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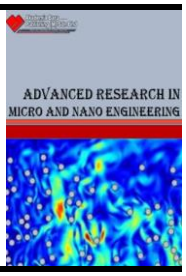


# Synthesis of Zeolite by Using Waste Cans as a Source of Aluminium and Testing its Performance as a Dye Adsorbent

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**ABSTRACT**

Aluminium metal has corrosion-resistant properties, light and easy to obtain, so it is widely used as a raw material for making soft drink cans. The aluminium content in used cans can be more than 90% and it can be used as raw material for making zeolite. In this study, zeolite was synthesized, and its performance was tested as an adsorbent of water-soluble (Congo red) and water-insoluble (Naphthol blue black) textile dye waste. The method used in this study includes three stages, namely (1) synthesis of zeolite from waste cans as a source of Al and waterglass as a source of Si, (2) characterization of zeolite produced using XRD, FTIR, and SEM, and (3) performance test of zeolite on Congo red and Naphthol blue black dyes with adsorbent weight variation: 0.1 to 0.5 gram in 10 mL of a prepared sample at a concentration of 20 mg/L; contact time variations: 30 to 120 minutes at a speed of 150 rpm; pH variations: 5, 7, and 9; and variations in the initial concentrations of dyes of 10 to 40 ppm. The decrease in dye concentration was measured using a UV-Vis spectrophotometer. The Al content in the used cans obtained is 96.5%. The zeolite was successfully synthesized, and the yield obtained from the zeolite formation reaction was 92.67%. XRD characterization results show the diffraction pattern of a mixture of zeolite-X and zeolite-NaP1 with an average crystal size of 24,08 nm. The purity of the synthetic zeolite for zeolite X was 55.44% and zeolite Na-P1 was 44.53%. FTIR characterization shows the intensity of functional groups T-O, T-O-T or O-T-O, and T-H, where T is Si or Al. Characterization by SEM, synthetic zeolite has the same morphology as Faujasite zeolite. The maximum adsorption capacity of zeolite on Congo red and Naphthol blue black was 321,2921 mg/g and 77,5354 mg/g based on Freundlich isotherm. The optimum conditions for adsorption of Congo red and Naphthol blue black dye were at 0,4 gram and 0.1 gram of adsorbent weight, 30 minutes of contact time, 5 and 7 of pH and 40 mg/L initial concentrations of dyes.

## 1. Introduction

The chemical industry is developing rapidly along with the increase in the number of the world's population. Industries that use synthetic dye stuffs in the manufacturing process of their products are also increasing [1]. In developing countries, the textile industry plays an important role in

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economic development. But the textile industry also causes quite serious environmental problems, one of which is from the waste of the dyeing process of materials or clothes [2]. Synthetic dyes contain compounds with a complex aromatic molecular structure, it makes dyes difficult to decompose naturally when discharged into the environment [3]. Synthetic textile dyes can be categorized into water-soluble and insoluble dyes in water. Based on the physical and chemical properties, Congo red, Crystal violet, Auramine, Methylene blue, Azin, and Rhodamine B are dyes that soluble in water and Naphthol is one of the dyes that are insoluble in water.

Congo Red dye is widely used in the cellulose industry such as the cotton fabric industry and paper industry. Congo red can cause allergies such as anaphylactic shock and can even cause cancer Naphthol blue black dye has been widely used by the textile industry for fabrics, nylon, silk, and batik dyeing [4]. This compound is not easily degraded, if degraded, it takes quite a long time. If it is too long in the biosphere, it will become a source of pollutants due to its carcinogenic and mutagenic nature [5].

Various methods are used to treat waste with synthetic dyes, including biological, physical, and chemical methods including adsorption, biosorption, coagulation/flocculation, advanced oxidation, ozonization, membrane filtration and liquid extraction [6]. Adsorption is one of the physics methods that are widely used to treat waste with dye content because it has the properties of being easy to use, efficient and low in energy needs, and it contains various types of adsorbent materials [7]. Adsorbents commonly used are zeolite, activated carbon, nanoparticles, and polymers. Zeolites are a highly porous material made up of crystals of aluminium silicates [8].

One of the wastes that poses problems for the environment is used cans that are hoarded in landfills. The aluminium content in the can can be used as a raw material for making zeolite. This research was conducted by synthesizing zeolite from waste cans as a source of aluminium and waterglass as a source of silica. Waterglass is a common name for the chemical sodium silicate ($Na_2SiO_3$). Synthesis of zeolite is carried out by the sol-gel and hydrothermal methods. The sol-gel process first produces amorphous gel from an interaction between aluminate and silicate. The hydrothermal method is used due to the growth of crystals of good quality with a controllable composition [9]. Furthermore, zeolite is characterized by XRD, FTIR and SEM. To find out the performance of the zeolite, it was tested as an adsorbent of water-soluble textile dyes, namely Congo red and insoluble water textile dyes, namely Naphthol blue black. Adsorption optimization is carried out by varying the weight of zeolite, contact time, pH and dye concentration.

In the research conducted by Abdelrahman [10], zeolite was synthesized via a hydrothermal method using waste aluminium cans as an aluminium source and four silicon sources namely, fumed silica, sodium metasilicate, silica gel, and tetraethyl orthosilicate, the maximum adsorption capacities of the malachite green dye on the as-prepared zeolite nanostructures (ZF, ZM, ZS, and ZT) adsorbents were 226.757, 239.234, 29.744, and 25.221 mg/g respectively. In the research conducted by Iryani *et al.,* [2], zeolite ZSM-5 was synthesized from kaolin bangka and adsorption was carried out on textile dyes. Adsorption capacity for dyes Congo red, Auramine, Methylene blue, Azin and Rhodamin B respectively 129, 157, 134, 205 and 210 mg/g. In both n both experiments it can be seen that zeolite has good adsorption in synthetic dyes.

## 2. Methodology

### *2.1 Materials*

The research was conducted from August to October 2022 in the laboratory of PT Organo Science Laboratory which is at Plaza Amsterdam, Jl. MH. Thamrin Blok A No.7, Citaringgul, Babakan Madang, Kabupaten Bogor, West Java, Indonesia. The tools used are a set of laboratory glass tools, sandpaper,

porcelain crucible, mortar, magnetic stirrer, hotplate, spatula, analytical balance, oven, furnace, pH meter, spectrophotometer UV-Vis, ICP-OES, FTIR, XRD and SEM. The materials used are waste cans, waterglass ($Na_2SiO_3$), NaOH, HCl, $H_2O_2$, demineralized water, filter paper, Congo red dye, and Naphthol blue black dye.

### *2.2 Methods*

#### *2.2.1 Determination of aluminium content and synthesis of sodium aluminate*

The method of determining the concentration of aluminium in the waste can and the synthesis of sodium aluminate was carried out by modifications to the IEC 62321 methods [11]. Waste cans are cleaned with sandpaper, and cut into small pieces. The can is weighed as much as 1 gram, and 5 mL of concentrated nitric acid is added, stirred and covered with a watch glass. Heated the sample above the hotplate at 95 ± 5 $^oC$, then the sample was cooled, and 10 mL of concentrated hydrochloric acid was added, and reheated for 30 minutes. The solution is added 1 mL of 30% peroxide acid, then reheated until the sample condition does not change, heating is carried out until the solution volume is 5 mL. The filtrate is filtered and put into a 100 mL measuring flask, demineralized water is added and stamped to a volume of 100 mL. The solution is measured using ICP-OES. Furthermore, after the concentration of aluminium is known, it is continued with the synthesis of sodium aluminate. Weighed 2 gram waste cans that had been cleaned with sandpaper and cut into small pieces, put in 50 mL of NaOH solution 1 M. Reaction is allowed to occur until all the aluminium is dissolved. Then, the mixture is dried in the oven for 12 hours at 100 °C and calcined at 650 °C for 3 hours in a furnace.

#### *2.2.2 Zeolite synthesis*

The zeolite synthesis has been prepared as follows: 14.5 gram sodium silicate (waterglass) 58%, 3 gram sodium aluminate, 2 gram NaOH and demineralized water are mixed into a porcelain container. Sodium aluminate is mixed with waterglass (sodium silicate) gradually. The mixture was stirred for 1 hour in a closed container and then continued by heating at a temperature of 100 $^oC$ for 24 hours in the oven. The synthesis results were washed to pH 8 with demineralized water and dried at 100°C in the oven for 12 hours.

#### *2.2.3 Characterization of zeolite*

Characterization of zeolite with X-ray diffraction (XRD), 1 - 2 gram zeolite platted into the sample holder and analyzed using XRD. Characterization of zeolite by Fourier Transform Infra Red (FTIR), 10 mg zeolite is grinded together with 300 mg of dry KBr powder. Next, it is placed on a preparate and pressed with a press to form a pellet. The formed pellets are placed in a sample holder and analyzed using FTIR at a wavelength of 4000-400 $cm^{-1}$.

Characterization of zeolite with SEM, zeolite is taken 5 grams and placed in a sample holder, then the sample is analyzed morphologically using a magnification of 100 – 20000 times until the surface morphology and particle shape are clearly visible.

### *2.2.4 Congo red and naphthol blue black adsorption optimization*

First, the standard series of Congo red and Naphthol blue black was created in 10, 20, 30, 40 and 50 mg/L. Choose one of any standard to determine the maximum wavelength value with a range of 400-725 nm using a spectrophotometer UV-Vis. Then, the standard series is measured at the maximum wavelength value.

Determination of optimum weight of Congo red and Naphthol blue black adsorption. Zeolite adsorbent with weight variations of 0.1, 0.2, 0.3, 0.4 and 0.5 grams was added in 10 mL of Congo red and Naphthol blue black samples with a concentration of 20 mg/L, at pH 7, the solution was stirred for 30 minutes at a speed of 150 rpm and allowed the solution to stand for a while until the zeolite settled. The upper of the solution is slowly taken and its absorbance is measured with a spectrophotometer UV-Vis.

Determination of optimum contact time of Congo red and Naphthol blue black adsorption. 10 mL of Congo red and Naphthol blue black samples with a concentration of 20 mg/L, at pH 7, added with the optimum weight of zeolite. The mixture is stirred at a speed of 150 rpm for 30, 60, 90 and 120 minutes, and allowed the solution to stand for a while until the zeolite settles. The upper of the solution is slowly taken and its absorbance is measured with a spectrophotometer UV-Vis.

Determination of optimum pH of Congo red and Naphthol blue black adsorption. 10 mL of Congo red and Naphthol blue black samples with a concentration of 20 mg/L, set the pH solution at 5, 7, and 9 with the addition of NaOH or HCl, then added with the optimum weight of zeolite. The mixture is stirred at a speed of 150 rpm with optimum time, allowing the solution to stand for a while until the zeolite settles. The upper of the solution is slowly taken and its absorbance is measured with a spectrophotometer UV-Vis.

Adsorption process of various concentrations at optimum conditions. 10 mL of Congo red and Naphthol blue black samples with a concentration of 10, 20, 30 and 40 mg/L, set at the optimum pH, then added with the optimum weight of zeolite. The mixture is stirred at a speed of 150 rpm with optimum time, allowing the solution to stand for a while until the zeolite settles. The upper of the solution is slowly taken and its absorbance is measured with a spectrophotometer UV-Vis.

### *2.2.5 Determination of adsorption capacity*

The adsorption capacity is calculated based on the following equation

$$q_e = \frac{(C_o - C_e)V}{m} \quad (1)$$

The determination of the maximum adsorption capacity can use the Langmuir isotherm equation as follows

$$\frac{C_e}{q_e} = \frac{1}{K_L q_{max}} + \frac{C_e}{q_{max}} \quad (2)$$

While the Freundlich isotherm equation is used as follows

$$Log\ q_e = Log\ K_f + Log \frac{1}{n} C_e \quad (3)$$

Where:
m = mass of adsorbent used (grams)
V = volume of solution (Liters)
qe = number of dye molecules adsorbed at equilibrium state (mg/g)
qmax = maximum adsorption capacity (mg/g)
KL = Langmuir constant (L/g)
Kf = Freundlich isoterm constant (L/g)

## 3. Results

### *3.1 Determination of Aluminium Concentration and Synthesis of Sodium Aluminate*

Determination of the concentration of aluminium in the waste cans needs to be known before making sodium aluminate. The analysis was carried out in three repetitions and it obtained aluminium concentrations of 96.88%; 96.72% and 95.90%, with an average of 96.50%. The large enough aluminium content can be used as a raw material for the synthesis of sodium aluminate. Sodium aluminate is synthesized by the reaction between aluminium, NaOH and water, the reactions that occur are as follows

$$2Al + 2NaOH + 6H_2O \rightarrow 2NaAl(OH)_4 + 3H_2$$

Sodium aluminate is calcined at a temperature of 650 $^oC$, the calcination process is carried out to increase the peak of crystallization formed, so that the zeolite that will be synthesized obtains a purer result [12]. The yield resulting from the experiment was 93.53%.

After sodium aluminate is obtained, it is mixed with sodium silicate (waterglass), NaOH and demineralized water for the zeolite synthesis process. The reaction of the formation of zeolite according to Arnelli *et al.,* [13] is

$$NaOH + NaAl(OH)_4 + Na_2SiO_3 \xrightarrow{25\ ^oC} Na_a(AlO_2)_b(SiO_2)_c.NaOH.H_2O\ (gel)$$

$$Na_a(AlO_2)_b(SiO_2)_c.NaOH.H_2O\ (gel) \xrightarrow{25-175\ ^oC} Na_x(AlO_2)_x(SiO_2)_y.mH_2O\ (zeolite)$$

### *3.2 Synthesis of Zeolite*

The reaction of the formation of zeolite according Adamson and Gast [14] is

$$2NaOH + 10Na_2SiO_3 + 6NaAl(OH)_4 + 10H_2O \rightarrow Na_6Al_6Si_{10}O_{32}.12H_2O + 22NaOH$$

Sodium aluminate is mixed with sodium silicate gradually, the solution slowly becomes in the form of a half-viscous gel. The solution occurs due to the fairly strong interaction between aluminate and silicate, which marks the beginning of the formation of zeolite. Then, continued by hydrothermally process, when the solution is completely mixed, it is heated in the oven at 100 $^oC$ for 24 hours. This heating is carried out to perfect the growth of zeolite crystals [15]. After the heating process, zeolite is washed off until obtaining a pH of 8. This washing process is done to remove the remains of bases and impurities attached to zeolite, the initial pH is around 12 and the washing is carried out around 11 times. Next, the zeolite is dried to evaporate the water in the pores of the zeolite. Based on the zeolite synthesis reaction according to Adamson and Gast [14], the zeolite yield produced from the experiment was 92.67%.

### *3.3 Zeolite Characterization*

#### *3.3.1 XRD (X-Ray diffraction)*

Characterization of zeolite with X-ray diffraction (XRD) is used to analyze the structure, purity and size of crystals. The analysis was carried out with an Empyrean Series 3 Panalytical X-ray diffractometer (XRD), data collector software with a range of 10$^o$ to 90$^o$, using Kα radiation of 1.54060 Å, a voltage of 40 kV and a current of 30 mA. Figure 1 shows the zeolite diffraction pattern.

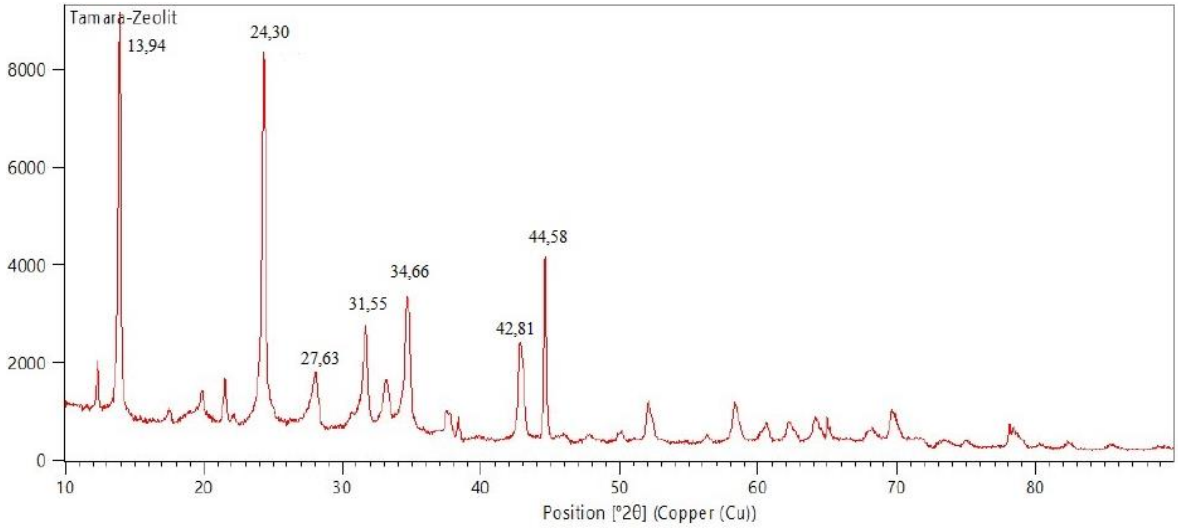


**Fig. 1.** Zeolite XRD Diffraction

Based on the XRD intensity data of zeolite synthesis, it can be known the composition of the constituents of zeolite. The peak amount of zeolite is related to the purity and crystallinity of the synthesis result. The more distinctive peaks of zeolite that appear, the greater the purity and crystallinity [17]. The purity of synthetic zeolite can be determined by knowing the similarity of the angular peak of 2θ as the Table 1 below, then calculating the purity of synthetic zeolite by comparing the zeolite intensity (%).

**Table 1**
Comparison of Diffraction XRD Zeolite

| Zeolit Sintesis | | Zeolit-X [16] | Zeolit-X [17] | Zeolit Na-P1 [16] | Zeolit Na-P1 [18] |
|---|---|---|---|---|---|
| 2θ (°) | Int. (%) | 2θ (°) | 2θ (°) | 2θ (°) | 2θ (°) |
| 12,30 | 7,84 | 6,12 | 13,86 | 12,46 | 12,46 |
| 12,39 | 7,04 | 10,00 | 24,18 | 17,66 | 17,66 |
| 13,94 | 30,79 | 11,73 | 26,57 | 21,67 | 21,67 |
| 19,78 | 5,26 | 15,43 | 29,86 | 28,10 | 28,10 |
| 21,48 | 6,78 | 23,31 | 34,12 | 30,84 | 33,38 |
| 24,30 | 100,00 | 26,65 | | 33,38 | 38,01 |
| 27,63 | 6,04 | 30,94 | | 35,76 | 42,20 |
| 28,00 | 5,81 | 33,59 | | 38,01 | 46,08 |
| 31,55 | 14,89 | | | 44,18 | |
| 34,66 | 31,68 | | | 46,08 | |
| 37,54 | 8,14 | | | | |
| 38,33 | 5,06 | | | | |
| 42,81 | 21,07 | | | | |
| 44,58 | 37,19 | | | | |
| 49,86 | 4,05 | | | | |
| 52,04 | 10,45 | | | | |

Based on the research of Treacy and Higgins [16], synthetic zeolite has a similarity of zeolite diffraction X at an angle of 2θ = 12.30°; 12.39°; 24.30°; 27.63°; 31.55°; and 34.66°, the purity of zeolite X obtained 55.44%. Synthetic zeolite has a diffractional similarity of zeolite NaP1 at an angle of 2θ = 12.30°; 12.39°; 21.48°; 27.63°; 28.00°; 31.55°; 34.66°; 37.54°; 38.33°; 44.58°; and 49.86°, zeolite purity obtained 44.53%. The determination of zeolite size is determined from the results of the diffractogram according to Table 2.

**Table 2**
Zeolite Crystal Size Calculation Results

| 2θ (°) | Cos θ | K (rad.Å$^{-2}$) | λ (nm) | FWHM (°) | FWHM (rad) | Crystal Size (nm) |
|---|---|---|---|---|---|---|
| 13,9397 | 0,9926 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 22,10 |
| 24,3042 | 0,9776 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 22,44 |
| 27,6315 | 0,9711 | 0,9 | 0,15406 | 0,3011 | 0,0053 | 27,17 |
| 28,0014 | 0,9703 | 0,9 | 0,15406 | 0,3011 | 0,0053 | 27,19 |
| 31,5500 | 0,9623 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 22,79 |
| 34,6624 | 0,9546 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 22,98 |
| 42,8096 | 0,9310 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 23,56 |
| 52,0449 | 26,0225 | 0,9 | 0,15406 | 0,3622 | 0,0063 | 24,41 |
| Average (nm) | | | | | | 24,08 |

### *3.3.2 FTIR (Fourier transform infrared)*

Characterization of zeolite with Fourier transform infrared (FTIR) to determine the functional groups contained in zeolite. Characterization of FTIR is carried out in the wave number range of 4000 - 400 cm$^{-1}$. Figure 2 shows the resulting FTIR spectrum.

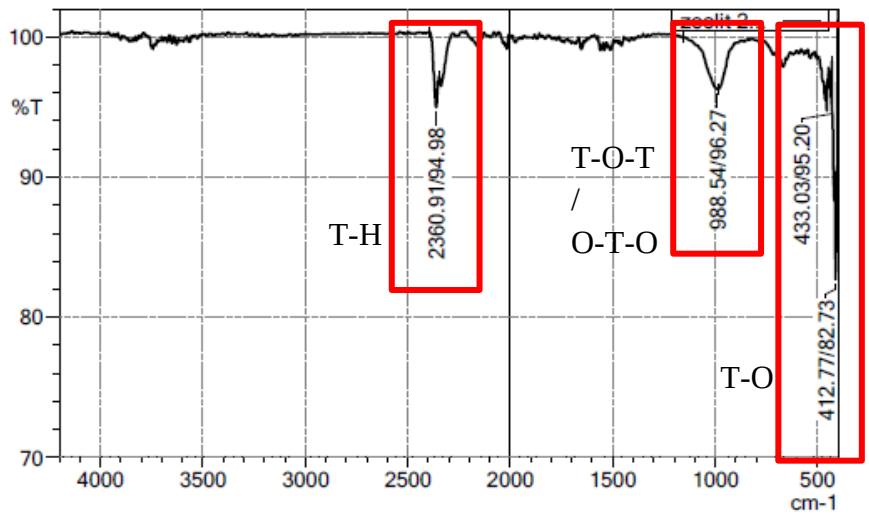


**Fig. 2.** FTIR Spectrum of Zeolite Synthesis

The absorption that occurs is compared with the results of previous research in Table 3, the absorption at a wavelength of 500 – 400 cm$^{-1}$ is related to the absorption of T-O bending vibrations. Absorption in the 988 cm$^{-1}$, showing vibrational absorption of external asymmetric ranges (T-O-T) and internal (O-T-O) [19]. The absorption that occurs around 2360 cm$^{-1}$ shows T-H adsorption. T is Si or Al. The typical infrared absorption region for zeolite X is at 1400 - 400 cm$^{-1}$, shown in Figure 2 there is absorption in that area [20].

**Table 3**
FTIR Zeolite Synthesis Test Results

| Functional Group | Wave Number ($cm^{-1}$) | |
|---|---|---|
| | Standard | Result |
| T-O | 500 – 400 [19] | 412 & 433 |
| T-O-T & O-T-O | 1250 – 950 [19] | 988 |
| T-H | 2400 – 2100 [20] | 2360 |

### *3.3.3 SEM (scanning electron microscope)*

Characterization of zeolite with SEM is used to determine particle morphology (surface) and particle size. SEM results of zeolite synthesized from canned waste and waterglass, with various magnifications are presented in Figure 3-6.

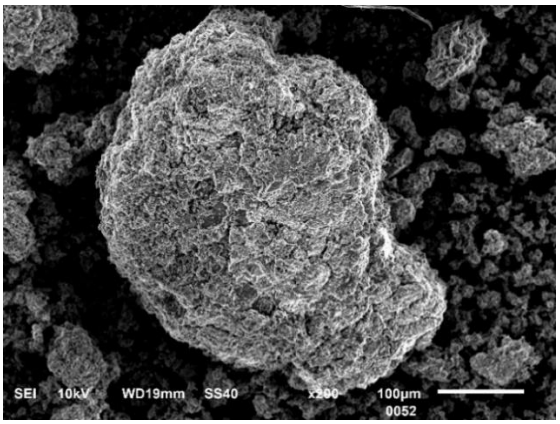


**Fig. 3.** Zeolite SEM Magnification 200x

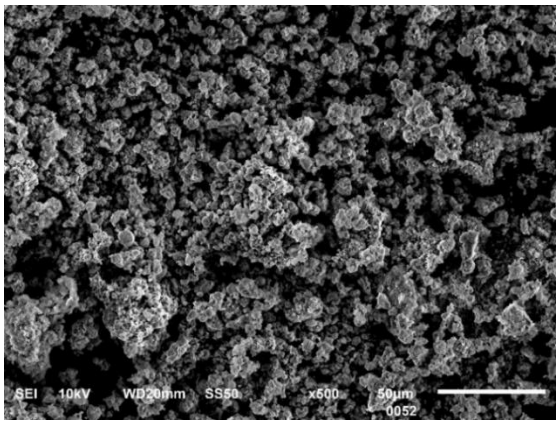


**Fig. 4.** Zeolite SEM Magnification 500x

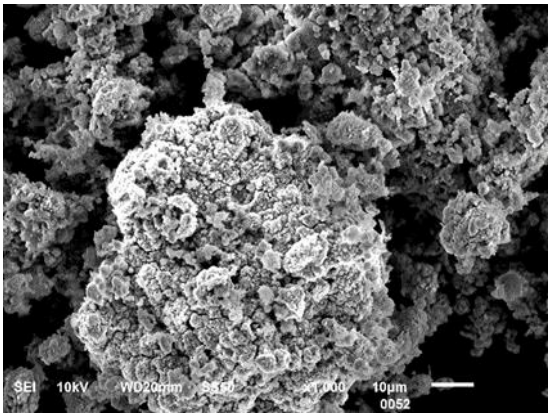


**Fig. 5.** Zeolite SEM Magnification 1000x

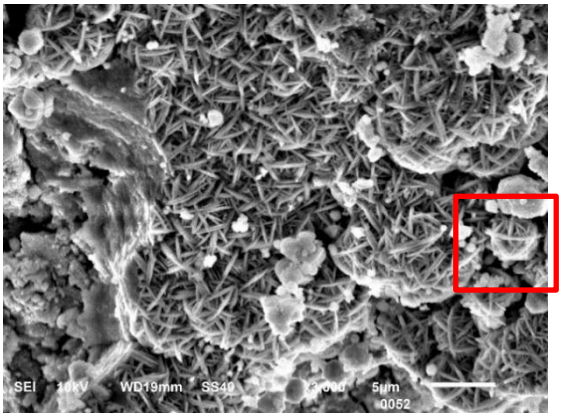


**Fig. 6.** Zeolite SEM Magnification 3000x

SEM analysis at 200 – 1000 x magnification shows that there are scattered lumps, and the surface looks rough and porous. At a magnification of 3000 x, it can be seen that the shape of crystals is round and elongated like fiber and it piled to form a hexagonal polytype that looks as faujasite zeolite with various sizes. Faujasite zeolite type (FAU) is a mineral group in the zeolite family of silicate minerals, an example of zeolite that includes FAU zeolite is zeolite X. FAU zeolite is one of the important zeolites and can be an ideal candidate for dye removal [21]. Thus, it can be known that zeolite crystallization has been formed and can be used as an adsorbent.

### *3.4 Adsorption Optimization of Congo Red and Naphthol Blue Black Dyes*

After the zeolite has been characterized, the performance of zeolite was tested in the Congo red and Naphthol blue black dyes. The maximum wavelength obtained for Congo red compounds is 499.88 nm and Naphthol blue black is 618.01 nm. Standard curves of Congo red and Naphthol blue black are carried out at 10, 20, 30, 40, and 50 ppm, the linearity obtained with a $r^2$ value of Congo red 0.9986 and Naphthol blue black 0.9989. The value of the variation in adsorption capacity for both dyes can be seen in Tables 4 and 5.

**Table 4**
Congo Red Adsorption Capacity on Various Optimization Variations

| Zeolite Weight (gram/10 mL) | Contact Time | pH | Dye Concentration | Adsorption Capacity |
|---|---|---|---|---|
| 0,1 | 30 minutes | 7 | 20 mg/L | 0,0399 mg/g |
| 0,2 | 30 minutes | 7 | 20 mg/L | 0,0297 mg/g |
| 0,3 | 30 minutes | 7 | 20 mg/L | 0,0312 mg/g |
| 0,4 | 30 minutes | 7 | 20 mg/L | 0,0551 mg/g |
| 0,5 | 30 minutes | 7 | 20 mg/L | 0,0390 mg/g |
| 0,4 | 60 minutes | 7 | 20 mg/L | 0,0511 mg/g |
| 0,4 | 90 minutes | 7 | 20 mg/L | 0,0458 mg/g |
| 0,4 | 120 minutes | 7 | 20 mg/L | 0,0427 mg/g |
| 0,4 | 30 minutes | 5 | 20 mg/L | 0,0617 mg/g |
| 0,4 | 30 minutes | 9 | 20 mg/L | 0,0580 mg/g |
| 0,4 | 30 minutes | 5 | 10 mg/L | 0,0262 mg/g |
| 0,4 | 30 minutes | 5 | 30 mg/L | 0,0849 mg/g |
| 0,4 | 30 minutes | 5 | 40 mg/L | 0,0994 mg/g |

**Table 5**
Naphthol Blue Black Adsorption Capacity on Various Optimization Variations

| Zeolite Weight (gram/10 mL) | Contact Time | pH | Dye Concentration | Adsorption Capacity |
|---|---|---|---|---|
| 0,1 | 30 minutes | 7 | 20 mg/L | 0,1991 mg/g |
| 0,2 | 30 minutes | 7 | 20 mg/L | 0,1085 mg/g |
| 0,3 | 30 minutes | 7 | 20 mg/L | 0,0969 mg/g |
| 0,4 | 30 minutes | 7 | 20 mg/L | 0,0713 mg/g |
| 0,5 | 30 minutes | 7 | 20 mg/L | 0,0547 mg/g |
| 0,1 | 60 minutes | 7 | 20 mg/L | 0,1876 mg/g |
| 0,1 | 90 minutes | 7 | 20 mg/L | 0,1708 mg/g |
| 0,1 | 120 minutes | 7 | 20 mg/L | 0,1831 mg/g |
| 0,1 | 30 minutes | 5 | 20 mg/L | 0,1615 mg/g |
| 0,1 | 30 minutes | 9 | 20 mg/L | 0,1767 mg/g |
| 0,1 | 30 minutes | 7 | 10 mg/L | 0,0885 mg/g |
| 0,1 | 30 minutes | 7 | 30 mg/L | 0,2729 mg/g |
| 0,1 | 30 minutes | 7 | 40 mg/L | 0,3028 mg/g |

In the first condition, which is varying the weight of zeolite to determine the optimum condition, the weight of zeolite used ranges from 0.1 to 0.5 grams. The optimum weight of zeolite for the absorption process of Congo red dye is at 0.4 grams with an adsorption capacity of 0.0551 mg/g and Naphthol blue black at 0.1 grams with an adsorption capacity of 0.1991 mg/g. Zeolite adsorbents have active sides, when all the active sides of the adsorbent have been filled by the adsorbate, the highest adsorption capacity can be obtained. The more adsorbent used, the wider the surface area will be, so the possibility of adsorption will be greater. However, when the optimum weight is

obtained, there will be a decrease in the levels of trapped dye, because not all sides are actively filled by adsorbate. The active side in large numbers takes a longer time to reach an equilibrium state [22]. Therefore, in this study, samples with high adsorbent weights did not produce optimal adsorption capacity.

The determination of the optimum contact time is used to determine the time required to reach dye adsorption equilibrium by zeolite. Generally, the longer the contact time, the more adsorbate it absorbs. The optimum adsorbate contact time on Congo red and Naphthol blue black was both at 30 minutes, then decreased slightly afterwards. At 30 minutes, the active side contained in the surface of the zeolite has been fully filled by a number of adsorbates, therefore the increase in contact time will only affect a slight increase in adsorption capacity and even tends to be constant. The longer the adsorption time, the adsorption stability effect will be disrupted so that the adsorption capacity after the optimum contact time can decrease [23].

In determining the optimum pH, changes in pH can affect the chemical and surface properties of the adsorbent, ions competing in the absorption process, and the solubility of the adsorbate [24]. pH variations are carried out at pH 5, 7, and 9. The optimum pH was obtained for Congo red dye at pH 5 and for Naphthol blue black dye at pH 7. pH variations at 5, 7 and 9 do not provide significant changes in adsorption capacity.

Figure 7 below shows a diagram of determining the optimum concentration with various variations in dye concentration.

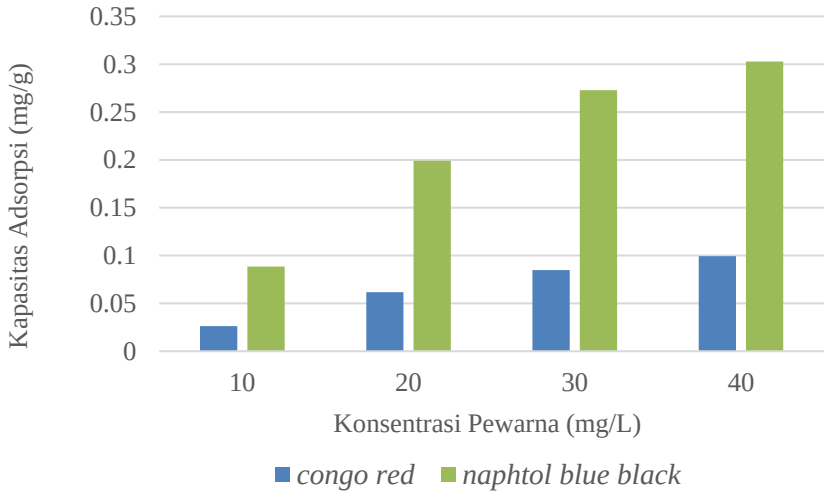


**Fig. 7.** Diagram of Determination of Optimum Dye Concentration in Adsorption

A dye concentration of 40 mg/L has the greatest adsorption capacity. An increase in the concentration of Congo red dye and Naphthol blue black is followed by an increase in the amount of substance adsorbed (adsorption capacity). The higher the adsorbate concentration will increase its adsorption capacity. This happens because the increase in adsorbate concentration will increase competing and movement of cations towards the active site on the surface of the adsorbent so that the absorption process will be faster [25]. The adsorption mechanism consists of three stages, namely external diffusion (diffusion of the adsorbate to the outer surface of the adsorbent), intraparticle diffusion (diffusion into the pores of the adsorbent), and physical or chemical adsorption at the active site [25]. The positive charge on the dye will interact on the surface of the negatively charged zeolite so that the intensity of the color will decrease. The bond between the zeolite adsorbent and the dye adsorbate that occurs during the reaction is a Van der Waals bond. This bond is defined as the force of attraction between molecules due to dipole-dipole attraction. Dipolar molecules tend to combine

with closes molecules until the negative pole of one molecule approaches the positive pole of another. This zeolite contains a certain amount of negative charge and positive charge [26].

### *3.5 Determination of Dye Adsorption Isotherms*

Determination of adsorption isotherms are using two models, namely Langmuir and Freundlich. Langmuir isotherms are based on monolayer adsorption on the active side of a homogeneous adsorbent, while Freundlich isotherms describe adsorption on heterogeneous surfaces. The isotherm values are presented in Table 6.

**Table 6**
Data Isoterm Langmuir dan Isoterm Friendly

| Parameter | | Congo Red | Naphthol Blue Black |
|---|---|---|---|
| Langmuir | $q_{max}$ (mg/g) | 357,1429 | 131,5789 |
| | $K_L$ | 0,7568 | 0,1934 |
| | $R^2$ | 0,9825 | 0,9361 |
| Freundlich | $K_f$ (mg/g) | 321,2921 | 77,5354 |
| | n | 1,01833 | 1,1095 |
| | $R^2$ | 0,9929 | 0,9636 |

In the adsorption process, both dyes show a greater correlation value of Freundlich isotherm than Langmuir isotherm. This shows that the adsorption process that occurs is multilayer adsorption (heterogeneous). The Freundlich isotherm equation shows that adsorbents have a heterogeneous surface and each adsorbent molecule has a different absorption potential. The Freundlich equation can also show adsorption processes that are reversible or irreversible, and it does not rule out the possibility that a monolayer adsorption process will form. The adsorption process of dyes dominated by Freundlich isotherm, indicates that the interactions that occur between the adsorbate and the adsorbent are dominated by physical interactions with weak bonds and only involve Van der Waals interactions [27,28].

## 4. Conclusions

Zeolite is synthesized from waste cans as a source of Al and waterglass (sodium silicate) as a source of Si. The synthesis was carried out using the sol-gel method followed by the hydrothermal method, and the yield of the zeolite formation reaction was 92.67%.

Zeolite was successfully synthesized, with XRD characterization results showing a mixed diffraction pattern of zeolite-X and zeolite-NaP1 with an average crystal size of 24.08 nm. The purity of the synthesis results for zeolite X is 55.44% and zeolite Na-P1 is 44.53%. FTIR characterization shows the intensity of functional groups T-O, T-O-T or O-T-O, and T-H, where T is Si or Al. Characterization with SEM, synthetic zeolite has the same morphology as Faujasite zeolite.

Synthetic zeolite can act as an adsorbent of water-soluble dye Congo red and water-insoluble dye Naphthol blue black. The maximum adsorption capacity of zeolite in absorbing Congo red reached 321.2921 mg/g and Naphthol blue black reached 77.5354 mg/g based on Freundlich isotherms. The optimum state of absorption of Congo red and Naphthol blue black occurs at weights of 0.4 g and 0.1 g, the contact time of both is at 30 minutes, the optimum pH is at 5 for Congo red and 7 for Naphthol blue black and 40 mg/L initial concentrations for both dyes.

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