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기름/물 분리를 위한 제올라이트 기반의 세라믹 분리막에 대한 총설

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A Review on Zeolite-based Ceramic Membrane for Oil/Water Separation

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요 약: 정유소와 석유 공장에서 발생하는 폐수는 심각한 환경오염으로 이어진다. 기름이 있는 물을 정수 처리하는 데에는 많은 방법이 존재하지만, 가장 효과적인 방법은 막을 이용한 기술이다. 물에서 기름기를 제거하는 데 사용되는 유기재료로 만들어진 고분자 막은 파울링이라는 고질적인 문제를 가지고 있다. 무기성 막은 수명이 길다는 점에서 유기성 분리막보다 효율적이다. 제올라이트 막은 우수한 화학적 안정성을 갖고 있으며 오랜 기간 재활용할 수 있다. 막에서 친수성의 존재는 막의 수 투과량을 증가시킨다. 제올라이트로 만들어진 세라믹 분리막은 물과 기름을 분리하는 데 사용되는 효율적인 무기막 중 하나이다. 본 리뷰논문은 i) 순수 제올라이트 막과 ii) 다른 물질과 혼합된 제올라이트 복합막, 2가지로 분류되는 제올라이트 기반의 무기막을 사용하는 물과 기름 분리 기술을 중점으로 다루고 있다.

Abstract: Wastewater from refineries and petroleum plant lead to severe environmental pollution. There are various existing processes applied for oily water treatment, but membrane-based technology is one of the most efficient methods. Polymeric membranes prepared from organic materials for the separation of oil in water often face chronic problem of membrane fouling. Inorganic membranes are considered to be more efficient due to longer lifetime than organic membranes. Zeolite membranes have better chemical stability and long-term recyclability. The presence of hydrophilicity enhances the water flux of membrane. Ceramic membranes prepared from zeolites are another efficient class of inorganic membranes applied for oil water separation. This review is focused on oily wastewater separation based on zeolite membrane which classified into two categories, i) neat zeolite and ii) zeolite composites with other materials.

Keywords: *zeolite, oil water separation, wastewater, zeolite composites*

1. Introduction

Growing demand of oil leads to increasing activities in refineries and petrochemical plant. Oily wastewater is released from these plants as well as from mining, transportation and various sectors[1-5]. The appropriate treatment of wastewater is highly essential to control environmental pollution along with reuse of treated water in agriculture, plants and other purposes. This will help to reduce the demand of clean water which is

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mainly due to unusual weather condition resulting from global warming. Existing method for oily water treatments is induced air flotation, coagulation and continuous electro-oxidation process. However, membrane technology is considered one of the most efficient and cost effective processes for oily wastewater treatment.

Membranes are generally classified into two types; i) organic-based membrane and ii) inorganic-based membrane[6-10]. Oil in water separation membrane based on organic polymeric materials often faces severe problem of membrane fouling despite various membrane modification techniques. Inorganic membranes have several advantage for oily wastewater separation process such as great chemical stability, high physical stability and long term reusability. Ceramic membrane are usually made up of metal (silica, alumina, zirconia) oxides, metal-organic framework or zeolite.

Advantage of zeolite membrane is low cost, the ease of membrane fabrication, higher surface area, excellent separation properties based on size exclusion mechanism and presence of different porosities[11-13]. It consists of silicone, aluminium and oxygen arranged in crystalline form with tetrahedral framework, resulting in the formation of porosity with various sizes. The hydrophilicity of zeolite membrane enhances the water flux of membrane. The coating of zeolite on the membrane substrate also enhances the membrane performance of membrane. This review is mainly classified on two types; neat zeolite and zeolite composites with other materials. The schematic and performance of zeolite-based membranes for oil/water separation are shown in Fig. 1 and Table 1, respectively.

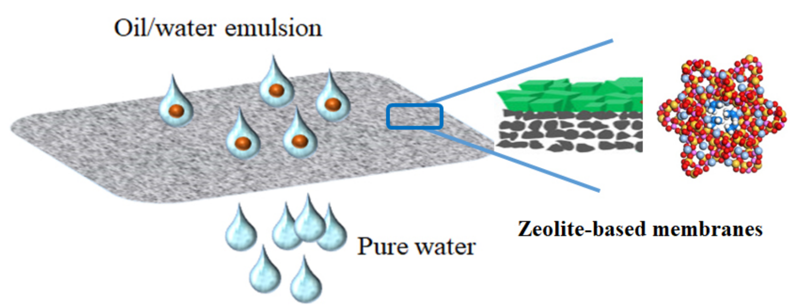

Fig. 1. Schematic illustration of zeolite-based inorganic membranes for oil/water separation.

Table 1. Summary of Zeolite-based Inorganic Membranes for Oil/Water Separation

Membrane	Pure water flux (L/m^2 h)	Oil or TOC rejection (%)	Reference
Silane-grafted sand	100 - 150	99.1	[14]
Silane-grafted sand	144	99.4	[14]
SiC	~162	98.52	[17]
MZ ceramic	-	95 - 99	[18]
MAZ ceramic	-	95 - 99	[18]
MA ceramic	176 - 349	98.4 - 97.2	[20]
MAZ ceramic	549 - 136	97.1 – 96.3	[20]
MZ ceramic	48 – 40	98.7 – 95.9	[20]
SSN@F-FAU	> 50000	98.8	[22]
SSN@F-LTA	> 40000	> 99.0	[22]
Cellulose acetate/nano-zeolite Electrospun nanofiber composite	–	97	[24]

TOC: total organic carbon; SiC: silicon carbide; MZ: mullite-zeolite; MAZ: mullite-alumina-zeolite membrane;
MA: mullite-alumina; SSN: stainless steel net; FAU: Faujasite zeolites; LTA: Linde type A;
SSN@F-FAU: zeolite FAU powder and SSN@FAU coating; SSN@F-LTA: zeolite LTA powder and SSN@LTA coating.

2. Zeolite-based Ceramic Membranes

On account of the merits of inorganic membranes, the use of neat zeolite and its composites materials for oil/water separation has been considerably intensive. Very recently, Aloulou *et al.* prepared hydrophobic silane-grafted sand membrane to separate water from oily wastewater[14]. Various experimental techniques such as Fourier transform infrared (FT-IR), scanning electron microscope (SEM) and nitrogen adsorption/desorption isotherm were used to check the perfluorinated groups, membrane morphology, and average pore size distributions, respectively. Air-gap membrane distillation process was used to purify oily water at trans-membrane pressure (TMP) of 1 bar and at 85℃ and compare the performance with other membranes. The silane-grafted sand membrane showed the best stabilized flux with a huge gap compared to other membranes while maintaining oil retention over 99%. Two other similar, but hydrophilic, membranes were also tested: zeolite microfiltration membrane (MFZ) and titania-smectite nanocomposites ultrafiltration membrane (UFTiSm). Among them, the UFTiSm needs highest TMP, which is 3 bar, while the other two only need 1bar. Overall, three membranes showed similar oil rejection of over 99% and similar permeate flux in the range of 100~150 L/m^2 h.

In another study, Jafari *et al.* synthesized three cheap tubular ceramic microfiltration membranes, i.e. mullite, mullite-zeolite, and mullite-zeolite-activated carbon membranes, from kaolin clay, natural zeolite and activated carbon powder[15]. The oil emulsion droplets size in the feed solution was characterized using dynamic light scattering (DLS). The crystalline structure of fabricated membranes was confirmed by X-ray diffraction (XRD). The permeation flux and total organic carbon (TOC) rejection were checked to analyze the performance of membranes. Through a laboratory scale cross-flow test, they found out that putting activated carbon and natural zeolite in the synthesis of ceramic membranes improved the porosity, permeation flux and TOC rejection. Also, they found out that the permeation flux and TOC rejection are directly proportional to the presence of salt in feed solution and high sintering temperature decreases porosity, linear radial, and longitudinal shrinkage due to the change in phase and release of free silica. Moreover, they found that using alkali as a cleaning agent recovered the membranes up to 65.8% which could be improved. Since the experiment was handled in a small scale, they used an exponential triple smoothing model to forecast the permeation flux of membranes when they are operated longer and obtained the result that the flux of the membrane was still good after two and half hours of purifying oily wastewater. Considering their low cost and use of chemicals and high efficiency, the ceramic membranes fabricated from natural zeolite and active carbon may be a good substituent for conventional methods of treating oily wastewater. Especially, it could be used for oil desalter discharge treatment.

Jafari *et al*. fabricated new two-layer tubular ceramic membranes with better performance from cheap and familiar raw material[16]. First, they synthesized a porous support tubular clay-based ceramic support membrane from 50% of kaolin (< 50 μm), 30% of alumina (< 50 μm), 20% natural zeolite (< 50 μm) along with 32 wt% of water and 2 wt% of starch as a binder by using extrusion technique. Then, they coated the support membrane with natural zeolite and activated carbon by using the cross-flow filtration process to make the two-layer microfiltration membranes. SEM was used to measure the surface and cross-sectional microstructure of the support and MF membranes. No phase transformation was observed, as characterized by wide-angle X-ray diffraction (WAXD). Thermogravimetric analysis (TGA) was also used to check whether the difference in treating temperature influences the thermal behavior of the support membrane. Fig 2 shows the SEM images of the zeolite membranes. From the fabrication, they found that increasing the treating temperature makes the mechanical and thermal properties of the support membrane better, and around 1150°C is the best for treating the ceramic support. Also, they found that the support membrane treated by activated

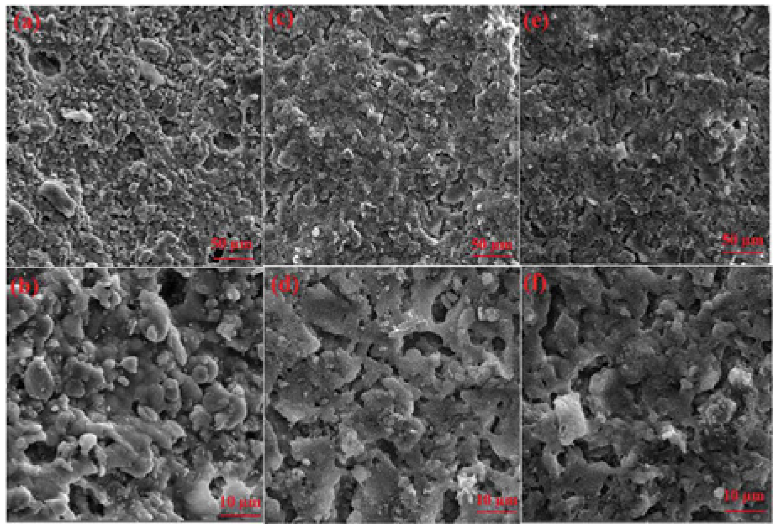

Fig. 2. SEM images of the surface of (a and b) support membrane, (c and d) MF-Z membrane and (e and f) MF-C membrane (Reproduced from Jafari *et al*., 16, Taylor & Francis).

carbon showed better rejection and permeate flux than those by zeolite.

Jiang *et al.* lowered the sintering temperature of porous silicon carbide (SiC) membranes which is usually exceeds 2000℃[17]. In order to do that, they put residues, NaA(r), came out from producing NaA zeolite in sintering process. By doing so, they achieved lower sintering temperature which is about 1000℃ with no noble gas protection. SEM was used to identify the microstructure of the membrane. XRD and X-ray photoelectron spectroscopy (XPS) were used to analyze the sample phase. DLS was used to measure the oil droplet size distribution. NaA(r) additives not only lowered the sintering temperature but also enhanced the performance such as rejection rate of oil droplet and water permeance. Also, they found out that simply utilizing ultrasound process could regenerate the membranes, which made the membranes to be used in long-term filtration. The pure water flux of the membrane reached 3700 L/ m^2 h.

Rasouli *et al.* used extrusion method with low-cost easily accessible raw materials (i.e., kaolin clay, natural zeolite and a-alumina powder) for MF[18]. Then, in-line coagulation in the hybrid process was done by aluminum and iron salts ($Al_2(SO_4)_3$, $FeCl_3$, $Fe_2(SO_4)_3$ and $FeSO_4$) with different concentrations, which produces mullite-alumina-zeolite (MAZ) and mullite-zeolite (MZ) ceramic MF membranes. DLS was used to analyze the size of oil emulsion droplets. SEM was used to see the surface morphology of the synthesized MZ and MAZ membranes. XRD was used to check the presence of the phase transformation. To evaluate the performance, they measured the membrane flux and the oil rejection percent and found out that that $FeSO_4$ with a concentration of 200mg/L was the best coagulation agent for the flux of both of MAZ and MZ membranes, and $Al_2(SO_4)_3$ for oil rejection percent.

Rasouli *et al.* prepared tubular and symmetric mullite, mullite-alumina, mullite-alumina-zeolite and mullite-zeolite membranes, which are cheap and have good permeation flux and TOC rejection, with kaolin clay, natural zeolite and α-alumina powder[19]. Then, they applied adsorbent agents, powdered activated carbon and natural zeolite powder, with different concentrations to the in-line adsorption–MF process. By doing so, they achieved improvement in TOC rejection for all membranes by adding either powdered activated carbon or natural zeolite powder, in permeation flux of all membranes except for mullite and mullite-alumina ceramic membrane by adding 800 mg/L of powdered activated carbon, and in permeation flux of all membranes except mullite-alumina membrane by adding

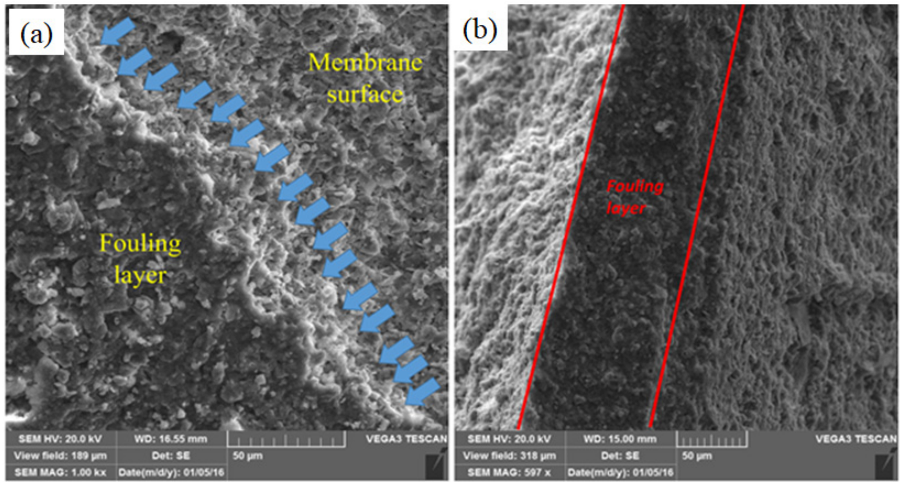

Fig. 3. (a) SEM micrograph of surface of mullite-alumina membrane after oily wastewater treatment in the hybrid adsorption–MF process using 800 mg L^{-1} natural zeolite. (b) SEM micrograph of cross-section of mullite-alumina membrane after oily wastewater treatment in the hybrid adsorption–MF process using 800 mg L^{-1} natural zeolite (Reproduced from Rasouli *et al.*, 19, IWA Publishing).

natural zeolite powder. SEM was used to analyze the surface morphology of the membranes, as shown in Fig. 3. XRD was used to detect any phase transformation. These membranes can be used in washing applications or flash tank water by reusing the treated wastewater.

Rasouli *et al.* fabricated four different ceramic microfiltration membranes (i.e., mullite, mullite-alumina (MA), mullite-alumina-zeolite (MAZ), and mullite-zeolite (MZ) membranes) from local clays such as kaolin clay, α-alumina powder and natural zeolite powder and evaluated the pure water flux and oil rejection of these membranes[20]. SEM was used to analyze the surface morphology of the mullite and MZ membranes. XRD was used to anlayze the different phases in the membranes. TGA was used to detect weight loss steps. Fig. 4 represents the SEM image of the membrane.
While all four membranes showed high flux and oil rejection performance, the highest flux was 549 L/m^2 h from MA membranes with 50 wt% alumina and the highest oil rejection performance was 98.77% from MZ membranes with 50 wt% zeolite. Moreover, they used Hermia's models to compare their experimental data with theoretical data and found that the flux decline shown during the experiment matches well with the prediction made from Hermia's models.

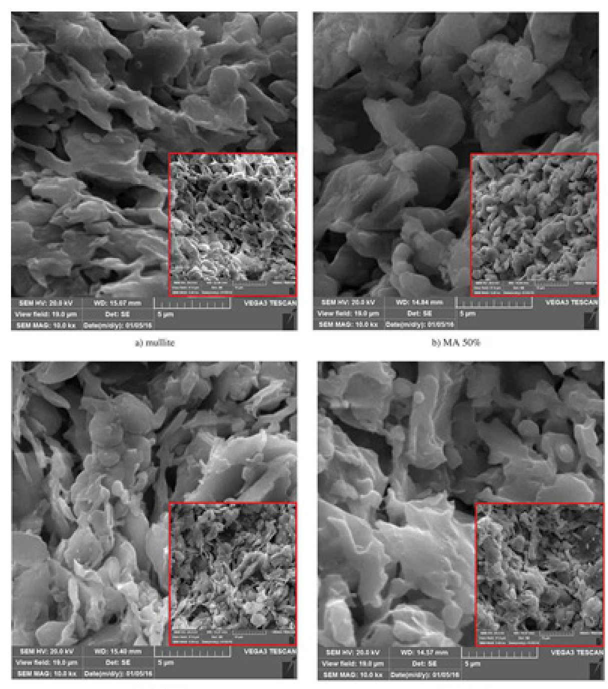

Fig. 4. SEM images of membranes (5000 X): (a) mullite, (b) MA 50%, (c) MAZ 20% and (d) MZ 20% (Reproduced from Rasouli *et al.*, 20, Taylor & Francis).

Rasouli *et al.* studied the wastewater purifying performance of four composite membranes [mullite, mullite-alumina (MA 50%), mullite-alumina-zeolite (MAZ 20%), and mullite-zeolite (MZ 40%)] that they made from cheap local raw materials such as kaolin clay, natural zeolite, and alpha-alumina powders[21]. SEM was used to analyze the surface and cross-section morphology of the synthesized membranes. XRD was used to find any changed or produced phase in the membrane structure. DLS was used to measure the oil emulsion droplet size. Fig. 5 represents the schematic of membrane separation process. They found out that the flux, oil rejection, and ion rejection is influenced by the salt concentration of the wastewater from the desalter. The increase in salt concentration leads to higher ionic strength, resulting in slightly higher oil and ion rejection. This membrane showed very good oil rejection that varied in between 96.2 to 99.2%.

Chen *et al.* fabricated a hydrophobic SSN@F-FAU membrane. Zeolite FAU layer was grown on stainless steel net (SSN) which was modified with aminopropyltriethoxysilane (APTES). This surface was further post modified with perfluorooctyltriethoxysilane (POTS) to enhance hydrophobicity of the surface[22]. By the post modification, the water contact angle of the SSN@FAU changes from 15° to 132° while both structure and morphology of the SSN@FAU did not change, so it becomes hydrophobic. SEM was used to analyze the microstructure of the zeolite powder and coating. XRD was used to check the crystallinity of the membrane support and its coating property. The SSN@F-FAU coating showed good separation efficiency which exceeds 99.1% even after 50 cycles of experiments. Also, they found that simply changing SSN@FAU to SSN@LTA in post modification of POTS guarantees similar performance with SSN@F-FAU coating. These shows that the membranes can be prepared universally and have high potential to be applied in oil/water separation in industrial scale.

Peyravi *et al.* fabricated polyester/poly(vinyl alcohol) dynamic membrane incorporating photocatalytic zeolite/TiO_2 by two different ways: self-forming and precoating method[23]. FTIR was utilized to detect the presence of TiO_2. XRD was used to verify the presence of TiO_2. SEM was utilized to analyze the surface morphology of the fabricated membranes. Utilizing the

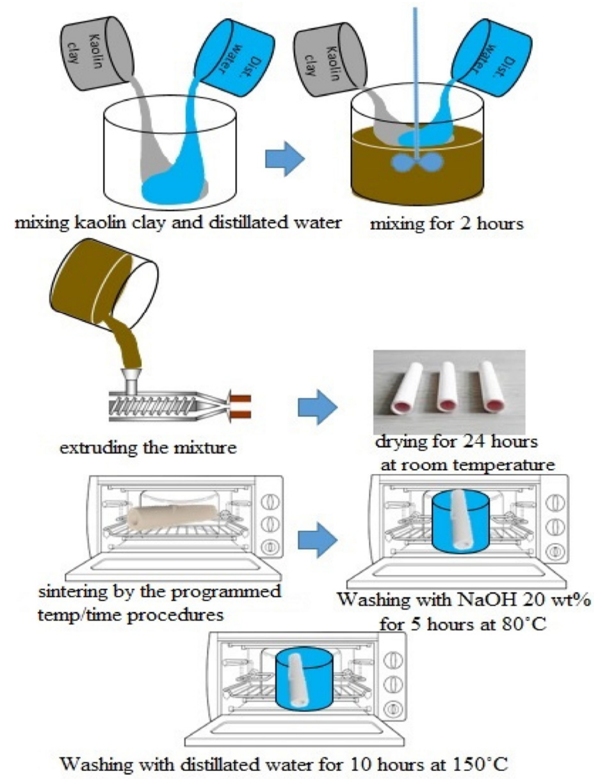

Fig. 5. Schematic representation of mullite membrane fabrication (Reproduced from Rasouli *et al.*, 21, MDPI).

dynamic membrane as the support membrane not only enhanced wastewater treating performance but also lowered the cost of manufacturing. Between two methods, pre-coating showed better fouling reduction because it forms uniform protective layer of zeolite/TiO_2 dynamic material on the support membrane. Also, utilizing UV irradiation process with cleaning agent, sodium dodecyl sulfate improved membrane flux recovery rate by 83.6% while maintained the oil rejection rate by 85.3%.

Sultana *et al.* fabricated optimized uniform nano- and microfiber electrospun composite membranes from electrospinning 16% (w/v) cellulose acetate in a dual solvent, acetic acid mixed with acetone, with incorporation of 10% w/w zeolite into the cellulose acetate membranes[24]. SEM was utilized to see the morphology of the synthesized nanofiber membranes. XRD was used to analyze what elements are in the electrospun membranes. Energy dispersive X-ray spectroscopy

(EDX) mapping was utilized to find the position of specific elements. Water contact angle measurement was used to analyze the contact angle of the cellulose acetate membranes with and without zeolite. They found its super-hydrophilic properties from the membranes and its high filtration efficiency (97%) from optical micrographs. The membranes they made have potential of being used in oily wastewater treatment because aside from its properties mentioned above it is biodegradable, non-toxic, and cost-effective.

3. Conclusion

Inorganic membrane is alternative to organic polymeric membrane due to it better antifouling property, chemical stability and long term usability in oily wastewater treatment process. Polymeric membrane is chemically or physical modified to enhance the antifouling property. Although the modified organic polymeric membranes are successful in various separation process, but it is still not highly effective for oily water treatment. Carbon-based membranes are another class of successful inorganic membrane for oil in water separation process. Zeolitic membranes are considered best among various types of inorganic membrane due to its lower cost, easier process ability and lower antifouling property due to the hydrophilic property of the materials.

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