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Utilization of Yogyakarta natural zeolites to reduce NH_4 and NO_2 levels in shrimp pond water and its kinetic rate study

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Abstract. Natural zeolite is an unique crystal that has a very small and uniform pore size when compared to other adsorbents, such as activated carbon and silica gel. This study aims to determine the potential of natural zeolite as an adsorbent, to absorb toxic gases in pond water, especially shrimp ponds and to study its kinetics rate of NH_4 and NO_2 adsorption. This study consisted of two stages: activated zeolite using NaOH, and tested the absorption power of NH_4 and NO_2 gases. Zeolite was activated using NaOH where 25 grams of zeolite were mixed with 0.5N NaOH and heated for 2 hours at 70°C, then stirred and dried in an oven at 110°C for 4 hours. Then zeolite was cooled in a desiccator. The results of NH_4 and NO_2 gas adsorption showed that natural zeolites can reduce NH_4 and NO_2 gas levels in shrimp pond water. The rate constant of pseudo-second-order, and the initial adsorption rate at any given time t, can be predicted from the equation which is full zeolite obtained the best perform of NH_4 and NO_2 adsorption.

1. Introduction

Development of cultivation systems from traditional to intensive with the majority shrimp ponds have potential towards increasing environmental pollution. Less optimal utilization of excessive feed will cause accumulation of organic matter. Decomposition of organic matter requires oxygen in the process, so that the availability of oxygen for the biota in it is reduced. If this happens continuously it will cause death for shrimp and other biota. Pollution materials that are difficult to decompose by microorganisms also cause hoarding and result in damage to the environment which will directly disrupt organisms that live in these environments. Organic pollution materials that function as fertilizers are actually detrimental due to algae blooms and aquatic plants causing oxygen competition in the waters. The above factors are the cause of the decline in the body's resistance to the attack of the disease because of the poor quality of the environment, if this is left continuously then mass death will occur so that the population will decline [1].

Traditional methods for nitrogen removal from wastewater are denitrification, nitrification, chemical coagulation, adsorption, selective ion exchange, ammonia stripping, electro dialysis, filtration, reverse osmosis [2, 3, 4, 5]. Removal processes of dissolved nitrogen compounds, can vary. For intensive RAS (Return Activated Sludge), the most common practice involves utilizing nitrifying bacteria to convert ammonia-nitrogen to nitrate-nitrogen [6]. The rate limiting step of this process is the oxidation of ammonia [7]. Incomplete nitrification occurs when a lack of NOB (Nitrite Oxidizing Bacteria) productivity is present, leading to increased concentrations of nitrite. The adsorption process



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is more widely used in industry because it has several advantages, which are more economical and also does not cause toxic side effects and is able to eliminate organic materials. Adsorption is a process that occurs when molecules from substances liquid or gas accumulates on a solid / liquid surface, forming a thin layer formed from molecules or atoms. The most important thing in the adsorption process is the selection of a good type of adsorbent [6].

The adsorption process of solutions theoretically generally lasts longer than the adsorption process on gas, steam or pure liquid [8]. This is due to the adsorption of the solution involving competition between the solution components and the adsorption site. The solution adsorption process can be estimated qualitatively from the polarity of the adsorbent and the constituent components of the solution. The tendency of polar adsorbents to more strongly absorb polar adsorbates than non-polar adsorbates, and vice versa. The solubility of the adsorbate in the solvent is the determining factor in the adsorption process, generally the hydrophilic substance is difficult to adsorb in a dilute solution. Basically, an adsorbent must have a high specific surface area, which has small diameter pores so that the adsorbate retention process by the adsorbent takes place more effectively. Specifically, the pore size also determines the adsorption of a particular compound in solution. If the adsorbent's pore size gets smaller, the adsorption capacity is greater, assuming that the adsorbed component can enter the porous cavity. The greater number of adsorbents will provide a larger surface area for the adsorbate to be desorbed. In addition, the more the amount of adsorbent will also provide greater opportunity for contact with the adsorbate molecules [9].

One of the most potential adsorbents is zeolite. Zeolite has several properties such as having dehydration properties, high cation exchange, a good catalyst, and as an agent for other compounds [10]. Increased efficiency or optimization of zeolites as adsorbents can be done through activation. The activation process aims to cleanse the pore surface, remove disturbing compounds and increase the specific surface area [11]. At the activation stage, the adsorbent is first soaked using an activating material, including HCl, HNO₃, H₂SO₄ and H₃PO₄ [12].

This study aims to improve the quality of water using natural zeolite as an adsorbent so that the levels of NH₃ and NO₂ contained in water can be reduced and safe for shrimp breeding and also to aims kinetics rate of NO₂ and NH₃ adsorption using zeolite catalyst. In this study, filters were made from PVC pipes with a pipe length of 100 cm and a diameter of 6 inches (15.24 cm). The results of this study are expected to reduce the level of Fe to 96% and Mn decreased to 84.3%.

2. Experimental

2.1 Zeolite Activation

Zeolite is activated using NaOH where 25 grams of zeolite are mixed with 0.5N NaOH and heated for 2 hours at 70°C while stirring and then dried in an oven at 110°C for 4 hours. The zeolite is then cooled in a desiccator. After drying, zeolite is ready for use. This study used 3 variations of zeolite, volume zeolite 1/3 part of pipe, volume zeolite 2/3 part of pipe and full volume of zeolite that were filled in the pipe of filter.

2.2 Nitrite Levels Analysis

Analysis of nitrite levels was carried out using Diazotasi-Spectrofotometry method. In this study, samples with sulfanilic acid and N- (1-Nepthyl ethylene diamin) dihydrachloride in an acidic atmosphere (pH 2 - 2.5) formed a complex compound, then placed on a spectrophotometer to be analyzed at a wavelength of 420 nm.

2.3 Ammonium Levels Analysis

Analysis of ammonium levels was carried out by the Nessler-Spectrophotometry method. In this analysis, the sample in an alkaline atmosphere is reacted with Nessler reactants to form complex compounds that are yellow to brown. The intensity of the color that occurs is measured its absorbance at a wavelength of 420 nm.

3. Results and Discussion

3.1. NH_4 and NO_2 decreasing results

Figure 1 shows that the NH_4 concentration in shrimp pond water decreased from 300 minutes in all the amount of zeolite used. In the use of 25 grams a decrease in NH_4 concentration from 4.276 ppm to 3.879 ppm or around 9.284%. On the use of 50 grams and 100 grams there was a decrease in NH_4 concentration of around 15.084% and 18.569% respectively. This shows that the more amount of zeolite used, the greater the amount of NH_4 gas adsorbed.

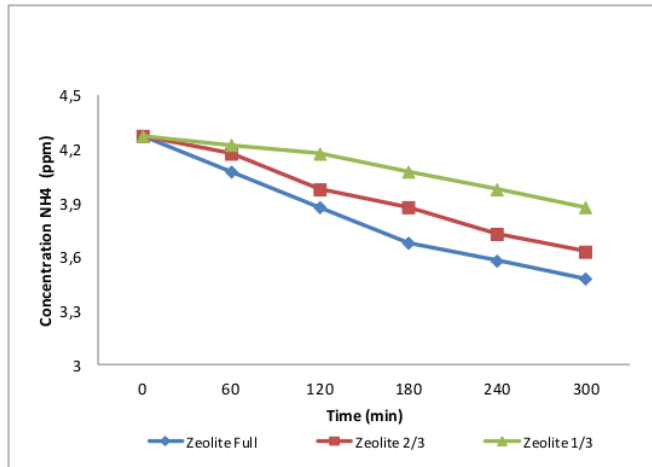


Figure 1. Decrease in NH_4 concentration with different amounts of zeolite

While Figure 2 shows that the NO_2 concentration in shrimp pond water decreased from 300 minutes in all the amount of zeolite used. In the use of 25 grams a decrease in NO_2 concentration from 0.457 ppm to 0.354 ppm or around 22.538%. On the use of 50 grams and 100 grams there was a decrease in NO_2 concentration of around 33.917% and 40.700% respectively. This shows that the more amount of zeolite used, the greater the amount of NO_2 gas adsorbed.

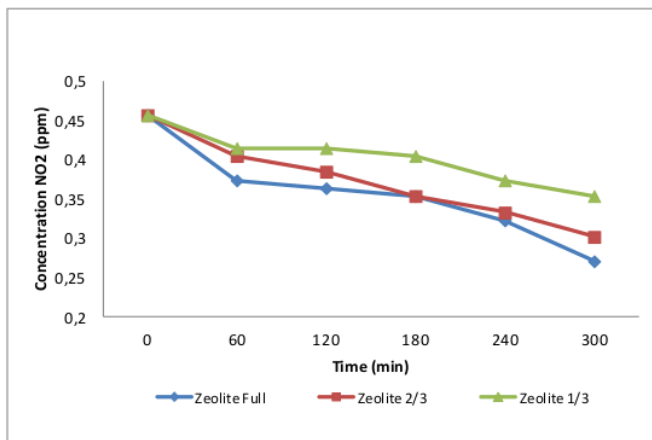


Figure 2. Decrease in NO_2 concentration with different amounts of zeolite

3..2. Kinetics rate of NH₄ and NO₂ adsorption

There are many mathematical models are reported in the literature, all attempting to describe quantitatively in the kinetic behaviour during the adsorption process. Among these models, the pseudo-first-order, pseudo-second-order and intraparticle diffusion model were used to test kinetic experimental data.

3.2.1. Pseudo-second-order model

The rate constant of pseudo-second-order adsorption is determined from the following equation given by Lagergren [14, 15]:

$$\frac{1}{C_A} - \frac{1}{C_{A0}} = k t \tag{1}$$

where, C_A and C_{A0} are the amounts of NH₄ and NO₂ adsorbed (mg/g) at equilibrium and at time t (min), respectively, and k (min⁻¹) is the rate constant of the pseudo-second-order adsorption.

Results of k value for NH₄ adsorption can be shown in table 1 below.

Table 1. Adsorption kinetic parameters of NH₄

Zeolite	NH ₄ before	NH ₄ after	K	R ²
Full	4.276	3.435	0.000188	0.98717
2/3	4.276	3.581	0.000138	0.99194
1/3	4.276	3.824	0.000072	0.98273

Results of k value for NO₂ adsorption can be shown in table 1 below.

Table 2. Adsorption kinetic parameters of NH₄

Zeolite	NO ₂ before	NO ₂ after	K	R ²
Full	0.457	0.222	0.004441	0.89637
2/3	0.457	0.250	0.003601	0.98858
1/3	0.457	0.297	0.002010	0.98002

Table 1 and table 2 showed that the utilization of full zeolite obtained the higher adsorption of NH₄ and NO₂ gases. It means that the more zeolites are used, the more NH₄ and NO₂ gases are adsorbed. It is because of the activated zeolite has a surface area and acidity that is easily modified so it can absorb more NH₄ and NO₂ gases because the framework of the zeolite has been substituted with ions from the gas.

4. Conclusion

Full zeolite utilization obtained the best perform of NH₄ and NO₂ adsorption, respectively gave a decrease in the concentration of 18.569% NH₄ and 40% NO₂. Adsorption kinetic data were analysed using the pseudo-second-order model. The advantage of using pseudo-second-order model is that the adsorption capacity, the rate constant of pseudo-second-order, and the initial adsorption rate at any given time t, can be predicted from the equation that respectively obtained 0.000188 and 0.004441 of k value for NH₄ and NO₂.

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