



Utilization of Coal Fly Ash as a Raw Material for the Synthesis of Zeolite like Substance

G. Elavarasan¹, P. Rajakrishnamoorthy¹, D. Karthikeyan² and C.G. Saravanan³

¹Research Scholar, Department of Mechanical Engineering, Annamalai University, Annamalai Nagar (Tamil Nadu), India.

¹Research Scholar, Department of Mechanical Engineering, Annamalai University (Tamil Nadu), India.

²Associate Professor, Department of Mechanical Engineering, Annamalai University (Tamil Nadu), India.

³Professor, Department of Mechanical Engineering, Annamalai University (Tamil Nadu), India.

(Corresponding author: P. Rajakrishnamoorthy)

(Received 05 April 2019, Revised 04 June 2019, Accepted 10 June 2019)

(Published by Research Trend, Website: www.researchtrend.net)

ABSTRACT: Fly ash is considered as a waste byproduct of thermal power station that was disposed into the landfill or ponds which results in environmental pollution. In this paper, we have attempted to utilize that waste fly ash as a raw material for the synthesis of zeolite like material, which is having a wide variety of applications. Naturally occurred zeolites from volcanic fly ash takes more time for the formation, but in our laboratory, the same type of zeolites can be formed with less period. The fly ash was collected from the thermal power station and was examined using the characterization techniques X-ray diffraction spectroscopy analysis (XRD), and Scanning electron microscopy(SEM). Then the fly ash was subjected to the acid treatment of Hydrochloric acid (HCL) to remove metal oxides in fly ash, then the process synthesis of zeolite like material was successfully carried out and its characters resembles like commercial zeolite.

Keywords: Flyash characterization, Physical properties, raw material for zeolite, synthesis of zsm-5; zsm5 comparison.

INTRODUCTION

Around 71% of electricity generation in India is supplied from the Thermal power station that uses coal or lignite [1]. These thermal power stations operated by coal produce tons of fly ash, bottom ash and other polycyclic aromatic hydrocarbons while burning [2]. In that around 65% to 95% were as fly ash and remaining were bottom ash [3]. It has been found that around 131 tons of fly ash particles have been disposed into the landfill of 65000 acres and this is expected to increase in the future [4-5]. The fine particles of fly ash disposed into the landfill may suspend in the air and form haze or smog like substance that during inhalation causes serious health problems [6]. Recycling of fly ash particle is expensive, but these fly ash particles can be utilized for many applications as a raw material in various fields [7]. It has been found that the fly ash particle finds its successful application in the field of construction works [8-12] we have utilized this fly ash for the synthesis of zeolite like material [13] as it is more active catalyst material. The study clearly shows that the two types of zeolites namely zeolite P and faujasite can be formed from the coal fly ash [14]. Keka Ojha et al, have successfully synthesized the X type zeolite from the coal fly ash and examined its characterization and compared it with the commercial 13X zeolite [15]. Anand Srinivasan *et al*, have synthesized various types of zeolites using hydroxide solution and they have successfully tested its ability to absorb the SO₂ using the synthesized zeolite [16]. Miki Inada et al, have involved in the research of the effect of the silica addition in the formation of the zeolite Na-PI from coal fly ash by the hydrothermal treatment in the presence of alkaline solution [17]. Various types of zeolites were formed by alkali fusion with fly ash followed by the hydrothermal treatment, In that, the effect of the

silicon/aluminum ratio, cation exchange capability, acid treatment, and calcination temperature was analyzed in the research work of Vikranth Volli *et al* [18]. As this conversion of flyash to zeolite is very much useful for the solid waste management of the thermal power station, we can reduce the waste landfill disposal[19]. Apart from this, the zeolites can be used for various purposes like removing metals from contaminated water, manufacturing of ceramics and glass, as a catalyst and in the synthesis of geopolymers, etc. [20]. The zeolite X can be even prepared in a rapid time of 20 minutes by ultrasound treatment after the alkali fusion of fly ash [21]. Further, the fly ash particle utilization for the production of zeolite in a large scale manner was carried out to check the possibility of the conversion [22].

II. CHARACTERIZATION OF COLLECTED FLY ASH

The samples of fly ash and bottom ash were collected at different Thermal Power station (TPS) from Neyveli Lignite Corporation India Ltd, namely TPS-I, TPS-I Expansion, and TPS-II, TPS- II Expansion situated Neyveli. After finding the various characterization of collected fly ash, it is understood that SiO₂ content in the TPS-I Expansion fly ash is slightly high compared with other fly ash. Hence TPS -I expansion fly ash is chosen for this project work. Bottom ash is not considered because its generation is very low compared to fly ash generation. Figure 1 shows the XRD pattern of the collected fly ash from the TPS I. The mineralogical characterization of received coal fly ash from the TPS I power station was conducted by the Philips Spectrometer PW1404 is used. Induction source is constituted by an XRD lamp among dual anode (Cr-Au) with the highest power of about 2kW [23].

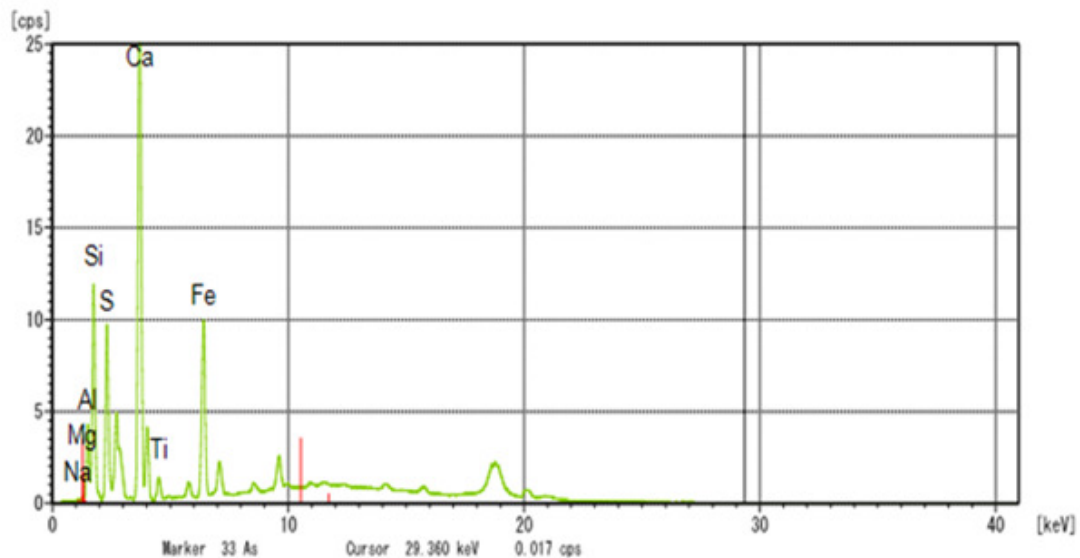


Fig. 1. XRD pattern of TPS-I flyash.

Scanning Electron Microscopy analysis is carried out with a JEOL-JSM 6610LV electron microscope. The samples are initially coated through a thin layer deposition of platinum to avoid charges into the

samples. Micrographs of the samples are recorded with a 10-20 kV accelerating voltage. Fig. 2 shows the SEM image of TPS-I flyash.

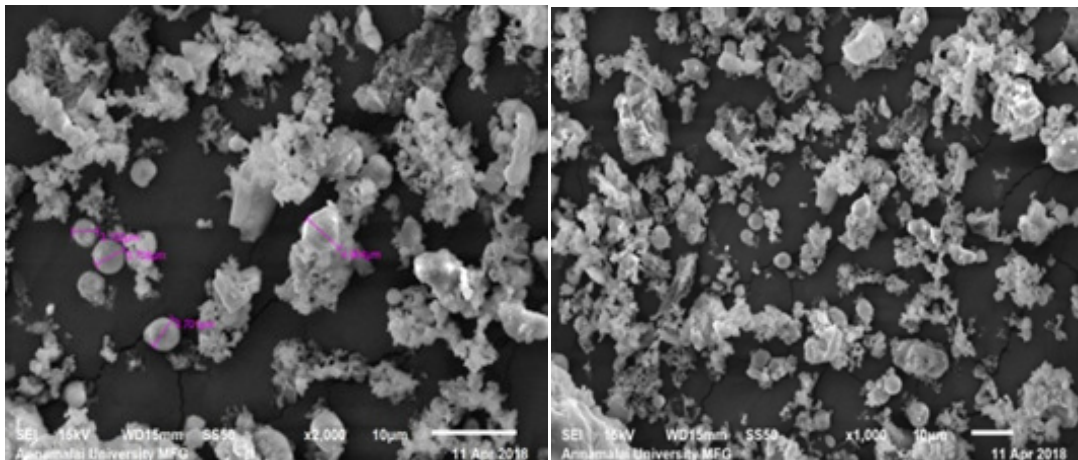


Fig. 2. SEM images of TPS – I Fly ash.

III. CONVERSION OF FLY ASH TO ZEOLITE LIKE MATERIAL

Fig. 4 and 5 shows the block diagram of the synthesis of zeolite like material from fly ash. Figure 6.a to 6.e shows the photographic view of various types of equipment used for the synthesis process of zeolite. Flyash sample is subjected to sieving to remove coarse particles by a sieve for about 180 microns [24]. The fly ash is then treated with Hydrochloric acid to reduce some undesirable components like CaO, Al₂O₃, SO₃, etc. which are present in the sample. The acid treatment of fly ash particles is carried out by taking concentrated HCl of 200ml and de-ionized water of 800ml and the fly ash sample of about 50g [25]. The solution is powered in a round bottom flask and heated at 80deg Celsius in a reflux assembly for 8 hours and allowed to cool at room temperature for about 4hours. Then the solution is filtered, added with 200ml of de-ionized water and placed in a magnetic stirrer/hotplate and continuously

stirred for 15 minutes. The solution is then filtered and added with 200ml of de-ionized water and stirred for 15 minutes. This process is repeated until the solution reaches the pH of de-ionized water and then dried at 120°C in a Muffle furnace for about 4hrs. The acid Treated Fly Ash (TFA) of about 1.25 g was mixed with fumed silica of 0.75 g in 20 ml of de-ionized water and it was placed in a stirrer for mixing. Then 0.5g of sodium hydroxide and 1.5g of tetrapropylammoniumbromide (TPABr) was added to the mixture while stirring. After aging, the final mixture was kept in the autoclave and heated for 120°C for 12 hours and it was allowed to cool down [26]. The synthesized product was filtered, washed with deionized water and dried in an oven at 70 °C and the temperature was increased up to 550°C in the ramping rate of 15 °C/min to burn off TPABr. After cooling down, the product was washed with deionized water and dried in an oven at 70 °C for about 4 hours, then the zsm-5 like zeolite was obtained.

Table 1: Various Equipment used in the synthesis process.

Sl. No.	Equipment	Make
1	Muffle furnace	Sandy scientific Instruments & Co Chennai. (900deg Celsius max)
2	Sieving Machine	Micro Mech Instruments Pvt Ltd, Chennai
3	Magnetic Stirrer	Remi 2 Ltr Stirrers with Hotplate 2 MLH Dimensions (WxDxH) (mm) : 200 × 225 × 185 m
4	Weighing scale	VS weighing System, Chennai (0.01g to 1200g)
5	Reflux Assembly	Technico Laboratory Products Pvt. Ltd, Chennai

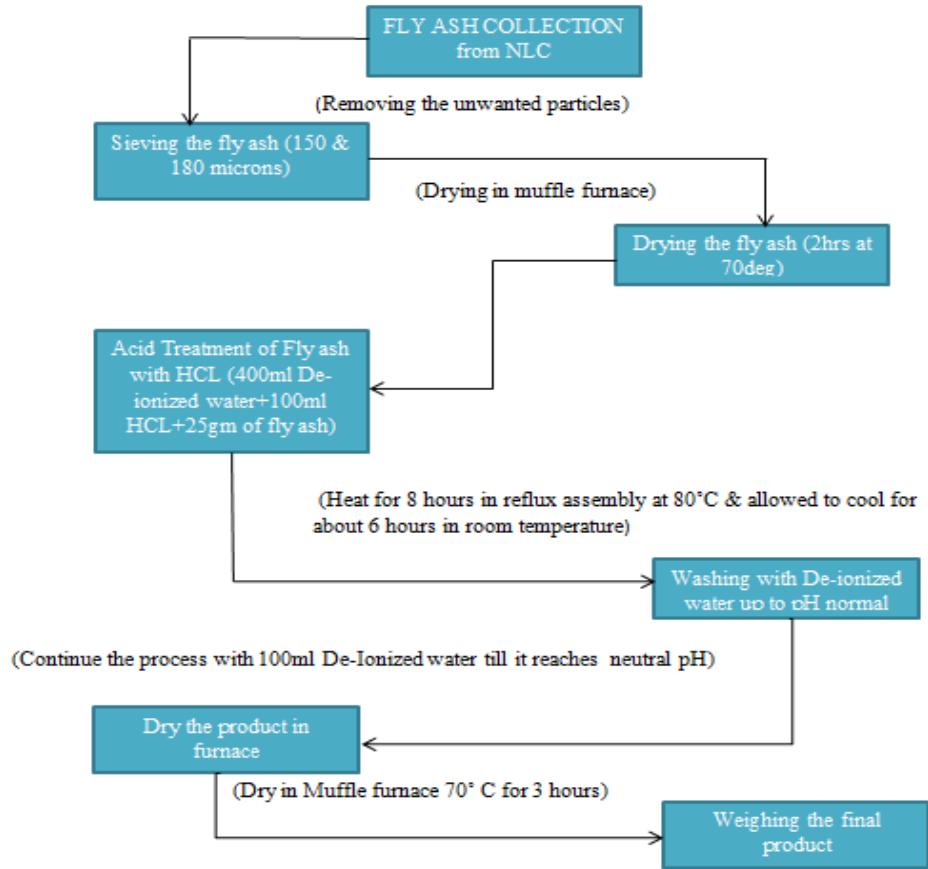


Fig. 3. Block Diagram of Acid Treatment of fly Ash.

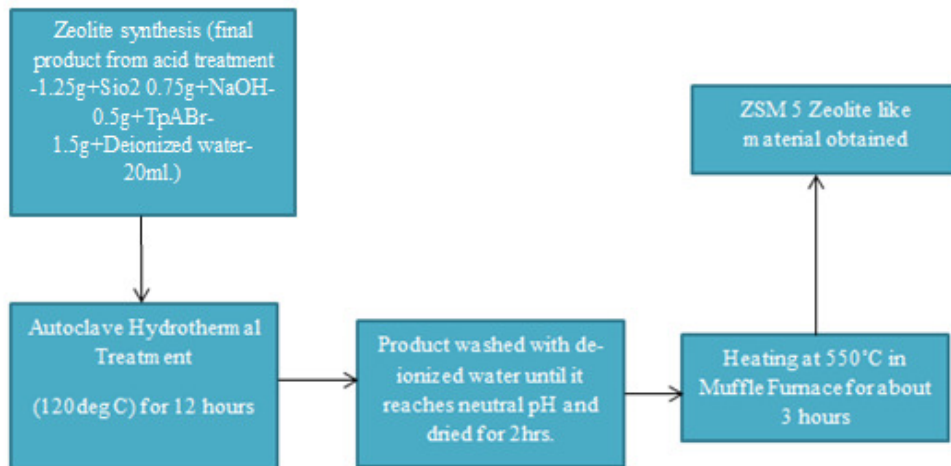


Fig. 4. Block Diagram of Zeolite synthesis process.

IV. CHARACTERIZATION OF SYNTHESIZED ZSM-5 ZEOLITE

The zeolite like material was synthesized from the flyash and it was subjected to the various characterization techniques like XRF, SEM and the same was compared with the commercial zeolite. Figure 7 shows the XRD pattern of the synthesized zeolite like material. This

pattern will be helpful to identify the components that are present in the zeolite like substance. Figure 8 shows XRD of the commercial zeolite and this was useful to compare the content of the synthesized zeolite like substance to the commercial one. Figure 7 and 8 shows the SEM image of the synthesized zeolite and the commercial zeolite.

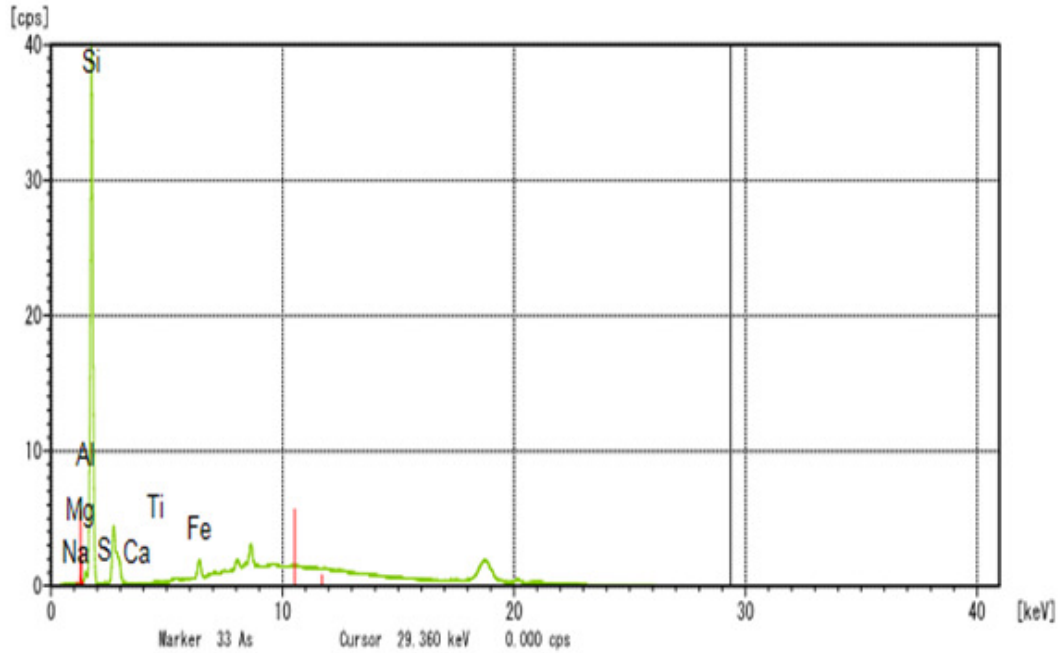


Fig. 5. XRD pattern of Synthesized Zeolite.

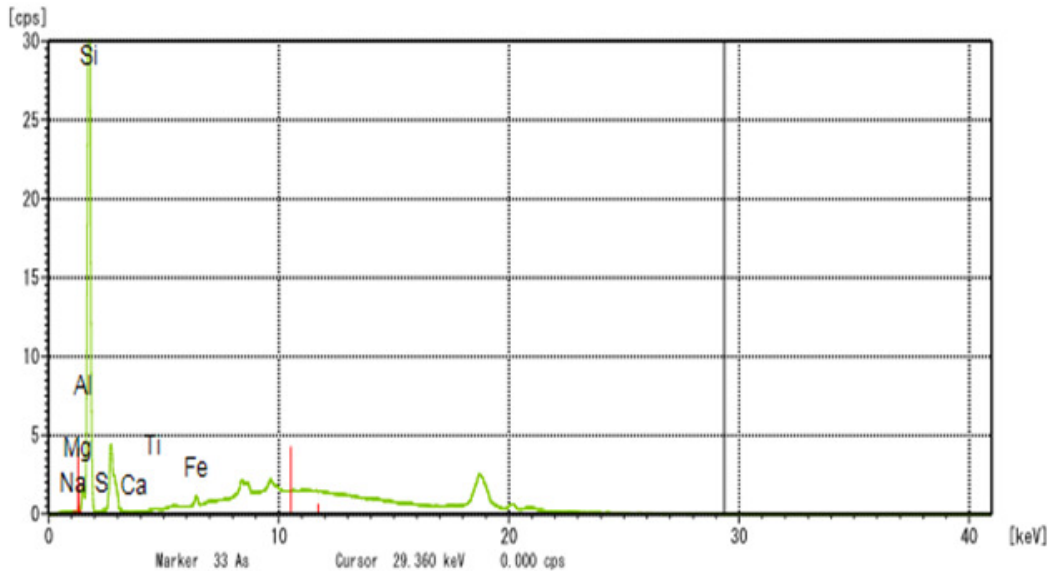


Fig. 6. XRD pattern of Commercial Zeolite.

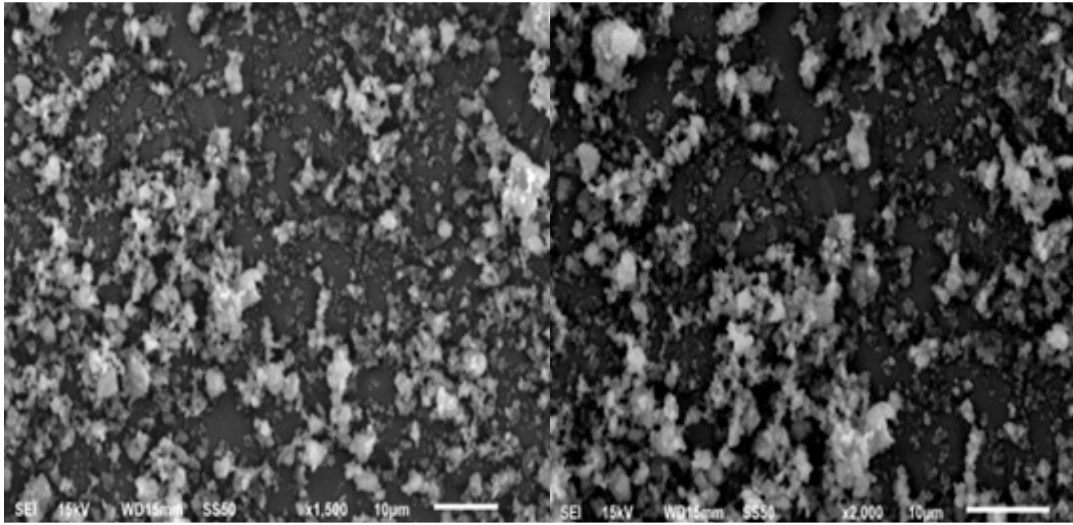


Fig. 7. SEM of synthesized Zeolite.

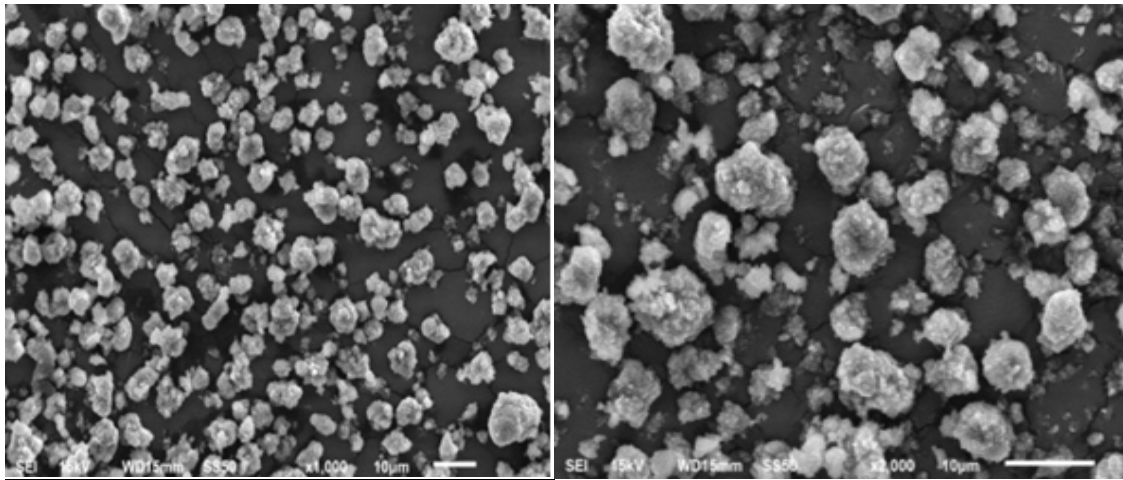


Fig. 8. SEM of Commercial Zeolite.

V. RESULTS AND DISCUSSION

Table 2 shows the result of XRF analysis of fly ash, synthesized zeolite and commercial zeolite

(i) It is noticed from table 2 shows that there is a marked increase in the percentage of Na₂O in the zeolite material compared to fly ash. The weight of Na₂O is found to increase from 2.7 to 6.9 this is due to the capture of Na⁺ ions needed to neutralize the negative

charges on the aluminate in the zeolite during the synthesis process.

(ii) It is observed from the table that the percentage of SiO₂ in the zeolite material is increased to 40.979 from the fly ash 88.372 this is due to the addition of SiO₂ powder during the synthesis process.

(iii) The percentage of other materials (Al₂O₃, MgO, SO₃, CaO, TiO₂, Fe₂O₃) are considerably reduced. This is due to the acid (HCL) treatment of raw fly ash before the synthesis process.

Table 2: Chemical composition of fly-ash, Commercial Zeolite and Synthesized Zeolite samples (weight %).

Composition	NLC Fly-ash (TPS-I – Fly ash)	Commercial Zeolite	Synthesized Zeolite
Na ₂ O	2.753	6.774	6.930
MgO	4.523	1.127	1.155
Al ₂ O ₃	24.534	4.538	3.256
SiO ₂	40.979	87.452	88.372
SO ₃	11.366	0.053	0.134
CaO	13.983	0.006	0.014
TiO ₂	0.531	0.005	0.025
Fe ₂ O ₃	1.331	0.044	0.114

VI. CONCLUSION AND FUTURE SCOPE

Zsm-5 like material was successfully synthesized from fly ash samples and its characters were found for selecting the correct ash for synthesis. The zeolite like material was synthesized from the selected fly ash and it was compared with the commercial zsm5-zeolite. From the comparison it has been found only a slight variation is observed between the synthesized and commercial zeolite. This work can have a significant economic and environmental impact in India if the synthesis process is scaled up as it provides a cheaper way to produce a catalyst that has wide application in various fields, and it promotes the valorization of Indian coal fly ash that considered by many as a waste disposal material. The future scope is that the synthesized zsm-5 like zeolite can be utilized as a catalyst to reduce the harmful exhaust emissions in the Automobiles. As the existing catalyst in catalytic convertor was very costly, the use of zsm5 zeolite as a catalyst for the reduction of emission can be a cost effective method. Thus this work can have an important application in the field of automobile exhaust catalyst used for emission control.

ACKNOWLEDGEMENT

First and foremost, it is a great pleasure for us to express our heartfelt gratitude to our beloved Head of Department, Dr. C.G. Saravanan, Mechanical Engineering, Annamalai university, for his unflinching support, endless hours of help and constant guidance through every stage of this project despite his busy schedule. It has been a great zest and honor to work with him. We are deeply grateful to Dr. D. Karthikeyan for his continuous support, and to our parents and friends who have supported us throughout the period to complete successfully complete this project.

REFERENCES

- [1]. Ranganatham, M.V.S. "Energy Statistics (2018)". *Central Statistics Office Ministry Of Statistics and Programme Implementation, Government Of India, New Delhi*, **25**,1-7.
- [2]. Harkness, J.S., Ruhl, L.S., Millot, R., Kloppman, W., Hower, J.C., Hsu-Kim, H., Vengosh, A., (2015). "Lithium isotope fingerprints in coal and coal combustion residuals from the United States", *Procedia Earth Planet. Science*,**13**, 134–137.
- [3]. Jayaranjan, M.L.D., Van Hullebusch, E.D., Annachhatre, A.P., (2014). "Reuse options for coal fired power plant bottom ash and fly ash". *Rev. Environ. Sci. Biotechnol.*, **13**(4),467–486.
- [4]. Anil Kumar Dixit and Lokesharappa (2011). "Disposal and Management of Fly Ash". *International Conference on Life Science and Technology, IPCBEE, IACSIT Press, Singapore*, **3**, 11-14.
- [5]. Banerjee, S., Sharma, G.C., Chattopadhyaya, M., Sharma, Y.C., (2014). "Kinetic and equilibrium modeling for the adsorptive removal of methylene blue from aqueous solutions on of activated fly ash (AFSH)". *J. Environ. Chem. Eng.*, **2**(3): 1870–1880.
- [6]. Liu, J., Dong, Y., Dong, X., Hampshire, S., Zhu, L., Zhu, Z., Li, L., (2016). "Feasible recycling of industrial waste coal fly ash for preparation of anorthite-cordierite based porousceramic membrane supports with addition of dolomite". *Journal of European Ceramic Society*, **36**(4), 1059-1071.
- [7]. FarwaMushtaq, Muhammad Zahid, Ijaz Ahmad Bhatti, Saqib Nasir, Tajamal Hussain. (2019). "Possible

applications of coal fly ash in wastewater treatment". *Journal of Environmental Management*, **240**: 27-46.

- [8]. Aljerf, L., (2015). "Effect of thermal-cured hydraulic cement admixtures on the mechanical properties of concrete". *Interceram-International ceramic review*, **64**(8): 346–356.
- [9]. Hoy, M., Horpibulsuk, S., Rachan, R., Chinkulkijniwat, A., Arulrajah, A., (2016). "Recycled asphalt pavement–fly ash geopolymers as a sustainable pavement base material: strength and toxic leaching investigations". *Sci. Total Environ.*, **573**: 19–26.
- [10]. Xu, G., Shi, X., (2018). "Characteristics and applications of fly ash as a sustainable construction material: a state-of-the-art review". *Resources Conservation & Recycling*, **136**, 95–109.
- [11]. Ebrahimi, A., Saffari, M., Milani, D., Montoya, A., Valix, M., Abbas, A., (2017). "Sustainable transformation of fly ash industrial waste into a construction cement blend via CO₂ carbonation". *Journal of Cleaner Production*, **156**: 660–669.
- [12]. Fukasawa, T., Karisma, A.D., Shibata, D., Huang, A.N., Fukui, K., (2017). "Synthesis of zeolite from coal fly ash by microwave hydrothermal treatment with pulverization process". *Advanced Powder Technology*, **28**, 798–804.
- [13]. Eliyas Majeed and Deepak, (2018). "An Experimental Investigation of Rice Husk ash and Waste Paper Sludge ash as Partial Replacement of Cement in Concrete". *International Journal on Emerging Technologies*, **9**(2): 23-26.
- [14]. Shih, W.H., Chang, H.L. Shen, Z. (1995). "Conversion of Class- F Fly ash into Zeolites". *Mater. Res. Soc. Symp. Proc.* **371**, 39-44.
- [15]. Keka Ojha, Narayan C. Pradhan and Amar Nath Samanta, (2004). "Zeolite from fly ash: synthesis and characterization". *Bull. Mater. Sci.*, **27**(6): 555-564.
- [16]. Anand Srinivasan and Michael W. Grutzeck, (1999). "The adsorption of SO₂ by zeolites synthesized from flyash", *Environmental Science & Technology*, **33**: 1464-1469.
- [17]. Miki Inada, Yukari Eguchi, Naoya Enomoto, Junichi Hojo (2005). "Synthesis of zeolite from coal fly ashes with different silica-alumina composition". *Fuel*, **84**, 299-304.
- [18]. Vikranth Volli, M.K. Purkait, (2015), "Selective preparation of zeolite X and A from flyash and its use as catalyst for biodiesel production". *Journal of Hazardous Materials*, **297**.101-111.
- [19]. M.W. Kasture, V.V. Bokade, P.N. Joshi, R. Kumar (2007). "Synthesis and characterization of value added catalysts zeolite beta using environmentally detrimental flyash". *Studies in Surface Science and Catalysis*. **170**: 438-443.
- [20]. R.S. Blissett, N.A. Rowson (2012). "Review article-A review of the multi-component utilization of coal fly ash". *Fuel*, **97**: 1-23.
- [21]. Sivamani Sivalingam, Sujit Sen (2018). "Rapid ultrasound assisted hydrothermal synthesis of highly pure nanozeolite X from fly ash for efficient treatment of industrial effluent". *Chemosphere*, **210**: 816-823.
- [22]. X. Querol, J. CUmaña, F. Plana, A Alastuey, ALopez-Soler, A Medinaceli, A Valero, M. J. Domingo, E. Garcia-Rojo, (2001). "Synthesis of zeolites from fly ash at pilot plant scale.Examples of potential applications". *Fuel*, **80**(6): 857-865.
- [23]. Yingchao Dong, Stuart Hampshire, Jian-er Zhou, Zhanlin Ji., Jiandong Wang, Guangyao Meng, (2011). "Sintering and characterization of flyash-based mullite with MgO addition". *Journal of the European Ceramic Society*, **31**(5): 687-695.

- [24]. Arkadiusz Derkowski, Wojciech Franus, Elzbieta Beran, Adriana Czimerova (2006). "Properties and Potential applications of zeolite materials Produced from fly ash using simple method of synthesis". *Powder technology*, **166**: 47-54.
- [25]. Hidekazu Tanaka, Atsushi Fujii (2009). "Effect of stirring on the dissolution of coal fly ash and synthesis of pure-form Na-X and X-Zeolites by two-step process". *Advanced powder technology*, **20**(5): 473-479.
- [26]. M. Chigondo, U. Guyo, M. Shumba, F. Chigondo, B. Nyamunda, M. Moyo, T. Nharingo (2013). "Synthesis and Characterisation of Zeolites from Coal Fly Ash (CFA)". *IRACST-Engineering Science and Technology: An International Journal (ESTIJ)*, **3**(4): 714-718.

How to cite this article: Elavarasan, G., Rajakrishnamoorthy, P., Karthikeyan, D. and Saravanan, C.G. (2019). Utilization of Coal Fly Ash as a Raw Material for the Synthesis of Zeolite like Substance. *International Journal on Emerging Technologies*, **10**(1): 176-182.