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Research Paper

Effective Ammonia Removal from Hospital Wastewater by Using a Combination of Filtrations and Bio-Adsorbent from Tea Waste

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Abstract
Hospital wastewater contains a high concentration of ammonia that exceed the predetermined quality standards and can cause pollution to the environment. Therefore, the treatment of hospital wastewater is required prior to being released to the environment. Effective ammonia removal process has been developed by using a combination of sponge filtration pretreatment, followed by adsorption using activated carbon from waste of tea leaves, and nanofiltration using ceramic membrane as the final process. Experiment results show the highest effectiveness of ammonia removal of 70% by using filtration only. Effectiveness is increases up to 95% by a combination of filtration and adsorption. A maximum effectiveness of 100% is achieved when using a combination of filtration, adsorption, and nanofiltration. This combination produces an optimal condition for the ammonia removal by using 3 L/min flowrate, adsorbent mass 140 g, and operating time 15 minutes at which the ammonia concentration is 0.08 mg/L. This concentration is below the standard of allowable ammonia concentration of 0.1 mg/L.

Keywords
Adsorption, Ammonia, Tea Waste, Medical Liquid Waste, Nanofiltration

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1. INTRODUCTION

Hospitals produce a large quantity of wastewater from various hospital activities that involve the utilization of pharmaceuticals, chemicals, and liquid excretion from human bodies in the form of urines, feces, blood, etc (Zhang et al., 2020). Therefore, hospital wastewater is the source of dangerous substances that can cause harmful effect to the environment (Khan et al., 2021). Ammonia is discarded in hospitals wastewater as part of medical waste (Meo et al., 2014). Ammonia in water can be in the form of $NH_4{}^+$ and NH_3, both can irritate the eyes, respiratory tract failure, and skin due to its alkaline nature (Edwards et al., 2024). The biological effects of ammonia and nitrates in humans after acute exposures are dose-related and depend on their concentration (Dey et al., 2021). Ammonia concentrations are not significantly reduced by conventional treatment, as typical hospital wastewater treatment processes only involve physical processes (Khan et al., 2021). Hence, it is crucial to look for alternative treatments that are successful in practice. Ammonia can be removed from wastewater by adsorption using activated carbon (Ren et al., 2021) and bentonite (Cheng et al., 2019). Application of nanofiltration was also reportedly successful to remove pharmaceuticals compound from wastewater (Radjenovic et al., 2008). To meet wastewater quality standards, ammonia concentration can be reduced by combining adsorbents and filtration, which is one of the available methods (Huang et al., 2018).

Tea waste is an industrial waste that is available in large quantities (Amarasinghe and Williams, 2007). Tea waste takes a long time to biodegradation; however, it can be converted into bio adsorbent (Uddin et al., 2009). Therefore, tea waste can be used as an adsorbent rather than considered as environmental pollutant (Kabir et al., 2021). The tea leaf waste was efficiently applied as an adsorbent for cationic and anionic dyes in water solutions (Jain et al., 2020). Under optimal conditions, up to 98% of the dye can be removed from solution by adsorbent from tea waste (Mariah et al., 2023). The physical properties of tea waste, such as its large surface area and rapid adsorption rate (Zhou et al., 2018), make it suitable for use as an environmentally friendly adsorbent with minimal capital investment and easy availability of raw materials (Katha et al., 2021). Furthermore, it can be utilized as a alternative to environmental management by reducing tea waste. The tea solid waste could potentially be an active carbon material (Song et al., 2019). This material can be used as a low-cost adsorbent for removing effluent from wastewater (Ahmaruzzaman and Gayatri, 2010). Tea leaf pulp contains 37% cellulose, 14% hemicellulose and lignin, and 25% polyphenols 25% (Madrakian et al., 2012). Due to its high carbon content, tea waste has been proven to be effective precursor for active carbon production (Zhang et al., 2021).

In present study, a combined of adsorbents and nanofiltration membrane was employed for liquid waste hospital treatment. The advantages of using this membrane technology are relatively low operating costs, environmental friendliness, space efficiency, and a continuous separation process (Oatley-Radcliffe et al., 2017). Nanofiltration membranes have a pore size of 0.001μm which can filter waste with high organic content (Söderlind, 2020). The study aims to utilize nanofiltration membranes to treat the residual ammonia that cannot be absorbed by tea waste adsorbent. The use of nanofiltration membranes has gained popularity in wastewater treatment, drinking water, and clean water production processes (Yang et al., 2019). The use of nanofiltration membranes in full-scale wastewater management has been shown in several studies to be very efficient in removing residues in hospital liquid waste, including those from pharmaceutical activities (Radjenovic et al., 2008). In research to determine the morphological structure of tea waster carbon and the composition of the elements contained in the carbon, the SEM - EDS method was used, where one way to determine the morphological condition of the adsorbent is to use a Scanning Electron Microscope (SEM) (Inkson, 2016).

2. EXPERIMENTAL SECTION

2.1 Materials

Hospital wastewater was obtained from the wastewater treatment plant (WWTP) of the Ogan Ilir Regional General Hospital, South Sumatera, Indonesia. The wastewater characteristic is presented in Table 1. The activated carbon adsorbents were prepared using tea waste provided by PT Sinar Sosro Palembang. Sponge filter with 10 μm pore size for pretreatment filtration was obtained from local market. Nanofiltration was conducted using Kusatsu Toray Nanofiltration Membrane RO Cleaner NF-1812-150.

Table 1. Hospital Wastewater Characteristic

Parameters	Values	Standard
Temperature (°C)	25	$\leq$ 30
pH	9.1	6-9
Amonia (mg/L)	8.26	0.1

2.2 Methods

2.2.1 Preparation of the Carbon Adsorbent

The tea waste was washed and dried in an oven at 110°C for 4 hours, then heated in a furnace at 350°C for 1 an hour. The carbon powder from tea waste was being sieved through 100 mesh. Tea carbon powder was activated using 8% sulfuric acid for 24 hours, filtered, and washed with distilled water until the pH was between 5 and 7. The tea waste carbon is then dried in an oven at 110 oC for 8 hours. Tea waste carbon powder was analyzed for moisture content, ash content, and volatile matter content. The Discorena hisp (Gadung) flour was added to the activated carbon. The mixture powder was made into tablets with a diameter of 1.5 cm and a thickness of 1 cm, and then dried in an oven at 110°C for one hour.

2.2.2 Treatment of Hospital Wastewater

Treatment procedure of the hospital wastewater was described in Figure 2. Wastewater from the wastewater treatment tank was transferred using a pump and then flowed into a column containing filter with 10 μm pores. Subsequently, it proceeded to flow into the adsorption column that contained activated carbon from tea waste adsorbent, then it was filtered through the nanofiltration membrane column. The pH and ammonia concentrations were evaluated using operating times that varied from 15 to 75 minutes. The sample flow rates were 2 L/min and 3 L/min, and adsorbent weight were 50, 100, and 140 g, respectively.

2.2.3 Analysis Methods

Activated carbon quality was determined using proximate analysis according to ASTM D7582-10. Ammonia content was measured using ASTM D1426-15 test method A Direct Nesslerization. Measurement of pH conducted using pH meter. The morphological structure of tea waste carbon and the elements contained in the carbon were probed using SEM/EDX. Effectiveness of ammonia removal was calculated using formula (Equation (1)):

$$\text{Effectivenes}\,(\%) = \frac{C_{Ai} - C_{Af}}{C_{Ai}} \times 100 \quad (1)$$

C_{Ai} = initial ammonia concentration (mg/L)
C_{Af} = final ammonia concentration (mg/L)

3. RESULT AND DISCUSSION

3.1 Filter Column Pretreatment of Hospital Wastewater

Pretreatment of hospital wastewater is conducted by using a 10 μm sponge filter as an initial filter to remove impurities in medical liquid waste such as solid particles, sludge, and organic substances, which are may interfere with the performance of the adsorbent. As shown in Figure 3, a small variation of pH value is occurred at flowrate of 2 L/min and 3 L/min. Pretreatment using sponge filter is not significantly affect the pH of wastewater that indicates a low adsorption of minerals in liquid waste by sponge filter.

Meanwhile, it can be seen in Figure 4 that a downward trend in ammonia concentrations is observed at the output of the sponge filter, both at flow rates of 2 L/min and 3 L/min. The ammonia concentration is decreasing as the

(a) **(b)** **(c)**

Figure 1. Preparation of the Carbon Adsorbent : (a) Tea Waste (Raw Material) (b) Tea Waste Carbon Powder (c) Tea Waste Carbon Tablets

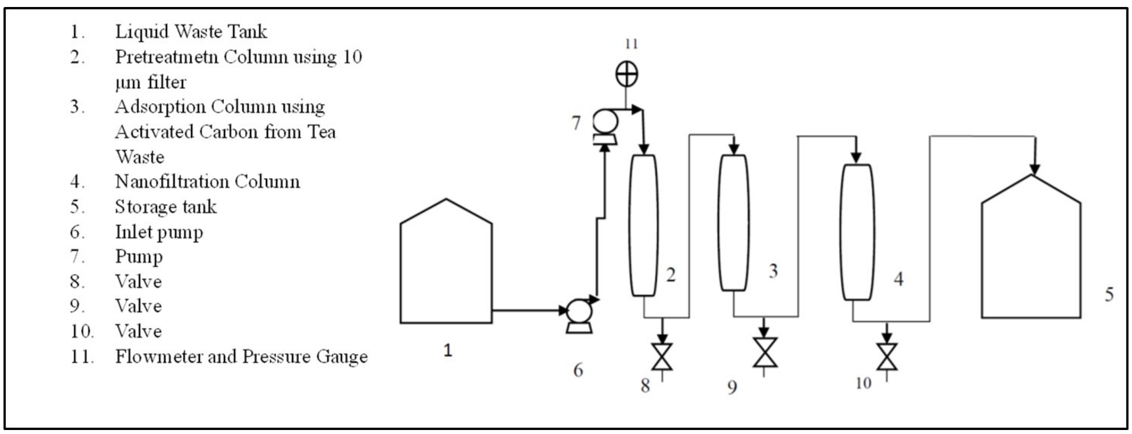

Figure 2. Flowsheet of Hospital Wastewater Treatment

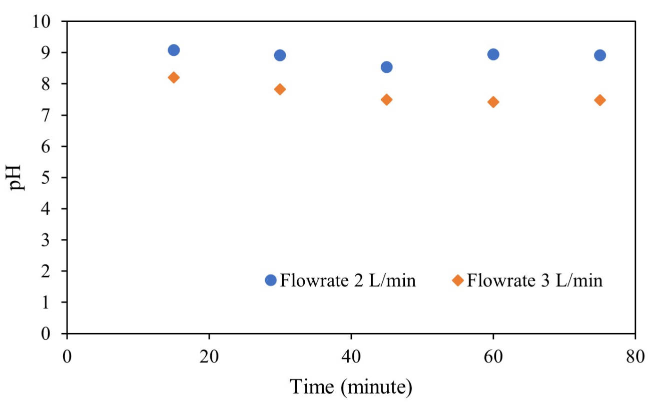

Figure 3. Effect of Filtration on pH Value

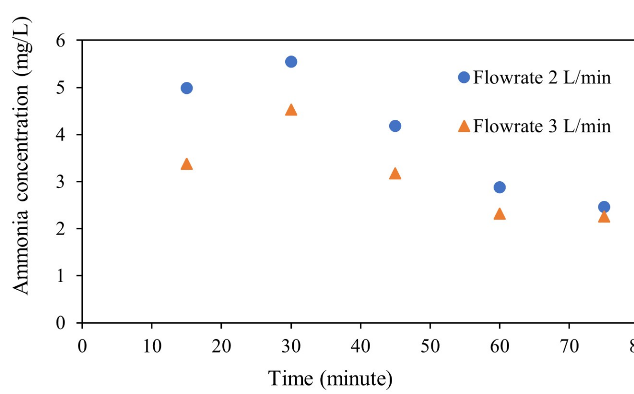

Figure 4. Effect of Filtration on Ammonia Concentration

operating time increases. The optimum condition occurs at a flow rate of 3 L/min and an operating time of 75 minutes, at which the ammonia concentration drops to 2.26 mg/L. However, this value is still above the permissible ammonia concentration for safe water discharge to the environment. The capability of sponge filter to adsorb ammonia indicates that the filter surface is negatively charge. However, the available surface is not sufficient to remove the remaining

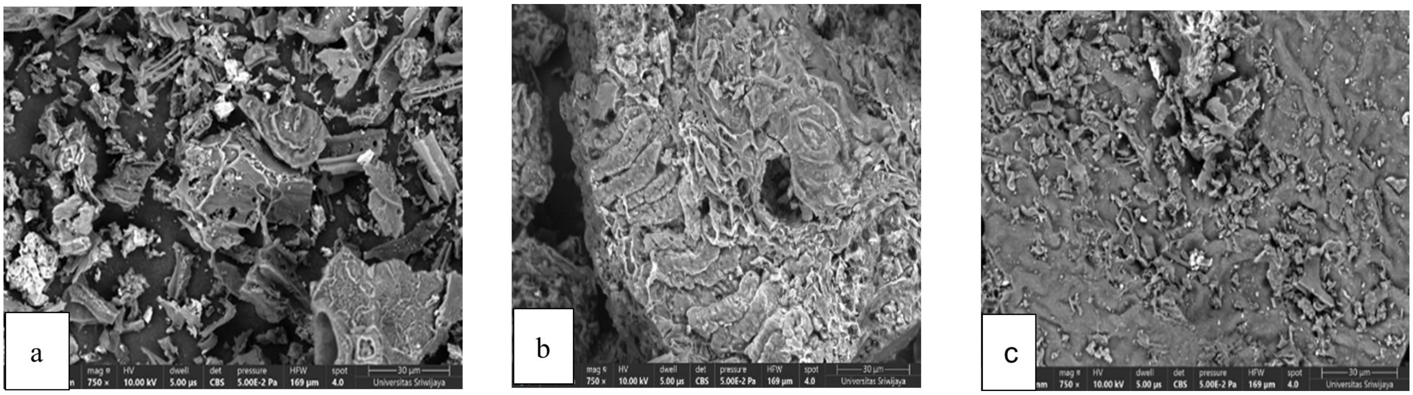

Figure 5. SEM Analysis Results on Tea Waste Carbon (a) Before Activation (b) After Activation (c) After Adsorption Process

Table 2. Characteristic of Activated Carbon from Tea Waste

Parameters	Standard	Values
Water (%)	Max. 15	7.21
Ash (%)	Max. 10	9.48
Volatile matter (%)	Max. 25	24.74

ammonia in liquid waste as the filter also removes other impurities.

3.2 Activated Carbon Adsorption Treatment of Medical Liquid Waste

3.2.1 Characteristic of Activated Carbon

The standard quality of activated carbon is represented by water content, ash content, and volatile substance. Therefore, those parameters are measured to determine the characteristic of activated carbon from tea waste as shown in Table 2. It can be seen that the quality of activated carbon from tea waste meets the required standard of water, ash, and volatile matter content.

Activation also increases pore formation as can be seen in Figure 5. The basic structure of carbon is thought to resemble the structure of pure graphite which is composed of hexagonal layers composed of carbon atoms (Pinem et al., 2020). Meanwhile, activated carbon shows a smoother surface with widened pores (Figure 5b). This is expected because the activation has been done with H_2SO_4 concentrations of 8% so that the opening of the pores will cause the process of absorption of molecules optimally. Activation of tea waste carbon adsorbent using sulfuric acid causes the morphology of the surface structure of the biosorbent to become porous and causes the surface area of the biosorbent to become larger (Duran et al., 2011). Surface of activated carbon after adsorption process becomes denser as the surface is fully covered by adsorbed molecules (Figure 5c).

Elemental composition in Table 3 shows an increasing nitrogen content of adsorbent as the result of ammonia adsorption.

Table 3. Elemental Composition of Activated Carbon

Compound	Before Activation (%)	After Activation (%)	After Adsorption (%)
Carbon (C)	62.3	62.0	62.2
Nitrogen (N)	6.8	9.3	12.3
Oxygen (O)	26.5	27.7	24.9
Sodium (Na)	0.2		
Magnesium (Mg)	0.6	0.1	
Alumunium (Al)	0.6	0.1	0.3
Silica (Si)	0.1	0.1	
Phosphor (P)	0.3		
Sulfur (S)	0.4	0.4	0.2
Pottasium (K)	0.2		
Calcium (Ca)	2.0	0.3	0.2
Tantalum (Ta)			0.1

Elemental composition of activated carbon before and after sulfuric acid activation is depicted in Figure 1c wich are summarized in Table 3. Based on SEM-EDX results, the dominant element in tea waste carbon is the C atom, because tea waste is a polymer in the form of cellulose chains. The cellulose-based polymer structure of tea leaves results in a relatively strong chemical adsorption ability on metal ions and organic bases (Madrakian et al., 2012). There is slight increase of oxygen and nitrogen after activation as an indication that sulfuric acid activation increases the acid sites to promote ammonia adsorption.

3.2.2 Effect of Operating Time on Ammonia Adsorption

The decrease of ammonia concentration is observed as the operating time increases to 75 minutes. Ammonia concentration is sharply reduced from 2.26 mg/L to as low as 0.35 mg/L as depicted in Figure7. Slight variation of the ammonia concentration may indicate the adsorption process has reached the equilibrium state. The lowest ammonia concentration is 0.35 mg/L after operating time of 60 minutes.

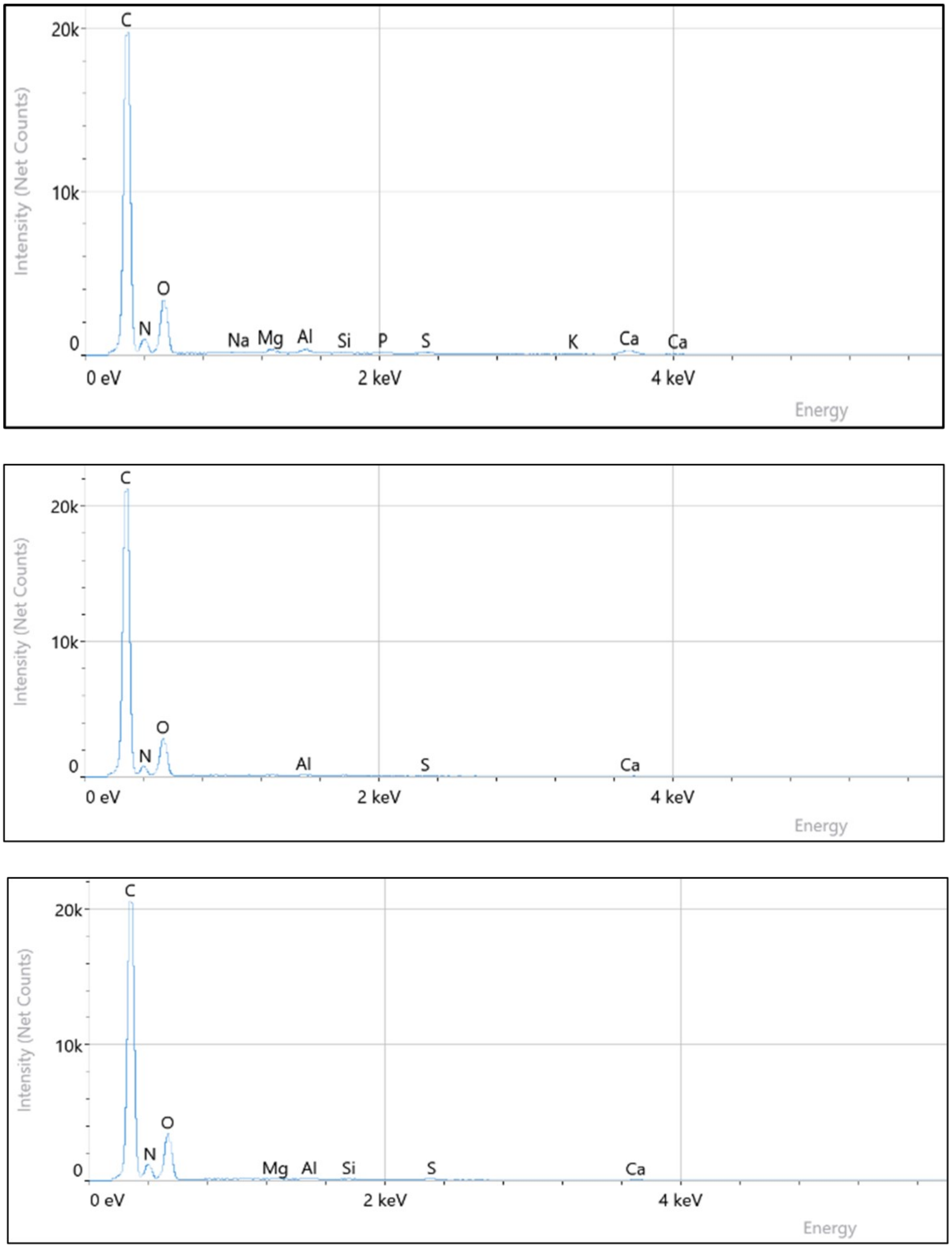

Figure 6. SEM-EDX Analysis Results on Tea Waste Carbon (a) Before Activation (b) After Activation (c) After Adsorption Process

Ammonia dissolved in medical wastewater can raise the pH of the water to alkaline. The activated carbon adsorption decreases the pH value of medical liquid waste from 8.8 to 7.15 as the effect of lower quantity of dissolved ammonia in liquid waste. Operating time is not affecting the pH value of liquid waste significantly because a low concentration of ammonia has been achieved after 15 minutes operating time as described in Figure 8. Therefore, longer adsorption time

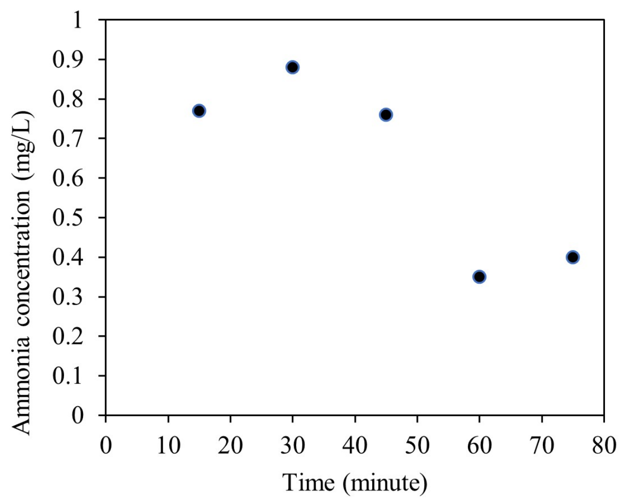

Figure 7. Effect of Filtration on Ammonia Concentration

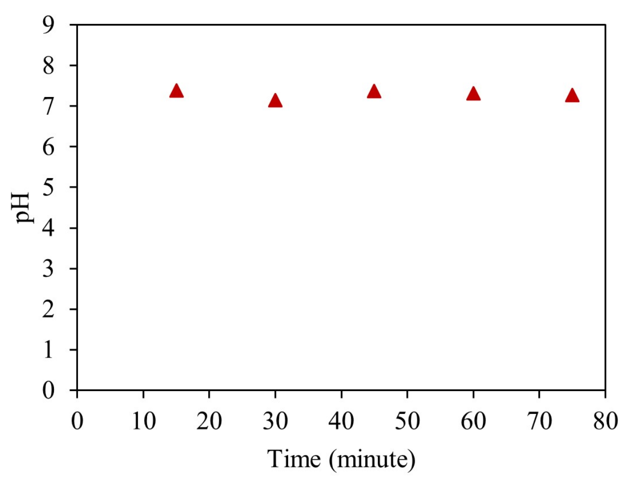

Figure 8. Effect of Operating Time on Adsorption Results on pH (Flow rate 3 L/min, Adsorbent 140 g)

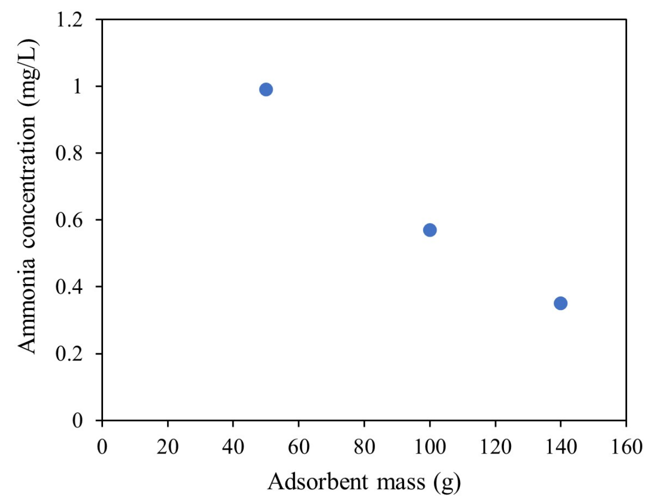

Figure 9. Effect of Adsorbent Mass on Ammonia Concentration (Flowrate 3 L/min, 60 Minutes)

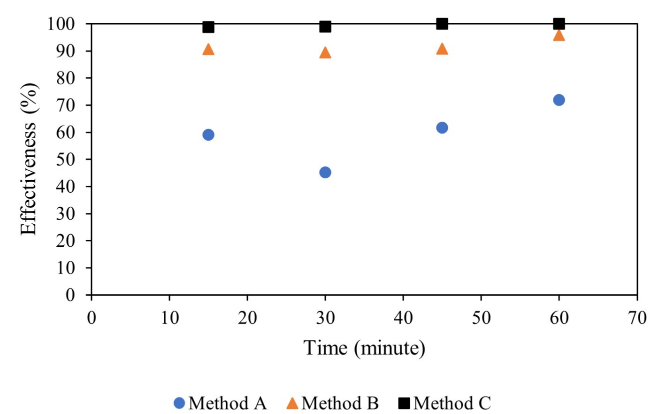

Figure 10. Effect of Adsorbent Mass on pH (Flowrate 3 L/min, 60 Minutes)

will not produce significant effect on ammonia reduction and pH variation.

3.2.3 Effect of Adsorbent Mass Ratio

The effect of adsorbent mass variation can provide an overvie w of the adsorption ability of the adsorbent, where in general the adsorption ability will increase with the addition of the given adsorbent mass (Mouni et al., 2018). The effect of the amount of adsorbent on adsorption ability is because the number of active sides on the surface of the adsorbent will increase if the increasing amount of adsorbent mass is given (Celebi et al., 2020). As shown in Figure 9, increasing adsorbent mass from 50 g to 140 g reduces ammonia concentration from 0.99 mg/L to 0.35 mg/L. In Figure 10, as expected, the pH value shows a slight decreasing trend at higher adsorbent mass ratio.

3.2.4 Effect of Flow Rate

Flow rate variation from 2 L/min to 3 L/min results in decreasing concentration of ammonia and pH as presented in Table 4. Increasing flowrate will improve mass transfer between adsorbent and adsorbate as the adsorbate can penetrate deeper into the adsorbent and reaches higher surface area of adsorbent. Using 60 min operating time, the ammonia concentration is decreasing to 0.7 mg/L at 2 L/min and 0.35 mg/L at 3 L/min indicates significant effect of flowrate on adsorption process. Similar trend also is observed for pH value that reduces to 7.25 at 2 L/min and 7.15 at 3 L/min

However, the ammonia concentration after adsorption treatment is still above the required standard 0.1 mg/L. Apparently, ammonia in water faces a competitive adsorption with other cations on activated carbon. Ammonia adsorption decreases when other cations are present at high

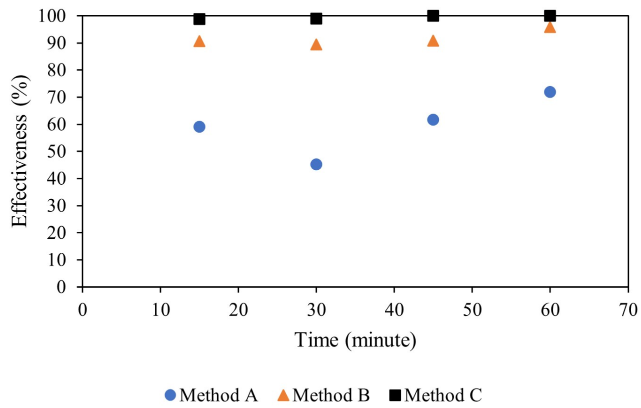

Figure 11. Effectiveness of Treatment Methods (Adsorbent Mass 140 g, Flowrate 3 L/min) Using Method : (A) Filtration (B) Filtration + Adsorption (C) Filtration + Adsorption + Nanofiltration

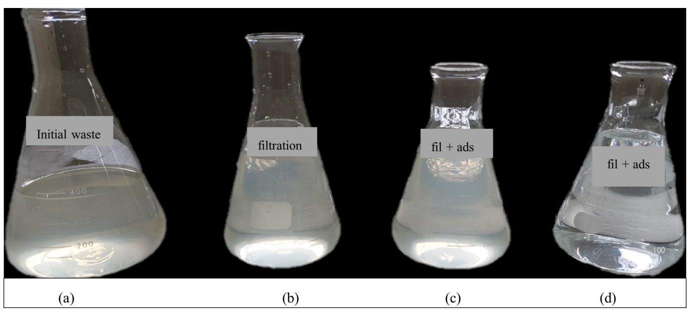

Figure 12. Products of Treatment : (a) Initial Waste (b) Filtration (c) Filtration + Adsorption (d) Filtration + Adsorption + Nanofiltration

Table 4. Effect of Flow Rate on Adsorption Results (Adsorbent 140 g, 60 Minutes)

Flow Rate (L/min)	pH	Ammonia Concentration (mg/L)
2	7.25	0.71
3	7.15	0.35

concentration (Ren et al., 2021).

3.3 Nanofiltration Membrane Treatment of Liquid Medical Waste

The output of the adsorption treatment has not met the ammonia standard; therefore, the treatment is continued using nanofiltration. As presented in Table 5, nanofiltration of hospital wastewater at all operating conditions can reduce the ammonia concentration below the required standard 0.1 mg/L. The capability of nanofiltration membrane to remove traces component from water is related to nanometer size pores that can reject ions in the water. Therefore, nanofiltration can be applied to remove a wide variety of ions using various membrane materials. Hence, operation parameters such as flowrate and operating time do not significantly affect filtration results.

Table 5. Ammonia Reduction by Nanofiltration Treatment

Flow Rate (L/min)	Operating Time (minutes)	pH	Ammonia Concentration (mg/L)
2	15	7.28	0.08
2	30	7.33	0.04
2	45	7.34	0.00
2	60	7.41	0.00
3	15	7.5	0.1
3	30	7.26	0.08
3	45	7.36	0.00
3	60	7.37	0.00

3.4 Effectiveness of Medical Liquid Waste Treatment

Effectiveness of method combinations for ammonia removal from medial liquid waste can be seen in Figure 11. Sponge filtration process shows ammonia removal effectiveness in the range of 40%–70% during 60 minutes operation. The highest effectiveness t of 70% is obtained after 60 minutes operating time. Combination of filtration and activated carbon adsorption increases the ammonia removal effectiveness up to 90%-95%. Where the increase in adsorbent mass causes an increase in surface area on the adsorbent which allows contact between adsorbent particles and adsorbate molecules to be higher (Nsi et al., 2017). Complete removal of ammonia is achieved using combination of filtration, adsorption and nanofiltration. Ammonia concentration is already meet the required standard after 15 minutes operation.

Visual water quality after each method also very different as shown in Figure 12. Water turbidity is still observed after sponge filtration (Figure 12b) and combination of sponge filtration and activated carbon adsorption (Figure 12c). High clarity water is produced after combination of sponge filtration, adsorption, and nanofiltration as shown in Figure 12d. Nanofiltration treatment removes not only ammonia, but also other remaining impurities in wastewater (Arola et al., 2019).

4. CONCLUSIONS

Hospital wastewater treatment using a combination of sponge filtration, activated carbon adsorption, and nanofiltration is capable of removing ammonia completely to meet the required ammonia concentration standard. Treatment using sponge filtration is only remove 70% ammonia. Combination of filtration and activated carbon adsorption produces a better effectiveness of 95%. Activated carbon from tea waste is effectively enhances the removal of ammonia from wastewater. Addition of nanofiltration process is successfully produces clean water with high clarity and zero ammonia concentration. Therefore, a combination of sponge filtration, activated carbon adsorption, and nanofiltration can be applied for hospital wastewater treatment.

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