

Metal Oxide based Antibacterial Membrane

M Zunita*, I G B N Makertihartha, F A Saputra, Y S Syaifi, and I G Wenten

Department of Chemical Engineering, Institut Teknologi Bandung, Ganesha Street 10, 40132, Bandung, West Java, Indonesia

* Email: m.zunita@che.itb.ac.id

Abstract. Biofouling is one of the major problems that hamper the application of membrane processes. Biofouling causes performance deterioration and higher energy consumption due to membrane pore blockage. In addition, it also results in a higher cleaning frequency and a shorter membrane lifetime which increases membrane operating and maintenance cost. Preparation or modification of membrane with antibacterial properties has been considered as one of the attractive strategies to control biofouling. Recently, the combination of polymeric with biocidal materials has attracted a great attention in the preparation of antibacterial membrane. A lot of metal oxide nanoparticles which have biocidal characteristics have been used to prepare nanocomposite membrane with remarkable antibacterial properties and significant biofouling reduction. According to studies that have been reported, the enhancement of anti-biofouling of the membrane was due to an interaction between the metal oxides and microorganism on the membrane surface which prevents biofilm formation. The enhancement of metal oxide nanoparticles antibacterial effect has been studied, including the elaboration of nanostructured oxide consisting of two or metallic components. In this paper, the metal oxide-based antibacterial membrane is comprehensively reviewed. In addition, mechanism of metal oxide particles in preventing biofilm formation will be discussed.

Keywords: antibacterial membrane, biofouling, biofilm, metal oxide, nanoparticle

1. Introduction

Several technologies have been used for water and wastewater treatment, such as filtration, sedimentation, flocculation, coagulation, activated carbon adsorption, and biologically active sludge technology [1, 2]. Among all of water and wastewater treatments, membrane processes are more favorable as they are cheap, fast, highly selective, and flexible to be integrated with other processes [3-10]. Although there are so many major benefits of membranes over conventional technologies, their worst facing problem is biofouling [11]. The main consequence of fouling problem is blockage of the pores of membranes, which contributes to the tremendous decrease of membrane flux and more energy necessity for filtration [12-15]. Therefore, low biofouling membranes are of major interests in numerous fields including water and wastewater treatment or food industry [16]. This could be achieved by changing membrane's chemical structure as bulk and surface modification or using nanoparticles through the membrane matrix or on the surface [17-21].

Several types of nanoparticles and their derivatives like nanometals have given high attention for some researchers because of their potential antibacterial effects to remove biofilm on the membrane surface [22]. Furthermore, morphological and physicochemical characteristics of the nanometals have been proven to exert an effect on their antimicrobial activities [23]. The exact mechanisms for the



antibacterial effect of nanometals are still being investigated. But, there are two proposed possibilities those are free metal ion toxicity arising from the dissolution of the metals from surface of the nanoparticles and oxidative stress via the generation of reactive oxygen species (ROS) on surfaces of the nanoparticles[24]. In this article, we focused on the latest findings of antimicrobial activity of membranes with metal oxides as nanoparticles for wastewater treatment.

2. Characteristic of Metal Oxide as Antibacterial Agent

In general biofilm formation consist three stages: planktonic and initial interaction stages, development and maturation stages, bacteria detachment stages[25]. Biofilm formation can be stopped by blocking of the initial interaction stages from bacteria toward membrane surface. The attachment and development of biofilms on membrane surfaces is usually controlled by physical washing, such as air scrubbing, crossflow filtration, and backwashing and or using chemical substance such as dispersants and biocides[26, 27]. One of biocides that can be used to prevent the growth of biofilm is metal oxide nanoparticles. This addition of metal oxide on membrane surface aims to prevent initial interaction and development stages of biofilm formation. There are three synthesis methods of metal oxide nanoparticles, namely solution-based, vapor based, and biological based. The choice of synthesis methods will determine physicochemical properties of metal oxide nanoparticles[28]. Many metal oxide nanoparticles have been reported to have a remarkable antibacterial property, such as titanium oxide (TiO_2), silver oxide (Ag_2O), silica oxide (SiO_2), cuprum oxide (CuO), and much more[28, 29].

Many studies have reported how the metal oxides can have biocidal activity. But, there are two more popular proposed antibacterial mechanism of metal oxide nanoparticles: 1) Free metal ion toxicity arising from the dissolution of the metals from surface of the nanoparticles, 2) Oxidative stress via the generation of reactive oxygens species (ROS) on the surfaces of nanoparticles[24]. The antibacterial properties of metal oxide nanoparticles were affected by morphological and physicochemical properties of nanomaterials[22, 30]. Size and shape of metal oxide nanoparticles also affected its antibacterial activity[29]. Table 1 shows the antibacterial activity of each nanoparticle and its physical properties that affect their antibacterial activity.

Table 1. Antibacterial mechanism of several metal oxide nanoparticles.

| Nanoparticles | Mechanism | Characteristic | Influence factor | Ref. |
|-----------------------|--|---|--------------------------------------|-------------|
| Ag_2O | DNA loses its replication ability and the cell cycle halts at the G2/M phase owing to the DNA damage | High antimicrobial activity against both bacteria and drug-resistant bacteria, antifungal activity on spore-producing fungal plant pathogens, high stability, nontoxicity. | Particle size and shape of particles | [31] |
| ZnO | ROS generation on the surface of the particles; zinc ion release, membrane dysfunction; and nanoparticles internalization into cell. | Photocatalytic activity; high stability; bactericidal effects on both Gram-positive and Gram-negative bacteria; antibacterial activity against spores which are resistant to high temperature and high pressure | Particle size and concentration | [23, 32-36] |
| TiO_2 | Oxidative stress via the generation of ROS; lipid peroxidation that cause to | Suitable photocatalytic properties; high stability; effective antifungal for | Crystal structure, shape and size | [35, 37-40] |

| | | | | |
|-------------|---|--|--------------------------------------|--------------|
| | enhance membrane fluidity and disrupt the cell integrity. | fluconazole resistant strains. | | |
| CuO | Crossing of nanoparticles from the bacteria cell membrane and then damaging the vital enzymes of bacteria. | Effective against Gram-positive and Gram-negative bacteria; high stability; antifungal activity. | Particle size and concentration | [34, 41, 42] |
| MgO and CaO | Damaging the cell membrane and then causing the leakage of intracellular contents and death of the bacterial cells. | Effective against both Gram-positive and Gram-negative bacteria; high stability; low cost; availability. | Particle size, pH and concentration. | [43-47] |

3. Modification Membrane with Metal Oxide Nanoparticles

Biofouling is the result of interactions of biomass (including bacterial cells, cell debris, extracellular polymeric substances (EPS, etc.) with the membranes, so that it makes biofilms formation or accumulation of microbial products on the membrane surfaces even in the membrane pores[26]. To prevent this biofouling, we need to modify the membrane using the antibacterial agent, like metal oxide. Many researches show the modified membrane using a metal oxide that gives the membrane antibacterial properties and also improved the membrane properties. Nanoparticles play two critical roles in membrane forming: 1) acting as structurally strong members with greater compact and fouling resistance and 2) acting as materials to produce desirable membrane morphology [41, 48-50]. One of the common methods to synthesize metal oxide doped membranes is to use well-controlled and homogenized metal oxide particles as additives to the casting solution prior to synthesis procedures such as phase separation/immersion precipitation[51]. Metal oxides represent an important class of materials used for developing matrix membranes with enhanced properties in comparison with the purely polymeric membranes[52]. The result of polymeric membranes using metal oxides additives can be shown in Table 2.

Table 2. Features of metal oxide based antibacterial membrane.

| Matrix | Nanoparticles | Result | Ref. |
|--------------------------------|--|--|--------------|
| Polyvinylidene fluoride (PVDF) | TiO ₂ | High antifouling properties, increased photocatalytic properties | [31] |
| | Al ₂ O ₃ | Enhanced antibiofouling ability and flux recovery ratio | [23, 32-36] |
| Polyether sulfone (PES) | TiO ₂ , Al ₂ O ₃ and ZrO ₂ | Higher affinity, reduced membrane fouling resistance, increased hydrophilicity and rejection | [23, 32-36] |
| Polyvinyl chloride (PVC) | TiO ₂ | Greater water flux, enhanced antifouling properties, increased hydrophilicity | [35, 37-40] |
| Polysulfone (PSF) | ZnO | Lower contact angle, higher hydrophilicity, greater water flux | [34, 41, 42] |
| | TiO ₂ | Enhanced mechanical strength, hydrophilicity, antifouling ability and permeability | [34, 41, 42] |
| | SiO ₂ | Improved water flux, protein rejection and antifouling tendency | [34, 41, 42] |
| | ZnO | Enhanced water flux, hydrophilicity, permeability, porosity, rejection tendency and fouling propensity | [34, 41, 42] |

As we know from Tabel 2, that all modified polymeric membranes have improved its performance from its pristine membranes. Some result using polysulfone membrane which added with Zinc Oxide, Titanium Oxide, and Silica Oxide can be seen in Table 3 respectively.

Table 3. Filtration Properties of Pristine PSf Membrane and Modified PSf membrane with Metal Oxides[53-55].

| Parameter | Pure PSf | PSf-ZnO | PSf-ZnO-GO | PSf-TiO ₂ | PSf-TiO ₂ -GO | PSf-SiO ₂ | PSf-SiO ₂ -GO |
|--------------------------------------|----------|---------|------------|----------------------|--------------------------|----------------------|--------------------------|
| Concentration (% wt) | - | 2 | 0.6 | 0.5 | 0.5 | 0.3 | 0.3 |
| Porosity (%) | 75.2 | 86.7 | 90 | N/A | N/A | N/A | N/A |
| Mean RMS (nm) | 5.09 | 3.45 | 4.03 | 27.02 | 22.53 | N/A | N/A |
| Contact angle (degree) | 65.9 | 39.6 | 38.5 | 70.51 | 68.39 | 64 | 63 |
| Permeability (L/m ² hbar) | 0.89 | 2.83 | 5.11 | 0.55 | 0.58 | N/A | N/A |
| Pure water flux (L/m ² h) | 11 | 35 | 52 | 140.52 | 297.65 | 241 | 379 |
| Flux recovery ratio (%) | 85 | 97 | 99 | N/A | N/A | 69 | 72 |

From Table 3 shows that the addition of metal oxide nanoparticles successfully improve the performance of polysulfone membrane. This result also shows, that metal oxide not only gives membrane antibacterial properties but also increase filtration characteristic of the membrane. Antibacterial activity from the modified polymeric membrane using metal oxide can be observed using Scanning Electron Microscope (SEM).



Figure 1. The illustration of antibacterial activity on the membrane surface. Adopted from[53, 56]

Based on Figure 1 (the illustration of antibacterial activity on the membrane surface) shows that the addition of metal oxide can decrease microorganisms attachment on the membrane surfaces. The metal oxide on the polysulfone membrane surface can remove almost all bacteria colonies by using ZnO-GO and Ag-GO antibacterial agents. Other than that, the addition of Ag-SiO₂ and Ag@TiO₂-CNTs make the bacteria colonies die. The antibacterial activity of metal oxide will vary depending on its antibacterial mechanism, from Figure 1 emphasizes that the number of bacterial colonies on the membrane surface due to the amount or type of different metal oxides.

4. Challenge on Preparation of Metal Oxide Based Antibacterial Membrane

To improve membrane performance, many modifications have been made to the membrane preparation [51, 57-60]. Membrane preparation with the addition of metal oxide can increase membrane antibacterial, antifouling, and membrane filtration properties. Antibacterial mechanisms in the metal oxide involve the release of metal ions into the water which causes the amount of metal oxide in the membrane surface decreases [22]. The improvement of interaction between metal oxide and membrane surface is required to reduce the rate of metal ions released from the membrane surface so that maintain the antibacterial activity can go longtime, moreover can reduce soluble metal ions concentration in water. From Table 5 shows that combining two metal oxides or with another metal nanoparticle can increase membrane performance. There many studies reported that optimized metal

nanoparticles exhibited better antibacterial properties than a single metal oxide [61-65]. The other challenge is to combine the metal oxide with another natural resource, to give better antibacterial activity and friendly to the environment. Some research has shown the combination from ZnO and Eugenol to give antibacterial activities in polysulfone membrane [66]. Next challenge to improve the antibacterial property of membrane is combine various polymeric membranes with nanoparticles to find the most appropriate combinations and applications of the membrane fabricated. For example, in drinking water application, metal oxide must handle in carefully due to potentially its toxicity toward the human. Moreover, the antibacterial agent activity that only prevents microorganism growth on membrane surface. Beside of that, the polymeric materials and nanoparticles researches need to be optimized so that cost of production more competitive, give high performance and longtime antibacterial activity of membranes.

5. Conclusion

This current review is aimed to discuss about the utilization of metal oxide nanoparticles in membrane to prevent biofouling activity in the membrane surface. Based on these and earlier studies, Membrane is an advanced treatment technology that has many benefits such as cheap, highly selective, using less chemicals and energy, simple design, and easy to maintenance. Membrane also shows great performance for wastewater treatment application in many industries by using physical filtration processes or membrane bioreactor. In other hand, biofouling causes the blockage of the membranes pores and reduces the membrane performance. But, it can be removed by modifying the membrane with antibacterial agent like metal oxide nanoparticles. The proposed antibacterial mechanism of metal oxide nanoparticles are free metal ion toxicity from dissolution of metal oxide nanoparticles and oxidative stress via the generation of ROS on the surface of nanoparticles. The antibacterial properties of metal oxide nanoparticles was affected by morphological and physicochemical properties of nanomaterials such as crystal structure, shape, size, concentration, and pH. The modified membrane using a metal oxide nanoparticles has better antifouling properties and also improved other properties such as mechanical strength, water flux, hydrophilicity, permeability, porosity, and rejection tendency. It is expected that this review may be able to enhance further research into the development of metal oxide nanoparticles in membrane technology for many application, especially wastewater treatment.

6. References

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