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Conversion of Fly Ash to Zeolites A and X and an Assessment of Their Ion Exchange Capacity to Treat Water Containing Mercury

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Abstract: Over the next quarter century, the US government and private industry will allocate billions of dollars per year to remediate sites contaminated with various forms of heavy metals and petroleum products. Over the past 35 years, approximately 750,000 sites with potential contamination have been reported to federal and/or state authorities. Of these, about 300,000 sites still require remediation and additional contaminated sites are identified each year. The main contaminants are heavy metals, solvents, and petroleum products. This study focuses on removing mercury from water. Zeolites are known for their important physiochemical properties such as ion exchange, adsorption, and molecular sieving. Due to their molecular structure, they can be an ideal medium for separation of ions and small molecules from aqueous solution. Our team used a modified fusion process to convert fly ash from four U.S. power plants into zeolites A and X. The ion exchange capability (IEC) of mercury ions of the various samples made with the four types of fly ashes were determined. Based on our results, the fused fly ash (both zeolite A and X) from the Bowen power plant, North Carolina showed the highest IEC. Using the Bowen fused fly ash, it was determined that the IEC for Hg ions containing zeolite X was markedly higher than that containing zeolite A. This study shows that fly ash, a coal burning by-product, can be converted to zeolite with an IEC that has the potential of removing mercury, and potentially other heavy metals, from water and possibly soil.

***Keywords**:* Contaminated Water, Ion Exchange, Heavy Metals, Fly Ash, Zeolite A and X

1. Introduction

Industrial processes often generate waste by-products that can contaminate soil and water. Among the most concerning pollutants are heavy metals, solvents, petroleum products, and organic compounds. This paper focuses on the impact of heavy metals, particularly mercury on human health and their removal from water. Heavy metals are generally defined as elements with atomic weights between 63 and 200 and a specific gravity above 4 [1].

While small amounts of elements like copper, iron, and zinc are essential for life, others such as cadmium, lead, arsenic, and mercury are toxic even in trace amounts. The U.S. Environmental Protection Agency has identified mercury, lead, cadmium, arsenic, and chromium as priority contaminants due to their persistence and toxicity [2]. Mercury, in particular, cannot be broken down or neutralized and is harmful at very low concentrations. Its widespread presence in the environment poses serious risks to ecosystems and can cause severe neurological and physiological damage in humans.

In 2021, U.S. coal-fired power plants consumed around 800 million tons of coal, generating about 70 million tons of coal ash [2]. Rather than sending fly ash to landfills, one hopeful solution is converting it into zeolites a material used for ion exchange, adsorption, and molecular sieving. This not only reduces waste but creates valuable engineering products. According to ASTM C-618, fly ash is classified as Class F (low calcium) or Class C (high calcium). This study focuses on Class F. Four samples from U.S. power plants were chosen to represent a range of compositions:

- Conemaugh Generating Station, Pennsylvania
- Bowen Power Plant, Georgia
- Belews Creek Power Plant, North Carolina
- Oak Grove Power Plant, Texas

These specific fly ashes have been selected as representative of the broad range of fly ash compositions generated from various coal burning operations, see Table 1.

Table 1. Percent (%) Composition of Class F Fly Ash for Zeolite Construction Materials

	Conemaugh	Bowen	Belews Creek	Oak Grove
SiO_2	47.0	63.2	56.8	60.1
Al_2O_3	23.0	19.4	26.4	18.0
Fe_2O_3	21.4	5.4	6.0	7.9
CaO	2.1	4.3	2.9	3.7
MgO	0.8	1.3	1.9	2.4
Alkali	1.4	1.1	1.0	0.2
SO_3	0.05	1.4	0.9	1.0
LOI(%C)	4.0	2.1	0.6	1.5

Currently, the main uses of fly ash have been using as filler in concrete or as partial replacement for portland cement in construction. However, there is a limit that beyond which can be damaging to the overall properties of the hardened concrete. During the past three decades, production of synthetic zeolite from fly ash have been reported by many researchers [3-6]. Zeolites are ideal material as molecular sieve which possess large surface area with a uniform molecular pore. In this work fly ashes from the four power plants are converted to zeolite A and zeolite X by modified fusion process [7-12]. The detail of this process is shown below.

2. Conversion of fly ash to zeolite A and zeolite X.

Fly ash obtained from each of the four power plants listed above were placed in an oven at 100 OC for 8 hours to ensure there is no excess moisture in the ash as received from the power plants. Solid NaOH powder were mixed with each fly ash in a plastic bottle using a weight ratio of 6:5. These bottles were placed into a V-mixer for a period of 30 minutes to insure through mixing. The resulting mixture was ball-milled and heated at 575 OC for 90 minutes. After cooling for 6 hours followed by additional grinding, then distilled water was added to the fused fly ash/NaOH material using a weight ratio of 1:5. The mixture was placed into a plastic bottle and V-mixed for one day at room temperature to assure uniformity.

To synthesis zeolite X, the mixture is placed in a glass flask and cured for 8 hours at 60 OC. After curing, the mixture is washed with distilled water and centrifuged. The solids then air dried at 80OC for 10 hours. To synthesis zeolite A, prior to curing, Aluminum Hydroxide Hydrate (50% Al2O3) was added to fused flay ash at 1:10 weight ratio and proceed with the same steps as described in the previous paragraph.

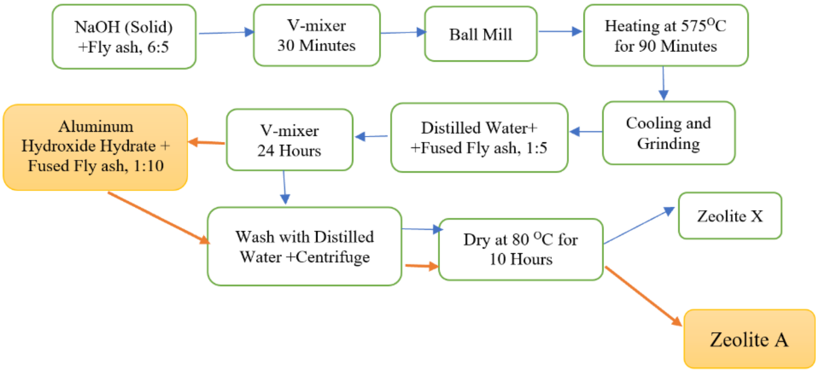

Figure 1. Flow Chart of the Fusion Method to Synthesize Zeolites A and X from Fly Ash.

The surface area and pore volume of the commercial zeolites and the treated fly ash are shown in Table 2. The surface area of treated fly ash from Bowen power plant was highest among the four and was chosen for this investigation.

Table 2: Surface Area and Pore Volume of Commercial and Synthetic Zeolites (cm^3/g)

Zeolites	**Surface Area (m^2/g)**	**Pore Volume (cm^3/g)**	**Pore size (Å)**
UOP 4A	400 - 500	0.25–0.30	4
UOP X-60	600-700	0.30–0.35	7.8
Conemaugh (Zeolite X/Zeolite A)	220/60	0.21/0.11	8/4
Bowen (Zeolite X/Zeolite A)	380/220	0.25/0.15	7.5/4
Belews Creek (Zeolite X/Zeolite A)	310/75	0.22/0.12	8/4
Oak Grove (Zeolite X/Zeolite A)	200/65	0.2/0.1	8.5/4.5

3. Methodology

Materials and Reagents: Zeolite X and zeolite A were produced from 4 fly ashes but the zeolites with the highest surface area produced from Bowen power plant was used in this investigation. Mercury nitrate ($Hg(NO_3)_2$) was used to produce Hg^{2+} ions. Deionized water and analytical grade chemical were used in this experiment.

Preparation of Mercury Solutions: Solution concentration of Hg 2+ with concertation ranging from 10 ppm to 5,000 ppm was prepared. To obtain these concentrations deionized water was used to dilute a stock solution containing 10,000 mg/L Hg^{2+}.

Ion Exchange Capacity Experiments: To determine the ion exchange capacity of the zeolite X and zeolite A, batch experiment was conducted. In this work, the effect of initial Hg2+ concentration, contact time, pH and temperature were investigated.

Isotherm: Ion exchange capacity efficiency of zeolite A and zeolite X for Hg2+ with concentration 10 ppm, 100, ppm, 1000 ppm and 5,000 ppm were conducted.

Kinetics: To determine adsorption kinetics, Samples were collected at intervals (5, 10, 20, 30, 60, 120, 240, 480 min).

pH Effect: To determine the effect of pH on ion exchange capacity of zeolites, the pH of the mercury solution was varied between 2 and 10.

Temperature Effect: To determine the effect of temperature on ion exchange capacity of zeolites, the experiments were conducted at 20, 30, and 40°C.

4. Data Interpretation

The results show that both synthetic zeolite A and zeolite X possess significant ion exchange capacity for Hg^{2+} ions. While zeolite X with larger pore system (about 8 A) showed consistently higher ion exchange capacity. Figure 2 shows the ion exchange capacity of synthetic and commercially available zeolite A and zeolite X for Hg^{2+}. IEC (in meq/g) represents how many cations a zeolite can exchange per gram. Since **Hg^{2+} is a divalent ion**, each ion counts as **2 equivalents** in ion exchange.

To evaluate the ion exchange capacity for Hg^{2+}, it is essential to distinguish ion exchange from adsorption and precipitation. A more accurate measure of true ion exchange is the corresponding release of Na^+ into solution. Figure 2 presents data at the optimal pH of 6, showing the ion exchange capacity in a 1000 ppm Hg^{2+} solution at 40 °C.

A more detailed study, which includes results from all four fly ash sources, will be published in a later paper. That work examines how pH, mercury concentration, and temperature affect the ion exchange behavior of Hg^{2+}

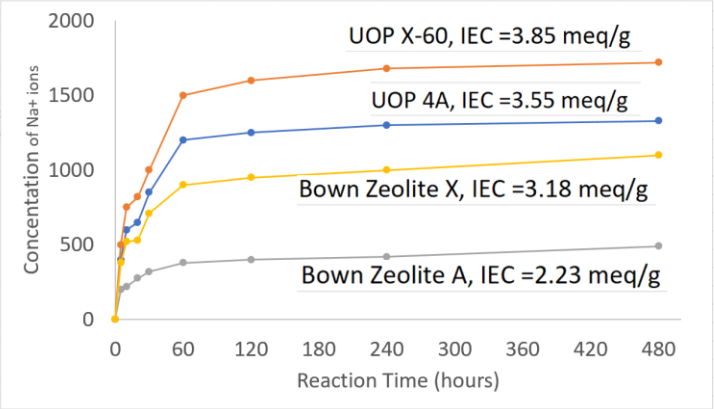

Figure 2. Mercury ion-exchange behavior and IEC of synthetic and commercial zeolite A and zeolite X.

5. Conclusion

Since the Industrial Revolution, the production of heavy metals such as lead, cadmium, chromium, arsenic, mercury, copper, and zinc has increased exponentially. Exposure to heavy metals has been linked with developmental delay, various cancers, kidney damage, and even death in some instances of exposure to very high concentrations.

Fly ash a coal burning by-product from four different power plant were used to synthesize zeolite A and zeolite X with a fusion process. Fly ash obtained from Bowen power plant was used in the work because it produced zeolites with the largest surface area.

Zeolite X produced from Bowen fly ash, with an estimated pore size of 8 Å, showed an ion exchange capacity of 3.18 meq/g for Hg^{2}+ about 85% of that of the commercial zeolite X (UOP X-60). In contrast, the fused fly ash converted to zeolite A had an IEC of 2.33 meq/g for Hg^{2+}, with a smaller pore size of around 4 Å, achieving roughly 68% of the capacity of its commercial counterpart (UOP 4A). The higher performance of zeolite X is likely due to its larger pore size, allowing for efficient mercury ion exchange

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