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A Review of Biopolymer-Based Membrane and Its Application in Oil Emulsion Wastewater Treatment

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Abstract - *This review paper will conduct mainly about oil-water emulsion treatment using biopolymer-based ultrafiltration membrane. Oil emulsion wastewater mainly use ultrafiltration process for its treatment due to its continuable process, high efficiency and low energy usage but they are relied on conventional non-degradable membrane which is made from synthetic polymer. The usage of conventional non-degradable membrane has given another waste problem in which reduces the sustainability of ultrafiltration process based on environmental perspective, therefore the degradable membrane material should be developed to increase its sustainability and reduce another waste problem. Biopolymer development has reach numbers in several years, it developing within year to year. Biopolymer such as chitosan, alginate and polylactic acid can be applied on ultrafiltration system in which it can be degradable through bio-degradation with or without modification. Modification through biopolymer in membrane fabrication for ultrafiltration will improve some characteristic that can lead to high efficiency and compability in ultrafiltration process especially for oil emulsion wastewater treatment.*

Keywords *– Oil emulsion, wastewater, ultrafiltration, biopolymer membrane.*
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1. Introduction

Oil emulsion wastewater classified as hazardous waste to the environment. The wastewater that contains oil emulsion usually came from various industrial processing in metal industries, automotive industries, transportation industries, petrochemical industries, food industries and oil gas industries [1]. Oil emulsion wastewater has high complexity in its composition (benzene, toluene, ethylbenzene, xylene, phenol, poli-aromatic hydrocarbon, fat, triglycerides) therefore, it can lead to high COD rate without any advanced treatment [2].

Wastewater treatment with high efficiency, high capacity, and low-cost method should be applied due to demand of wastewater treatment in industries would be a continuing process and it may affect to industries cost. Wastewater treatment method, especially for oil emulsions are filtration and absorption. Membrane technology is classified as filtration method and it has been used as an advanced method to process wastewater, especially oil emulsion wastewater. There are two mainly kinds of materials are used in membrane filtration, polymeric membrane and inorganic membrane. Polymeric membrane known as most efficient material at cost for membrane filtration rather than inorganic membrane, but polymeric membrane gives a challenge in disposal problems regarding to its characteristic that is non-degradable [3], therefore the wastewater treatment would give another waste from disposable membrane used for the filtration.

Biopolymer development in recent years will potentially substitute non-degradable membrane material and reduce waste problem due to disposed membrane. Biopolymer-based membrane mainly build from polyglycolic acid, polycaprolactone and polysaccharides [4]. Cellulose-based is the one of polysaccharide-based membrane that is potentially used for nanofiltration and ultrafiltration. Chitosan-based membrane is the one of potential cellulose-based membrane for nanofiltration and ultrafiltration [5] and extensively used in a number of application, it is produced from deacetylation process of chitin and it is soluble in water with small amount of acetic acid [6]. Chitosan is the most important biopolymer used for several applications, especially in biomedical application and separation technology in which membrane technology [7]. Alginate is another polysaccharide-based membrane that commonly used to build membrane material. Alginate is made from copolymers guluronic acid and mannuronic acid, therefore alginate membrane is not classified as cellulose-based membrane, alginate has more instability in water rather than chitosan. Modification of alginate mainly come for increasing stability of alginate regarding to its characteristic that is easy to be modificated with another functional group [8].

2. Membrane technology for wastewater treatment

Membrane technology is mainly used for wastewater treatment due to its capability, cost and efficiency. Membrane technology for filtration is classified as microfiltration (MF), ultrafiltration (UF) and nanofiltration (NF) [9]. Ultrafiltration and nanofiltration are mainly used as advanced method for wastewater treatment due to its efficiency related to pore size, UF uses membrane with pore size 1 – 100 nm and NF uses membrane with pore size < 2 nm either UF or NF use pressurized driving force between 1 – 10 bar [9].

Ultrafiltration (UF) commonly known as a well-established wastewater treatment with good permeate flux and high rejection of macromolecules. There are several factors related to UF mechanism such as size exclusion, molecule shape, charge and hydrodynamic conditions, those can affect the efficiency of rejection in UF process [10]. UF research for wastewater treatment has been conducted by [11] using polyethersulfone (PES) membrane to process high-salinity organic wastewater with crossflow system at 2 bar. It conducted that UF operation is influenced by foulant size, thus the presence of high-salinity affected rejection rate therefore characteristic of wastewater should be considered to choose the method of membrane filtration. Presence of fouling could not be avoided but can be prevented, by considering membrane pore size into wastewater characteristic [12]. UF should be applied to bigger particle size and high complexity of wastewater rather than NF, therefore organic and oily wastewater should be treated by UF method. UF membrane can be modificated through dispersion to increase its flux and rejection rate, [13] modificated polyvinylidenefluoride (PVDF) membrane with TiO_2-LiCl nanoparticles in refinery produced wastewater. It is shown that TiO_2-LiCl nanoparticles on PVDF gives higher hydrophilicity, smaller pore size, higher porosity, higher flux and higher rejection. It concludes that membrane modification through UF method can give better performance on wastewater treatment.

Nanofiltration (NF) is relatively known as newly advanced method in wastewater treatment, NF membrane pore size is nominally ~1 nm [14]. NF is known as more advanced filtration method than UF, it can be used to separate small colloids to small molecular ion therefore NF is mainly applied on smaller molecular weight of the component [15]. NF has been applied in number of wastewater treatment method: pharmaceutical wastewater [16]; textile wastewater [17]; phenol removal in wastewater [18]; concentrated chromium removal in tannery wastewater [19] and dairy wastewater [20]. In general NF is used either to remove non-macromolecules (phenol, azo-dyes, paracetamol and fertilizer) or to enhance UF process in two-stage filtration, due to its pore size NF is not recommended for macromolecules or in this case oil emulsion wastewater regarding to its molecular weight and complexity.

3. Biopolymer-based membrane for ultrafiltration and nanofiltration

Biopolymer development for membrane in ultrafiltration system has reached numbers on recent years. The development mainly focused about to replace conventional UF/NF membrane that is non-biodegradable thus reducing waste problem that is caused by disposal UF/NF membrane. Conventional polymer-based membrane cannot be considered as a sustainable process based on environmental perspective [21]. Chitosan and alginate are the most commonly used as membrane material rather than another biopolymer such as polylactic acid [22] it conducts that chitosan and alginate have similar selective properties to polivinyl acetate, the most synthetic membrane used therefore chitosan and alginate are considered to replace the synthetic membrane.

Chitosan ([1-4]-2-amino-2-deoxy-D-glucon) is classified as polyaminosaccharide with glucosamine and N-acetylglucosamine unit. Chitosan is made from deacetylation process of chitin, chitin itself can be extracted from *crustacea* family [23]. Chitosan as biopolymer-based membrane are applied into antibacterial filter [24]; palladium removal [25]; mercury and arsenic removal by ultrafiltration [26]; dyes purification by nanofiltration [27]. In general, chitosan can be applied as ultrafiltration membrane and nanofiltration membrane also can be modificated with another specimen such as metal ions, polymer and another biopolymer also, chitosan is potentially can be used to substitute conventional non-degradable polymeric membrane that is recently used in UF/NF system.

Alginate ([1-4]-β-D-mannuronic-α-L-guluronic acid) are polysaccharides isolated from brown algae and known as copolymer from D-mannuronic acid and L-guluronic acid in alternating blocks [28]. Alginate as biopolymer-based membrane are applied into dye removal [29]; seawater desalination through pervaporation [30]; oil emulsion separation [31]; ethanol pervaporation [32]; hemodialysis membrane [33]. Alginate is generally used in salt form such as sodium alginate or calcium alginate, it is rely on the characteristic of alginate that consists of acid unit also it can increase solubility of alginate in water [34]. Alginate consists of acid functional group, therefore it can be modificated into ester form to give more stability and dependency of the molecules when it interacts with water [33]. In general, alginate can be applied either as ultrafiltration membrane or nanofiltration membrane due to its modification, also alginate is potentially can be used to substitute conventional non-biodegradable polymeric membrane that is recently used in UF/NF system.

4. Oil emulsion wastewater treatment

Oily wastewater recently being a serious problem due to its impact on environment caused by evaporation of oil and hydrocarbon contents. Oily wastewater contains emulsion form and mixture of oil and water, the oil fraction can be fats, hydrocarbons, and petroleum compound such as diesel oil, gasoline and kerosene [35]. It demands an advanced process to treat the wastewater due to its complexity in compound. Number of research about the treatment of oil emulsion wastewater has been conducted, electrochemical [36]; coagulation and flocculation [37]; biological treatment [38]; membrane ultrafiltration [31]. Every method gives their advantages and disadvantages, electrochemical offers significant decreasing on COD number but it demands high maintenance due to existence of electrode; coagulation and flocculation offers cheaper cost and simple process but it gives lower selectivity and effectivity; biological treatment offers high mobility of COD and TOC removal but demands longer process and more utilities; membrane ultrafiltration offers high mobility in process and gives rapid-continuous process with high selectivity but it demands high cost on investment [37–40].

Membrane ultrafiltration currently developed to be an advanced treatment for oil emulsion wastewater treatment. It gives high selectivity, rapid process and low energy consumption [31]. Huang *et al.* (2015) conducts about oily wastewater treatment by UF using PVP (polivinylpyrrolidone) grafted PVDF (polivinylidenefluoride) membrame, it gives that PVDF-PVP membrane gives improvement on oily wastewater separation process rather that PVDF membrane [41]. Salahi *et al.* (2015) conducts about oily wastewater treatment by UF using PES (polyethersulfone) membrane, it gives that PES UF process on oily wastewater treatment gives TOC rejection about 96.3%, COD rejection 83.1%, turbidity 99.3%, OGC 99.7% with high permeation flux of 84.1 L/m^{2}h and fouling resistance around 63.0% [42]. Gholami, *et al.* (2020) conducts about oily wastewater treatment by UF using CuBTC-PES membrane, it gives that CuBTC-PES gives higher performance on oil rejection (99% rejection rate) rather than conventional PES membrane. Other than that, CuBTC addition on PES membrane will give better performance due to its anti-fouling properties with flux recovery ratio on 81% (tested by 8000 mg/L milk powder solution) and flux recovery ratio on 99% (tested by 4000 mg/L oily-water emulsion). It compiles that PES modification through CuBTC mixed matrix membrane modification will give better performance especially with oil-emulsion separation [43]. In short, membrane ultrafiltration can be used as oil emulsion wastewater treatment with several polymers used as membrane material but most of them are non-degradable material. Therefore, there should be a recommendation for using a bio-degradable material to reduce more waste produced from membrane material to reach a sustainable process based on environmental perspective.

5. Biopolymer-based membrane capability in oil emulsion wastewater treatment

Due to waste problem caused by usage of non-degradable membrane for ultrafiltration process in oil emulsion wastewater treatment, development of biopolymer-based membrane has been conducted in recent years. Gao *et al.*, (2018) conducts about preparation of modified alginate UF membrane with Cu^{2+} construction to aim superwetting property for highly efficient oil-water emulsion separation. Crude oil-water emulsion has ability to increase fouling in UF process due to its highly adhesive property, therefore it would be problem for continuing process in UF. To prevent fouling in UF membrane, a nanosized membrane pores with controllable anti-fouling properties by Cu^{2+} construction through alginate hydrogel multilayer membrane has been fabricated, Cu^{2+}/alginate multilayer UF membrane gives biomimetic hydrophilicity, underwater superoleophobicity, and anti-fouling ability from crude oil from addition of Cu^{2+}. It has been proven in its capability to crude oil-water emulsion with efficiency of 99.8% and high-water flux of 1230 L m^{-2} h^{-1} bar^{-1}. It conducts that alginate has capability in oil-water emulsion separation thus it will be potential to be applied on oil-water wastewater treatment [31]. Fan *et al.* (2020) conducts about preparation of superhydrophobic filter made from biopolymer-based polylactic acid (PLA) used for oil-water separation. Oil-water separation can be performed by hydrophobic-oleophilic interaction, oil can be adsorbed by oleophilic layer as permeate thus water can flow out as retentate. In that study, nonwoven PLA filter is made with non-solvent assisted phase separation method. The capability of PLA in oil separation has reached 99.5% (soybean oil), 98% (n-hexane), 97.5% (styrene), 97.5% (diesel oil), 96% (n-heptane), and 95% (silicone oil), respectively. It found that silicone oil has low separation efficiency due to its relatively large density, thus it resulted in blocking some of pores in PLA nonwoven filter [44]. Therefore, PLA is not recommended to be used for separation of high-density component. Ghorbani, *et al.*, (2021) conducts about preparation of polylactic acid (PLA) based membrane blended with several polymer: polybutylene succinate (PBS); polypropylene carbonate (PPC); and polyhydroxy butyrate (PHB) with and without silica nanoparticles (SNPs) addition. Two membranes are produced in that research, M1 (contains: 38.67% PLA; 16.57% PPC; 25.78% PBS; 9.67% PHB; 4.297% TEC and 5% SPNs) and M2 (contains: 40.71% PLA; 17.45% PPC; 10.18% PBS; 27.14% PHB; 4.523% TEC and 0% SPNs) both of M1 and M2 membranes give 98.6% efficiency in oil separation, also TDS removal of 11.3%, manganese removal of 14.17%, iron removal of 22.56%, and turbidity removal of 89.15%. M1 membrane gives more improvement in thermal and mechanical properties due to addition of silica nanoparticles with high porosity and uniform surface. Addition of SNPs can improve the surface of PLA based membrane with high porosity [45], therefore PLA can be used in oil-water emulsion with SNPs modification thus

Ghorbani, *et al.*, (2021) research has been improved Fan *et al.* (2020) research about usage of PLA in oil-water separation.

6. Conclusion

Oil emulsion wastewater treatment mainly uses ultrafiltration process due to its continuable process, high efficiency and less energy usage. UF process still mainly uses non-degradable membrane material which can reduce its sustainability in environmental perspective, therefore a biopolymer-based membrane should be developed for reducing the usage of non-degradable membrane material which can lead to increase its sustainability. Modification of biopolymer-based membrane will give high efficiency and reduce fouling due to the complexity of oil emulsion wastewater. There has to be more research to develop biopolymer-based membrane regarding to its capability that can substitute conventional non-degradable membrane to increase high sustainability in ultrafiltration process, especially in oil emulsion wastewater treatment.

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