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A Review on Membrane Filtration and Its Advantages over Conventional Approaches

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Abstract: Clean water supply has become one of the biggest challenges of the 21st century; therefore, water source protection is of increasing importance. Beyond environmental protection reasons, economic concerns—derived from increasing costs of processing water and wastewater discharge—also prompt industries to use advanced wastewater treatment methods, which ensure higher purification efficiency or even the recycling of water. Therefore, highly effective treatment of oily wastewaters has become an urgent necessity because they are produced in high quantities and have harmful effects on both the environment and human population. However, high purification efficiency can be difficult to achieve, because some compounds are hard to eliminate. Conventional methods are effective for the removal of floating and dispersed oil, but for finely dispersed, emulsified and dissolved oil advanced methods must be used, such as membrane filtration which exhibits several advantages.

Keywords: membrane fouling, modified membrane, oil contamination, oil-in-water emulsion.

1. Introduction:

Large amounts of oily wastewaters are produced by many different industries, such as oil refining, oil storage, transportation, metal, lubricant, oil, petroleum, and food industries.1–3 The treatment of oily wastewater is necessary because its contaminants can negatively affect both the ecosystem and human population, lowering the quality of superficial and groundwater, compromising aquatic lives and human health, affecting the quality of soil and crop production, and polluting the atmosphere with volatile contaminants.4–6 In addition, there are economic reasons—derived from increasing costs of water processing and wastewater discharge—that make the development of efficient treatment methods necessary, while also ensuring high purification efficiency or even water recycling. It is difficult to carry out this task due to several inorganic and organic substances, that is, dissolved minerals, dispersed and emulsified oils, and dissolved organic compounds, gases, and traces of chemicals used in the industries.7,8 The treatment of oily wastewater can be carried out by conventional methods, such as skimming,9,10 flotation,11,12 chemical destabilization using coagulation/flocculation methods,13,14 electrocoagulation,9,15,16 centrifugation,5,17 and biological treatments.18 These are effective for the elimination of floating oil (d > 150 μm) or even dispersed oil (d > 20 μm), but for finely dispersed, emulsified, and dissolved oily contaminants, advanced methods must be used. Membrane filtration is a promising method to treat these kind of pollutants due to several advantages like easy integration, and high removal efficiency.6,19,20 With the combination of one or more conventional methods and membrane filtration, the desired high purification efficiency can be achieved. Nowadays, the treatment tend to still be too expensive and/or too time-consuming,21 because the major limitation to the application of membrane-based oily wastewater treatment is fouling, that causes severe flux decline and reduces membrane performance.8,21–23 Besides, regular filtration shutdowns—to clean the membrane and recover the permeability—increase the costs and complexity of the system. The chemicals used for cleaning the membrane surface also increase the costs and reduce membrane performance and lifespan.24

Researchers all over the world carry out in-depth investigations into possible solutions to make the method both technically and economically feasible. In order to reduce the disadvantages, it is necessary to use appropriate pretreatments and/or increase membrane hydrophilicity to decrease the fouling properties.25 Based on this, the development of ultra-hydrophilic membranes with structures containing nanomaterials is revolutionizing the separation of oily wastewater by avoiding the attachment of oil droplets on the surface and stabilizing the filtration resistance at a low level, resulting in membranes without significant fouling properties.6,26,27 Photocatalytic nanomaterials promise further advantages for the preparation of highly hydrophilic, self-cleaning membranes, as these materials can decompose not only the organic pollutants from the surface when activated by artificial or solar irradiation28,29 but also the organic contaminants of the fouled pores as well, converting them into small (or even nontoxic) substances—without the formation of secondary pollutants.28–30 The most widely investigated photocatalytic material is titanium dioxide (TiO_2) due to its low cost, availability in large quantities, high chemical stability, and photocatalytic activity, etc.31 However, despite the numerous advantages of TiO_2, it can be activated mainly by ultraviolet (UV) light, which makes up a small fraction of solar light; therefore, solar systems that use only pure TiO_2 have limited efficiency in the degradation of hydrocarbons.32 Considering this, and the fact that artificial

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UV-light-based activation needs significant electrical energy, the development of visible-light active photocatalytic materials and their use in the preparation of solar-light active membranes has a huge potential to achieve high efficiency during pollutant removal and/or membrane surface cleaning.33 Solar-light
active superhydrophilic photocatalytic membranes could be the future's novel solution for advanced oil-in-water emulsion separation as they will be able to minimize the fouling problems, and be cleaned in a chemical- and energy-free manner, by simple solar light irradiation. The present work aims to provide an overview of the characteristics and effects of oily wastewaters and their suitable treatments, starting with conventional methods explaining their advantages and disadvantages. This study also deals with the membrane filtration process and its limitations related to flux reduction and fouling problems. The possible solutions, that is, modified membranes and the development of hydrophilic nanomaterials, to enhance membrane performance are described in detail. Furthermore, photocatalytic nanocomposite-modified membranes are also discussed as future perspectives.

2. Related Work:

Oily wastewaters consist of mainly oil, salt, and surfactants, but they also contain numerous harmful compounds: saturated straight-chain and branched hydrocarbons, cyclic hydrocarbons, olefins, aromatic hydrocarbons, and other non-organic substances, such as sulfur- and nitrogen-containing compounds, and heavy metals.8,34,35 The damage caused by oilcontaminated waters depends on the type, volume, and quality of the polluting oil, but also on the place and conditions of the discharge. Oily wastewaters can have harmful effects on organisms due to coating, asphyxiation, poisoning, or causing sublethal and stress effects, reducing the abundance and diversity of the fauna and flora.36 Soils can also be affected by oil contamination, reducing bacterial activity, killing earthworms, reducing
plant growth, affecting root elongation, and germination,37 which also affects crop production and groundwater quality.4,5 In relation to animals and human beings, effects of oil contamination can range from acute symptoms to chronic diseases, and by the accumulation in food chain, it can cause DNA damage, genotoxic, carcinogenic, and mutagenic effects, where the possible consequences include allergies, respiratory problems, autoimmune disorders, spontaneous abortion, or even cancer.34,38,39 There are regrettable examples, where nature has been severely damaged by years of oil contamination, and most of the population has been constantly exposed to crude oil through the water, air, and soil, and the number of degenerative diseases increased and life expectancy decreased.34,40 These examples highlight the importance of developing efficient oily wastewater treatments.

To reduce the chemical oxygen demand (COD) and biochemical oxygen demand (BOD) in oily wastewaters, the most common methods are skimming, centrifugation, flotation, and chemical destabilization. Biological decomposition, which uses anaerobic and aerobic bacteria, must also be listed here as it is a conventional wastewater treatment method; however, it is a novel and dynamically developing technique in this field. These conventional methods are often not efficient enough to achieve the newest limit values of treated wastewater, due to the complexity of the mixture and the presence of emulsified/dissolved oil and/or non-biodegradable organic contaminants.5,21,41,42 But, due to low operation costs and high purification efficiency in the case of floating/dispersed oils and biodegradable organic compounds, highly efficient combined treatments usually start with (or contain) one or more of these methods as pretreatment(s).43,44.

One of the most conventional oil separation method is skimming, a simple gravity separation method based on the density difference between the oil and water, in which the oil rises to the top of the device, and the suspended solids sink to the bottom of the separator.45 The API tank—designed according to American Petroleum Institute standards—is a widely used and simple separator that can eliminate droplets bigger than 150 μm.46 There are parallel and/or corrugated plate separators that can enhance gravity separation and remove oil droplets bigger than 50 μm.47 However, these skimming devices generate a large amount of sludge and are not efficient at eliminating the finely dispersed and dissolved oil, which makes it necessary to combine this method with other treatments.

Centrifugal forces can be utilized to increase the flow rate and/or purification efficiency, which can be achieved using centrifuges or hydrocyclones. The advantages of centrifugal separation are high throughput capacity, smaller equipment, and shorter residence time compared with simple gravity separation.10,14 During the application of this method, two forces are acting on the oil droplets: (a) buoyancy, which is responsible for the upward movement of the droplet as a result of the density difference between oil and water, and (b) drag force, which opposes buoyancy until the rise velocity reaches a terminal value when the two forces are equal.10 This terminal velocity is used as a separator design criterion and determines the droplet sizes that can be separated at a given resident time and throughput capacity.

According to Benito et al.,14 centrifugation can be used to treat both mineral and semi-synthetic oil containing water with >90% purification efficiency; however, the treated water still can still contain up to 1500 mg/L oil. Separation efficiency can be improved by increasing the buoyancy force and/or the droplet diameter.10 These aims can be achieved by coagulation/flocculation methods and bubble production-based flotations, which are detailed in the following sections.

In contrast with conventional centrifuge machines, hydrocyclones have no moving parts, just a cylindrical and a conical part. The fluid is injected tangentially through the inlet in the upper part, resulting in strong swirling motion and therefore, high centrifugal forces. As the fluid passes through in a spiral fashion, dense particles are forced against the wall and migrate downwards to the underflow; meanwhile, fine or low-density particles move upward to the overflow. Despite their advantages, hydrocyclones have low separation efficiency when the dispersed oil droplet diameter is smaller than 15 µm.48 In order to increase the oil removal efficiency, special hydrocyclones have been developed, such as the bubble enhanced hydrocyclone, in which the oil droplets and the bubbles collide with each other, and thus are carried out of the cyclone.48

Chemical destabilization involves the usage of chemicals (coagulants and flocculants) to neutralize the surface of colloids and to agglomerate them into bigger particles and flakes, which can be more easily removed by other separation techniques such as skimming or flotation. This method is widely used to treat wastewater because of its simplicity, low-energy consumption, easy operation, and versatility.13,49 The most widely used coagulants for the treatment of oily wastewaters are aluminum sulfate, ferric chloride, and polyaluminum chloride,17,42,50,51 but the development of novel, more efficient, and cost-effective coagulants is also an important field of research. For instance, Zeng et al.52 combined a coagulation/flocculation system with polyzinc silicate and anionic polyacrylamide to remove oil from heavy oily wastewater and removed more than 99% of the suspended solids and oil. Zeng and Park53 observed higher coagulation/separation performance by using zinc silicate and anionic polyacrylamide, compared with conventional coagulants. According to the authors, the addition of zinc more favorably neutralizes charges of the colloidal particles in oil-contaminated water, more effectively reduces turbidity, suspended solid content, and COD in a broader pH range.

Electrocoagulation is also a possible method to treat oily wastewaters, which requires smaller amounts of reagents compared with conventional coagulation, forming a smaller volume of sludge.15,54 The process is based on the in situ generation of coagulants by electrically dissolving aluminum or iron ions, which then attract fine, negatively charged droplets and particles. Due to the reduced surface charges and resulting coalescence of the droplets, they can be easily separated.50 The metal ions are generated at the anode, and hydrogen gas is released from the cathode. Hydrogen gas also helps to float the flocculated particles to the top of the water.55 According to Ögütveren et al.,16 this method can be effective to destabilize oil-in-water emulsions and the use of aluminum is more effective and requires less energy.

After the application of the previously detailed conventional methods, wastewaters often still contain significant amounts (few to hundreds of mg/L) of microscale and/or nanoscale oil droplets, requiring further treatment before discharge or reuse. Membrane filtration can be a good choice because it minimizes additional costs, it does not require chemical additions, it is easy to handle, its energy requirement is low, and it can still reach high removal efficiency.8,19,20,22,66 Therefore, this process is more frequently used to treat oily wastewater and to overcome the deficiencies of previously mentioned conventional methods.

During membrane filtration the membrane separates the contaminants from the water with a physical barrier that allows water to flow through the membrane, while the other substances are retained by the membrane surface. The water crosses the membrane due to a driving force, such as concentration difference, electric potential, partial pressure, or hydraulic pressure. The membrane separation behavior depends on adsorption, sieving, and electrostatic phenomena.1,19,67 In general, membrane separation by itself is effective to remove (oily) contaminants, but it is beneficial to minimize the accumulation of them on membrane surfaces to provide higher fluxes, so conventional methods combined with membrane filtration can be used, such as flocculation with membrane microfiltration (MF),68 or filtration and centrifugation followed by ultrafiltration (UF).14 Another technique to increase the membrane filtration efficiency is to combine different membrane processes, such as MF with UF or UF with reverse osmosis (RO), etc.5

Membrane filtration processes can be characterized according to the pore size or molecular weight cut-off (MWCO) value that defines the size of particles/ droplets/molecules/ions, which are retained by the membrane surface. This value decreases from MF to UF (UF), nanofiltration (NF), and RO, and the hydrodynamic resistance for water to pass through the barrier increases in this order.19,20

MF is usually used when the aim is to remove bacteria, suspended soils, or substances with sizes between 0.1 and 10 µm.19,69 László Kiss et al.70 used an MF membrane to separate the oil content of an oil-in-water emulsion and achieved higher retention rates in the case of relatively high oil concentration. Nandi et al.71 used a low-cost MF membrane and removed 98.8% of the oil.

Wang et al.72 used MF membrane to treat oily wastewater and recovered the membrane with simple cleaning, and aeration at regular intervals significantly improved performance.

Currently, by using MF, satisfactory water purity can be achieved when it is combined with other technologies or when the water contains low concentrations of oil compounds.21,73

UF can separate particles of 0.001–0.1 µm, macromolecules, and colloids from water; however, most of the dissolved ionic species still pass through the membrane. The application of UF to separate oily compounds has been widely studied and shows high efficiency in total organic carbon (TOC) removal and can achieve up to 98–99% of oil removal.8,19,69,73 Bodzek and Konieczny74 investigated a UF tubular membrane to treat oil emulsion and, with the pre-elimination of suspended oil, they achieved 99% COD removal efficiency. Srijaroonrat et al.25 used a UF membrane to treat oil-in-water emulsions, applying cyclic backflushing to recover the original performance and measured 50–120% higher steady-state fluxes (depending from the used transmembrane pressure) by using very brief (0.7 s) backflushing every minute.

NF can separate divalent ions, small organic molecules, and inorganic molecules with a size of 0.0005–0.001 µm. Therefore, this method is commonly used in desalting procedures, drinking water generation, textile and paper industry, etc. The rejection of solutes can achieve >99% efficiency.19, 21, 69 Among the various types of membrane separation processes, RO is the one which can eliminate even the finest molecules except water. It requires larger amounts of energy compared with the others, because water molecules have to be pushed through the membrane against a high osmotic difference with a high-pressure pump.21 The disadvantages of NF and RO—when treating oily wastewater—compared with MF and UF, are the higher fouling tendency, more difficult fouling recovery, and lower fluxes.1 These technologies can be used if the salt content is high in the oily wastewater.41

5. Conclusion:

The effective treatment of oily wastewater is necessary because these wastewaters are produced in high quantities and oil discharge causes damage to the natural environment and endanger human health. To meet the stringent emission limits, conventional techniques such as skimming, centrifugation, chemical destabilization, flotation, and biological treatment are not enough, because these methods are not able to remove small (submicron and nano-scaled) emulsified oil droplets. Membrane filtration plays an important role to complement the conventional treatment methods of oily wastewaters and to effectively remove these finely dispersed oils. There are different suitable types (MF, UF, NF, and RO) and materials (polymer and ceramic) of membranes; however, membrane fouling and flux reduction are still the major limitation to the application of these technologies.

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