes in chemistry, morphology and mineralogy of the fly ash.

Singh & Kumar (1998) carried out open column percolation leaching experiment on the coal ash samples to determine its leachate chemistry. Coal ash samples were collected from Chandrapura thermal power station of Damodar Valley Corporation in Bihar, India. In the experiment deionized water was percolated through a packed column in the presence of oxygen at a rate, which depends on the natural permeability of the material. The open column of leaching experiment was made from PVC pipe, 4 inch in diameter and 2 feet in length. The top end of the column was exposed to atmosphere and the bottom end was connected to quarter inch tubing. The leachate was collected through this tubing in 250 mL polypropylene beakers. The obtained leachate was analysed for physico-chemical properties and trace elements. The obtained results indicated that pH, conductivity, total dissolved solids and trace elements were within permissible limit or at below detectable limit. It was observed that no significant leaching occurred

and toxicity was manageable with respect to trace elements. The study concluded that out of nine metals only calcium and magnesium were leached in the entire period. In case of other seven metals (iron, lead, copper, zinc, manganese) leaching was some time intermittent.

Georgakopoulos et al. (2002) assessed the leachability of major and trace elements of fly ash from Ptolemais power station located at Northern Greece. Synthetic groundwater leaching procedure (SGLP) and leaching column test was conducted. Following the SGLP, 5g fly ash sample was mixed with 100 mL of purified Milli-Q type water in the ratio of 1:20 in a high density polyethylene bottle and shaken on a rotator shaker for 18 hour at room temperature. Leachate was decanted and filtered through membrane filters using Millipore GS of 0.22 μm , while in leaching column test, purified water was passed through the fly ash (100 g) in a vertical column with an internal diameter of 5 cm and a fillable height of 15 cm. The permeation procedure was controlled with a Gilson Minipuls 2 peristaltic pump. The flow rate of the pump was 2 mL/h throughout the duration of test, which was about 200 h (5 pore volumes). The column effluent was accumulated in 0.06 pore volume increments using a RediFrac fraction collector. The relative cumulative mass leached in both the tests was very close in both cases for Si, Al, Ca, Mg, Na, K, S, B, Ba, Be, Bi, Cs, Hf, I, Li, Ni, Rb, and rare earth elements Se, Th, Y and Zr. It was higher in SGLP leaching for Ti, Fe, Ag, As, Br, Co, Cr, Cu, Ga, Mn, Nb, Sc, Ta, Tl, V and Zn. While the mass leached was higher in the column test for Cd, Ge, Mo, Pb, Sb, Sn, Sr, U and W. The major elements showed that higher leaching rates were for S, K and Ca in both the leaching procedures. Taking as reference the column leaching test, the leaching rates for trace elements were higher than 5% only for Br, I, Mo, Sb, Sr and W. The relative mass leached for Sr and Mo exceeds 90%. A leaching rate between 5 and 0.5% was shown for Ag, B, Ba, Cd, Cr, Cs, Li, Pb, Rb, Se, Sn and Zn. Elements showing leaching rate lower than 0.5%, in most of the cases lower than 0.1%, were As, Be, Bi, Co, Cu, Ga, Ge, Hf, Mn, Nb, Ni, and rare earth metals Sc, Ta, Th, Tl, U, V, Y and Zr. On the basis of obtained results it was concluded that relative mass leached from Ptolemais fly ash was low as shown by both the column and SGLP leaching test. Only Sr and Mo were lost during the first five pore volumes of its leaching.

Das et al. (2012) investigated the leaching characteristics of coal combustion byproducts. Fly ash and pond ash samples were collected from Mejia thermal power plant, India for open column percolation leaching experiment to determine the long term leaching characteristics. pH, conductivity and total dissolved solids in leachate were within the permissible limit, only in some cases Ca,

Na and Mg was supported by high value of conductivity, TDS and salinity. K, Na and Ca were found regularly in leachates of both fly ash and pond ash samples through the entire study period, but their concentration decreased considerably with time of leaching period. Other elements like Fe, Ni, Zn, Pb, Cd, Co, Cu and Mn recorded as showing irregular pattern throughout the study period. Initially, Fe concentration was observed higher in case of fly ash and reduced gradually with the leaching time. Ni and Zn were within the permissible limit in both the samples of fly ash and pond ash. The lead concentration was recorded higher than permissible limit at early stage and then found to be decreasing and lastly it was declined to below detectable limit. Cd and Co was occasionally observed in leachates of both types of ash samples. Cu and Mn showed intermittent leaching trend. It was concluded that open column percolation leaching experiment might be used to predict the long term leaching behaviour of coal combustion byproducts because it reflects the real field conditions. This procedure can be predicted as the most important long term leaching experiment that can predict the environmental impacts of leachate on the surface and groundwater.

Singh et al. (2014) conducted a batch extraction leaching test on fly ash samples with three different leaching medium, acidic solution (pH 4.93 ± 0.05), neutral solution (pH 7) and alkaline solution (pH 10-11) in triplicate to identify pH dependence of leaching behaviour of some metals from fly ash. Fly ash sample was collected from Rajghat thermal power plant located in Delhi, India. Total number of six elements (Fe, Cu, Cr, Cd, Ni and Pb) was analysed in leachate obtained from different extractions. The leachability of Fe and Cd were found maximum in the alkaline solution, whereas the leachability Cu, Cr, Ni and Pb were found maximum in acidic solution. The leachability of trace element in fly ash was dependent on the time of leaching and pH values of the leachant.

Ibrahim (2015) studied the leaching characteristics, speciation and fractionation of trace and major elements from fly ash. Five liquid-to-solid ratios (L/S) were selected, ranging from 0.5 to 1. The releasing of trace and major elements from fly ash increased as L/S ratio increased over time (at pH 2.88). The study reported that $\text{Al}(\text{OH})_3$, $\text{Cd}_3(\text{PO}_4)_2$, azurite, jarosite, MnCO_3 , $\text{Pb}(\text{OH})_2$, quartz (SiO_2) and ZnCO_3 are the solid phases that control Al, Cd, Cu, Fe, Mn, Pb, Si and Zn activities in fly ash leachates. The sequential chemical extraction tests showed that Al, Cd, Cu, Fe, Mn, Pb, Si and Zn were mainly present in Fe-Mn oxide fraction, organics fraction and residual fraction and their leachability could be well- controlled in common natural environment.

Table 4: Literature review of leaching behavior of fly ash.

References	Fly ash samples	Leaching Test Methods	Observations
Praharaj et al. (2002)	Fly ash from electrostatic precipitator of thermal power plant in Orissa, India combusting sub bituminous coal.	Serial Batch Leaching Test	Trace elements were found to be in maximum concentration in leachate at low L/S ratio and decreased at higher L/S. On increasing the infiltration of rainwater through ash pile concentration of trace elements will be reduced.
Shivpuri et al. (2011)	Fly ash from the dust hoppers of electrostatic precipitator of six thermal power plants in Maharashtra, India.	Sequential Extraction Method	Leaching behavior of elements in coal fly ash was found to dependent on different leaching conditions like leaching medium and pH. Ca, Ni & Fe rich fly ash showed greater leachability in acidic or ion exchangeable conditions.
Yuan (2009)	Fly ash samples from pulverized and circulated fluidized bed combustor in thermal power plant of North China.	Sequential Extraction Method	Most active metals in fly ash were V, Cu, Zn & Cd. More than 80% elements in fly ash was in most stable residual fraction i.e. harmless to environment.
Baba and Kaya (2004)	Bottom and fly ash samples from Yenikoy, Kemerkooy, Yatagan thermal power plants in western Turkey.	Extraction Procedure Toxicity characteristic leaching procedure ASTM extraction procedure	It was observed that all three procedure (EP, TCLP, ASTM) gives different concentration of elements in extraction. EP procedure gives the highest concentration whereas ASTM gives the lowest concentration indicating that extraction results are pH dependent.
Singh et al. (2007)	Fly ash from ash pond of Bokaro & Durgapur thermal power plant, Fluidised Bed Combustion Plant, Fertilizer Corporation of India (India)	Open column percolation method	Significant concentration of nine trace elements i.e. Na, K, Mg, Ca, Fe, Pb, Mn, Cu & Zn in the leachate.
Singh and Kumar (1998)	Coal ash samples from Chandrapura thermal power plant, Bihar, India.	Open column percolation leaching experiment	No significant leaching observed, toxicity was manageable with respect to trace elements.
Georgakopoulos et al. (2002)	Fly ash from Ptolemais thermal power station	Synthetic Groundwater Leaching Procedure & Leaching Column Test	Very low percentage of relative mass was leached. Maximum relative mass leaching was observed for Sr and Mo only.
Das et al. (2012)	Fly ash and pond ash from Mejia thermal power plant, India.	Open column percolation leaching experiment	pH, EC, TDS were below IS limit, N, K & Ca found regularly leaching, all the trace elements were within permissible limit at the end of leaching.
Singh et al. (2014)	Fly ash from Rajghat power house, Delhi, India.	Batch extraction leaching test	In acidic medium leachability of elements was observed maximum as compared to alkaline and neutral medium.
Ibrahim, 2015	-	Sequential Extraction Method	Al, Cd, Cu, Fe, Mn, Pb, Si and Zn were found to be leached.
Sonawane et al. 2016	Fly ash from Eklahare thermal power plant, Maharashtra, India	Column leaching test	Heavy metals like Zn, Ni, Cu, Fe, Pb, Mn, Mg and Cd were found to be leached.

According to risk assessment code (RAC); Cd, Cu, Mn and Pb showed a medium risk, while Zn and Fe showed low risk level, but Al and Si were considered safe to the environment.

Sonawane et al. (2016) studied the leaching characteristics of coal ash generated from Eklahare thermal power plant, Maharashtra, India. During the study period, leaching behaviour of various heavy metals like Zn, Ni, Cu, Fe, Pb, Mn, Mg and Cd from leachate was studied from the column leaching test. The study concluded that when coal ash comes in contact with water, the alkaline elements present on its surface get dissolved rapidly and move into solution. When this solution percolates through groundwater, creates problem like groundwater contamination.

CONCLUSION

High ash coal produces large amount of fly ash and low thermal efficiency of the boiler including slogging, choking and scaling of the furnace. Fly ash contains trace amounts of toxic metals that results in negative effects on human health and on plants. During transport, disposal and storage phases, the residues from coal combustion are subjected to leaching effects of rain and part of the undesirable components in the ashes may pollute both ground and surface waters. These solid residues (fly ash) can be leached in higher concentration to the drinking water sources. The leachability of trace elements present in the ash increases with decreasing pH. The leachability of Fe and Cd is maximum under alkaline condition, whereas the leachability of Cu, Cr, Ni and Pb is maximum under acidic condition. Also the concentration of most elements such As, Ca, Mg, Mn, Na, S and Zn in leachate is maximum at lower L/S ratio and it decreases at higher L/S ratio. Methodologies of batch and sequential leaching test study being conducted by various investigators designates that open column percolation leaching experiment can be used to predict the long term leaching behaviour of coal combustion byproducts because it reflects the real field conditions.

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