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Research Article
Characteristics, Biofouling Properties and Filtration Performance of Cellulose/Chitosan Membranes

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Abstract

Background and Objective: As consequence of the increasing demand of membrane technology use, efforts to use natural resources for membrane material are gaining more and more importance. The purpose of this study was to increase fouling resistant of Cellulose Acetate (CA) membranes. **Materials and Methods:** The CA membranes prepared from water hyacinth were modified by addition of chitosan via either blending in phase separation method or via post-modification by surface coating. The membranes were characterized in term of water permeability, surface morphology, surface chemistry and surface hydrophilicity. The biofouling property was examined using Gram-positive and Gram-negative bacteria and protein. **Results:** The results showed that addition of chitosan changed the membrane characteristics, biofouling behavior and filtration performance. Inhibition zone method could not show clearly the antimicrobial activity of chitosan added against both *E. coli* and *S. aureus*. However, visualization of membrane surface clearly showed the antimicrobial activity. **Conclusion:** Overall, the addition of chitosan increased the resistance of CA membrane towards microbial fouling but it did not increase the resistance towards protein fouling.

Key words: Cellulose acetate membrane, chitosan membrane, microbial fouling, antimicrobial activity, biofouling

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

As a crucial material for life, water is becoming an increasingly scarce resource in worldwide. An increasing number of pollution infiltrating into sources of water worsens the quality of water resources. Thus, technology that can improve quality and quantity of raw water is urgently needed. Membrane technology has been proposed as a technology that improves not only water quality but also quantity through recovery. This technology has become a popular separation method in the last two decades for its ability to treat water with very small dosage of chemicals and relatively less energy consumption. The use of membranes in water treatment is focused on two areas of application, namely for drinking water production and coupled with bioreactors for waste water treatment[1-4]. However, membrane applications especially in some countries like Indonesia is still restricted by the high capital and operation cost because the availability of membranes still depends on import. Therefore, the use of local materials for membrane preparation is very important to reduce both capital and operational costs.

Cellulose as one of the most abundant natural polymers found in Indonesia, is very attractive to be used as a membrane material. Cellulose can be converted into Cellulose Acetate (CA) via acetylation process before being used as a membrane material. Recently we have synthesized CA membranes prepared from water hyacinth[5]. The CA is one of the polymers, which are often used as a membrane material. This is because its hydrophilic character, excellent film forming property, biocompatibility and biodegradable[1,6-9]. However, along with its good properties, CA has some weaknesses of poor thermal and chemical resistance, mechanical strength, biodegradability and greater compaction phenomena[10-13]. Another infirmity of CA is easily susceptible to microbial fouling, which eventually reduces the performance of membrane[14] and hinders the applications in water and wastewater treatments. Therefore, efforts to overcome this problem are strongly needed. One of the simple ways is by incorporating chitosan into CA membranes.

Chitosan is another natural polymer obtained by deacetylation of chitin, which its existence in Indonesia is also very abundant. Because its high biodegradability, biocompatibility, nontoxicity and antimicrobial properties, chitosan is widely used as antimicrobial agent either alone or blended with other natural polymers including CA[15,16]. Incorporating chitosan into polymer blends is frequently used to obtain new materials with antimicrobial properties[17].

To improve the antimicrobial property of cellulose film, cellulose/chitosan blend films have been prepared[18]. The antibacterial assessment using *Staphylococcus aureus* proved that addition of chitosan slightly increases antibacterial properties of the films. Hu *et al.*[19] blended quaternized chitosan and carboxymethyl cellulose to form blend films for food packaging. These films improved significantly the microbiological safety of foods. To improve antimicrobial activity of CA/chitosan composite films, silver nanoparticle was conjugated to those CA/chitosan films[20]. All those previous studies were concerned on the preparation of CA/chitosan films and their performance examination.

Recently, Waheed *et al.*[21] synthesized CA membranes with addition of chitosan. The membranes containing chitosan was claimed to be able to inhibit the microbial growth. Nevertheless, this study was dedicated to dense membrane preparation and their filtration performance has not been studied. They also used two different solvents, which will be difficult to be practically applied. Very recently, Akbari *et al.*[22] improved anti-fouling properties of polyamide nanofiltration membrane by coating of chitosan on top of tight NF membrane surface. It is reported that the hydrophilic character of chitosan enhanced water flux without affecting selectivity. The fouling experiments using cetyltrimethylammonium bromide showed that modified membranes by chitosan polymer have relatively better anti-fouling properties than un-modified membrane. To the best of our knowledge, no study has been reported for the preparation of porous polymeric CA/chitosan membranes. The biofouling behavior was mostly examined using bacteria. The biofouling behavior examined using other bio-foulants such as protein has not been conducted. Furthermore, the membrane filtration performance has also not been evaluated. This study was intended to introduce chitosan into CA membrane for minimizing biofouling of porous polymeric membranes. The performance examination was performed using bacteria and protein as model of foulants.

MATERIALS AND METHODS

Materials: Cellulose acetate was prepared by Istirokhatun *et al.*[5]. Commercial cellulose acetate as a reference was purchased from Aldrich, Germany. Chitosan, $(C_6H_{11}NO_4)_n$ was purchased from Biotech Surendo, Indonesia. *Escherichia coli* and *S. aureus* bacteria cultures were obtained from Microbiology Laboratory, Diponegoro University, Indonesia. The Bovine Serum Albumin (BSA) powder was purchased from ICN Biomedicals, Inc. (California, USA). Potassium dihydrogen phosphate (KH_2PO_4) and disodium hydrogen phosphate dehydrate ($Na_2HPO_4.2H_2O$) were purchased from Fluka Chemie AG (Buchs, Germany).

Nitrogen gas was purchased from CV. Rejo Makmur, Semarang, Indonesia. Distilled water produced from home-made pure water unit was used for all experiments.

Methods

CA-chitosan membrane preparation: Two different methods to study the microbial activity of chitosan on/in CA membranes were conducted. The methods included modification by blending of CA and chitosan as antimicrobial additive and post-modification of CA membrane by surface coating with chitosan as outer layer.

Membrane modification by polymer blend using phase separation: Preparation of CA-chitosan membrane by blending included: (1) CA and chitosan were dissolved in acetic acid solution as solvent by stirring, (2) The homogenous polymer solution was left without stirring until no bubble was observed and (3) CA-chitosan membranes were prepared by non solvent induced phase inversion method. The polymer solution was cast on a glass substrate using a casting knife. Thereafter, the cast membrane was solidified in a coagulation bath containing water for 30 min to remove acidity. The resulting membrane was washed and rinsed by soaking in water for at least 24 h before drying.

Membrane modification by surface coating: Modification of CA membranes by surface coating included: (1) CA membranes (without chitosan) were prepared, (2) Thereafter, chitosan solution with a certain concentration was coated on top surface of the already prepared CA membrane in step 1. The membranes were allowed to dry at room temperature for 5 h followed by drying in the oven at temperature of 40°C overnight.

Membrane characterizations: The membrane characterizations included water permeability, surface morphology, surface chemistry and surface hydrophilicity measurements. A JEOL JSM-6510 LA Scanning Electron Microscope (SEM) with 10 kV applied voltage was used to visualize membrane surface morphology. The samples were dried at room temperature and then attached on the sample supports and coated with gold sputtered for 1 min before analysis.

The membrane surface chemistry was observed by using the IR-Prestige-21 Shimadzu, Japan. A total of 32 scans were performed at a resolution of 4 cm^{-1} and the temperature of 21±1°C over the wavelength range of 500-4000 cm^{-1}. The IR solution 1.5 was used to record the sample spectra versus the corresponding background spectra.

Water permeability measurements were carried out using a dead-end stirred cell filtration system (Amicon model 8010), which was pressurized by nitrogen gas. Each membrane was firstly compacted by filtration of pure water at high pressure for at least 0.5-1 h to avoid effects of compaction. Thereafter, the pressure was reduced to the desired pressure for water permeability measurements (1 bar). The flux was gravimetrically measured.

The surface hydrophilicity was observed by measuring the contact angle. The protocols followed our previous studies[23]. Sessile drops static contact angle was measured using a goniometer (First Ten Angstroms, USA). Five microliters of water were dropped on the membrane surface from a micro-syringe with a stainless-steel needle in room temperature (25±3°C) with ~75% RH. At least five measurements of drops at five different locations were averaged to obtain contact angle for one membrane sample.

Antimicrobial activity test: The antimicrobial activity of the membrane was examined by an inhibition zone method, wherein *Staphylococcus aureus* and *Escherichia coli* were used representing Gram positive and Gram negative bacteria, respectively. This method has already been described by Ma *et al.*[24]. To determine antimicrobial activity, all membranes were sterilized in autoclave at 121°C and 2 bars for 15 min. Sterilized membranes were then placed on the surface of treated nutrient agar and incubated at 37°C for 24 h. The diameters of inhibitory zones surrounding the membrane disks were observed. The plates were photographed and the average inhibition zone diameters were observed. Clear zone formed near membrane is noted as an indicator of antimicrobial activity.

Filtration performance: The filtration performance examination was conducted by investigation of adsorption fouling and cross-flow filtration. The experimental set-up and procedures have already been described in detail in the previous studies[25]. The adsorptive fouling experiments were carried out by using a dead-end stirred cell filtration system (Amicon cell models 8010 from Millipore). To avoid the effects of compaction, each membrane was firstly compacted by filtering pure water at 4 bars for at least 0.5 h. Pure water flux (J_0) was then measured for each membrane sample at a pressure of 1 bar. Either *E. coli* or *S. aureus* solution with the concentration of ~10^3 CFU mL^{-1} was added to the cell. Thereafter, the outer membrane surface was exposed for 3 h without any flux at a stirring rate of 100 rpm. Afterwards, the solution was removed and the membrane surface was rinsed twice by filling the cell with pure water (5 mL) and shaking it

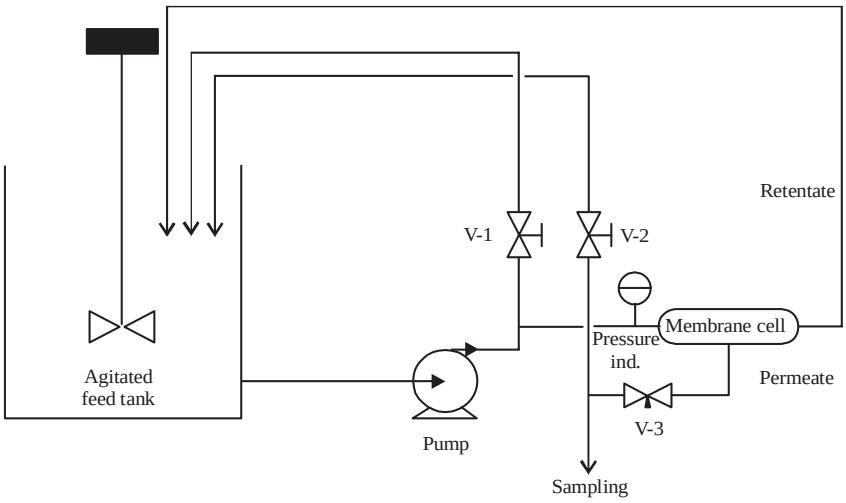

Fig. 1: Schematic diagram of the cross-flow filtration set-up. Source: Susanto and Widiasa[25]

for 30 sec. Pure water flux (Ja) was again measured. The extent of adsorptive fouling was expressed in term of relative water flux reduction (Eq. 1), which was calculated from the water fluxes at the same pressure before and after adsorptive fouling:

$$RFR = \frac{J_0 - J_a}{J_0} \quad (1)$$

A self-home-made laboratory scale filtration test was used in cross flow experiments (Fig. 1). The set-up consisted of a feed thank, a pump, a pressure indicator connected to feed side of membrane to determine the trans-membrane pressure and a flat sheet membrane cell. In each experiment, a new circular membrane disk with effective area 3.14 cm^2 was used. The bacterium culture (either *E. coli* or *S. aureus*) containing approximately 1.86×104 CFU mL^{-1} was diluted up to a concentration of ~10^2 CFU mL^{-1} and then used as a feed during crossflow filtration. In order to maintain constant feed concentration, the retentate and permeate were returned to the feed tank. All experiments were conducted at room temperature (28±2°C). As done in adsorption experiments, the membrane was firstly compacted by filtering the water for at least 0.5 h at a pressure of 3 bar. During UF experiments, the flux profile over time was gravimetrically monitored and the solute rejection was calculated.

RESULTS AND DISCUSSION

CA-chitosan membrane preparation: The CA-chitosan (CA-Ch) membranes were prepared by either polymer blending (CA-b-Ch) or surface coating (CA-c-Ch). Figure 2 shows water permeability of CA-Ch membranes prepared by blending and coating methods.

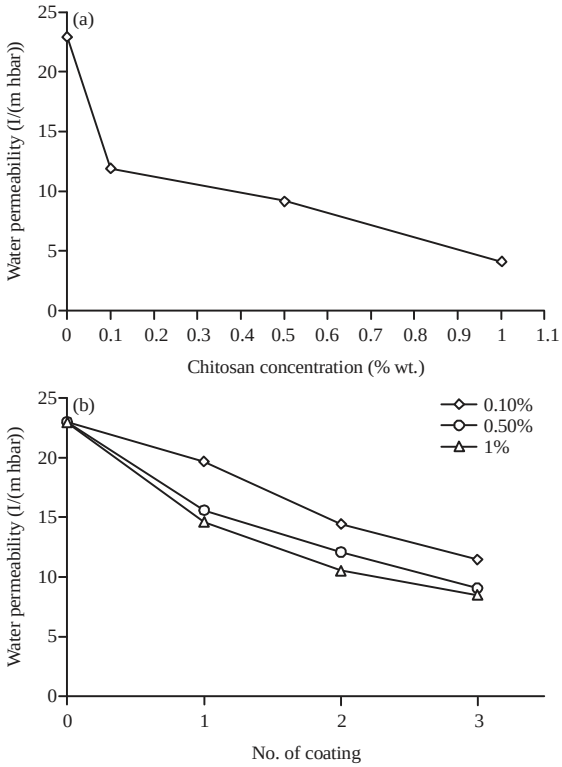

Fig. 2(a-b): Water permeability of CA-Ch membranes, (a) As a function of chitosan concentration (CA-b-Ch) and (b) No. of coating (CA-c-Ch)

It is clearly observed from Fig. 2a that the addition of chitosan decreased water permeability of CA

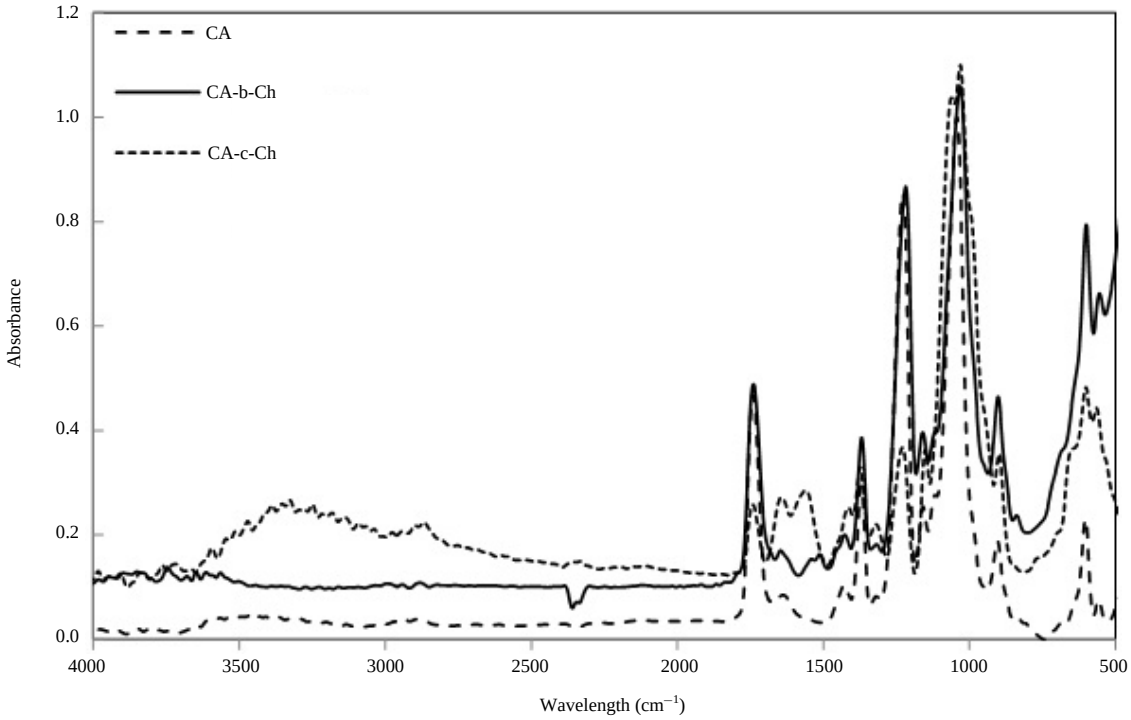

Fig. 3: FTIR spectra of CA and CA-Ch membranes

membranes. As the concentration of chitosan was increased, the water permeability decreased. This phenomenon can be explained that addition of chitosan increases polymer concentration and solution viscosity. The increase in polymer concentration increases polymer fraction in polymer solution, which will decrease the rate of liquid-liquid demixing resulting a membrane with lower porosity. The increase in solution viscosity causes reduction of mutual diffusivities between non-solvent and solvent in the system during solidification of the casting solution. Similar results have been reported in previous studies[26-28].

Coating of CA membrane by chitosan solution decreased the water permeability of CA membrane (Fig. 2b). For the same coating number, water permeability decreased with increasing chitosan concentration. As the number of coating was increased, the decrease in water permeability was more significant. The increase in chitosan concentration and number of coating caused the membrane top layer becomes thicker, which finally increases the membrane resistance towards water permeation. Coating of chitosan on top of CA membrane surface adds membrane resistance via pore narrowing, pore blocking and/or surface layer formation. In addition, surface coverage by chitosan can alter membrane surface hydrophilicity, which also influences water permeability. Different result was reported by Akbari *et al.*[22]. It is showed that the increase in chitosan concentration coated on top of polyamide membrane increases the water flux. The reason was due to chitosan is more hydrophilic than polyamide leading to more water absorption. It should be noted that Akbari *et al.*[22] prepared tight NF membranes therefore membrane surface hydrophilicity has a significant influence on permeate flux.

CA-chitosan membrane characterizations: In order to confirm the presence of chitosan on the membrane surface, the surface chemistry of CA-Ch membranes was examined by using FTIR spectroscopy. The absorption bands of functional groups within the wave number range of 500-4000 cm^{-1} were identified. The results are presented in Fig. 3.

Typical cellulose acetate could be observed from the peaks at 1730 cm^{-1} representing the presence of carbonyl group (C=O), the highest peak at 1025 cm^{-1} representing C-O-C bond and the peaks at 1360 and 1220 cm^{-1}, which indicate C-H bond from (CH_3). In addition, CA could also be observed by minor peaks at ~3500 and ~2840 cm^{-1} indicating O-H stretching and C-H bond stretching, respectively. This result agrees well with previous studies[21,29,30]. The addition of chitosan via either blending or coating could clearly be observed. The indication of chitosan presence was stronger for CA membrane coated by chitosan (CA-c-Ch) than for CA membrane blended with chitosan (CA-b-Ch). The presence of chitosan could be seen by the peaks at

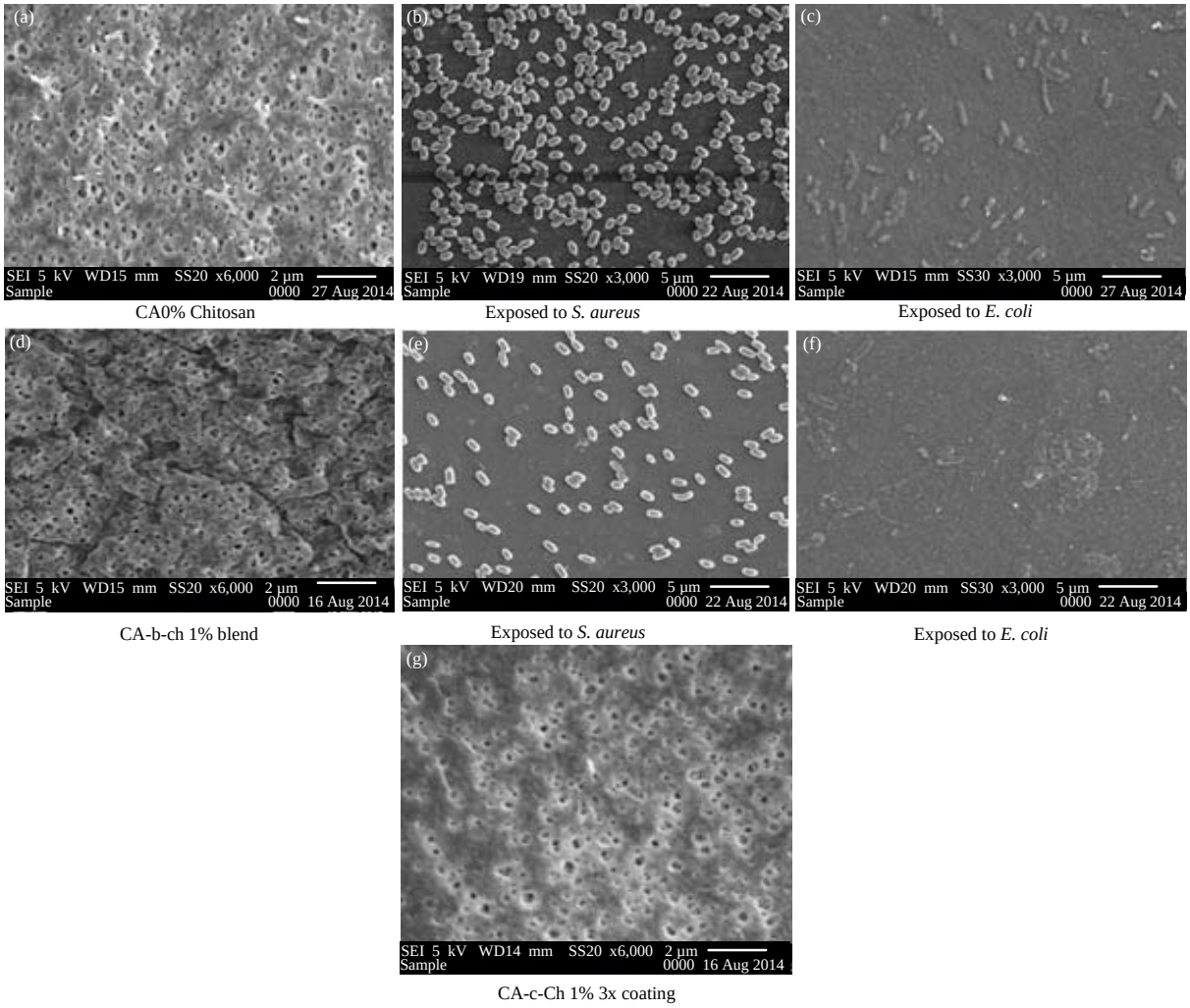

Fig. 4(a-g): SEM images of membrane surface morphology; virgin membrane, after exposing to *S. aureus* and *to E. coli*, respectively

~1550 cm^{-1} for CA-c-Ch and ~1510 cm^{-1} for CA-b-Ch. These peaks represent absorption band of amine bond (NH_2). Slight movement of peak for CA-b-Ch (1550-1510 cm^{-1}) is believed because both CA and chitosan present on the membrane surface. Similar results were reported by previous studies[29,31-34].

Visualization of membrane surface by using SEM (Fig. 4) supports the preceding explanations. It was observed qualitatively that addition of chitosan by blending decreased membrane pore size as well as pore density. In addition, CA-chitosan membrane prepared by blending (CA-b-Ch)showed a rougher surface than both CA membrane (without chitosan) and CA membrane coated by chitosan (CA-c-Ch). The smaller pore density and pore size as well as rougher surface should be the reason for the decrease in membrane water permeability. Visualization of coated membrane showed that both pore narrowing and blocking seemed to occur in some location with pore narrowing was more dominant.

Examination of membrane hydrophilicity by contact angle measurement suggests that addition of chitosan via blending and coating decreased membrane hydrophilicity as evidenced by their higher contact angle (Fig. 5). The reason behind this phenomenon is that pure chitosan has a higher contact angle than pure cellulose acetate meaning that cellulose acetate should be more hydrophilic than chitosan. This explanation is in agreement with Wang *et al.*[35], who showed that the addition of chitosan increases the contact angle of cellulose membrane. Addition of chitosan via surface coating should actually result in higher contact angle than by blending. It should be noted that the hydrophilicity of the membrane is influenced not only by the membrane material but

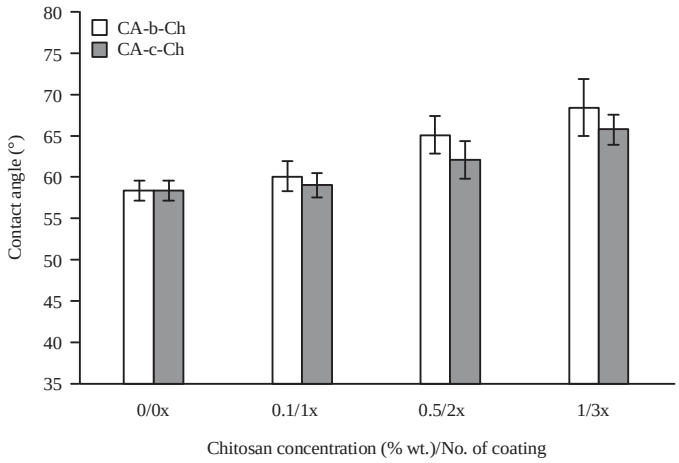

Fig. 5: Effect of chitosan concentration (for blend) or number of coating on contact angle of CA-Ch membranes

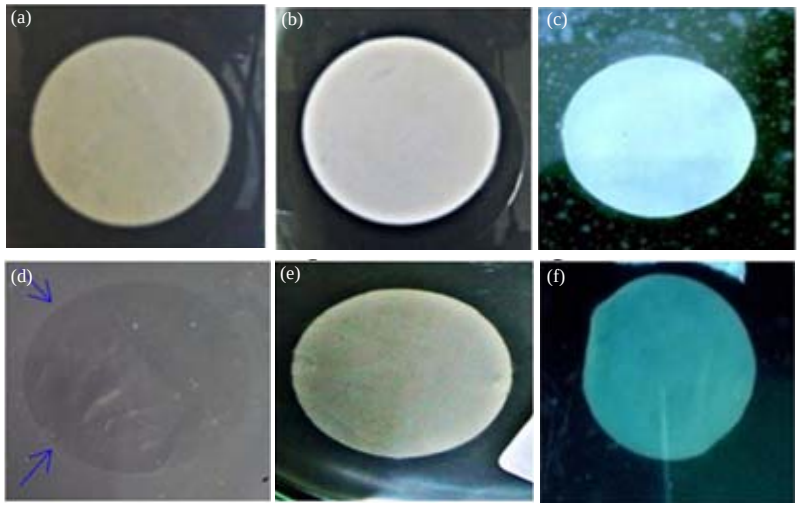

Fig. 6(a-f): Antimicrobial activity by inhibition zone test of different membranes against *E. coli* and *S. aureus*, (a) CA membrane exposed to *E. coli*, (b) CA-b-Ch (1%) membrane exposed to *E. coli*, (c) CA-b-Ch (1%) membrane exposed to *S. aureus* (d) Chitosan membrane exposed to *E. coli*, (e) CA-c-Ch (1x) membrane exposed to *E. coli* and (f) CA-c-Ch (1x) membrane exposed to *S. aureus*

also by the membrane structure such as membrane porosity and surface roughness[28]. The lower porosity, the higher contact angle of membrane. Further, contact angle is increased by increasing membrane surface roughness.

Antimicrobial activity test: The effect of chitosan on antimicrobial activity of CA-Ch membrane was examined by an inhibition method. Sterilized CA and CA-Ch with various chitosan concentrations (0.1, 0.5 and 1%) membranes were exposed to *S. aureus* (Gram-positive bacteria) and *E. coli* (Gram-negative bacteria) for 24 h. The results are presented in Fig. 6.

It is shown that CA membrane and all CA-Ch membranes (both blend and coating) did not exhibit clearly inhibition zone around the membranes. These results are in line with previous studies[31,32,36] but contrasts with previous studies[19,20]. In the study of Hu *et al.*[19] and Lin *et al.*[20] clear inhibition zone was observed. These phenomena can be explained as follow. The effectiveness of antimicrobial activity of chitosan is influenced by type of chitosan used and its concentration. Hu *et al.*[19] reported that quaternized chitosan has stronger antimicrobial activity than pure chitosan[19,37]. Incorporating of nanoparticles, which have antimicrobial activity, into chitosan may also another reason for the observed inhibition zone[20]. The other explanation is that the

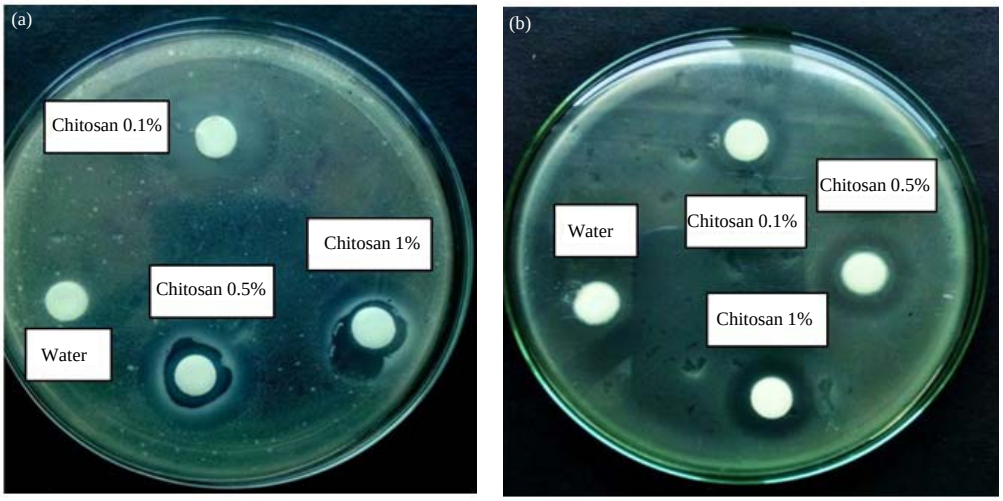

Fig. 7(a-b): Antimicrobial activity of membrane examined by inhibition zone dropped by either water or chitosan solution against (a) *S. aureus* and (b) *E. coli*

chitosan content is significant aspect to obtain clear inhibition zone. In this study, the chitosan content in the membranes prepared in this study is not high enough to produce inhibition zone. This explanation is supported by clear inhibition zone demonstrated by pure chitosan membrane.

Because chitosan as antibacterial agent is well known[21,38-41], further investigation of antimicrobial agent of chitosan was conducted. First, the experiments were performed similar with inhibition test using CA membrane. Thereafter, either water or chitosan solution with different concentration was dropped and flattened on the surface of CA membranes. The results are presented in Fig. 7.

It is shown that the antimicrobial activity of chitosan solution (in liquid form) against both *E. coli* and *S. aureus* was clearly seen. The degree of inhibition zone increased with increasing concentration of chitosan. These results suggest that beside type and concentration of chitosan, mobility of chitosan has also influence on the degree of antimicrobial activity. Liquid chitosan should be more mobile than solid chitosan.

As complement for the antimicrobial test by inhibition method, visualization of bacteria on the membrane surface was performed by SEM. The results are presented in Fig. 4. The (pure) CA membrane demonstrated a significant amount of bacteria on the membrane surface. It was observed that attachment of *S. aureus* on the (pure) CA membrane is more significant than attachment of *E. coli*. The blend membrane with 1% chitosan (CA-b-Ch) showed much less adhesion than the pure CA membrane for both *E. coli* and *S. aureus*. It was also observed that clustering bacteria was observed for *E. coli*.

The SEM images showed that the antimicrobial activity of CA-Ch membrane against Gram positive bacteria (*S. aureus*) is slightly better than against Gram negative (*E. coli*). This result agrees with previous results conducted by No *et al.*[42] and Tao *et al.*[43]. Tao *et al.*[43] reported that chitosan was more effective as an antibacterial for *S. aureus* compared with *Pseudomonas aeruginosa*, which is a Gram-negative bacterium. Using quaternized chitosan, Hu *et al.*[19] reported that the antimicrobial activity for *S. aureus* was more significant[19] than *E. coli*. The reason is because *E. coli* has a relatively less permeable, lipid-based outer membrane. This explanation is supported by the study reported by Azizi *et al.*[44]. Different results were reported by Alishahi and Aider[45] and Prescott *et al.*[46]. They reported that chitosan was more effective as antimicrobial for Gram-negative than for Gram-positive bacteria. The reason was because Gram-negative bacteria are more hydrophilic than Gram-positive bacteria. The cell surface of Gram-negative bacteria has negative charge due to lipopolysaccharides containing phosphate and pyrophosphate groups. The cell wall of Gram positive bacteria is thicker than Gram-negative bacteria[47]. Different explanation has been proposed by Xing *et al.*[48]. They stated that cell wall of *E. coli* consists of thin membranes (peptidoglycan) and an outer membrane composed of lipopolysaccharide, lipoproteins and phospholipids. Meanwhile, the peptidoglycan layer of the *S. aureus* cell wall consists of a network with many pores. This causes foreign molecules can enter into the cell easily. Because *S. aureus* has no Outer Membrane (OM) to prevent the entry of foreign molecules, so *S. aureus* was more sensitive toward chitosan than *E. coli*. The explanation, that

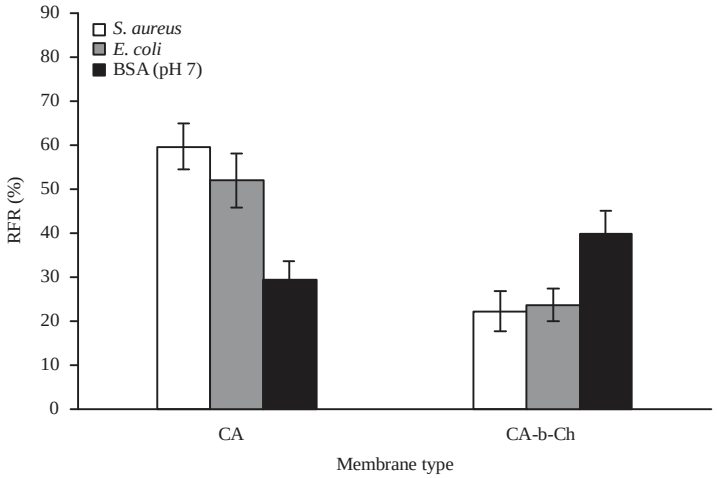

Fig. 8: Relative Flux Reduction (RFR) of CA and CA-b-Ch membranes after exposing to *S. aureus*, *E. coli* and BSA. The error bars represent standard deviation

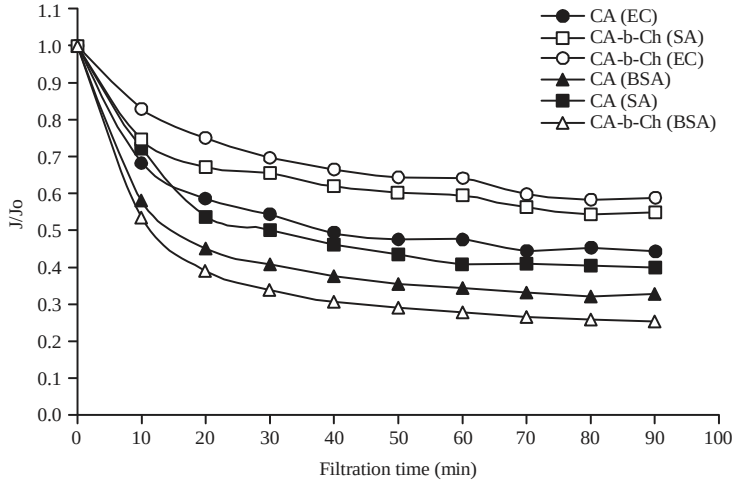

Fig. 9: Normalized flux (J/Jo) profile of CA and CA-b-Ch membranes with different feed solutions at a constant pressure of 1 bar

can be used to explain these different results in different studies, is the different characteristics of chitosan used (solubility, molecular weight, degree of deacetylation, concentration and pH medium).

In general, the antimicrobial activity of chitosan can be explained by several mechanisms[16,20]. The interaction of positively charged amine groups of chitosan with negatively charge bacterial cell membranes causing leakage of essential intracellular components such as proteinaceous, which eventually causes the death of bacteria[18,49-51]. Mei *et al.*[52] reported that the antimicrobial activity of chitosan due to the ability of chitosan to form a polymer layer on the surface of the bacteria. This layer hinders the transport of nutrient leading to the death of bacteria. Because chitosan used in this study is in solid phase entrapped in cellulose polymer, the mechanism proposed by Mei *et al.*[52] should not be the reason for this study.

Filtration performance: The filtration performance of membrane was investigated by adsorptive fouling and cross-flow filtration experiments. The results are presented in Fig. 8 and 9.

It was clearly observed that CA membrane showed a higher RFR after exposing to both *S. aureus* and *E. coli* solutions than CA-b-Ch membrane. For the CA membrane, RFR after exposing to *S. aureus* was the highest among all the foulants used indicating that the interaction of CA membrane *S. aureus* was the strongest among others. The effect of chitosan on RFR could be seen clearly. Interestingly, for bacterial foulant chitosan was able to reduce RFR

Table 1: Membrane rejection for different foulants

	Rejection (%)		
Membrane	*E. coli*	*S. aureus*	BSA
CA membrane	91.4±2.1	89.8±3.1	73.4 ±2.9
CA-b-Ch	96.3±3.2	98.2±2.6	78.8±4.2

indicating less fouling. The effect of lowering RFR was slightly higher for *S. aureus* (59.5-22.2%, which corresponds to 63% reduction) than *E. coli* (51.7-23.6%, which corresponds to 54% reduction). The RFR experimental results are consistent with the results obtained by visualization of membrane surface, where the effect of antimicrobial activity was slightly higher for *S. aureus* than for *E. coli*. Surprisingly, the presence of chitosan in the membrane increased RFR for protein fouling. This means the presence of chitosan increases the extent of adsorptive fouling by protein solution. The charge interaction between positively charge of membrane (amine groups from chitosan) and negatively charge of BSA (at pH 7, BSA should has positive charge) is the possible reason.

Cross-flow filtration results are plotted as permeate flux relative to initial water flux vs. filtration time (Fig. 9). Rapid flux decline in the early stage of filtration followed by gradual decrease was observed for all membranes examined as reported in many previous studies[23,53,54], this rapid flux declined in the early stage of filtration indicated contribution of concentration polarization. However, the difference in J/Jo for different membranes suggests that fouling also contributed to this flux decline. This explanation is supported by the results obtained from the flux measurements after stopping the filtration (for 5 min), which could increase the J/Jo but it was only within the range 5-13%. Overall, the results observed in cross-flow filtration experiments are in a good agreement with the results obtained from adsorptive fouling. The presence of chitosan could increase the J/Jo for the feed containing both *E. coli* and *S. aureus*. These results suggest that addition of chitosan in CA membrane increased the membrane resistance toward microbial fouling. The increase for J/Jo of CA-b-Ch membrane compared to CA membrane was slightly higher for the feed containing *S. aureus* (37%, 0.40-0.55) than for the feed containing *E. coli* (33%, 0.44-0.59). The presence of chitosan slightly increased membrane rejection for all foulants (Table 1). There is no significant different rejection for *E. coli* and *S. aureus* for both CA and CA-b-Ch membranes.

Different result was observed for the feed containing protein (BSA) solution. The CA-b-Ch membrane showed lower fluxes than the CA membrane indicating more severe fouling has taken place. Analogous to the adsorptive fouling, the charge interaction between negative charge of protein and positive charge of membrane followed by protein deposition forming a gel layer would be the possible reason. This means that the addition of chitosan did not increase the membrane resistance toward protein fouling.

CONCLUSION

Addition of chitosan into CA reduced the water permeability of CA membranes as well as the membrane hydrophilicity. Using inhibition zone method, the antimicrobial activity of CA-Ch membrane could not be clearly seen. However, visualization of membrane surface showed significantly the effect of chitosan on the attachment of both *E. coli* and *S. aureus* bacteria. The presence of chitosan increased the membrane resistant towards bacterial adsorptive fouling and increased the resistance of CA membrane towards microbial fouling but it did not increase the resistance towards protein fouling.

SIGNIFICANT STATEMENTS

- The biofouling behavior of cellulose acetate/chitosan (CA-Ch) membranes was studied
- Chitosan increased the membrane resistance against microbial fouling
- Chitosan did not increase the membrane resistance against protein fouling
- Antimicrobial activity of chitosan could not be seen by inhibition zone method
- Antimicrobial activity of chitosan could be seen via membrane surface visualization

ACKNOWLEDGMENTS

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