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Effectiveness of Activated Zeolite in Reducing Iron (Fe) and Zink (Zn) Metal Levels in Borehole Water

Zul Alfian*, Ronaldo Sensini Siregar
Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sumatera Utara, Medan, 20155, Indonesia
*Corresponding Author: zul20@usu.ac.id

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ABSTRACT

Research has been carried out on the adsorption of iron (Fe) and zinc (Zn) metal levels using activated zeolite with acid and base in healthy water. Characterization was carried out on activated zeolite using X-ray Fluorescence (XRF). Samples were taken from borehole water from Puji Mulyo area, Deli Serdang, and preserved with HNO_3 until pH < 2, then digested. Determined the concentration of iron (Fe) and zinc (Zn) before and after the addition of activated zeolite by NaOH and HCl using an Atomic Absorption Spectrophotometer (AAS) with length-specific waves are 248.3 nm for iron (Fe) and 213.9 nm for zinc (Zn) metal. The results showed that the concentration of iron (Fe) and zinc (Zn) in the borehole water after the addition of activated zeolite decreased. Zeolite activated by NaOH absorbs iron (Fe) metal by 68.60% and absorbs zinc (Zn) metals by 37.91%. Meanwhile, Zeolite activated by HCl absorbs iron (Fe) by 42.37% and absorbs zinc (Zn) metal by 15.17%. This research shows that the effectiveness of percentage (%) decreasing the concentration of iron (Fe) and zinc (Zn) metal using activated zeolite with NaOH was more significant and more effective than activated zeolite with HCl.

Keywords: Activated Zeolite, Adsorption, Atomic Absorption Spectrophotometer, Borehole Water, X-Ray Fluorescence

ABSTRAK

Telah dilakukan penelitian tentang adsorpsi kadar logam Besi (Fe) dan Zink (Zn) dengan menggunakan zeolit aktivasi dengan asam dan basa pada air sumur bor. Karakterisasi yang dilakukan pada zeolit teraktivasi menggunakan *X-Ray Fluorescence* (XRF). Sampel diambil dari air sumur bor warga di daerah Puji Mulyo, Deli Serdang dan diawetkan dengan HNO_3 sampai pH < 2 kemudian didestruksi. Ditentukan konsentrasi dari logam Besi (Fe) dan Zink (Zn) sebelum dan sesudah penambahan zeolit aktivasi oleh NaOH dan HCl dengan menggunakan Spektrofotometer Serapan Atom (SSA) dengan panjang gelombang spesifik yaitu 248,3 nm untuk logam Besi (Fe) dan 213,9 nm untuk logam Zink (Zn). Hasil penelitian menunjukkan bahwa konsentrasi logam Besi (Fe) dan Zink (Zn) pada air sumur bor sesudah penambahan zeolit aktivasi mengalami penurunan. Zeolit aktivasi dengan NaOH menyerap logam Besi (Fe) sebesar 68,60% dan menyerap logam Zink (Zn) sebesar 37,91%. Sedangkan zeolit aktivasi dengan HCl menyerap logam Besi (Fe) sebesar 42,37% dan menyerap logam Zink (Zn) sebesar 15,17%. Pada penelitian ini menunjukkan bahwa efektivitas persentasi (%) penurunan konsentrasi logam Besi (Fe) dan Zink (Zn) dengan menggunakan zeolit aktivasi dengan NaOH lebih besar dan lebih efektif daripada zeolit aktivasi dengan HCl.

Kata Kunci: Adsorpsi, Air Sumur Bor, Spektrofotometer Serapan Atom, *X-Ray Flourescence*, Zeolit Aktivasi

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1. Introduction

Water is a natural resource that is very important for life and is often used by living things, especially humans. Other compounds cannot replace the function of water. Therefore, water resources must be protected to be appropriately still utilized. Water utilization for various purposes must be done wisely by considering the interests of present and future generations. The aspect of saving and preserving water resources must be instilled in all water users [1].

Water has a variety of uses in household interests, especially to meet the needs of drinking water, which must meet the requirements set by the Government of the Republic of Indonesia. Drinking water requires strict requirements because it is directly related to the body's biological processes that determine the quality of human life. More than 70% of the human body consists of water, and more than 90% of the body's biochemical processes require water as the medium. If human drinking water is of poor quality, it will disrupt the body's biochemical processes and result in functional disorders [2].

Water, the source of human life, is very easily contaminated, which can cause toxicity in humans. Water that contains heavy metals that exceed the threshold value can be toxic to the human body and other living things when consumed. Excessive mineral content will also endanger human health. Sources of water pollution by metals mostly come from mining, metal smelting, and other industries. They can also come from domestic waste that uses metals and agricultural land that uses fertilizers containing metals [3]

Water pollution can be caused by contamination of human activity wastes from households, industrial waste, agriculture, etc. In Deli Serdang Regency, precisely on the Medan–Binjai Km 12 road, Puji Mulyo Village, Sunggal District, Deli Serdang Regency, many industries produce various kinds of products such as plastic factories, kitchen utensils, coffee and so on. Waste from factories in the form of liquids is not entirely appropriately managed, resulting in pollution. In addition to industry, there are also agricultural areas that have the potential as a source of water pollution, both from using fertilizers and pest control and plants. Waste from industry and agriculture that seeps into the ground will eventually contaminate groundwater. At the same time, people in Puji Mulyo village have not received clean water from PDAM (Regional Drinking Water Company). Hence, they use boreholes as a source of clean water, and some use borehole water as drinking water. Water used for daily living needs must meet the requirements stipulated in the Regulation of the Minister of Health of the Republic of Indonesia Number 32 of 2017 concerning Environmental Health Quality Standards and Water Health Requirements for Sanitary Hygiene Purposes, namely, clean water is water that is odorless, tasteless and colorless.

Based on initial observations, the boreholes of residents in the area have a cloudy water color and smell. Metals that are usually found in borehole water are Iron (Fe), Zinc (Zn), Copper (Cu), Lead (Pb), and Chromium (Cr) and other metals and these metals are very harmful to the human body if in excessive levels and can endanger human health. One way to treat contaminated water is by adsorbing metal ions into an adsorbent to reduce metal levels. The types of adsorption media commonly used include zeolite, activated carbon, bentonite, and sawdust. Of the four types of adsorbents, zeolite is most often used as a medium for processing factory waste [4].

Zeolite is a porous material with several dominant mineral contents (SiO_4 and AlO_4). Activating strong acid or robust base solutions can increase its adsorption capacity. Zeolite has a very regular crystal shape with interconnected cavities in all directions, which causes the surface area of zeolite to be very large [5]. The activity of natural zeolites tends to be low because there are still many impurities. Therefore, activation is necessary. According to the research results of [6], the activation process can improve some physical and chemical properties of zeolites, such as surface acidity and porosity, so that they are more effective as adsorbents. Increasing the usability of zeolite as an adsorbent can be done through physical and chemical activation [7]

The physical activation process is done by heating (calcination) to evaporate the water trapped in the pores of zeolite crystals so that the number of pores and specific surface area increases. Chemical activation can be done using a solution of hydrochloric acid or sulfuric acid and a base that aims to clean the pore surface, remove interfering compounds, and reorganize the location of interchangeable atoms [8].

The determination and reduction of metal levels is based on the Regulation of the Minister of Health of the Republic of Indonesia No.32 of 2017 concerning Environmental Health Quality Standards for Water Media for Sanitary Hygiene Purposes where the allowable levels are 1 mg/L for Iron (Fe) metal and 1 mg/L for Zinc (Zn) metal.

2. Materials and Methods

The tools used in this study include beaker glass, measuring cup, glass funnel, erlenmeyer, measuring flask, drop pipette, volume pipette, rubber ball, label paper, sample container, black isolation, pestle and mortar, 200 mesh sieve, stirring rod, spatula, porcelain cup, oven, muffle furnace, analytical balance, filter paper, hot plate stirrer, magnetic bar, universal indicator, desiccator, aquadest bottle, magnetic bar, Atomic Absorption Spectrophotometer (SSA), X-Ray Flourencence (XRF). The materials used in this study include borehole water samples, commercial zeolite, HNO_3 65%, HCl 1N, NaOH 1N, distilled water, Fe standard solution 1000 mg/L, and Zn standard solution 1000 mg/L.

2.1. Sample Collection and Preservation Method

Samples were taken from boreholes around the industrial and agricultural areas of Puji Mulyo Village. Sampling points are taken from 5 residents' houses with a distance of +10-50 meters, where sampling is done through the tap where the water comes out according to (SNI 6989.57: 2008) with a borehole depth of about 10-25 meters, which is then taken and mixed into one container. The sample water is then taken using a 600 mL plastic bottle, rinsed with distilled water until dry, and then rinsed with sample water. Then, the water was taken as much as 600 mL and preserved by adding 65% of HNO_3 until pH < 2, then tightly closed and covered with black insulation until the sample container was covered entirely.

2.2. Preparation and Activation of Commercial Zeolite

First, 25 g each of natural zeolite sieve size 200 Mesh was mixed with 250 mL of NaOH and HCl with a concentration of 1N. 0Then allowed to stand for 24 hours, then the mixture was heated while stirring for 2 hours at a temperature of 70°C. Then, the mixture was cooled, and the cooled mixture was filtered precipitate with Whatman filter paper No.42, after which the residue was washed with distilled water until pH 6-7 (neutral). Then, the residue was dried in an oven at 105ºC to remove the water content. After that, it was annealed at 300ºC to 400ºC for one hour. Then, the activated zeolite will be characterized by X-ray fluorescence to determine the content of elements in zeolite.

2.3. Sample Preparation (SNI 6989.4: 2009)

The test sample was homogenized with a pipet of 50 mL of the test sample into 100 mL beaker glass. Add 5 mL of HNO_3, and cover the beaker with a watch glass. Heated with a hotplate until the remaining volume is 15- 20 mL. If the destruction is not perfect (unclear), add another 5 mL of HNO_3, then cover with watch glass and heat again (not boiling). Repeat until all metals dissolve and the test sample becomes clear. Rinse the watch glass and put the rinse water into a beaker glass. Transfer the test sample into a 50 mL volumetric flask, filter, and add mineral-free water to the upper limit of the flask and homogenize.

2.4. Heavy Metal Adsorption by Static Method (batch)

A total of 5 grams of activated zeolite using NaOH was put into a glass beaker. Added 100 mL of borehole water sample, then allowed to stand for 3 hours and filtered using Whatmann filter paper No. 42. The resulting filtrate was re-analyzed using an Atomic Absorption Spectrophotometer (AAS) to see the percentage (%) decrease. The same was done for HCl-activated zeolite on borehole water samples.

3. Results and Discussion

3.1. Zeolite Compound Data Before and After Activation

From the results of the research conducted obtained in the two tables, it can be seen that the content of zeolite composition before activation is Si/Al (Si = 61.584) (Al = 11.863) and the content of zeolite composition after activation with NaOH is Si/Al (Si = 48.859) (Al = 7.910) and the content of zeolite composition after activation with HCL is Si/Al (Si = 52.035) (Al = 7.651). Where zeolite activation is carried out to increase the adsorption power of zeolites, activation with NaOH is to remove impurities in zeolites such as Na +, K +, Ca^{2+}, Mg^{2+} ions, and other metals where ion exchange occurs with NaOH so that it can bind and remove impurities in zeolites. At the same time, activation with HCL will donate protons into zeolites. So that zeolites experience dealumination and decationization, namely the release of Al and cations in the zeolite framework so that the alumina content is reduced and replaced with protons. The decationization process causes an increase in the surface area of the zeolite due to the reduction of impurities that cover the pores of the zeolite. As a result, the ratio of silica and alumina is large, so the adsorption power is greater.

Table 1. Zeolite Compound Data Before and After Activation with NaOH

Before NaOH Activation	Element	After NaOH Activation
11.863	Al	7.910
61.584	Si	48.859
1.58	P	0.52
0.27	S	0.118
2.639	Ca	2.551
1.794	Fe	1.087
0.005	Zn	0.004
1.68	Sr	1.54
0.009	Zr	0.0005
1.574	Na	1.042
0.048	Ba	0.058
0.008	Ga	0.00
0.004	Ni	0.002
0.064	Mn	0.037
0.129	Ti	0.116
3.096	K	2.489
0.691	Mg	0.301
0.003	Cu	0.003

Table 2. Zeolite Compound Data Before and After Activation with HCl

Before HCl Activation	Element	After HCl Activation
11.863	Al	7.651
61.584	Si	52.035
1.58	P	1.58
0.27	S	0.109
2.639	Ca	1.974
1.794	Fe	0.854
0.005	Zn	0.003
1.68	Sr	1.748
0,009	Zr	0.0003
1.574	Na	0.494
0.048	Ba	0.052
0.008	Ga	0.001
0.004	Ni	0.002
0.064	Mn	0.019
0.129	Ti	0.113
3.096	K	2.466
0.691	Mg	0.196
0.003	Cu	0.003

3.2. Effect of Addition of Activated Zeolite Using NaOH and HCL on Percentage (%) Decrease in Iron (Fe) and Zinc (Zn) Metal Concentration

Determination of iron (Fe) and zinc (Zn) metal content in borehole water samples before and after the addition of zeolite activation with NaOH and HCl was carried out by measuring the absorbance value and concentration using an Atomic Absorption Spectrophotometer (SSA) with a specific wavelength.

From the results obtained, the percentage (%) decrease in the concentration of iron (Fe) and zinc (Zn) using NaOH-activated zeolite is greater and more effective than HCl Activated Zeolite. From the results obtained, the percentage (%) decrease in the concentration of Iron (Fe) and Zinc (Zn) metals by using zeolite activated by NaOH is greater and more effective than zeolite activated by HCl. Iron (Fe) metal in borehole water before zeolite was added with a concentration of 1.685 mg/L. After adding NaOH-activated zeolite, the concentration was reduced to 0.529 mg/L; after adding HCl-activated zeolite, the concentration was reduced to 0.971 mg/L. In other words, the percentage (%) decrease in Iron (Fe) metal concentration is 68.60% for NaOH-activated zeolite and 42.37% for HCl-activated zeolite [9].

The results of the percentage (%) decrease in the concentration of Iron (Fe) metal in borehole water samples after the addition of activated zeolite using NaOH and HCl when plotted into chart form can be seen in Figure 1.

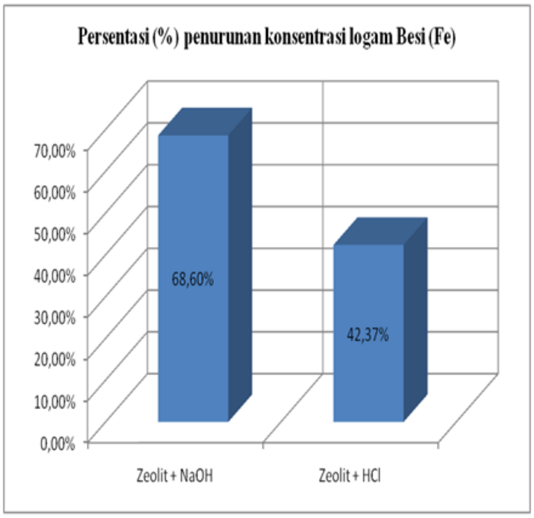

Figure 1. Percentage (%) Reduction of Iron (Fe) Metal Concentration after the Addition of Activated NaOH and HCl

For Zink (Zn) metal in borehole water before the addition of zeolite had a concentration of 0.3434 mg/L. After the addition of NaOH-activated zeolite, the concentration was reduced to 0.2132 mg/L, and after the addition of HCl-activated zeolite, the concentration was reduced to 0.2913 mg/L. In other words, the percentage (%) decrease in Zink (Zn) metal concentration is 37.91% for NaOH-activated zeolite and 15.17% for HCl-activated zeolite [9]. The percentage (%) decrease in Zink (Zn) metal concentration in borehole water samples after the addition of NaOH and HCl-activated zeolite when plotted into a chart can be seen in Figure 2.

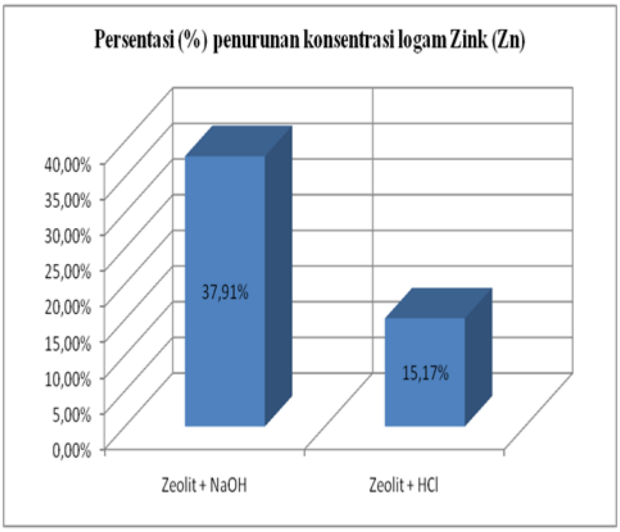

Figure 2. Percentage (%) Reduction of Zinc (Zn) Metal Concentration after Addition of Activated by NaOH and HCl

The chart shows the percentage (%) decrease in the concentration of Iron (Fe) and Zinc (Zn) metals using NaOH-activated zeolite has a percentage (%) decrease in concentration and effectiveness that is quite large compared to HCl-activated zeolite. Borehole water samples have Iron (Fe) metal content that exceeds the threshold value of quality standard water based on PerMenKes RI No.32 of 2017 concerning Chemical Parameters in Health Quality Standards Environment for Water Media for Sanitary Hygiene Purposes before being reduced by using NaOH and HCl activated zeolites. After adsorption using zeolite activation, the Iron (Fe) metal concentration decreased to 0.529 mg/L for NaOH-activated zeolite and 0.971 mg/L for HCl-activated zeolite. It can be seen in the chart from the results obtained that Iron (Fe) metal is mostly absorbed by adsorbents, namely zeolites activated using NaOH, with the effectiveness of percentage (%) decrease reaching 68.60%. Zinc (Zn) metal content in borehole water samples did not exceed the threshold value of quality standard water based on PerMenKes RI No.32 of 2017 concerning Chemical Parameters in Environmental Health Quality Standards for Water Media for Sanitary Hygiene Purposes before decreasing using NaOH and HCl activated zeolite. However, a decrease in Zn concentration was still carried out using NaOH and HCl-activated zeolites with the results of a reduction of the concentration of Zinc (Zn) metal to 0.2132 mg/L for NaOH-activated zeolites and 0.2913 for HCl activated zeolites. It can be seen in the chart from the results obtained that a lot of Zinc (Zn) metal is absorbed by the adsorbent on the NaOH activation zeolite with the effectiveness of the percentage (%) decrease reaching 37.91%. In the activation process with NaOH, there will be a process of dissolving silica, which is one of the components in the zeolite framework [10].

The dissolution of silica will cause changes in the zeolite structure and the reduction of silica in the zeolite framework so that the Si Al ratio decreases. A decrease in this ratio will increase zeolites' adsorption capacity and selectivity to polar molecules [11]. The absorption process can occur when the surface of the solid on the adsorbate molecules (substances to be absorbed) forms a solid surface so that some will stick to the surface of the absorbed solid. At first, the adsorption rate is quite large because the entire surface is still empty. But after the contact time gets longer, the surface filled by molecules is getting more and more, and the area of the empty area decreases, so the adsorption rate decreases [12].

4. Conclusion

According to the data from this study, iron (Fe) levels in borehole water samples have exceeded the threshold value of quality standard water based on PerMenKes RI No.32 of 2017 concerning Chemical Parameters in Environmental Health Quality Standards for Water Media for Sanitary Hygiene Purposes, where the threshold value for Fe is 0.3434 mg/L and zinc (Zn) levels have also exceeded the threshold value. Regarding Zink (Zn), the threshold value is not exceeded because the maximum level permitted for Zn is 15 mg/L. Iron (Fe) metal concentration was reduced following adsorption utilizing zeolite activation to comply with PerMenKes RI No.32 of 2017, specifically to 0.529 mg/L for NaOH-activated zeolite and 0.971 mg/L for HCl-activated zeolite. NaOH has activated the zeolite at 0.2132 mg/L for zinc (Zn) metal, whereas the zeolite that HCl has activated is 0.2913 mg/L. Iron (Fe) metal was absorbed by NaOH-activated zeolite by 68.60%, and zinc (Zn) metal was absorbed by 37.91%. In contrast, the ability of HCl-activated zeolite to absorb iron (Fe) and zinc (Zn) metals is 42.37% and 15.17%, respectively. These results suggest that zeolites activated with NaOH are more effective at removing metals from water.

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6. Conflict of Interest

Authors declare no conflicts of interest

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