

Synthesis of Zeolites from Class C and Class F Fly Ash Through Alkali Fusion: Focus on The Role of Main Impurities on Zeolite Formation

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Introduction

Although there is growing interest in renewable energy sources, coal remains a primary energy source globally, accounting for a substantial portion of energy generation. Unfortunately, coal combustion contributes significantly to carbon emissions, with coal-fired power plants alone responsible for approximately 45% of global carbon emissions (Belviso, 2018). One significant by-product of coal combustion in thermal power plants is fly ash, widely recognized as a hazardous and toxic waste material. The environmental risks posed by fly ash, containing toxic metals such as aluminum (Al), lead (Pb), cadmium (Cd), chromium (Cr), mercury (Hg), vanadium (V), boron (B), arsenic (As), and selenium (Se), underscore the considerable risks it poses to the environment (Belyaeva & Haynes, 2012). Therefore, there is a critical need for proper management to mitigate environmental contamination and associated health hazards.

Despite its hazardous nature, fly ash also contains valuable elements such as silica, alumina, iron oxide, and calcium oxide. Extracting these valuable components from fly ash through appropriate treatment processes, including acid leaching, presents an opportunity. One promising approach involves producing zeolites from leached fly ash, leveraging its silica and alumina content (Akın et al., 2021). Zeolites, renowned for their unique porous structure and adsorption capabilities, offer numerous environmental applications, including CO₂ capture, wastewater treatment, and soil remediation.

This study investigates the potential of recovering elements such as Fe, Al, and Ca from fly ash and converting residual coal fly ash into zeolites through a comprehensive process involving leaching and subsequent synthesis. The aim of this study was to transform hazardous fly ash into value-added zeolite materials while also shedding light on the influence of impurities originating from the source and properties of the investigated fly ash samples. This dual objective sought not only to mitigate the environmental impact of coal combustion but also to investigate how impurities in fly ash affect zeolite synthesis, thereby enhancing the applicability of fly ash sourced from diverse origins.

Methodology

Two types of fly ash samples, i.e., class F fly ash with high silicate (Si) and low calcium (Ca) content, and class C fly ash with low silicate (Si) and high calcium (Ca) content were obtained from Soma and Seyitomer thermal power plants located in Türkiye. The Class C fly ash underwent acid leaching with 1, 3, and 5 M HCl solutions at 25, 45, 65, and 90°C temperatures for 2 hours. Upon completion of the leaching process, the fly ash and leachate solutions were separated with centrifugation, and the fly ash residue was washed using vacuum filtration before being dried overnight in the oven. Subsequently, the leachates were analyzed using ICP-OES, while the fly ash residues underwent XRF analysis. Next, the selected leached fly ash samples were subjected to alkaline fusion-assisted hydrothermal synthesis to produce zeolites. The fly ash samples were mixed with ground NaOH at a mass ratio of 1:1.2, followed by exposure to a temperature of 600°C for 2 hours with a heating rate of 10°C/min for the alkaline fusion step. The resulting fusion products were mixed with double distilled water (DDW) at a ratio of 1:5 and stirred for 8 hours at room temperature for aging. Subsequently, the samples were left for the reaction at the temperature of 85°C for 18 hours. Finally, the obtained zeolite samples were washed three times with DDW via centrifugation, followed by drying overnight. Obtained zeolite samples were characterized by XRD analysis.

Results

Fig. 1 illustrates the effect of different leaching conditions on the distribution of Fe₂O₃ and CaO in Class C fly ash samples. From these results, it was seen that CaO is more soluble than Fe₂O₃. In Class C; it was observed that the Fe₂O₃ decreased from 8.29 to 2.33, while the CaO content remained the same (2.88 to 2.69%). It was noted that CaO exhibited greater solubility than Fe₂O₃, with approximately X% reduction in CaO with respect to X% in Fe₂O₃ upon HCl treatment. According to Fig. 1, fly ash samples labeled 3M-65 and 5M-90 demonstrated the most significant difference in the relative change of Fe₂O₃ while maintaining a nearly constant CaO level. Consequently, these raw materials were selected for further investigation in zeolite synthesis. The HCl treatment conditions applied to Class F fly ash samples were determined based on the chosen conditions for 3M-65 and 5M-90. The chemical compositions obtained for the selected conditions of both Class C and Class F fly ash samples are presented in Table 1.

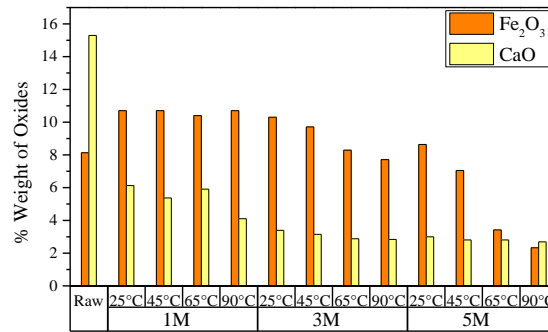


Fig. 1 Chemical composition of Class C and Class F fly ash samples subjected to different HCl leaching conditions

Table 1 Main chemical composition of selected fly ash samples (%)

Chemical composition	Class C			Class F		
	Raw	3M-65°C	5M-90°C	Raw	3M-65°C	5M-90°C
SiO ₂	36.4	61.5	70.5	55.2	63.5	67.8
Al ₂ O ₃	23.3	21.2	19.3	20.3	17.7	17
Fe ₂ O ₃	7.81	8.29	2.33	10.1	8.45	6.5
CaO	14.7	2.88	2.69	3.55	1.62	1.44
Other Oxides	17.8	6.13	5.18	10.8	8.73	7.26
SiO ₂ / Al ₂ O ₃	2.7	4.93	6.21	4.61	6.1	6.78

The table reveals that both fly ash samples respond to HCl leaching. In Class C fly ash, there is a noticeable increase in the solubility of Al₂O₃, Fe₂O₃, and CaO with increasing acid molarity and temperature. The most significant change was observed for Fe₂O₃ content in Class C, while only slight changes in Al₂O₃, Fe₂O₃, and CaO were observed with increasing HCl molarity and treatment temperature in Class F fly ash sample. To investigate the implications of this difference, the selected fly ash samples underwent alkali fusion-assisted hydrothermal synthesis under the same synthesis conditions to produce zeolites. The XRD analysis results of the obtained zeolite samples are presented in Fig. 2.

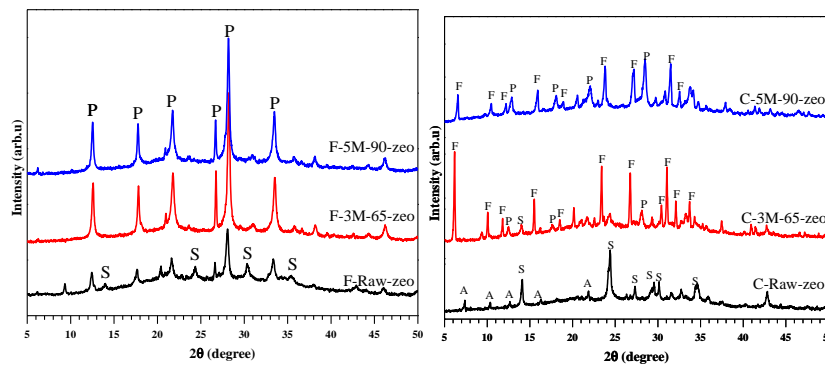


Fig. 2 XRD analysis results of obtained zeolites from Class F (left) and Class C (Right) fly ashes (P:Zeolite P, S:Sodalite, A: Zeolite 4A and F:Faujasite.)

The XRD analysis indicates that the raw fly ash transforms into zeolite P and a mixture of sodalite and zeolite A for Class F and Class C samples after hydrothermal synthesis. After acid leaching, sodalite transforms into zeolite P for Class F fly ash, while faujasite-type zeolite with a minor P phase is formed upon using Class C fly ash sample as raw material. These findings suggest that HCl leaching is more effective on Class C fly ash for removing impurities and obtaining the highly desirable faujasite-type zeolites.

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