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Research Article

Application of Electro kinetic technique to remediate fly ash for its sustainable use

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Abstract
Fly ash is a by-product of coal combustion in thermal power plants which is classified as hazardous waste and a serious threat to environment. The study was conducted to determine the potential and examine the efficacy of electro kinetic technique (EKT) using variables like pH, total dissolved solids (TDS), e concentration of chlorides (Cl^-), sodium (Na^+),magnesium (Mg^{2+}), potassium (K^+), ammonia (NH^{3+}) and calcium (Ca^{2+}) on fly ash as a low-cost treatment for enhancing the use of fly ash in a more sustainable manner. The probability of removing heavy metals and chlorides from fly ash suspended in water using electro dialysis was studied as they are highly dependent on pH and conductivity of the fly ash. The voltage gradient and duration indicated significant effect in the change of pH values showing a range from 4.6 to 7.7 at cathode and anode respectively, while the Total dissolves solids (TDS) varying from 72.33±5.6 to 146±5.4 showed the enhanced availability of ions post electro dialysis. In terms of chlorides, Cl^- the content was observed to be 265.06 mg/l which was high enough to cause corrosion problems in later stages of reuse of fly ash. The concentration of cations like Na^+, K^+, NH^{3+}, Mg^{2+}, Ca^{2+} were observed to be notably influenced by the duration of study and pH in electro dialysis. The experimental results of the study showed that the proposed technique based on the fundamentals of electro kinetics and dialysis could efficiently improve the remediation process which would remove metals by converting them to available form in fly ash.

Keywords: Electro kinetics, Electro migration, Fly ash, Redox reaction, Total Dissolved Solids

INTRODUCTION

Power generation using coal continues to be as the prime source of energy. Fly ash is produced when coal is burnt which is collected by electrostatic precipitator in stacks at power stations. Sustainable use of this fly ash remains to be one of the key concerns in recent years. There have been special ash utilization division that has been set up to ensure the maximum use of fly ash from the huge quantities of ash that is produced at the thermal power stations. Many studies have been done to utilize this by-product as a resource in various sectors. He *et al*. (2020) has shown the potential significance of using coal power plant produced fly ash to make absorbent zeolite. Fly ash quality largely depends upon the type of coal burnt. Generally fly ash is fine in nature, with low unburnt carbon and high pozzolanic activity. The use of fly ash in brick and cement manufacturing gives a double advantage of reduced cost and efficient disposal (Gupta *et al*., 2020). Fly ash is a proven resource material for many application of construction industries and currently is being utilized in the manufacturing of bricks/ blocks, tiles, road embankments, construction of low lying areas, in agriculture and for improvement of soil fertility etc. According to (Senapati, 2011) In India, approximately 90 million tons of fly ash is generated every year, which is largely responsible for environmental pollution. Currently, India utilizes only 3% of fly ash produced while in developed countries like Germany, 80% of it is consumed in various industries. (Mahzuz and Hasan, 2020) have experimentally proved that on the addition of fly ash content, up to 10% increases the compressive strength of concrete. Fly ash can prove to be a serious environmental pollutant if not managed properly. At the same time, various remediation and stabilization techniques have been studies for utilization of fly ash as it constitutes favourable characteristics for further applications (Ferreira *et al*., 2003, Lima *et al*., 2012). During the last decade electro dialytic process (EDR) has emerged as a technique to extract metals and chlorides from fly ash (Pedersen *et al*., 2005, Ottosen *et al*., 2006, Lima *et al*., 2009, Lima *et al*., 2012). Electro kinetic treatment enhances the potential of degradation of waste and increase the availability of pollutants at a comparatively lower cost. This technique includes passing of electricity to mineralize ionic species and compounds present in the fly ash to enhance the mobilization and efficiency of removal of metals (Huang *et al*., 2015). A direct current (DC) produces electric field of low voltage which induces polarization in the particles of fly ash which makes them charge or discharge electricity(Pedersen *et al*., 2005). The redox reaction that occur during the treatment mineralizes the organic compounds and increases the mobilization of inorganic compounds (Ribeiro *et al*., 2007, Lima *et al*., 2009). A review of the recent literature on electro kinetic process performance and efficiency is summarized and presented in this study. The main objective of the study was to evaluate the effect of electro dialysis on the various chemical characteristics of fly ash like pH, TDS, availability of ions like Na^+, K^+,NH^{3+},Mg^{2+},Ca^{2+} and chlorides for investigating the efficiency of EKT .

MATERIALS AND METHODS

Fly ash description: This study used the fly ash generated from a coal-burning power plant as experimental material to study the efficiency of electro kinetic technique to remediate fly ash. The fly ash was collected from the incinerator facility of a thermal power plant near Visakhapatnam, Andhra Pradesh. Only fine completely dried fly ash from the outlet of electrostatic precipitator (ESP) characterized by lower carbon content and higher fineness was collected for the study. The quality of fly ash is based generally on the type of coal (Bituminous/ sub-bituminous/ lignite) used for power generation. Based on the type of coal burned the fly ash is divided into two classes Class F and Class C. Class C fly ash is obtained by burning of sub-bituminous coal and consist mainly of Calcium alumina and free lime (CaO). Fly ash of Class C type is called commonly high calcium fly ash. Class F fly ash is generally produced from burning of bituminous and anthracite coals which consists of alumina- silicate glass, mullite and also some amount of magnetite present in it. In this study, Class C type fly ash has been used for the experiments collected from the ESP directly.

Electro kinetic technique: In this technique, a direct current (DC) mineralizes the organics present in the fly ash like VOC's (volatile organic compounds) and makes them available in an organic compound form to aid in the removal of metal contaminants (Elicker *et al*., 2014). When electric field is applied to fly ash many physical, chemical effects are observed. Electro kinetic treatment is widely known technique that fosters changes in pH, redox potential, concentration of electrolyte and various chemical properties of fly ash (Huang *et al*., 2015). According to (Meer and Nazir 2018), when electro kinetic treatment is given to fly ash, it alters the current, voltage drop, pH, conductivity, total dissolved solids, the concentration of anions and cations which influence the migration of ions, metals and other contaminants present in fly ash towards anode and cathode based on the charge of ions (Ferreira *et al*., 2003)

Experimental setup: Class C type Fly ash samples were collected from a power station near Visakhapatnam directly from the outlet of electrostatic precipitator using a shovel and stored in a plastic bag. The electro remediation assay was performed in a electro dialytic cell made of Plexiglas with 6 cm internal diameter. Two stainless electrodes of dimensions(10×20×5.5cm) were used as anode and cathode (Fig 1.) The distance between anode and cathode was kept constant at 15cm and the intermediate space was filled with fly ash mixed with water in the proportion of 1:4. The system used a low voltage DC power supply to operate under a constant voltage of 30V and 100 mA current. The experiment was carried out for 11 days for 6hrs every day. The samples were collected from both the sides of electrodes i.e anode and cathode after every alternate day i.e. on 1st, 3^{rd}, 5^{th}, 7^{th}, 9^{th}, and 11^{th} day. The parameters selected for analysis were pH, total dissolved solids, concentration of chlorides and cations like sodium, calcium, magnesium, ammonia, potassium to assess the treatment efficiency of Electro kinetic technique for fly ash. The setup for the experiment is shown in Fig.1 The samples collected were then dried in a thermostatic heater at 120^0C for two hours and sifted by a 200- mesh sieve.

Analysis of samples: 10g of sample was mixed with 50ml deionized water which was centrifuged and an aliquot was filtered using Whatmann filter paper no.41 (150mm) and pH was measured using Sigma-27 DP pH meter. Total dissolved solids were measured using the aliquot from the 1:5 sample and deionized filtered solution by KE-TDS-01 digital TDS meter. Chlorides (Cl^-) were measured (mg/L) using potentiometric titration as described in Danish standard DS 239($AgN0_3$ titration method). Calcium (Ca^{2+})(mg/L) as $CaC0_3$, Magnesium (Mg^{2+})(mg/L) as $CaC0_3$ were measured using complexometric titration with EDTA to EBT (Eriochrome black –T) as end point for total hardness and Mg^{2+} was obtained by difference (Pereira *et al* 2011) and sodium(Na^+), potassium(K^+) and ammonia (NH_3^+) in ppm (parts per million) were determined using IC metrohm (Ion chromatography)instrument.

RESULTS AND DISCUSSION

The results of the parameters studied are presented in Fig. 2-6. In the course of application of electric field, electrolysis reaction took place at electrodes, which resulted in alteration in pH, electrical conductivity and availability of various ions like Na^+, K^+, NH^{3+},Mg^{2+}, Ca^{2+} and chlorides as supported by the enhanced values of these post EKT treatment. Thus increased availability

Fig 1. *Experimental setup showing fly ash in electro dialytic cell, dual channel power supply.*

of ions was observed with an increased amount of dissolved solids after the treatment, which were further confirmed by the following results.

pH: The applied potential difference in each of the sample was kept constant during the entire process. As the time of treatment progressed, the pH values showed alterations in a significant manner. At the anode, the pH was observed to be 7.72±1.04 while at cathode it altered as 4.66±1.56. The trend of pH (Fig. 2) showed a pattern where the pH values at anode decreased at first and then increased. At the anode, oxidation of water took place, which resulted in the release of H^+ ions, while reduction reaction happened at the cathode and released OH^- ions. The transport of OH^- was comparatively slower than H^+ ions which created a difference between the electrodes leading to acidification of medium between the electrodes. (Acar *et al*.1996, Puppala *et al*., 1997). Production of ions at electrodes results in a rise in current density. According to the study conducted (Rutigliano *et al*. 2008), low pH favours electro removal as acidic pH, stimulates the release of metallic ions in the medium during the treatment. It has also been observed (Matsumoto *et al*. 2007) that removal of hydroxyl ions formed at cathode by metallic ions, followed by the formation of insoluble hydroxides.

Total dissolved solids: The range of Total Dissolves Solids varied significantly between 72 ppm to 210 ppm at anode and cathode, respectively. The results of our experiment have been supported by findings of (Ferreira *et al*., 2005) that highly mobile fraction was available in the initial days of the experiment. At the cathode, the dissolved solids showed a huge increase on Day 7 showing maximum availability of metallic species and ions. Pedersen *et al*. (2005) used platinum electrodes with ammonium citrate as assisting agent for electro dialytic experiments for a period of 5-20 days and observed finding that with increasing remediation time there is a dissolution of mineral salts such as alkali chlorides, the similar trend was observed in our findings (Fig 3). It can be explained through this that, the dissociation of ions was high on Day 5 and 7. The decrease in pH might also influence, the availability of metal species and ions, as it has been already mentioned that acidic medium helps in ion dissociation and that justifies the increase in TDS at the cathode to 146±54.3 ppm. TDS is an important parameter which also is an indicator of conductivity which implies that if conductivity is low, a high amount of energy is consumed per unit of mass of pollutant removed.

Chemical composition:

Chlorides: According to (Ottosen *et al*. 2006), chlorides presence enhances the degradation potential of organic pollutants due to formation of various species like HOCl, Cl_2 and ClO^- which enhances the potential of fly ash to be used as a soil ameliorant. In the present study, there was a drastic change in the chlorides concentration from Day 1 to Day 11, which reported the value of 447.3mg/L, and no change was observed in control over the days. There was a slight decrease in chloride concentration at anode on Day 5. The availability of chlorides increased over the treatment time at the cathode, which was favoured by the increase in the ionic activity due to the development of the acidic medium. According to Weibel *et al*. (2018), mobilization of heavy metals mainly of Pb (lead) and Cu (copper) was due to redox reaction and formation of metal-chloride complexes. In addition to the use of fly ash in concrete, it was the water-soluble fraction of chlorides that was of interest.

Sodium, ammonia, potassium – According to Hansen (2000), metal ions undergo electro deposition as per thermodynamic principle, depending on the metals and ion concentration present in fly ash. According to the results obtained in our experiment in case of sodium ions, it became more available at the anode during the treatment compared to that at cathode owing to the alkaline medium at the anode. As observed by (Kim *et al*., 2010) study to separate chlorides and sodium from tidelands using electro dialytic technique for 10 days, sodium ions were desorbed from fly ash surface by ion exchange between sodium and hydrogen ions which were transported by electro migration as seen in our results also. While the amount of sodium available reaches its peak of 55.978 ppm at Day 5 showing maximum concentration and effectiveness. Similarly, potassium was concentrated towards the anode, though its concentration slightly decreased at Day 7 it increased again during the rest of the treatment days (Table 1). Ammonia showed a very significant change at Day 9 with a high concentration of 174.29 ppm at anode though the value at cathode showed very less alterations.

Calcium and magnesium- In the experiment, initially calcium was unavailable but as the days of treatment progressed, it showed a concentration of up to 200mg/L at the cathode(Fig.5) indicating that the electro kinetic process helped in transforming the unavailable form of calcium into a detectable form which can

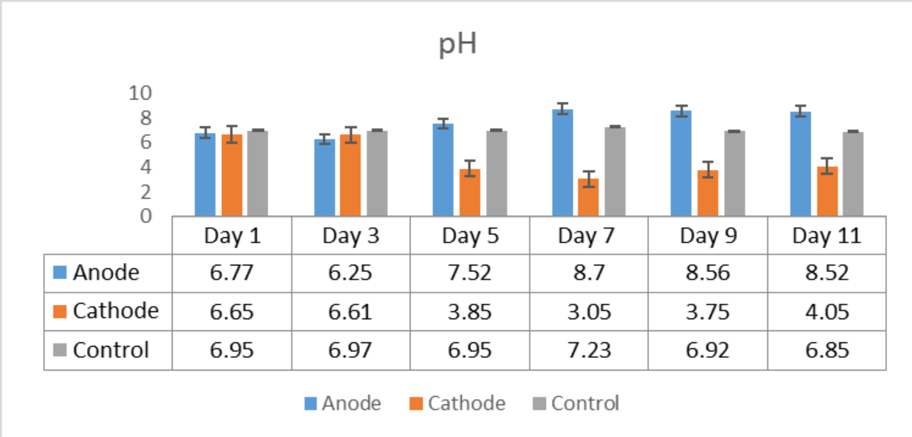

Fig 2. *Changes in pH at anode, cathode and control post electro dialysis.*

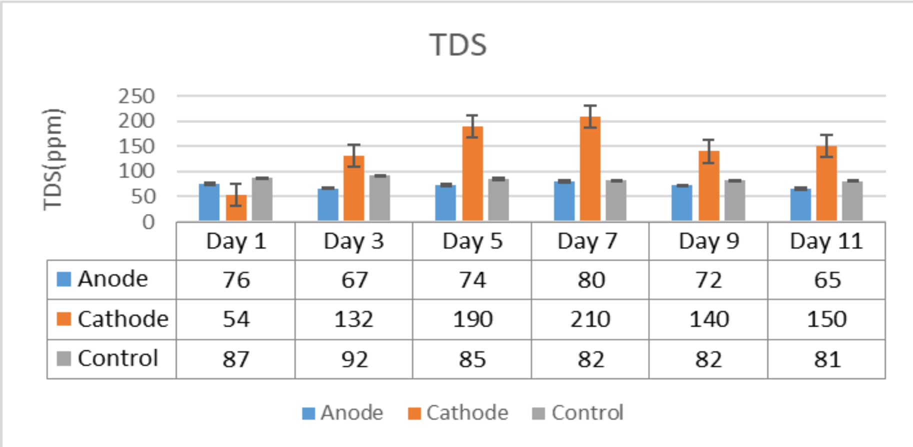

Fig. 3. *Changes of total dissolved solids after EKT treatment.*

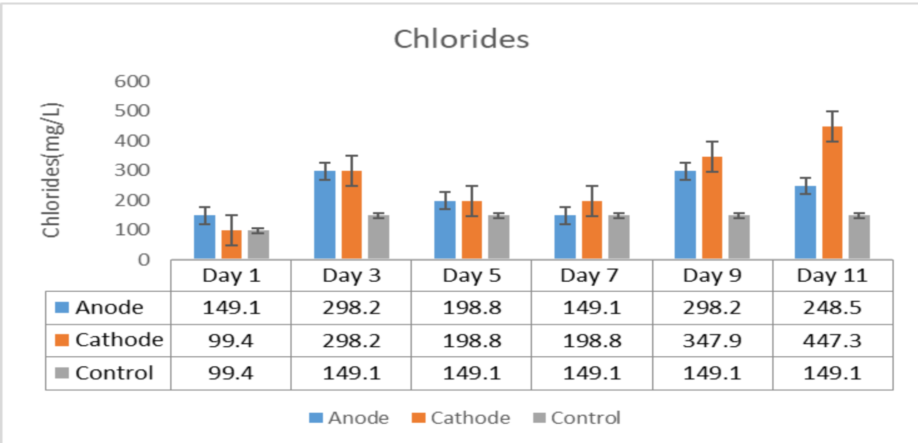

Fig. 4. *Changes in chlorides concentration after EKT treatment.*

be utilized for many purposes. The detection of calcium post electro dialysis was because of pH change in the fly ash as it enhanced the desorption of calcium. The highest calcium concentration of 178 mg/L was observed in the latter days of the experiment. Magnesium and calcium form complexes with chlorides which are observed by (Kim *et al*., 2010) in separating sodium and chlorides from tidelands using EKT technique for 10 days. In our study Magnesium, on the other hand showed increased value at the cathode and the peak of 200mg/L on Day 9 and 11(Fig.6).

Potential difference (Voltage): The initial voltage gradient was set to 30 volts on the first day of experiment. As the time increased, the potential difference had its impact on pH, conductivity and ionic availability. An increase in voltage has been noticed from Day 1 to day 11. The voltage of cell increases when electrical resistance increases as the current is kept at constant 100 mA as observed in our study, (Pedersen 2005) indicated the similar trend in his experiment to

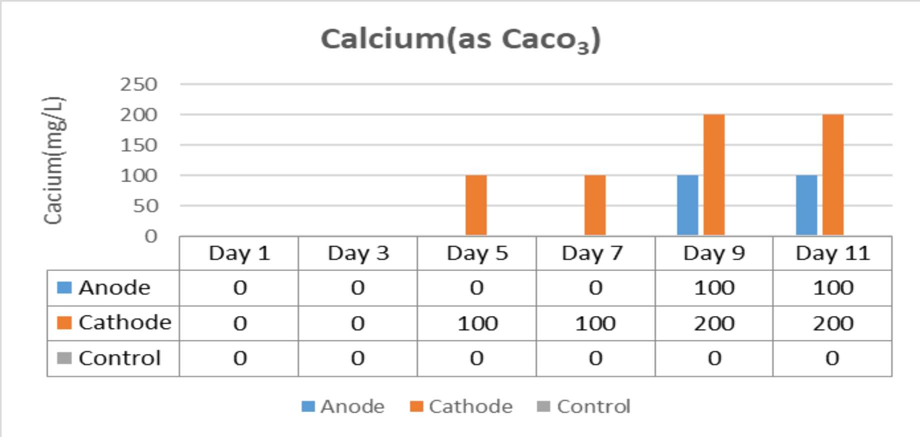

Fig.5. *Changes in calcium concentration in fly ash solution during electro dialysis.*

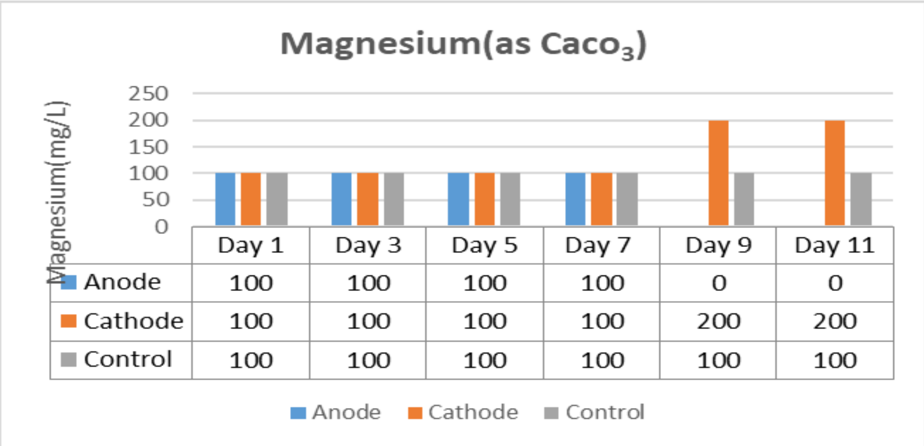

Fig. 6. *Changes in magnesium concentration in fly ash solution during electrolysis.*

remove heavy metals from fly ash using ammonium acetate as an assisting agent in EKT technique. Voltage increase was more prominent after day 5 at the cathode from (29.7 to 30.5 volts). There had been an overall increase of voltage and current flow at the cathode from Day 1 to Day 9 up to 33.7 volts According to a study of Zayas *et al* (2007), greater efficiency of ionic availability has been recorded with an increase in time of electrolysis and also on the different potential difference applied for the treatment.

Conclusion

The study was conducted to determine the potential and effect of electro kinetic technique for enhancing the use of fly ash in a more sustainable manner. With the increase in the concentration of desorbed ions, there was a decrease in electrical resistance. As the duration of the experiment increased, there was increased desorption of exchangeable cations like Ca^{2+}, Mg^{2+}. Sodium was removed by electro migration towards the cathode, but chlorides migrated towards the anode. Post EKT treatment soluble form of calcium was observed as desorption of calcium was affected by pH. The alterations in the pH and ionic species concentration were supported by total dissolved solids indicating the increase in the availability of ions and metal species for further treatment. These alterations in Na^{+}, K^{+}, $NH3^{+,}$ $Mg2^{+,}$ Ca^{2+} and chlorides clearly indi-

Table 1. Alterations in sodium, potassium and ammonia concentration during EKT.

	Sodium (mg/kg)		Ammonia (mg/kg)		Potassium (mg/kg)	
	Anode	**Cathode**	**Anode**	**Cathode**	**Anode**	**Cathode**
Day 1	53.394	5.185	25.146	5.483	26.074	23.378
Day 3	25.211	20.188	13.827	9.927	47.607	22.448
Day 5	3.629	55.978	19.052	14.629	23.875	23.407
Day 7	4.181	12.354	9.785	15.023	29.88	23.45
Day 9	7.822	10.547	12.311	15.616	30.298	22.054
Day 11	52.824	10.523	10.539	18.331	21.8	24.56

cated that the metals and ionic species present in fly ash were affected by electro kinetic treatment and it could be further studied to utilize these available ions for agricultural purpose. It could be inferred that when low voltage current was applied to fly ash, the acidic medium was developed at cathode denoting low pH resulting in increased TDS thus increasing their availability in the medium for further treatment. Electro dialytic remediation has shown good potential as a method for treating fly ash. On the other hand more research in future is expected to combine techniques like leaching, phytoremediation, thermal treatment, chelating agents, evaporation process etc. with electro dialytic technique for effective removal of hazardous elements from fly ash and make it more user friendly by-product for its transformation from waste to resource. Thus, fly ash as a by-product should not be looked upon as waste and immediate concerns are required to utilize this in various sectors for it to prevent becoming an environmental hazard in the coming years.

Conflict of interests

There is no conflict of interest regarding the publication of this article.

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