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REVIEW PAPER

Surface matrix functionalization of ceramic-based membrane for oil-water separation: A mini-review

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Abstract−Advanced filtration requires a cost-effective, highly efficient and environmentally friendly membrane fabrication to achieve excellent and extreme oleophobic and hydrophilic states with an appropriate surface functionalization approach. For an efficient oil-water separation, surface material and structure have been categorized as superoleophobic (high oil repellency) and superhydrophilic (strong affinity to water). This can be attributed to their selective mechanisms. In addition, it is very important to consider these key factors as they will imperatively provide a unique capability to exhibit surface roughness, stimulate low and high surface energy, and improve surface chemistry, which makes membranes function efficiently for the oil and water separation system. In this review article, the application of ceramic membrane for oily water treatment and the principle of superhydrophilic and superoleophobic ceramic membrane for oil-water separation are elaborated and, in addition, the current progress for the use of inorganic material to functionalize ceramic membrane surface based on physical and chemical methods is appraised. Finally, the challenges and outlook on the membrane for oily-water treatment are briefly examined.

Keywords: Ceramic Membrane, Superoleophobic and Superhydrophilic, Surface Functionalization

INTRODUCTION

Oily wastewater from industries such as oil/gas recovery, palm oil mill effluent (POME), mining, and petroleum refining is an extremely common pollutant worldwide as it affects the environment and aquatic life [1-6]. The effects can be attributed to the current market high demand for oil globally for domestic consumption and as well as industrial uses [7]. Oily wastewater can exist as stable, unstable, and free-floating oils [8]. For instance, effluent from palm oil mill has a higher proportion of water (95-96 v/v %) and with 0.6-0.7 v/v % unstable oil, and the remaining 4.5 v/v % contains solid suspensions [9]. This has generated tremendous a concentration of biological oxygen demand (BOD), chemical oxygen demand (COD), oil and grease, and as well as suspended solids. The level of these organic matters is due to the presence of unrecovered palm oil; its disposal to water bodies without pre-treatment poses a threat to both humans and the ecosystem [6,7]. However, the current strategy of using conventional sedimentation, skimming, biological anaerobic and aerobic systems, centrifugation and faulty ponds treatment relies on the application of bacteria, which are unsafe, uneconomical, and unsustainable [12]. This is because it requires highly proper maintenance and assessment, high labour and operational costs. The challenges of treating oily wastewater will require the use of an advanced separation strategy such as microfiltration (MF) and ultrafiltration (UF) techniques to treat a large proportion of water being lost from oily wastewater operation [10].

The MF process operates under a driving force (pressure) with the array of 0 to 2 bar for the removal of bacteria from feed solutions (liquid-liquid), colloid and suspended particles. MF makes use of porous membranes, which could be symmetric or asymmetric with a thickness of 10-150 μm, molecular weight cut-off (MWCO) largely above 100 kDa and pore size ranging from 0.1-10 μm, respectively. MF is primarily employed for biological treatment and in the design of bioreactors, which makes it the most appropriate process for oily wastewater application to remove active biomass, COD and BOD [13]. The disadvantage of MF is that it allows the permeation of bacteria, dissolved organic and inorganic components, and is consequently constrained to the pre-treatment of wastewater. The cost of producing MF is inexpensive compared with conservative treatment methods [14].

While, the UF process also operates under driving force (pressure) with an array of 1.4 to 6.9 bar for the removal of proteins, starch, viruses, colloid silica, organic, and dyes [13]. UF makes use of porous membranes, which could be symmetric or asymmetric with thickness 1-10 μm, MWCO largely above 1-300 kDa and pore size ranging from 0.01-0.1 μm, respectively [14,16-19].

It is, in this view, pertinent to protect the environment through developing systems that are economically viable and sustainable. Sev-

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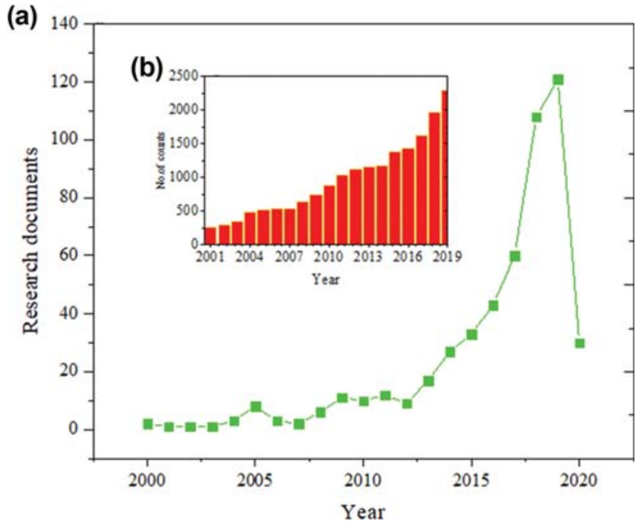

Fig. 1. Research outlook on (a) research publications on oil-water separation and (b) surface wettability (inset). Adopted from Scopus and accessed in February 2020.

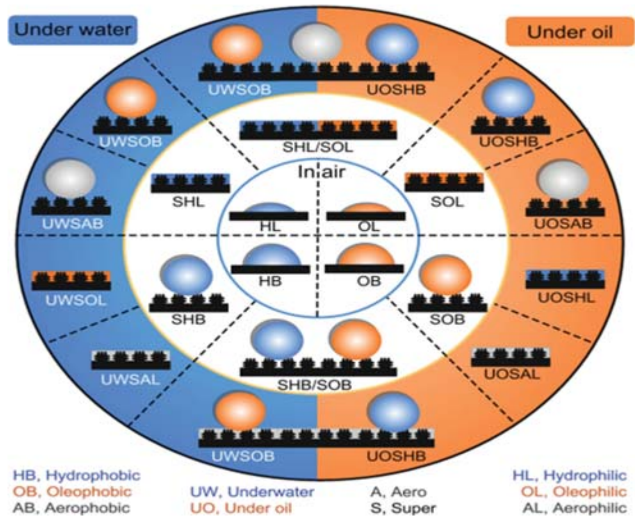

Fig. 2. The system of "superwettability". Reproduced with permission from Ref. [25], copyright, Wiley 2020.

eral articles have reported various strategies of oil and water separation as indicated in Fig. 1(a), most specifically the use of natural occurrence starting materials [19]. The application of natural ceramic membrane has been considered for presenting more advantages over the polymeric membrane in terms of excellent solvent resistance, long lifetime, high thermal stability and exhibit chemical inertness in the area of MF and UF processes [20]. This type of membrane is usually formed based on the initial fabrication of layer and multilayer materials such as alumina, silica, titania, zirconia, zeolite, and bentonite.

However, one such advanced strategy and cutting-edge at enhancing MF and UF membrane surfaces is the concept of surface wettability that usually indicates the ability of a material to become wetted or dewetted when liquid encounters it, which has been greatly explored. Fig. 1(b) (inset) reveals the research activity on the surface wettability, as the research output has increased tremendously over the years. Since the first reported article in 1929, scientific reports have increased significantly, confirming to the novice of anti-wettability approach for oil repellency. Superwettability, such as for superoleophobicity and superhydrophilicity, has properties of retaining oil at the surface of the solid and removes water from oily wastewater [23-26]. As indicated in Fig. 2, the middle part--hydrophilicity, oleophilicity, hydrophobicity and oleophobicity--illustrates the behavioral changes in the surface properties with respect to flat substrates in air. After introducing enough micro-/nanoscale two-tier roughness, superhydrophilicity, superoleophilicity, superhydrophobicity and superoleophobicity states can be generated in air. Left blue circle: Underwater superoleophobicity, superoleophilicity, superhydrophobicity and superaerophilicity upon rough substrates; Right orange circle: Under-oil superhydrophobicity, superhydrophilicity, superoleophobicity and superoleophilicity upon rough substrates. Diverse types of extreme wettability have served as important building blocks to construct a system named "superwettability" [25].

Superoleophobicity membrane material is still quite challenging to develop as oils with low surface tension tend to wet and spread across the membrane surface. This leads to a build-up of fouls and consequentially reduces flux rates, that ultimately is a decline in the operation and the performance of membrane [26-29]. It is important to note that the surface and matrix functionalization or modification of membrane would play a significant role either in water or air such as its ability in anti-blocking, chemical shielding, oil and water separation, self-cleaning and anti-oil capability [24,33-35]. This will also improve the membrane surface stability and thereby increase the superhydrophilicity and superoleophobicity in terms of oily wastewater separation [2]. As it will imperatively provide a unique capability to exhibit surface roughness, stimulate low and high surface energy, improve surface chemistry, which makes membrane function efficiently for the oil and water separation system [3,34,36,37].

CERAMIC MEMBRANE FOR OILY WATER TREATMENT

Ceramic membrane (inorganic membrane) is a form of microporous membrane used in a different area of MF, UF and other processes [35]. This type of membrane, as indicated in Fig. 3, is usually formed based on prefabrication of layer and multilayer materials such as silica, alumina, kaolin, titania, zirconia, zeolite, and bentonite. While, the configurations include plate, tube, hollow and multichannel ceramics [35]. These configurations possess different degrees of membrane surface area per unit of volume and with the ability to permeate fluids at the exact hydrodynamic conditions and operating transmembrane pressures.

The structures of ceramic membrane usually consist of support layer ranges from 1 to 20 μm with a porosity of 30-65%, and the transition layer ranges from 20-60 μm and 30-40% of the thickness and porosity, respectively. This layer is quite smaller in thickness and pores relative to the support layer. While the functional layer has a thickness (minimal) in nano-particles size, and also porosity ranges of 40-50% [37]. The various types of membrane are best distinguished based on their pore size and their application. As indicated in Table 1, the membrane types play a significant role in

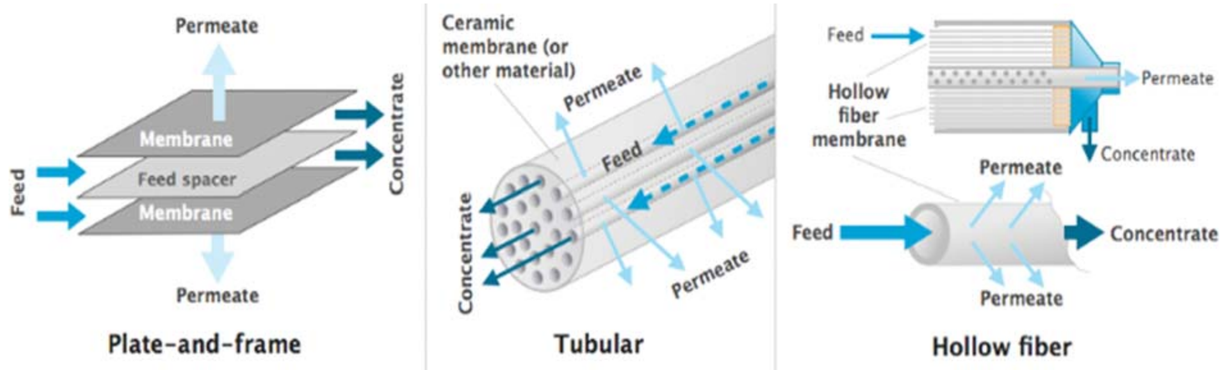

Fig. 3. Schematic representation of membrane configurations. Reproduced with permission from Ref. [36], copyrights MattwPlant Pte 2020.

Table 1. Types of membrane types, pores, and their applications: RO: Reverse osmosis; NF: nanofiltration; GS: gas separation [8]

Membrane pore type	Mechanism	Pore size (nm)	Membrane types	Species
Macroporous	Bulk diffusion	>50	UF, MF	Fungi, Bacteria, Oil emulsion
Mesoporous	Knudsen diffusion	2-50	UF, NF, GS	Virus, Colloidal solid, Protein
Microporous	Microporous diffusion	0.3-2	RO	Water, inorganic ions

oil-water separation due to the differences in the particle size of oil and water, and more so, the surface charges of the membrane surface. However, the choice of appropriate membrane system from the previous studies shows that the use of UF and MF is more effective and efficient in the oil/water separation due to the pore size and surface morphology [8].

In recent times, outstanding reports have been issued that focus on the development of low-cost ceramic membrane materials based on their natural abundance and availability, and these materials include kaolin, bentonite, ball, zeolite, bauxite, among others [38-40]. For instance, kaolin formation occurs during the geological transformation of rocks and exists in different colors, particle sizes and shapes [41]. Meanwhile, ball clay contains more fine particles as compared to kaolin with the presence of iron, and its plasticity is quite an excellent ceramic material [38]. For the bentonite which is formed from natural mineralogical earth's crust in the subgroup of smectite clay, primarily consists of montmorillonite and other minerals such as calcite, beidellite, quartz, illite and kaolinite [42].

The clay usually occurs in yellowish, brownish, black, grey, bluish, or greenish color, with the characteristic of swelling when in contact with water. These have the capability to exchange ions with a vast range of metals and other minerals. Bentonite also shows good thixotropy. This behavior can be attributed to the applied stress that makes it less viscous and then becomes highly viscous material again. These low-cost ceramic membrane materials (over the years) have metamorphosed into various areas of application (Table 2) because of overwhelming excellent thermal, chemical and microbiological stabilities, long service time, high hydrophilicity and low

Table 2. Summaries on different membrane material and configuration for oil/water separation. Note: N/A- not available

Ceramic material	Configuration/ module	Sintering temperature °C	Mean pore size, μm	Water permeability, L/m^{2}h	Mechanical strength (MPa)	Reference
Zirconia	Tubular membrane	1,500	1.8	NA	NA	[45]
Fly ash	Spherical membrane	NA	0.97	2,270	0.10	[56]
Kaolin	Flat membrane	1,100	0.35	36.51	12.53	[57]
Kaolin	Hollow fibre	1,500	2.26	320	105.34	[47]
Palm oil fuel ash	Hollow fibre	1,000	10.5	219.2	60	[40]
Kaolin, bentonite	NA	1,000	1.25	0.277	58	[58]
Calcined diatomite, bentonite, kaolin, talc	NA	1,000	0.4	0.09	32	[59]
Kaolin/alumina	NA	1,300	1.5	580	0.1	[60]
β-SiAlON	Hollow fibre	1,650	1.05	23,611	200	[61]
Zeolite	Hollow fibre	1,000	NA	40	46	[62]
Mullite	Hollow fibre	1,250	50.5±2.1	1,286±18	55.8±58	[63]
Alumina dross	Hollow fibre	1,275	<50	239.47	84	[64]
Yttria-stalilzed zirconia	Hollow fibre	1,475	NA	118.4	336	[65]

hydrophobicity [43].

Table 2 details some ceramic-based membrane materials, supports, configuration, operating parameters, and properties [44-52]. These materials stand out to be an efficient solution to separate or treat oil-water emulsion at low-cost, and on the other hand, can be used as a substitute for expensive ceramic membrane starting material. As a result of the high efficiency of the kaolin-based ceramic membrane for oily removal from wastewater, a few researches were able to attain 99.99% oil rejection and flux of about 320 $L/m^2 \cdot h$ after 20 min of filtration as reported by Hubadillah et al. [19]. While, good performance was recorded when the effect of hydrophilic of kaolin with 89.00% chemical oxygen demand (COD) rejection and less than 30% fouling reduction [53].

In a related work, the composite of kaolin was used to fabricate membrane with 99.98% oil rejection and 111.6 $L/m^2 \cdot h$ was obtained. Das et al. investigated the application of ceramic-based membrane for oil-water separation where the oil rejection was 95.4% and flux was obtained at 4.1056×10^{-6} $L/m^2 \cdot h$ [54]. While Table 2 summarizes various ceramic membrane materials and properties. The summaries show that low-cost ceramic materials with supports were explored to fabricate different membrane modules. The effects of sintering temperature on the ceramic revealed that membranes can be sintered between 1,000 °C and 1,600 °C. β-SiAlON membrane possessed the highest flexural strength and sintering temperature as compared to other membranes. The water fluxes showed that the membrane fabricated from calcined diatomite has the lowest flux of 0.09 $L/m^2 \cdot h$ and β-SiAlON has the highest flux of 23,611 $L/m^2 \cdot h$.

Furthermore, the separation mechanism of the ceramic membrane has proven to be a more cost-effective and efficient advanced technique in oily wastewater separation. Research findings have shown that ceramic membrane can be used successfully to separate oily wastewater [34,56,59]. However, the merits of using ceramic membrane in terms of excellent chemical and thermal stability as well as flexural strength have made it a better alternative choice when compared to the polymeric membrane [55]. Hence, the ceramic membrane technology can work without the addition of chemicals and little operational demands if the fabrication parameters are well designed.

PRINCIPLE OF SUPERHYDROPHILIC AND SUPEROLEOPHOBIC CERAMIC MEMBRANE FOR OIL-WATER SEPARATION

The principle of superoleophobic/superhydrophilic surfaces barrier on the oil-and-water separation is based on the idea of "oil rejection" and "water removal", in which water wets the surface and permeates through the membrane as stated in section 1. Such design and structure reduces the tendency of the surface to be fouled by oil and allows a gravity-driven separation process [66]. Several research outputs on membrane separation that is based on absolute "water-removal" methods possessed strategy of superhydrophilic surfaces that exhibit also superoleophobicity have, however, been fabricated from a wide range of organic and inorganic materials include polyelectrolytes, zwitterionic polymers, metal oxides, hydrogels, and graphene oxide [22,26,59,66-72]. Superoleophobic/superhydrophilic surfaces regularly exhibit high oil-water separation with low fouling and high efficiencies. However, for underwater superoleophobic/superhydrophilic, the design and structure require the substrate (membrane surface) to be prewet with water before the separation process to sidestep fouling [29,35,73,74]. Additionally, the surfaces displaying in-air superhydrophilicity and superoleophobicity are challenging to fabricate and require a cross-linker that concurrently exhibits surface energy that is lower than oil and higher than water. Hence, in practice, surfaces exhibiting both the wetta-

Table 3. Surface tension values of some common liquids for surface energy analysis [82]

Liquids	Perfluoro hexane	n-Hexane	Polydimethyl siloxane	Ethanol	n-Decane	Pyridine	Diiodo methane	Water
Surface tension@20 °C in mN/m	11.91	18.43	19.00	22.10	23.83	38.00	50.80	72.80

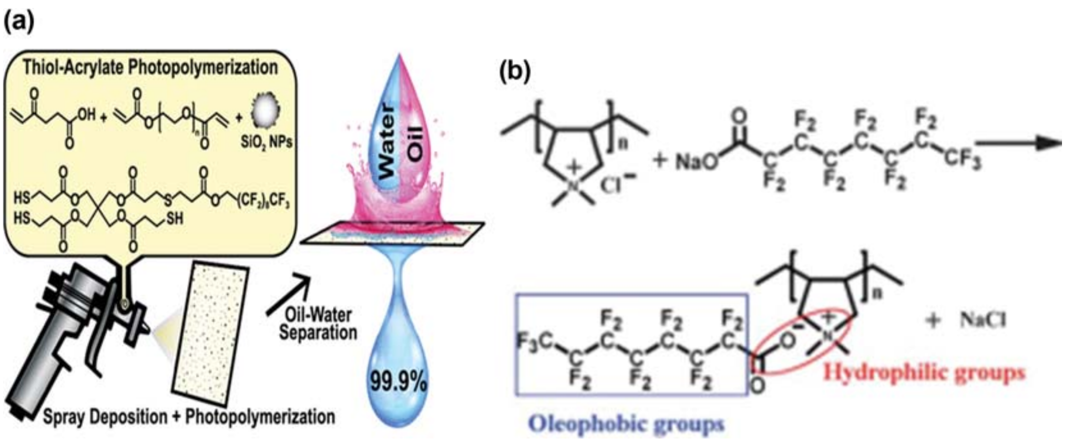

Fig. 4. Schematic representation of rational design of superhydrophilic/superoleophobic surfaces for oil-water separation via (a) Thio-acrylate with fumed silica nanoparticle [84], (b) PDDA-PFO (complex polymers) [74].

Table 4. Advantages and disadvantages of physical and chemical treatment strategies of membrane surface functionalization [86,87]

Treatment strategies	Advantages	Disadvantages
Physical treatment	• No chemical reaction involved • Entails the blending, coating, polishing and thermal treatment • It is a simple, cost-effective, and eco-friendly process • Most suitable for flat sheet configuration • Reduces membrane fouling	• Becomes unstable within a short time • Limited to inorganic materials • Low reusability • Low stability
Chemical treatment	• Simple chemical reactions are involved • Involves interfacial polymerization • Entails surface reaction, surface grafting, and post-treatment • Reduces membrane fouling • Applicable to both inorganic and organic membrane • Possess long-time stability during operation • Most suitable for hollow fibre and tubular configuration	• Changes the surface chemistry of membrane • High cost of fabrication and operation • Inconsistent in the modified surface

bility properties concurrently in-air are rare, as superoleophobic surfaces generally exhibit superhydrophobicity [35,75]. The relatively few papers that have described the fabrication of superoleophobic and superhydrophilic surfaces exploit a stimuli-responsive rearrangement of the material interface using complex polymer with fluorinated compounds [35,73,76,77].

There are vast challenges in fabricating superhydrophilic and superoleophobic surfaces when the majority of organic liquids possess ultralow surface tension [24,78-80]. Additionally, the materials having extremely low surface energy are preferable with high surface roughness also required as it tends to support oil repellency at the substrate surface [22]. As shown in Table 3 the typical surface tensions of common oils are within 18-33 mNm^{-1} [82]. To further achieve superoleophobicity, the required surface tension of the material surface must be as low as a few mNm^{-1}. The -CF_3 group, almost alone, has a quite low surface energy [25,73,79,81]. For example, Fig. 4(a) shows a schematic representation of superoleophobic and superhydrophilic spray coating process with photopolymerization of thio-acrylate laden SiO_2. Xiong et al. reported that their superoleophobic and superhydrophilic surface fabrication revealed oil-water separation efficiency of 99.9% and water flux of 699 L/m^2h [84]. Therefore, to fabricate superoleophobic surfaces, most materials need to be modified with a low-surface-energy layer after the formation of rough microstructures in order to minimize fouling associated with the membrane during operation. While Yang et al. designed a superoleophobic and superhydrophilic coated surface with low tilt angles [32]. The PDDA-PFO/SiO_2 (Fig. 4(b)) prepared by sol-gel method was coated on the stainless steel for oil-water separation [78]. The results revealed an excellent superoleophobic and superhydrophilic coated surface with a contact angle of 155±1^o for hexadecane (oil) droplets.

SURFACE FUNCTIONALIZATION OF CERAMIC MEMBRANE FOR OIL-WATER SEPARATION

Functionalization of the membrane surface enables alteration of the wetting properties (such as the membrane morphologies, surface energy and chemistry) of the membrane surface for proper oil and water separation [85]. One such functionalization process is the use of inorganic material, which is the most common because of its unique capability to exhibit surface roughness and make the membrane function efficiently for oil and water separation [34]. Surface functionalization or modification involves binding by physical or chemical approach with interactive materials, whose properties such as affinity and catalysis can improve the membrane performance. The treatment strategies for physical/chemical and their advantages and disadvantages are summarized in Table 4. Their performances are in terms of membrane selectivity, roughness, surface energy, surface charge, hydrophilicity, biocompatibility, functionality and separation, and also limiting the drawback associated with fouling between the membrane and unwanted particles from the feed [8,66,84].

1. Physical Treatment of Surface Functionalization of Ceramic Membrane

1-1. Coating Method

Membrane surface coating is a physical treatment which entails blending, coating, polishing, grinding and thermal treatment process which does not change the chemical composition of the membrane surface [88]. Hence, the coating is quite suitable for flat membrane and hollow fiber membrane because of its simplicity, cost-effectiveness and environmental friendliness [89]. For example, Hu et al. prepared a high performance porous ceramic membrane by a two-step coating and one-step sintering; the ceramic showed defect-free when sintered at 1,450 ^{o}C for 2 h after coating time of 11 s [89]. In a similar approached, Zhu et al. demonstrated pinhole defect-free ceramic membrane with a dip-coating velocity of 50 mms^{-1} and withdrawal speed of 4 mms^{-1} using a single coating and sintering [88]. Also investigated the reclaiming of oil from oily water using a porous ceramic membrane with a superhydrophobic and superoleophobic surface, Su et al. however, reported two-approach treatment by sol-gel and polyurethane- polydimethylsiloxane coating with four recycles time after annealing at 500 ^{o}C for 4 h [27].

1-2. Ion Beam Irradiation/Implantation

Ion beam or implantation involves the application of ions and light to improve the surface modification of a membrane. The technique creates process functionalization on the membrane such as macro-molecular destruction, cross-linking, free radical formation, carbonization and oxidation, which alters the binding structures of

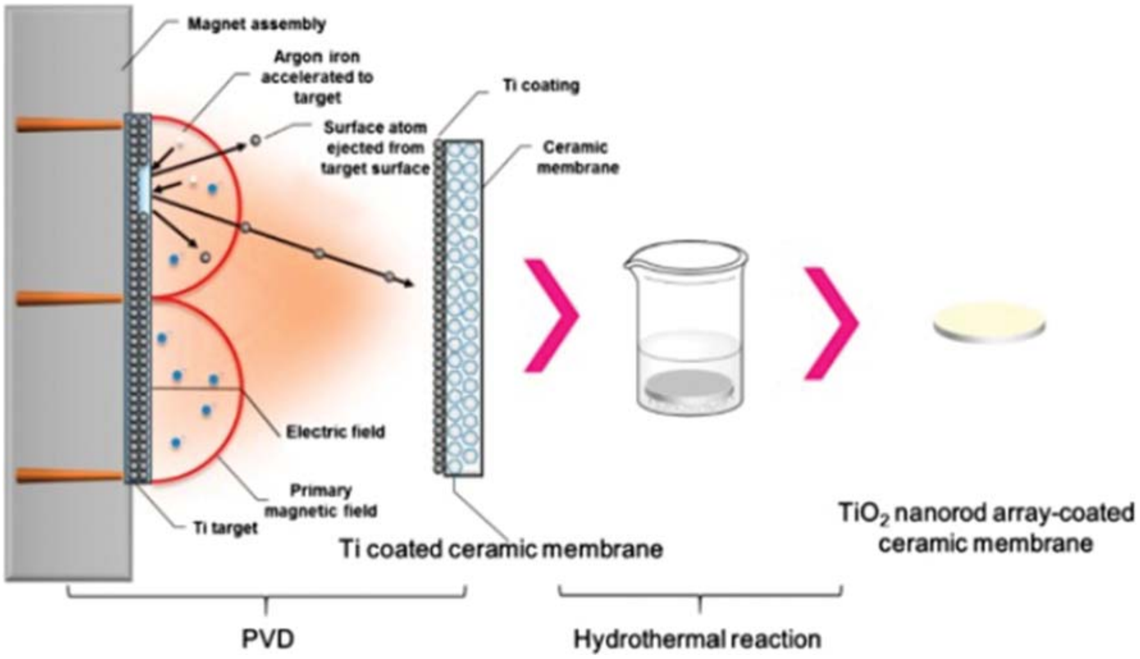

Fig. 5. Fabrication process of TiO_2 nanorod arrays on ceramic membrane [83].

the membrane layers [90]. There are still limited numbers of researches on ion beam irradiation for surface functionalization of ceramic membrane. Li et al. studied the site-selective local fluorination of graphene induced by ion beam irradiation. The investigation on the defect sites revealed that fluorine atoms strongly adhered to the surface structure of graphene [90].

1-3. Plasma Irradiation

Plasma irradiation works on the principle of subjecting a gas to a radio frequency in a vacuum chamber. This is, thereby, producing partially ionized gas and un-ionized radicals. This further produces more of the ions and radicals. The ions and radicals then react with the chemical groups on the membrane surface to form a functional group which leads to an ion functionalized membrane surface [71,90]. In 2016, Wang et al. examined surface functionalization of ultrathin graphene oxide supported by PVDF for oily water separation; however, the membrane showed an improved underwater oleophobicity and hydrophilicity, and moreso less oil adhesion to the membrane surface after four cycles treatment. Hence, these have shown to enhance the photocatalytics of the membrane and physical behavior of the membrane surface [92].

1-4. Vapor Phase Deposition

Vapor phase deposition (VPD) is a dry process which does not require the use of solvent. VPD is carried out in a vacuum chamber where the coating material is heated/or at reduced pressure until it vaporizes, while the membrane to be coated is be exposed to the suspended coating material, which allows a gradual settling and thus forms a uniform coating [93]. However, the coating thickness can be modified by controlling the heating process and using initiator to control the time required during the coating process [92,93]. For instance, Tran et al. carried out a surface modification of commercial tubular ceramic membrane with carbon nanotubes using catalytic chemical vapor deposition at the calcined temperature of 650 °C for 6 h. The rejection rate for the modified pristine was 90% while the flux was 1,033 L/m^{2}h. Irreversible adsorption of alginate was achieved after chemical cleaning backwashing [95]. Zhang et al. prepared a superhydrophilic and underwater superoleophobic with TiO_2 nanorod coated on Al_2O_3 porous ceramic membrane for the purpose of oil-water separation and effective surface fouling reduction. Fig. 5 illustrates the two-step method of preparing the TiO_2 nanorod using magnetron sputtering and oxidation reaction. The performance of the facilely synthesized surface showed 99.1% oil rejection, water flux of 41.8 L/m^{2}h, and oil contact angle more than 150° [83].

2. Chemical Treatment of Surface Functionalization of Ceramic Membrane

2-1. Membrane Surface Grafting

Surface grafting is a surface modification method that establishes covalent bonding between chemical (polymers) additives and membrane matrix through the initial stages such as pre-irradiation, peroxidation, and mutual irradiation [86]. Surface grafting can be done as either "grating from" or grafting to" on the membrane surfaces. Surface grafting is also appropriate for both flat and hollow fibre membrane configuration.

In 2015, Yang et al. reported the use of SiO_2 (~20 nm diameter) and a positively charged cationic polyelectrolyte as a surface grafting material to develop surface roughness on metal meshes by spray gun coating process (Fig. 6). The surface displayed both oil repellency and water penetration behavior with the efficiency above 99.0% and contact angle of 158±1° [78]. Similarly, Li et al. carried out dip-coating of SiO_2 (>1 μm diameter) with polystyrene (PS), then dissolved in tetrahydrofuran (THF) to produce required surface roughness [97]. Another work used inorganic functionalized material to develop the surface roughness and increase the contact angles with the application of TiO_2 particles [98]. Li et al. also

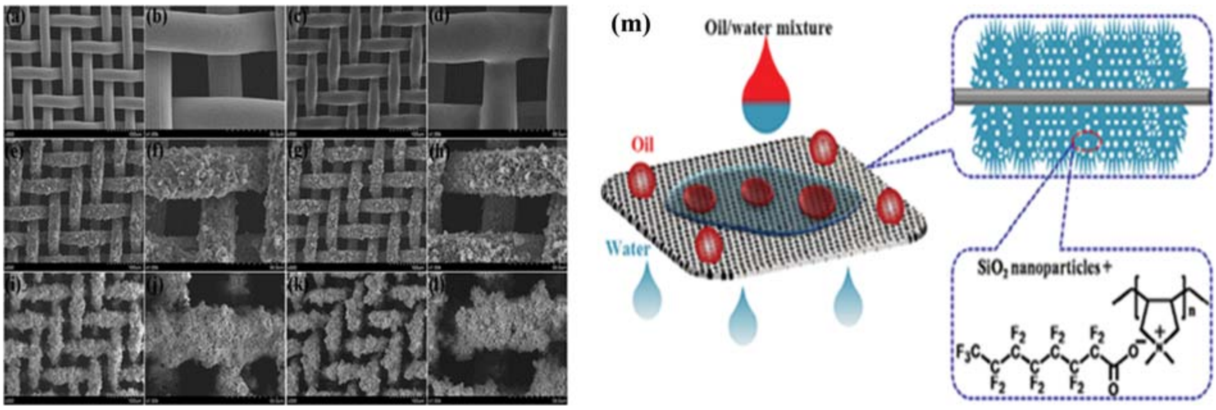

Fig. 6. **SEM images of the original mesh and schematic illustration of the superhydrophilic and superoleophobic polyelectrolyte-fluorosurfactant complex (PFC)-based mesh for oil/water separation (a) and (b), the PFC-coated mesh (c) and (d), the PFC/SiO_2-coated mesh with 25 wt% (e) and (f), 40 wt% (g) and (h), 50 wt% (i) and (j), and 57 wt% (k) and (l), SiO_2 surface microstructure and chemical structure (m) [78].**

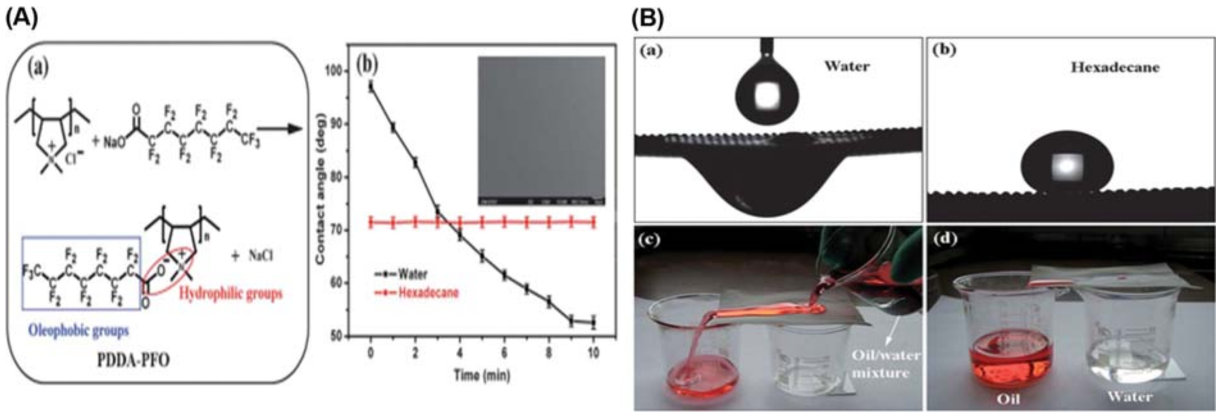

Fig. 7. **Poly (diallyl dimethylammonium chloride)- sodium perfluorooctanoate PDDA-PFO/SiO_2-coated mesh film shows special wettability, with both superhydrophilic and superoleophobic properties. (A) (a) Chemical reaction mechanism used for the synthesis of PDDA-PFO. (b) Time dependence of contact angles for water and hexadecane on a spin-coated PDDA-PFO film. The inset shows a FESEM image for the PDDA-PFO film. (B) (a) Water droplet spreading on and permeating through the mesh. (b) Shape of a hexadecane droplet on the mesh with a contact angle of 157°, (c) and (d) Oil/water separation experiment was performed on the PDDA-PFO/SiO_2-coated mesh [74].**

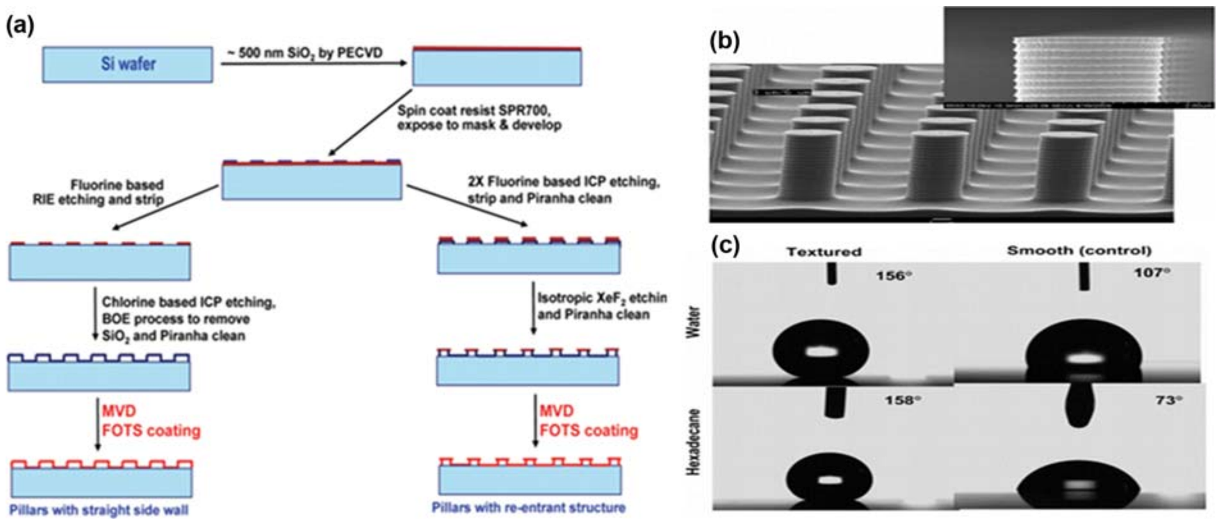

Fig. 8. **(a) Fabrication of surface textures comprising pillars with straight sidewalls with and without re-entrant structures, (b) SEM micrograph of the textured fluorosilane layer (FOTS) surface on Si wafer (inset: higher magnification micrograph showing details of the pillar structure), (c) Static contact angles for water and hexadecane on the textured FOTS surface (control: smooth FOTS surface on Si wafer) [107].**

applied TiO_2 (~100 nm diameter) to modify a mesh surface, and the results showed high water selectivity in a 40-times reusability and 97.5% separation efficiency [99]. Gao and co-researchers carried out a surface modification with a mixture of TiO_2 particles and Cu via a hydrothermal approach at low temperature, that allows the decomposition of blue dye in oily wastewater under UV light for 2 h [99].

Huang et al., on the other hand, reported the use of graphene oxide membrane as an inorganic material to separate water from oily wastewater, and the water selectivity was quite high [101]. In contrast, Liu et al. successfully employed boron nitride nanotubes with metal mesh for water separation from oil and water mixtures in a superhydrophilic surface [103]. Wen et al., Yu et al., Zhang et al. studied grafting of graphene oxide, zeolite-coated mesh and carbon nanotubes on the membrane, respectively, to obtain a functionalized surface [102-104]. Similarly, Wang et al. carried out surface modification of metal mesh with pure silica zeolite that consequently achieved high water selectivity, high flux rate and purity [106]. In related work, Yang et al. investigated the responsivity behavior of superhydrophilic and superoleophobic coated surface with fluorinated sodium perfluorooctanoate - poly (diallyl dimethylammonium chloride) and silica, PFO - PDDA/SiO_2, as illustrated in Fig. 7. The coated mesh showed a contact angle of 157±2^o for oil repellency and after a tilted angle of 2^o water penetration within 9 mins [74].

Zhao et al. extensively investigated the design and fabrication of silicon wafer for superoleophobic application. The wafer surface design was 3 μm×7 μm×6 μm, and tridecafluoro-1,1,2,2-tetrahydro octyl trichlorosilane was used as the coating agent. The superhydrophobicity and superoleophobicity toward water and oil possessed 156^o and 158^o at slide angle of 10^o respectively [107]. Fig. 8 The outcome was attributed to the textural structure and fluorinated compounds present.

Besides, various studies have been conducted using nanoparticles as a functionalized material. Table 5 provides the summaries of surface functionalization of the ceramic membrane, treatment and

Table 5. Surface functionalization of membrane, treatment and performances

Substrate material	Coating material	Coating methods	Oil rejection	Contact angle of oil	Application	Reference
Stainless steel mesh (200 μm)	PDDA-PFO/SiO_2 nanocomposite	Spray casting	NA	157±2^o	Anti-fouling for hexadecane water separation	[74]
Cotton fabric:	Fluoroethyl/silane/ PEG-phosphate	Wet-chemical coating	NA	150^o	Antifouling	[108]
Stainless steel mesh (34-380 μm)	Hydrogel	Dip-coating	99 %	155.3±1.8^o	Antifouling and hard recycling limitation: gasoline, diesel, vegetable oil	[23]
Stainless-steel mesh	Chitosan-based nanocomposites (PFO/SiO_2)	Spray casting	NA	157±1^o	Antifouling: hexadecane and water	[109]
Stainless-steel mesh (50 μm)	TiO_2 nanoparticle	Simple and fast dip-coating	98-99%	170^o	Antifouling: hexane, vegetable oil, n-hexane	[110]
Cotton fabric	PDDA, PFO	UV-irradiated dip coating	99%	150±1^o	Antifouling: hexadecane	[111]
Al_2O_3 membrane	TiO_2	Simple magnetron sputtering and hydrothermal oxidation	99.1%	150^o	High oil-water selectivity, Antifouling	[76]
Cellulose membrane	Zn-Al layered double hydroxide nanosheets	Surface grafting	99.94%	150^o	Oil-water separation	[112]
Membrane materials (kaolin, quartz, alumina oxide)	Silica	Dip coating	99.95%	157.8^o	Oily wastewater separation	[60]
Titanium plate and mesh	Titania nanowire	Immersion coating	99.7%	157±1.5^o	Oil repellence and oil-water separation	[113]
Titania foam	TiO_2-oxyfluoride	Hydrothermal	99%	156^o	Oil water emulsion separation	[76]
Co_3O_4/graphene oxide composite	TiO_2	Vacuum deposition	99.5%	150^o	Oil water emulsion separation	[68]
Cu mesh	Hydrogel	Layer by layer self-assembly	97%	159^o	Oil repellence and oil-water separation	[69]

performances. The results show that the alteration of the wetting properties of the membrane surface for proper oil and water separation is achievable. Different coating strategies were explored: dip, cast, vacuum deposition, spray, and wet-chemical; all have shown excellent adhesion onto the substrate. Oil rejection and contact angle from all the studies revealed that contact angle, CA above 150^o and oil rejection of more than 98% were achieved.

CONCLUSION, CHALLENGES AND FUTURE WORK

Oil-water separation requires cost-effective, highly efficient, and environmentally friendly membrane fabrication, to achieve an excellent superhydrophilic and superoleophobic with an appropriate surface functionalization approach. This review has discussed and elaborated ceramic membrane and the surface matrix functionalization that have considered both the physical and chemical treatment as an approach to membrane surface modification.

Furthermore, the setback and other associated challenges such as the processing of low-cost starting material and the fouling with severe flux decline over time, which also impedes long term operation of the membrane in oily wastewater treatment. This, however, can be addressed by exploring the use of other low-cost starting materials such as ball clay and bentonite clay for the fabrication of ceramic membrane and their applications in the oily wastewater separation as researches are still very limited. These materials are largely abundant, with high plasticity and low refractory as discussed in section 2. It will also require the use of appropriate surface functionalization materials in order to address the decline associated with membrane fouling, which will involve the use of different grafting materials, most especially titanium dioxide (TiO_2) and silicon dioxide (SiO_2) as a composite material. Hence, the goal is to make the physical structures (oleophobicity) of the membrane with the capacity to exhibit an excellent structural property such as improved particle size, crystalline quality, and specific surface area. Some of these materials have shown to be low-cost materials and exhibited good chemical and thermal stability, and non-toxicity. However, a ceramic membrane with appropriate grafting material will provide better tunable superoleophobicity and superhydrophilicity, high surface area and ion exchange capacities and pigmentary properties.

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