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Application of activated carbon/TiO_2 composite for ammonia degradation from leaching process in methyl iso cyanate production

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A B S T R A C T

The wastewater in the washing process in Methyl Iso Cyanate production line contains ammonia which causes air pollution. Therefore, the ammonia waste processing system is needed that can degrade ammonia so that its concentration decreases and reduces the pollution. Photocatalytic degradation of ammonia is an attractive technology because of its easy operation, high efficiency, low cost, and low secondary pollutants. The photocatalytic process produces hydroxyl groups from the decomposition of water which can oxidize ammonia into nitrate and nitrogen gas. This research aims to process ammonia waste by combining adsorption and photocatalysis processes to obtain better performance in degrading ammonia. The research began with the preparation of activated carbon as an adsorbent and the synthesis of activated carbon-TiO_2 composite material. Activated carbon is heated at 500º C for 6 hours for the activation process. Composite material synthesis is carried out using the slurry method. Activated carbon performance testing was carried out at weight variations of 10, 20 and 30 gram. Measurement of ammonia concentration was carried out using the spectrophotometry method. Optimum results were obtained at an activated carbon weight of 30 gram with a reduction in ammonia concentration of 1.01%. TiO_2 as much as 5% of the weight of activated carbon (30 gram) is dissolved in ethanol and activated carbon is added to obtain a composite material. In the composite material performance test, a maximum reduction in ammonia concentration of 45% was obtained using the ultrasonic stirring method.

A B S T R A K

Air limbah pada proses pencucian di lini produksi Methyl Iso Cyanate mengandung amonia yang menyebabkan pencemaran udara. Oleh karena itu diperlukan suatu sistem pengolahan limbah amonia yang dapat mendegradasi amonia sehingga konsentrasinya menurun dan pencemarannya berkurang. Degradasi amonia secara fotokatalitik merupakan teknologi yang menarik karena pengoperasiannya yang mudah, efisiensi tinggi, biaya rendah, dan polutan sekunder yang rendah. Proses fotokatalitik menghasilkan gugus hidroksil dari hasil penguraian air yang dapat mengoksidasi amonia menjadi nitrat dan gas nitrogen. Penelitian ini bertujuan untuk mengolah limbah amonia dengan menggabungkan proses adsorpsi dan fotokatalisis untuk memperoleh kinerja yang lebih baik dalam mendegradasi amonia. Penelitian diawali dengan preparasi karbon aktif sebagai adsorben dan sintesis material komposit karbon aktif-TiO_2. Karbon aktif dipanaskan pada suhu 500º C selama 6 jam untuk proses aktivasi. Sintesis material komposit dilakukan dengan menggunakan metode slurry. Pengujian kinerja karbon aktif dilakukan pada variasi berat 10, 20 dan 30 gram. Pengukuran konsentrasi amonia dilakukan dengan menggunakan metode spektrofotometri. Hasil optimal diperoleh pada berat karbon aktif 30 gram dengan penurunan konsentrasi amonia sebesar 1,01%. TiO_2 sebanyak 5% dari berat karbon aktif (30 gram) dilarutkan dalam etanol dan ditambahkan karbon aktif sehingga diperoleh material komposit. Pada uji kinerja material komposit diperoleh penurunan konsentrasi amonia maksimum sebesar 45% dengan menggunakan metode pengadukan ultrasonik.

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1. Introduction

The development of industrial sectors in various fields has given rise to many environmental problems. One of the negative effects of these problems is the pollutant ammonia. The presence of ammonia in the environment can lead to eutrophication. As a result, the growth of aquatic plants could not be controlled and the balance of the ecosystem was disrupted [1]. Excessive ammonia concentration in the air can damage the human respiratory systems. Ammonia concentrations above 300 ppm can cause respiratory tract damage, skin and eye irritation, and death [2]. Therefore, it makes sense to find ways to treat ammonia waste, such as converting it into less harmful compounds.

The treatment of ammonia can be done by various methods such as nitrification and denitrification [3], ion exchange [4], membrane technology [5], and adsorption[6-8]. An effective, easy, and inexpensive way to treat ammonia waste is adsorption. Ammonia adsorption uses an adsorbent that adsorbs ammonia, which reduces its concentration in the solution. Several different adsorbents, namely activated carbon, zeolite, alumina, silica, and graphene oxides (GO), have been studied in the literature for ammonia adsorption [9]. However, these methods are inefficient, and some produce secondary pollutants [10]. Photocatalysts are also being developed as an alternative method.

Photocatalytic ammonia degradation is an attractive technology due to its easy operating system, high efficiency, low cost, and low secondary pollutants. The process of photocatalytic produces the hydroxyl group from water decomposition that can oxidizes ammonia to nitrate and nitrogen gas [11]. But this technique has imperfections, one of which is the limited power of semiconductor adsorption of the pollutant. TiO_2 limited adsorption capability to contaminants resulting in low photocatalytic activity efficiency in its application. It needs a technology that permits photocatalysts to breakdown pollutants at high reaction rates by absorbing more pollutants. The adsorption-photocatalyst combination is a promising technique. When TiO_2 and adsorbent are coupled, a synergistic effect occurs, causing the reaction rate to rise [12]. The efficiency of the process by which pollutants are degraded in semiconductor catalysts depends on how well the adsorbent performs in adsorbing pollutants and transferring them to the surface of the photocatalyst.

In this research, active carbon will be combined with TiO_2 to obtain a composite material that can degrade ammonia in wastewater from the washing process, the remaining product of the Methyl Iso Cyanate production process. The aim of this study is to test the performance of the activated carbon-TiO_2 composite material for degrading ammonia. The effectiveness of composite materials is measured based on the efficiency of removing ammonia and the capacity to remove the ammonia. The research began with the preparation and characterization of activated carbon, followed by testing its performance and testing with photocatalysis by paying attention to the variables of adsorbent mass weight, wastewater contact time with the adsorbent, and the stirring method.

2. Methodology

2.1. Activated Carbon Preparation

The activated carbon particles used are 5-7 mm in size. The activation of activated carbon is carried out through a calcination process. This process is carried out to open the pores of activated carbon, evaporate the crystal water contained in the active carbon structure and increase thermal stability. Calcination was carried out at 500°C for 6 hours.

2.2. Activated Carbon Performance Test

The next step is to test the performance of activated carbon that has been activated and without activation. Activated carbon is placed in a container weighing 10 grams, 20 grams and 30 grams, then mixed with the diluted sample, the process is carried out closed using aluminum foil/plastic shrink to maximize NH_3 absorption. Samples that have been in contact with activated carbon are taken based on variable time, with a syringe that has been modified with a filter, then the samples are analyzed using a spectrophotometer.

2.3. Synthesis of Activated Carbon-TiO₂ Composite Material

The synthesis was carried out after obtaining the most optimal results from the activated carbon performance test. TiO_2 as much as 5% of the weight of activated carbon is dissolved in ethanol and 30 gram of activated carbon is added. The mixture was heated at 80°C, then dried in an oven at 100°C. The composite material is cooled at room temperature.

2.4. Composite Material Performance Test

This step is carried out using a batch reactor. The inside of the reactor is coated with aluminum foil to maximize UV radiation in the reactor. The dimensions of this reactor are 43 cm long, 28 cm high, 24 cm wide, and with a steroform 1 cm thick. This reactor is equipped with a Magnetic Stirer and Ultrasonic so that there is no TiO_2 photocatalyst deposition at the bottom of the beaker.

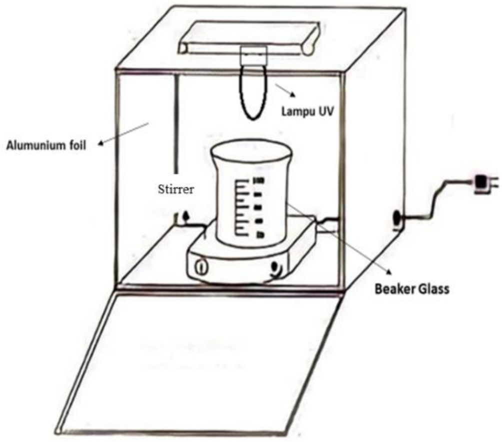

Figure 1. Schematic of photoreactor

This test will focus on the performance of the activated carbon-TiO_2 composite. NH_3 adsorption was analyzed with a UV Spectrophotometer with a wavelength of 640 nm. The pH value is measured using a pH meter. The performance of the composite material was tested by ammonia photodegradation using a photoreactor specifically designed for this research. It was conducted at PT XYZ using samples taken from wastewater samples from washing at the Methyl Iso Cyanate plant. Trials on the adsorption and degradation processes were carried out in the PT XYZ laboratory and the Chemical Engineering laboratory at Universitas Serang Raya.

3. Results and Discussion

3.1. The Activated Carbon Preparation Effects on Ammonia Adsorption

Several terms are used to make it easier to explain pictures and tables of research results. These terms are: A10 (activation with a loading of 10 gram), A20 (activation with a loading of 20 gram), and A30 (activation with a mass of 30 gram). The activated carbon used in this research is activated carbon with a surface area of 8-30 mesh. Adsorption of ammonia can be influenced by several factors, namely the surface area of the adsorbent, particle size, contact time, and pore size distribution. The greater the surface area of an adsorbent, the more adsorbate that can be absorbed, so that the adsorption process can be more effective. A longer contact time can enable the diffusion and contact process between activated carbon and ammonia to be more effective, so that the adsorption process will be more effective. Another factor is the pore distribution. The pore distribution will influence the size distribution of the adsorbate molecules that enter the adsorbent particles.

One of aims in this research is to find the optimum loading for ammonia adsorption. The activated carbon used is TA and A with loading of 10 gram. Preparation of activated carbon is conducted by heating the activated carbon in a furnace to open the pores and remove the water content in the activated carbon so that it can adsorb ammonia effectively [13]. The volume of ammonia used was 200 ml with an initial concentration of 1812 ppm. In Figure 2, activated carbon prepared by heating can adsorb more ammonia in 60 minutes than unprepared activated carbon, although the maximum adsorption capacity has the same value. This proves that the pores will open more, and the water content will decrease if the activated carbon is heated, making it easier to adsorb ammonia.

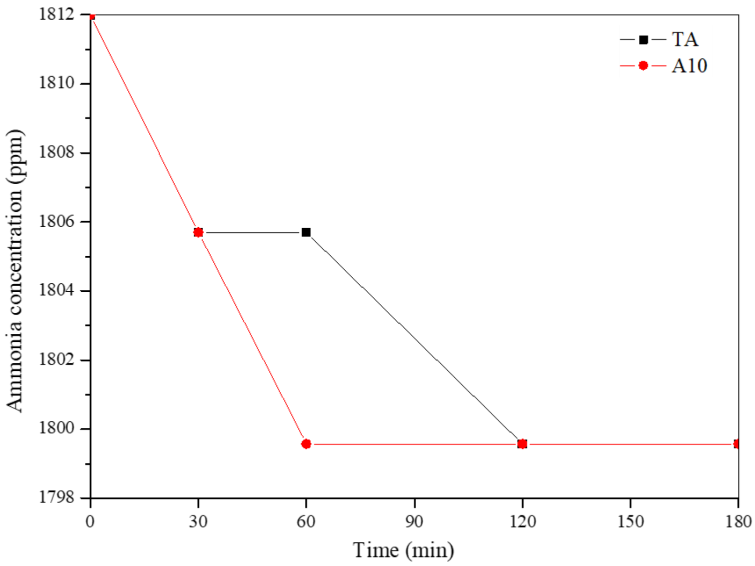

Figure 2. Effect of heating on activated carbon

Optimum conditions were also studied based on the amount of active carbon added to the waste as an ammonia adsorbent. Activated carbon loading varied from 10, 20, 30 gram. Figure 3 shows 30 gram of activated carbon can adsorb more ammonia than others. Even though the adsorption capacity only reaches 1.01%, this shows that the more activated carbon used, the more ammonia is absorbed. The small adsorption capacity is due to the high concentration of ammonia in the waste.

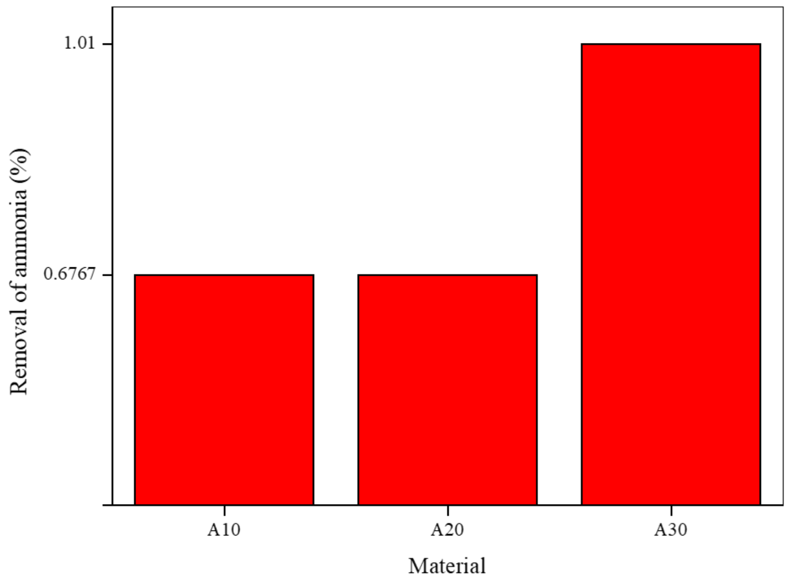

Figure 3. Effect of carbon activated loading on removal of ammonia

3.2. Effect of Combination of Activated Carbon and TiO_2 on ammonia degradation

In this study, activated carbon combined with titanium dioxide (TiO_2) photocatalyst into a composite material was used to degrade ammonia. The degradation test was carried out in a closed reactor which was irradiated with ultraviolet (UV) light and used 2 types of stirring media, namely using a magnetic stirrer and ultrasonic. Based on the tests that have been carried out, the results obtained are as shown in figures 4 and 5.

Figure 4 shows the decrease in ammonia concentration after activated carbon was combined with TiO_2. The presence of the TiO_2 photocatalyst on the surface of the activated carbon will have a synergistic effect in the adsorption and photocatalytic processes, so that the absorption life of the activated carbon will be longer. This result is like that reported by Elysabeth T, et al who used zeolite as an ammonia adsorbent. In this research, it was found that the addition of the TiO_2 catalyst to the adsorbent will cause a synergistic effect, where the process will take place simultaneously to reduce the occurrence of saturation in the adsorbent due to the accumulation of ammonia in the pores of the adsorbent[14]. This effect also reduces the desorption of ammonia compounds from the catalyst surface because ammonia absorbed in the adsorbent pores will be degraded by the TiO_2 photocatalyst.

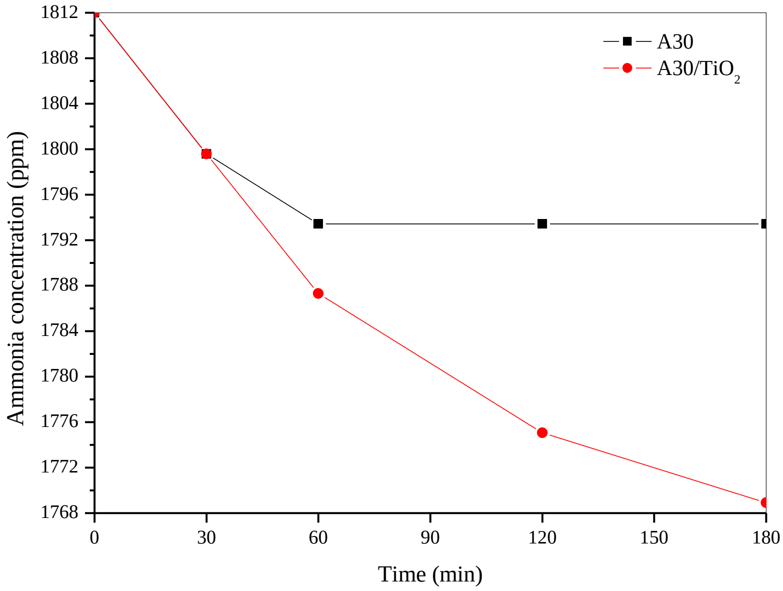

Figure 4. Effect of loading TiO_2 on ammonia photodegradation

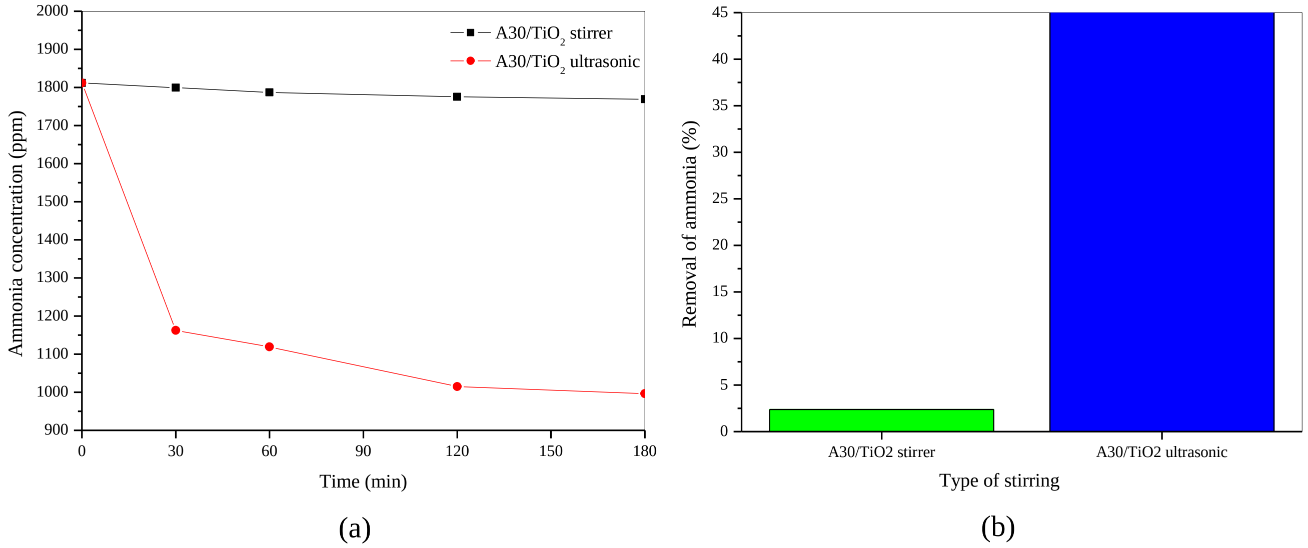

Figure 5. Effect of stirring type in ammonia photodegradation (a) the decrease of ammonia concentration and (b) removal of ammonia

Figure 5 shows a very significant difference in results when using magnetic stirrer and ultrasonic stirrers. Figure 5 shows that stirring using ultrasonics produces better results than stirring with a magnetic stirrer. The reduction in ammonia concentration reached 45%, this is higher than with a magnetic stirrer. Ultrasonic waves are more effective in mixing waste with composite materials so that the composite materials are distributed more evenly in the waste. This is because ultrasonic stirring uses vibration waves with an ultrasonic frequency above 20,000 Hz, whereas using a magnetic stirrer uses a rotation speed of 400 rpm so that the composite material becomes more stable and there is no accumulation at certain points.

4. Conclusion

In summary, the ammonia adsorption process takes place better if the activated carbon is prepared by heating at a high temperature. The more active carbon loading, the more ammonia will be absorbed. Although the amount of ammonia adsorbed was not significant, variations in loading showed differences in absorption capacity. The insignificant decrease in ammonia concentration was caused by the initial concentration being too high. Activated carbon combined with TiO_2 produces a composite material which has a synergistic effect in adsorbing and degrading ammonia. So that the ammonia removal process runs more effectively. The stirring process also influences the degradation process. Stirring using ultrasonics causes the ammonia concentration to decrease significantly. This is because with ultrasonics, stable and better stirring is obtained so that there is no accumulation at certain points.

Acknowledgements

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