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Synthesis of Zeolite from Fly Ash and Aluminum Can Waste as a Nickel (Ni) Adsorbent

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ABSTRACT

Palm oil boiler fly ash and waste aluminum cans are abundant industrial residues that remain underutilized. This study aims to synthesize Y-type zeolite from boiler fly ash and aluminum can waste for application as an adsorbent to reduce nickel (Ni^{2+}) content in industrial wastewater. The experiment employed variations in aluminum can mass (1 g, 2 g, 3 g) and boiler fly ash mass (60 g, 80 g, 100 g). Zeolite synthesis and its adsorption performance were evaluated in two stages. The obtained zeolite was characterized using FTIR and XRD to confirm Y-type zeolite formation, while AAS analysis determined nickel concentration. The optimal adsorption efficiency of 87.69% was achieved with 2 g aluminum can waste and 80 g boiler fly ash. These findings demonstrate that combining fly ash and aluminum waste can effectively produce zeolite Y with high potential as a sustainable adsorbent for heavy metal removal from wastewater.

Keywords: zeolite, boiler fly ash, aluminium foil waste, adsorbent

1. INTRODUCTION

The industrial sector currently represents one of the major pillars of Indonesia's economy, with the palm oil industry serving as a key contributor. During the processing of oil palm fruits, solid residues in the form of palm shells are generated. The utilization of various palm oil mill by-products has been widely studied, including the use of palm shells and fibers as boiler fuel to meet the plant's energy requirements. The combustion of these materials produces by-products such as fly ash (approximately 100 kg per week) and boiler bottom ash (about 3 to 5 tons per week).[1] Fly ash has attracted attention as a potential adsorbent due to its chemical composition, which contains approximately 31.45% SiO_2, 15.2% CaO, and 1.6% Al_2O_3.

In addition to fly ash, a considerable amount of aluminum waste is produced from used beverage cans. Although the recycling potential of aluminum cans reaches up to 95%, the increasing consumption of aluminum-packaged beverages has led to a significant accumulation of aluminum waste. Chemical composition analysis indicates that aluminum foil contains approximately 92–99% aluminum, making it a promising source of aluminum for the synthesis of aluminosilicate-based materials such as zeolites.[2]

Fly ash and aluminum cans can be utilized as precursor materials for zeolite synthesis through the hydrothermal method. Zeolites are crystalline aluminosilicates characterized by a three-dimensional framework of interconnected silicate ($[SiO_4]^{4-}$) and aluminate ($[AlO_4]^{5-}$) tetrahedra sharing oxygen atoms, which form a regular molecular-sized porous structure. These materials exhibit distinctive physicochemical properties, including high adsorption capacity, ion-exchange ability, and catalytic activity.[3,4]

A previous study conducted by Safitri and Jahro (2020) successfully synthesized zeolite X from boiler ash and aluminum foil waste for the adsorption of Zn(II) and Cd(II) ions. The synthesis involved variations in the Si/Al molar ratio (1.4, 1.5, and 1.6) and the addition of Na_2EDTA (2.0 g and 3.0 g). The highest crystallinity of 75% was achieved at an Si/Al ratio of 1.6 with 3.0 g Na_2EDTA. Building upon these findings, the present study aims to synthesize zeolite derived from fly ash and aluminum can waste for potential application as an adsorbent for nickel (Ni^{2+}) ions.

2. EXPERIMENTAL

2.1. Preparation of Waste Aluminum Cans

The waste aluminum cans were prepared through mechanical and chemical cleaning. The cans were first abraded using sandpaper to remove surface contaminants. They were then washed thoroughly with soapy water to eliminate impurities, rinsed with clean water, and air-dried. After drying, the cans were cut into small pieces approximately 0.5 cm in size.

2.2. Preparation of Fly Ash from Boiler

The fly ash was pretreated prior to silica extraction. It was first washed with hot water and filtered to remove floating impurities. The settled (non-floating) fly ash was separated and soaked in a 10% nitric acid (HNO_3) solution for 12 hours. After acid treatment, it was rinsed with distilled water and dried in an oven at 80 °C for 3 hours. Subsequently, 200 g of the dried fly ash were added to 2000 mL of 1.5% sodium hydroxide (NaOH) solution. The mixture was boiled using a hot plate for 30 minutes. The extract was then cooled to room temperature and left to stand for 12 hours before being filtered. The resulting filtrate, containing silica sol, was gradually added with 10% HNO_3 until gel formation occurred. The pH was adjusted to neutral (pH = 7) using a universal indicator. The resulting silica gel was left at room temperature and washed repeatedly with hot water while being filtered using a 200-mesh sieve until the filtrate was clean and white. The obtained silica was then dried in an oven at 80 °C and ground into a fine powder.

2.3. Zeolite Synthesis

Zeolite synthesis was carried out via a hydrothermal method. First, 11.2 g of NaOH were dissolved in 450 mL of distilled water, followed by the addition of 2.5 mL of tetrapropylammonium hydroxide (TPAOH). The solution was stirred until homogeneous. This precursor solution was divided into two parts: 250 mL and 200 mL, which were used to dissolve the silica and alumina, respectively, at different mass ratios of 60:1, 80:2, and 100:3 (silica:alumina, in grams). In the second stage, aluminum can pieces were gradually added to the 200 mL alumina precursor solution and stirred until homogeneous. In the third stage, the silica extract from fly ash was added to the 250 mL silica precursor solution and stirred until homogeneous. The silica and alumina

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solutions were then combined and mixed thoroughly. The resulting mixture was transferred into polypropylene bottles and subjected to hydrothermal crystallization in an oven at 100 °C for 48 hours. The zeolite gel was then washed with distilled water and dried at 80 °C. The dried zeolite crystals were ground and sieved using a 200-mesh sieve. The final product was calcined in a furnace at 550 °C for 5 hours to improve crystallinity and remove organic residues.[5]

2.4. Nickel (Ni) Ion Adsorption Test

The adsorption capacity of the synthesized zeolite was tested for nickel (Ni^{2+}) removal from wastewater. The concentration of Ni^{2+} ions before and after adsorption was measured using Atomic Absorption Spectroscopy (AAS). A total of 0.125 g of zeolite was added to 25 mL of wastewater sample. The mixture was stirred using a magnetic stirrer for 60 minutes and then allowed to stand for 45 minutes. The filtrate was collected, and the residual Ni^{2+} concentration was measured using AAS.[6]

3. RESULTS AND DISCUSSION

3.1. FTIR Spectrum Analysis

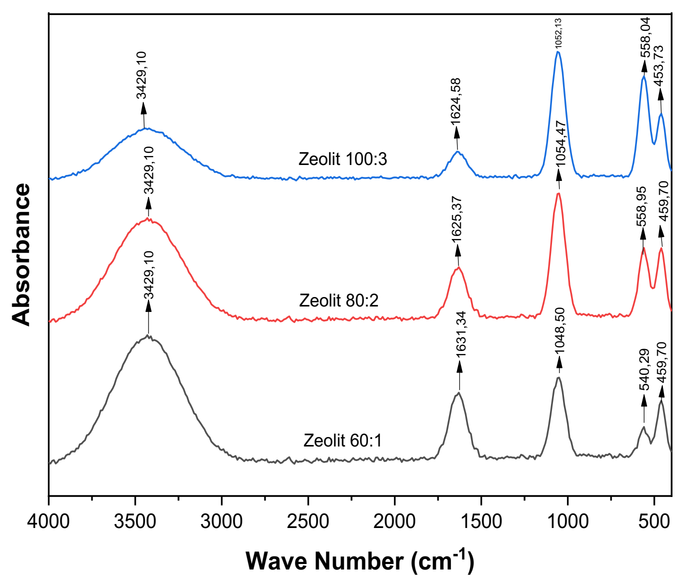

Figure 1. FTIR Analysis of Zeolite Samples with Various Rations (60:1; 80:2; and 100:3)

As shown in Figure 1, the FTIR spectrum analysis of the three synthesized zeolite samples derived from boiler fly ash (ratios 60:1, 80:2, and 100:3) showed the development of characteristic aluminosilicate structures. In the low wavenumber region (459–463 cm⁻¹), a band was observed, corresponding to the T–O–T (Si/Al–O–Si/Al) bending vibration. The intensity of this band increased with the synthesis ratio. The 60:1 sample exhibited absorption at 459.70 cm^{-1} with relatively low intensity, whereas the 100:3 sample displayed a sharper absorption at 463.73 cm^{-1}. This indicated a stronger bonding within the aluminosilicate framework.

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According to Wahyuni, a higher degree of regularity in the T–O–T band reflects the formation of a more stable zeolite network.[7]

In the range of 540–560 cm⁻¹, another band appeared, associated with the double six-membered ring (D6R) structure, which is a characteristic feature of zeolites. The 60:1 sample showed a weak band at 540.29 cm^{-1}, while the 80:2 and 100:3 samples presented more intense bands around 558 cm^{-1}. According to Prabowo (2020), absorption bands in this region indicate the development of crystalline zeolite structures from amorphous silicate materials such as fly ash. The increase in band intensity at higher synthesis ratios suggested that the zeolite framework became more developed and structurally ordered.[8]

A prominent peak was observed in the 1000–1100 cm^{-1} region, corresponding to asymmetric stretching vibrations, which was attributed to the homogeneity of Si and Al after synthesis. The 60:1 sample showed a peak at 1048.50 cm^{-1}, while the 80:2 and 100:3 samples displayed peaks around 1025 cm^{-1}. The shift of the peak toward lower wavenumbers, along with the increase in intensity, indicated a transition from an amorphous silica phase to a porous zeolitic structure. This observation is consistent with the findings of Lestari and Suryani (2019), who reported that the formation of zeolite from fly ash is characterized by the emergence of absorption bands in the 1000–1050 cm^{-1} range.[9]

Moreover, the bands detected at 1613–1625 cm^{-1} were assigned to the H–O–H bending vibrations of adsorbed water molecules. The lower intensity of this band in the 100:3 sample indicated a reduction in physically adsorbed water, which corresponds to higher crystallinity of the zeolite. Santosa (2017) stated that the reduction in adsorbed water reflects improved structural stability. Additionally, a broad band was observed at 3420–3430 cm^{-1}, corresponding to O–H stretching vibrations from surface and hydroxyl water groups. All samples showed absorption near 3429 cm^{-1}; however, its intensity decreased for the 100:3 sample. This suggested a reduction of free water and surface hydroxyl groups, implying the formation of a more mature zeolite framework, as also explained by Setyowati (2021).[10,11]

3.2. Nickel (Ni) Ion Adsorption

The AAS analysis results for Ni ion adsorption are presented in Table 1.

Table 1. AAS results of Ni ion adsorption

Experiment	Mass of Boiler Fly Ash Extract (g)	Mass of Scrap can (g)	Ni (%)
1	60	1	48,74
2	80	2	87,69
3	100	3	69,12

As shown in Table 1, Ni ion adsorption reached its optimum when 80 g of boiler fly ash extract and 2 g of scrap can material were used, and decreased when 100 g of fly ash extract and 3 g of scrap cans were applied. This occurred because an increase in active sites on the zeolite surface allowed more efficient adsorption of Ni^{2+} ions. However, further increasing the amount of fly ash extract altered the Si/Al ratio, leading to reduced adsorption efficiency. This behavior is consistent with the findings of Nurhasni et al. (2010), who reported that higher fly ash content in zeolite synthesis leads to greater adsorption efficiency for heavy metal ions.[12]

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3.3. XRD Analysis of Zeolitw

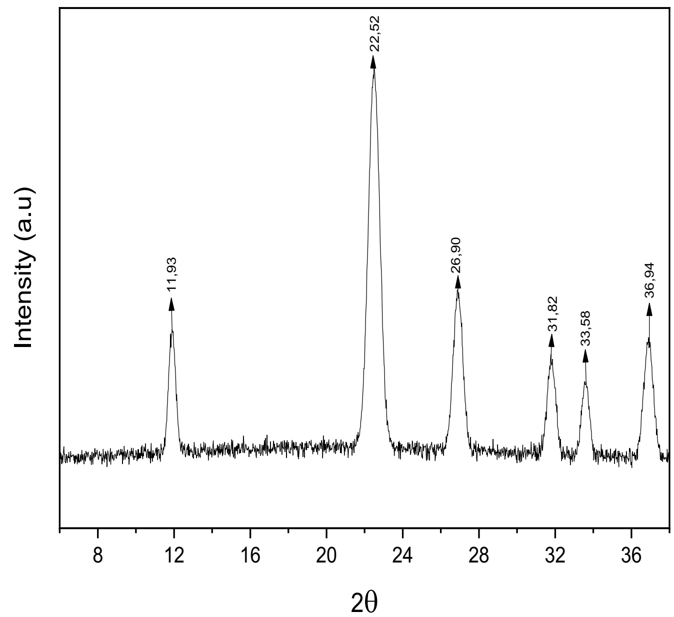

Figure 2. Diffractogram of Zeolite

The XRD diffractogram presented in Figure 2 revealed several crystalline peaks, indicating the formation of a porous aluminosilicate phase. The dominant peaks were observed at 2θ values of approximately 11–12°, 22–24°, 26–28°, 31–33°, and 36–37°. The most intense peak appeared in the 22–24° range, which represents the main reflection of the zeolite crystalline structure and indicates a high degree of crystallinity. The sharpness of this peak was attributed to the presence of SiO_2. This agrees with the results reported by [13] who observed that strong peaks at $2\theta = 22–25°$ are characteristic of SiO_2.

The low-angle peak (~11–12°) was associated with reflections from characteristic zeolitic planes, often related to the porous unit cell structure. The prominent peak in the 22–24° region served as clear evidence of a well-developed tetrahedral Si–O–Al framework. Furthermore, secondary peaks at approximately 27°, 31–33°, and 36–37° confirmed the presence of crystalline zeolite phases. According to the JCPDS No. 48-1862 database, the synthesized material corresponded to Y-type zeolite (modernite type), which is characterized by a higher Si content compared to Al.[14, 15]

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4. CONCLUSION

The synthesis of zeolite using 80 g of boiler fly ash and 2 g of aluminum can scrap achieved optimal Ni^{2+} ion adsorption with an efficiency of 87.69%. The enhanced adsorption capacity was attributed to the increased number of active sites on the zeolite surface, enabling stronger metal ion binding. XRD analysis confirmed the formation of Y-type zeolite, characterized by its porous structure and high adsorption ability. These results demonstrate that industrial and metallic wastes can be effectively reused as precursors for zeolite production, providing a sustainable approach for water purification and environmental remediation applications.

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