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# Synthesis of Citric Acid-Crosslinked Chitosan Membrane with Zeolite Filler and Its Application as Cu(II) Ion Separation Membrane

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**Abstract.** Heavy metal is a pollutant material that can harm the environment, especially Cu(II) metal. Various innovations have been made to remove Cu(II), one of which this by using chitosan membrane technology. However, chitosan has limited functional groups and is less porous. To overcome this weakness, cross-link modification was carried out with citric acid and the addition of zeolite. This study aims to synthesize and characterize citrate crosslinked chitosan membrane with zeolite filler and to determine the effectiveness of the membrane as Cu(II) ion separation membrane. The method used in this study is the phase inversion with the procedures a) zeolite activation, (b) chitosan crosslinking citrate with zeolite addition, (c) membrane printing, (d) membrane transport, (e) physicochemical test. The FTIR results showed the presence of a secondary amide at a wavenumber of 1653 cm<sup>-1</sup>. Modification of chitosan membrane can increase the thickness and weight, porosity, and hydrophilicity of the membrane. However, the addition of zeolite reduces water absorption and swelling. Based on this research, the best membrane for adsorbing Cu(II) ions is the Z0.25 membrane with a percentage of separation of 48.15% and a rejection percentage of 99.26%.

## INTRODUCTION

Environmental pollution is a serious problem for life, one of the problems is water pollution. Water pollution can be caused by heavy metals. Heavy metals are polluting materials that can harm the environment and organisms [1]. One of the heavy metals that are found in waters is Cu(II) ion [2]. The presence of heavy metal Cu(II) is commonly found in metal coating wastewater, wire industry, paint industry, metalworking industry, and the printed circuit board manufacturing industry [3]. The maximum Cu content allowed is 0.05-1.5 ppm [4]. To remove heavy metal Cu(II) from wastewater, membrane technology can be used.

The use of chitosan as material for separation membranes has much developed because chitosan has advantages, including selective properties and two reactive functional groups, such as amine group (-NH<sub>2</sub>) and hydroxyl group (-OH) which have strong chelation with ions [5]. However, chitosan has limitations, such as the low number of active groups to bind the target compound [6]. To overcome these weaknesses, modifications of chitosan were carried out, one of which was cross-linking. One of the crosslinking agents that can be used is citric acid. The cross-link reaction between chitosan and citric acid can increase the number of reactive groups so that it can increase hydrophilicity [7][8]. One of the other membrane modifications is the addition of zeolite [9]. Zeolite is a potential material because has a porous structure that can adsorb various pollutants such as heavy metal ions and organic molecules [10]. The addition of zeolite in the membrane can increase the membrane adsorption process against heavy metals [11].

This research was carried out on the synthesis of citrate crosslinked chitosan membrane with zeolite fillers with variations in zeolite weight, characterizing the physicochemical membranes through a functional group (FTIR), surface morphology (SEM), crystallinity (XRD), thickness and weight, water uptake, swelling degree, porosity, hydrophilicity and determine the effectiveness of the membrane as a separation membrane against Cu(II) ions.

## EXPERIMENT

### Materials

The materials used in this study were natural zeolite (Wonosari, Gunung Kidul, Yogyakarta), HCl 37% pa (Merck), chitosan (BM = 40,000 g/mol, DD = 87%, Biotech Surindo), acetic acid pa (Merck), citric acid pa (BM = 210.14g/mol, Merck), NaOH (BM = 40.00 g/mol, Merck), aquades, CuCl<sub>2</sub>.2H<sub>2</sub>O (Merck), K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> (Merck).

### Methods

#### Natural Zeolite Activation

The zeolite that had been sifted 150 mesh then washed with aquadest, stirred for 1 hour. The zeolite mixture was heated at 70-80 °C for 1 hour. The zeolite was left at room temperature overnight and filtered, then heated at 120°C for 4 hours. The heated zeolite is then activated by adding HCl. The mixing of zeolite with HCl was carried out for 1 hour. Then left it overnight. Zeolite which had been activated with HCl was then washed with aquadest until the pH was constant. Neutral zeolite was dried at 150 °C for 2 hours.

#### Synthesis of chitosan/citrate/zeolite solution

The synthesis process of chitosan/citrate/zeolite membranes was made in 4 variations of the ratio addition zeolite. The membrane synthesis was initiated by dissolving the chitosan powder in acetic acid and then adding citric acid. The cross-link reaction was carried out for 4 hours at 55°C. The chitosan solution that has been cross-linked with citrate is added with zeolite in a weight ratio of 1:0.5; 1:0.25; 1:0.125 and 1:0.0625. Zeolite was added to the chitosan/citrate solution with stirring for 2 hours. The membrane was printed in a petri dish and dried in an oven. The dry membrane was removed with NaOH then washed with aquadest until neutral.

#### Characterization of Membrane

The characterization of chitosan/citric acid/zeolite membranes was carried out using Fourier-transform Infra-Red Spectroscopy (FTIR) to determine the functional groups in the membrane, Scanning Electron Microscope (SEM) to determine the surface morphology and membrane cross-section, X-Ray Diffraction (XRD) to determine the presence of zeolite in the membrane.

#### Measuring the weight and thickness

Measurement of membrane thickness using a thickness meter (QITCO), membrane thickness was measured at 5 different points. The weight of the membrane in each variation was weighed using the OHAUS analytical balance.

#### Swelling Degree

The swelling degree was carried out by measuring the dry membrane diameter, the membrane was then soaked in aquadest for 24 hours and the wet membrane was measured with a ruler.

## **Water uptake**

The dry membrane was weighed using the OHAUS analytical balance, then the membrane was soaked in aquadest for 6 hours, and every 1 hour the weight of the membrane was weighed using OHAUS analytical balance.

## **Porosity**

The membrane was soaked in aquadest for 24 hours. The membrane was weighed using an OHAUS analytical balance. Then the membrane was dried in an oven at 100°C for 24 hours. The weight of the membrane was weighed using OHAUS analytical balance.

## **Hydrophilicity**

Membrane hydrophilicity was measured using a sessile drop method. The degree of hydrophilicity is determined from the contact angle formed between the membrane surface and water.

## **Determination of optimum pH**

Cu solution (20 ppm) was conditioned at pH 5, 6, and 7 as the feed phase. Place the membrane on the diffusion cell, and aquadest as the permeate phase on the left side of the diffusion cell. Then, it was tested using AAS to determine the concentration of Cu(II) transported by the membrane.

## **Determination of the optimum time**

Cu solution pH 6 as the feed phase. Put the membrane on the diffusion cell, for the permeate phase, aquadest are being added. Then, diffusion cell on a magnetic stirrer for 24 hours. The feed phase filtrate is taken every 4 hours. The results of the filtrate were tested using AAS to determine the concentration of Cu(II) transported by the membrane.

## **Determination of Optimum Membrane Variation**

Cu(II) solution pH 6 as the feed phase. Place the membrane (Z0,0625, Z0,125, Z0,25, Z0,5) in the diffusion cell, the permeate phase added by aquadest. Then, the diffusion cell is on a magnetic stirrer for 8 hours. The results of the filtrate were tested using AAS to determine the concentration of Cu(II) transported by the membrane.

# **RESULTS AND DISCUSSION**

## **Activated Zeolite**

Zeolite 150 mesh was washed with aquadest to remove impurities polar such as soil so that it dissolves in aquadest. Zeolite was dried at 120°C for 4 hours to remove the aquadest. The chemical activation of zeolite was carried out by adding HCl and soaked overnight to clean the zeolite pores from impurities and rearrange the positions of the exchanged atoms [12]. Zeolite was washed with aquadest until neutral. Zeolite was dried at 150°C to evaporate water in the zeolite pores so the number of pores and the surface area increased [13][14]. The increase in surface area of zeolite is expected to increase the ability of the zeolite to absorb heavy metals.

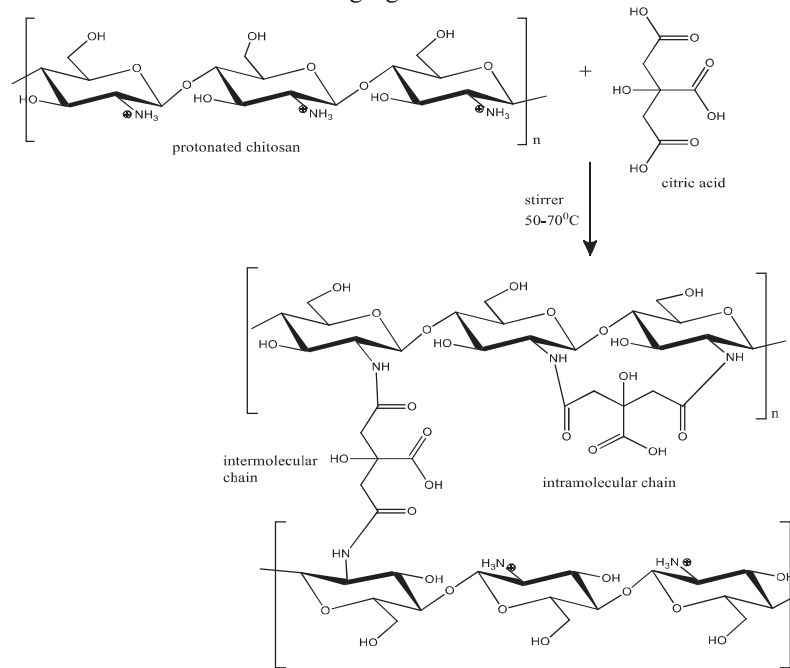
## **Synthesis of Chitosan/Citric Acid/ Zeolite Membrane**

The synthesis of chitosan membrane crosslinked citric acid with zeolite filler was started by making a chitosan solution. The dissolution of chitosan in acetic acid causes the  $-\text{NH}_2$  group on the chitosan to become  $\text{NH}_3^+$ , this reaction runs fast [15].

Chitosan modification was carried out by crosslinking with citric acid. Citric acid is used as a crosslinking agent and adds an active group to the membrane which serves as the active site of the membrane in the transport process.

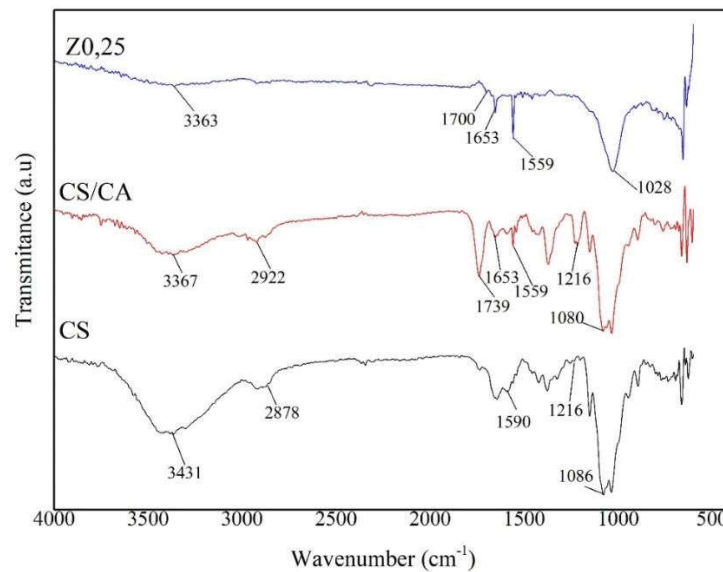
The negatively charged carboxylic group of citric acid is expected to be able to bind Cu(II) heavy metal and increase the interaction of permeate compounds through the membrane.

The crosslinking reaction is carried out at a temperature of 50-70 °C because heating can accelerate the crosslinking reaction. The types of reactions in cross-linking between chitosan and citric acid are intermolecular reactions that occur in two chitosan chains and intramolecular reactions in one chitosan chain [15]. The chemical reaction that occurs between chitosan and citrate is shown in the following fig. 1



**FIGURE 1.** The reaction of cross-linked chitosan citric acid

To determine the success of cross-linking between chitosan and citric acid FTIR test was performed. The resulting FTIR spectra are shown in Fig 2.



**FIGURE 2.** FTIR spectra of various membrane

Peak O-H spectra at 3431 cm<sup>-1</sup> (chitosan spectra) shifted to 3367.91 cm<sup>-1</sup> (CS/CA spectra). This shift indicates that there has been a change in the O-H group in chitosan and there has been a cross-linking between the  $\text{-NH}_2$  group in

chitosan and the –COOH group in citric acid. In addition, the peak in N-H spectra of chitosan shifted from 1590  $\text{cm}^{-1}$  to 1653  $\text{cm}^{-1}$  which was N-H spectra of amide. The C-N amide group appears at 1559  $\text{cm}^{-1}$ .

Modification of chitosan that has been cross-linked with citric acid then added with zeolite, the ratio between chitosan: zeolite is 1:0,5, 1:0,25, 1:0,125, 1:0,0625. The addition of zeolite did not create new groups on the membrane. The following research [16], [17], and [11] the intensity of the band at 3400  $\text{cm}^{-1}$  (-OH group) decreases on the Z0.25 membrane due to the interaction of –OH in chitosan with -OH group in the zeolite. Then the two bands at 1080  $\text{cm}^{-1}$  on the CS and CS/CA membranes shifted to the 1028  $\text{cm}^{-1}$  region on the Z0.25 membrane due to overlapping Si-O band with C-O stretch band.

## Thickness and Weight

Thickness and weight can show the effect of variations in the mass composition of membrane constituents in the same unit area on the thickness and weight membrane. The results of thickness and weight measurements on various membrane variations can be seen in table IV.1.

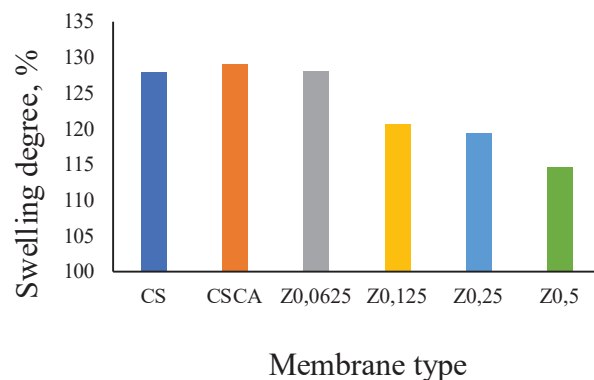
**TABLE 1.** The results of thickness and weight measurements on various membrane variations

Membrane type	Thickness ( $10^{-2}$ mm)	Weight ( $10^{-2}$ g)
CS	4,0	6,81
CSCA	6,2	6,90
Z0,0625	6,8	7,31
Z0,125	8,4	7,53
Z0,25	9,0	8,62
Z0,5	9,4	10,

Table 1 shows that membrane with modification of citric acid and zeolite can increase the weight and thickness of the membrane. The increase in weight and thickness of the resulting membrane was due to the same amount of chitosan as the main raw material for the membrane but the addition of citric acid as a crosslinking agent and zeolite as a filler so that the membrane becomes heavier and thicker.

## Swelling Degree

Measurement of the swelling degree aims to estimate the size of a substance that can diffuse into the membrane. The results of the swelling degree are shown in Fig. 3. Swelling degree increase after modification with citric acid and decrease with the addition of zeolite to the modified membrane. CS/CA membranes have a high swelling degree because citric acid has a hydrophilic COOH group and it can react with water molecules through intermolecular hydrogen bonds. The Swelling degree decreased with the addition of zeolite mass on the membrane. Similar results in the study [17], this is due to the addition of less hydrophilic zeolite particles and polymer chain stiffness caused by inorganic particle filling.



**FIGURE 3.** Percentage swelling degree on various membranes

## Water Uptake

The water uptake test aims to determine the membrane's ability to in absorbing water. The results of the absorption test for water can be seen in Fig. 4. From these results it can be seen that the CS/CA membrane has the highest water absorption value of 105.76%, this is due to the inclusion of citric acid in the membrane composition. Citric acid has three carboxylic groups (-COOH) which can increase the interaction between water and polymer [15]. The water absorption value decreases with increasing zeolite mass on the membrane. Similar results in the study [17], this is due to the addition of less hydrophilic zeolite particles and polymer chain stiffness caused by inorganic particle filling.

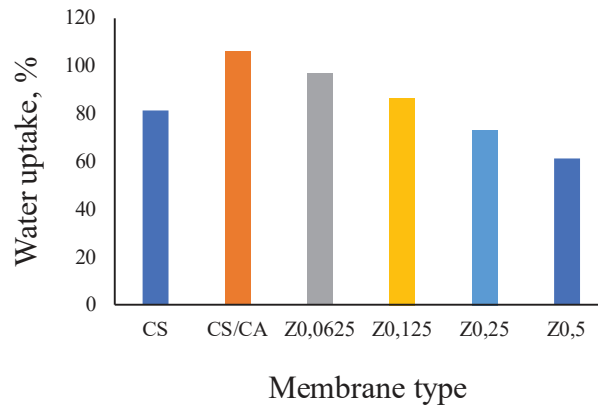


FIGURE 4. Percentage of water absorption on various membranes

## Porosity

The membrane porosity test aims to determine the volume of free space by the area between the materials in the membrane. The results of the porosity test can be seen in Fig. 5. It can be seen that the CS/CA membrane has a porosity the highest at 54%. The membrane porosity increases with increase zeolite on the membrane. In the study [18], the increase in the porosity of the membrane due to zeolite as a porous material contributed to the porosity of the membrane.

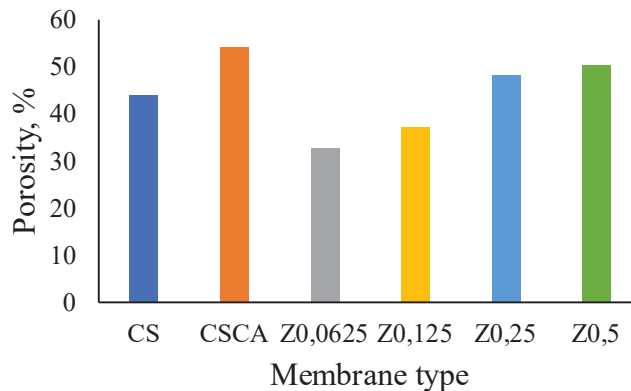


FIGURE 5. Percentage of porosity in various membranes

## Hydrophilicity

The hydrophilicity test on the membrane surface was carried out to determine the hydrophilicity of the membrane. The results of the contact angle measurements on various membranes can be seen in Table 2. From Table 2 can be seen that the contact angle increases with the addition of zeolite mass. The surface contact angle is an indicator for determining hydrophilicity or hydrophobicity of the membrane. If the contact angle value is below  $90^\circ$ , the membrane surface is called hydrophilic. From Table 2 can be seen that all membrane is hydrophilic which allows it to be applied to heavy metal transport. The contact angle increase with the addition of zeolite mass, the same trend was observed in swelling degree and water uptake tests. According to research [19], the membrane absorbs water through chitosan which interacts strongly with water constituents on the membrane.

TABLE 2. Result of contact angle on membrane

No.	Membrane type	Contact angle ( $^\circ$ )
1	CS	89
2	CS/CA	69
3	Z0,0625	70
4	Z0,125	71
5	Z0,25	72
6	Z0,5	80

## X-Ray Diffraction Spectroscopy (XRD) Analysis

The crystallinity of chitosan membrane crosslinked citric acid with zeolite filler was confirmed using X-Ray Diffraction (XRD). The results of the XRD analysis of the chitosan membrane crosslinked with citric acid and zeolite filler can be seen in Fig. 6. The peak of zeolite characteristics was not significantly observed at the Z0.25 membrane. According to research [9] pure zeolite produces main crystal peaks at  $7.05^\circ$ ,  $10.03^\circ$ ,  $12.3^\circ$ ,  $15.9^\circ$ ,  $21.5^\circ$ ,  $23.8^\circ$ ,  $25.9^\circ$ ,  $26.9^\circ$ ,  $29.8^\circ$ ,  $30.68^\circ$ ,  $34.03^\circ$ ,  $44^\circ$ , and  $52.4^\circ$ . So this shows that the zeolite becomes amorphous in the chitosan/citric acid matrix. The crystallinity of zeolite is limited due to the strong interaction between chitosan, citric acid, and zeolite. In this study, diffraction peaks were found at  $9.98^\circ$  and  $20.12^\circ$  which indicated the diffraction pattern of chitosan and at  $15.3^\circ$  indicating cross-linking between chitosan and citric acid. According to [11] the peaks of chitosan around  $10^\circ$  and  $20^\circ$  become weak on the membrane because the chitosan loses its crystallinity.

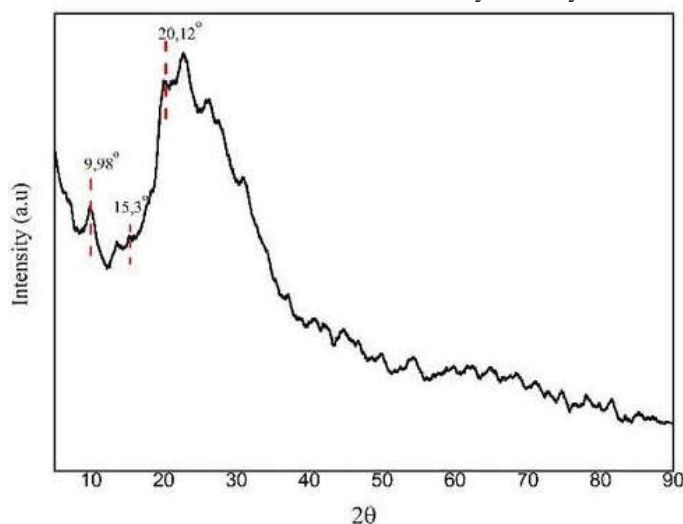


FIGURE 6. XRD Spectra of chitosan/citrate/zeolite membrane



## Scanning Electron Microscopy (SEM) Analysis

The surface morphology of chitosan and the chitosan membrane crosslinked citric acid with zeolite filler can be seen in Fig. 7. The Analysis was used a Scanning Electron Microscopy (SEM) instrument. Based on Fig. 7, it can be seen that the chitosan membrane has a solid, smooth, flat, and homogeneous surface and cross-section so that the chitosan membrane does not show pores because the surface is very tight. While the citric acid crosslinked CS membrane with zeolite filler has a rougher surface and tends to be porous than the chitosan membrane. This difference is possible because the presence of filler will make the membrane more expand so that it enlarges the pores on the membrane surface. In addition to the presence of functional groups in the membrane, the presence of pores on the membrane surface will affect the ability of the membrane to interact with the analyte and increase its ability to separate the desired analyte.

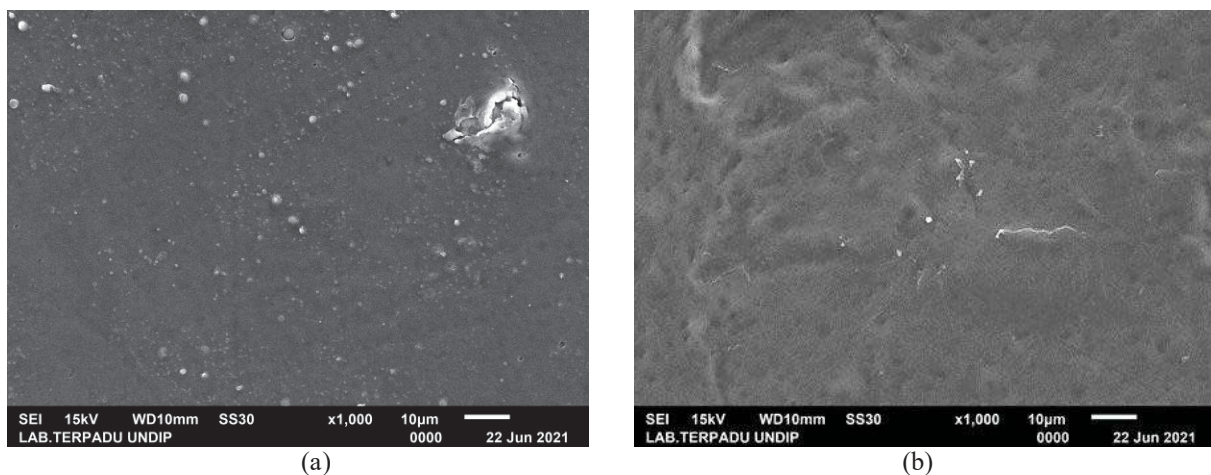


FIGURE 7. Morphology of membranes (a) chitosan (b) chitosan/CA-Zeolite

## Cu(II) Ion Separation Performance Test

The synthesized membrane will be used in the process of separating Cu(II) ions. This separation test was carried out to determine the ability of the membrane before modification and before modification with citric acid and zeolite. Determination of the optimum pH of Cu(II) was carried out by varying the pH in the feed phase 5, 6, and 7. The results of the separation of Cu(II) ions can be seen in Table 3.

TABLE 3. Percentage of Cu(II) separation at various pH

pH	Separation (%)
5	7,28
6	48,15
7	73,41

The optimum pH for Cu(II) ion separation is pH 6. At pH 7 although the percentage of separation is very high, at pH 7 another Cu ion species has been formed, it is  $\text{Cu}(\text{OH})_2$ . So that the separation of Cu(II) metal ions at a very high pH 7 can be influenced by the precipitation process of  $\text{Cu}(\text{OH})_2$  species. In this study, the separation activity of Cu(II) was carried out at pH 5-7 because at a pH below 5 the membrane was not very stable and at a pH above 7 Cu(II) it might have tended to be in molecular form, even at a more alkaline pH, it would form a precipitate of  $\text{Cu}(\text{OH})_2$ .

Determination of the optimum time of Cu(II) was carried out by varying the time for 24 hours. The results of the separation of Cu(II) ions can be seen in Table 4. Based on the table, it can be seen that the metal ion separation results fluctuated. Seen the percentage of separation, the increase in separation occurs every 8 hours. So that the 8 hour is the optimum hour for the separation of Cu(II) ions.

**TABLE 4.** Percentage of membrane separation against Cu(II) ions at time variation

Time (hour)	Separation (%)
4	34,91
8	48,15
12	44,48
16	48,68
20	55,22
24	40,25

Determination of the optimum membrane for Cu(II) separation was carried out with CS, CS/CA, Z0,0625, Z0,125, Z0,25, and Z0,5 membranes. The results of the separation of Cu(II) ions can be seen in Table 5. Based on the data in Table 5, it can be seen from the percentage of membrane separation The optimum is indicated by the Z0.25 membrane. This is because of the Z0.25. the membrane has many pores so that more metal ions are adsorbed on the membrane. From the percentage of separation, the more zeolite added to the membrane, the higher the percentage of separation. This is because the increase in zeolite will add pores and the active site on the membrane so that more Cu(II) ions are trapped in the membrane and bind to the active site in chitosan, citric acid, and zeolite.

**TABLE 5.** Percentage of separation of Cu(II) ions in membrane variations

Membrane type	Separation(%)
CS	4,26
CS/CA	21,49
Z0,0625	8,24
Z0,125	9,72
Z0,25	48,15
Z0,5	23,19

To determine the separation process that occurs, the percentage of rejection is determined. Based on the rejection result data in Table 6, it can be seen that the Z0.25 membrane has the highest percentage rejection by 99.26%. This shows that on the Z0.25 membrane more adsorbs Cu(II) ions than other membranes. According to [20], the ions are adsorbed as a result of concentration equilibrium in a two-chamber then Cu(II) ions can bind to the active groups on the surface and inside of the membrane. With the HSAB concept, Cu(II) ions belong to the middle group of acids that can bind  $-NH_2$  which is a hard base, and the ability to form complexes depends on the size and hardness of the metal ion. So this membrane is more suitable for use as an adsorption membrane.

**TABLE 6.** Rejection percentage of Cu(II) ions on membrane variations

Membrane type	R(%)
CS	98,67
CS/CA	87,64
Z0,0625	98,71
Z0,125	99,19
Z0,25	99,26
Z0,5	98,29

## CONCLUSION

Citric acid-crosslinked chitosan membrane with zeolite filler has been successfully synthesized as confirm by the FTIR results of the chitosan/citrate/zeolite membrane with the peak at wave number  $1653\text{ cm}^{-1}$  that formed amide secondary which is the modification of primary amine into a secondary amine. Modification of chitosan membrane increases the thickness and weight, porosity, and hydrophilicity of membrane, but with the addition of zeolite decreased water uptake and swelling degree membrane. The best membrane for adsorbing Cu(II) ions is the Z0.25 membrane with a separation percentage of 48.15% and rejection percentage of 99.26% so that this membrane is better used as an adsorption membrane.

## ACKNOWLEDGMENTS

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