Review

A Review on the Nanofiltration Process for Treating Wastewaters from the Petroleum Industry

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Abstract: Activities and/or processes in different segments of the petroleum industry, including upstream and downstream, generate aqueous waste streams containing oil and various contaminants that require treatment/purification before release/reuse. Nanofiltration (NF) technology has been approved as an efficient technology for treating wastewater streams from the petroleum industry. The primary critical issues in an NF treatment process can be listed as mitigation of membrane fouling; selection of appropriate pre-treatment process; and selection of a suitable, cost-effective, non-hazardous cleaning strategy. In this study, NF separation mechanisms, membrane fabrication/modification, effective factors on NF performance, and fouling are briefly reviewed. Then, a summary of recent NF treatment studies on various petroleum wastewaters and performance evaluation is presented. Finally, based on the gaps identified in the field, the conclusions and future perspectives are discussed.

Keywords: oily wastewater; produced water; refinery; membrane; nanofiltration; fouling

1. Introduction

Petroleum (crude oil and natural gas) is one of the world’s main energy sources; and it is an essential provider for many other industries [1,2]. Water is used in different segments of the petroleum industry including upstream and downstream for different applications such as production, cooling, washing, processing, etc. [3]. The exploration and production of petroleum, processing of hydrocarbons in refineries and petrochemical plants, and even other activities like storage, transportation, and distribution of petroleum products [2,4,5], can generate aqueous waste streams containing oil and various contaminants that require treatment/purification before release/reuse. If not suitably treated, the oily wastewater streams not only contaminates the environment and endangers water resources and human health but also decreases the reuse capability of oil and water [2,6–15].

Produced water, water produced as a byproduct during the extraction of oil and natural gas, from both oil and gas fields is the petroleum industry’s most massive waste stream by volume [16,17]. It has a complex composition consisting of various organic and inorganic compounds [18,19]. There are different approaches regarding the waste management of produced water including (i) avoiding the production of water onto the surface by polymer gels or downhole water separators; (ii) injecting into formations after probable treatment to decrease fouling and bacterial growth; (iii) possible discharging to the environment according to the discharge regulations; (iv) reusing within the petroleum industry operations with minimal treatment; and (v) remarkable treatment for beneficial uses [16,18,20].

In the petroleum industry, a range of wastewater treatment technologies, including primary treatment processes such as physical and physicochemical processes; secondary treatment processes such as suspended and/or attach growth biological processes; and tertiary treatment processes such as sand filtration, membrane processes, ion exchange,
chemical oxidation, advanced oxidation processes (AOPs), etc., have been utilized to treat wastewater streams [2,11–14,21–24]. There is an increasing interest in designing the new energy-efficient, cost-effective, reliable, resilient, and sustainable wastewater treatment systems [25].

Pressure-driven membrane processes are the most commercial membrane filtration technology [26]. Based on the membrane pore sizes, these processes have typically been classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [27]. These membranes have usually been utilized to treat wastewater streams from the petroleum industry by applying high pressure (high energy) across the membranes [28–32]. In comparison with the conventional treatment techniques, the membrane technology offer advantages such as effective removal of oil, compact design, less-necessity for chemical additives [33–37] and stable effluent quality [36]. The feed streams with high oil concentration cannot be treated by MF and UF due to their relatively larger membrane pores, whereas NF and RO, with higher rejection in comparison with MF and UF, suffer from high energy consumption [2,32,38–41]. In the tradeoff between the acceptable rejection and energy-consumption, NF has potential to replace RO membranes because of lower operating pressure and/or energy consumption [27,42–45], relatively lower investment, operation, and maintenance costs [44]. There is a need to comprehensively review treating wastewater streams from the petroleum industry using the NF process. Thus, this study intends to review the treatment of petroleum wastewater streams by NF technology and then, based on the gaps identified in the area, discuss the conclusions and future perspectives.

The remaining sections of this paper are organized as follows. In Section 2, NF separation mechanisms, membrane fabrication/modification, effective factors on NF performance, and fouling are briefly reviewed. Section 3 includes a discussion of recent NF treatment studies on various petroleum wastewaters and performance evaluation. Finally, Section 4 discusses conclusions and future perspectives of this study.

2. Nanofiltration
2.1. Nanofiltration Fundamentals

In the mid-1980s, Eriksson [46] used the term NF for the new class of membranes that their characteristics fall between UF and RO [2,26,46–49]. The pore size and molecular weight cut-off (MWCO) of NF membranes are 1–10 nm [50–52] and 100–2000 Da [26], respectively. The operating pressure is usually 5–35 bar [49]. These membranes are relatively impermeable to divalent ions, dissolved organic matter, pesticides, and other macromolecules, but tend to pass monovalent ions [2,27,51,53,54].

Wetted surface, preferential sorption-capillary, solution-diffusion, charged capillary, and finely porous rejection mechanisms have been presented by Macoun [55] as the major rejection mechanisms. Further information can be found in Macoun [55] and Shon et al. [27]. A combination of charge effect repulsion, solution diffusion, and sieving through micro/nano-pores have been reported as separation mechanisms [2,26,56]. Among the mentioned mechanisms, the sieving and charge effects are two dominant separation mechanisms of NF membranes. Uncharged or high molecular weight solutes are separated by sieving or size exclusion mechanism. Whereas the charged solutes are separated by both sieving and the electrostatic interaction between the solute species and the membrane surface (Donnan phenomenon) [26,37,47,49,52,57,58].

2.2. Nanofiltration Process Applications

NF membranes can relatively reject divalent ions, multivalent ions, organics, starch, sugar, pesticides, herbicides, and other macromolecules [16,54,56]. In comparison with MF and UF processes, this process has higher efficiencies in the reduction of chemical oxygen demand (COD) and total dissolved solids (TDS) and also operates under low pressure (i.e., low energy usage) conditions compared to RO process [59]. Thus, there has been an increasing interest to use NF technology as an effective process in a variety of applications:
• Food industries including dairy [60,61], beverage [62–64], sugar [65,66], vegetable oil [67,68] and plant extracts [49,69];
• Textile industry and dye concentration [49,70–73];
• Biotechnological/pharmaceutical industry [49,50,74–76];
• Water purification: Water softening, removal of natural organic matter, heavy metals, viruses and bacteria from water [37,49,50,77–81]; and
• Wastewater treatment: Olive mill wastewater [82,83], coke wastewater [84], municipal wastewater [85,86], leachate [87,88], car wash wastewater [89], pulp and paper wastewater [90], oily wastewater from the petroleum industry [59], etc.

2.3. Factors Affecting the Nanofiltration Process Performance

NF process performance is significantly influenced by membrane characteristics, feed characteristics, and operational conditions [26]. Membrane characteristics including MWCO, porosity, morphology, charge, and hydrophilicity can dramatically affect the NF process performance. Additionally, membrane performance is strongly influenced by feed characteristics such as the molecular weight, molecular size, geometry, charge, hydrophilicity of the solute and the feed water chemistry (e.g., pH) [26,42,91,92]. Furthermore, operational conditions such as temperature, pressure, and flow rate can impact the separation process [26]. Bellona et al. [42] and Mulyanti and Susanto [26] completely reviewed the effective factors on NF process.

Rahimpour et al. [47] investigated the effect of operating variables including temperature and trans-membrane pressure (TMP) on the permeate flux, COD, and electric conductivity (EC) in NF treating the oily wastewater generated by the washing of gasoline reserving tanks. The permeate flux, COD, and EC removal were enhanced with increasing TMP. COD and EC removal were reduced with an increase in temperature, whereas the permeate flux was increased. Pressures of 15–20 bar and temperatures of 20–30 °C were reported as the optimum conditions for the permeate flux and COD removal [47]. Additionally, Salahi et al. [36] reported the optimum permeate flux of 180.1 L/m²·h at feed temperature of 45 °C, TMP of 4 bar, the cross flow velocity of 1.3 m/s, pH of 10 and salt concentration of 11.2 g/L in NF treating the desalter effluent wastewater from Tehran refinery using nano-porous membrane [36]. Furthermore, Hedayatipour et al. [93] investigated the effect of temperature, pH and TMP on removal efficiency of Ba, Ni, Cr, NaCl, and TDS from the effluent of the dewatering process in an oil and gas well drilling industry by NF process. The temperature of 25 °C, the pressure of 170 psi and pH of 4 were reported as optimum conditions which 85.3%, 77.4%, 58.5%, 79.6%, and 56.3% removal efficiencies were obtained for Ba, Ni, Cr, NaCl, and TDS, respectively [93].

2.4. Fabrication and Modification of Nanofiltration Membranes

Surface chemistry, porosity, pore size distribution, physicochemical compatibility with process feeds, lifetime, and cost are key factors to fabricate the NF membranes [49]. In recent years, researchers have focused on fabricating and developing various polymeric, ceramic, and hybrid ceramic-based NF membranes [94]. Each NF membrane type has advantages, disadvantages, and specific applications; however, polymeric NF membranes have been extensively studied due to their availability, easy modification [94], and good film-forming property [95]. Recent advances and research trends in NF membranes fabrication and modification have been reviewed (e.g., Mohammad et al. [50]; Paul and Jons [96]; Oatley-Radcliffe et al. [97]; Ji et al. [95]; Rabbani Esfahani et al. [92]; and Merlet et al. [94]).

Material selection, additive concentrations, and modification techniques can play important roles in obtaining optimal NF membranes [52]. Different materials and techniques have been used to fabricate NF membranes. Materials such as polymer, ceramic, or a hybrid consisting of both may be used in the structure of a membrane from the active (selective) layer to the porous support layer(s). The porous ceramics for NF are composed of oxide materials [94]. Polysulfone (PSF) [98], polydimethylsiloxane (PDMS) [99], polyethersulfone (PES) [100,101], poly(ether ether ketones) (PEEK) [102], poly(vinylidene fluoride) (PVDF),
cellulose acetate (CA) [103], aromatic and semi aromatic polyamides [104,105], polybenzimidazole (PBI) [106], polyaniline (PANI) [107], and polyacrylonitrile (PAN) [52,108] have been reported as applied polymers to prepare polymeric NF membranes. Note that both polymeric and ceramic NF membranes used for treating wastewaters from the petroleum industry are discussed in detail in Section 3.

Interfacial polymerization (IP), phase inversion, UV/photo-grafting, electron beam irradiation, plasma treatment, layer-by-layer, etc. are several approaches to fabricate the polymeric NF membranes [50,96]. IP is the common technique to prepare thin film composite (TFC) NF membranes [44,109]. TFC membranes are made of one support layer and one thin active layer on the top of the support layer [110,111]. They are the main type of RO, NF, and forward osmosis (FO) membranes [112,113]. The incorporation of nanoparticles into the TFC membranes results in thin-film nanocomposite (TFN) membranes. Different techniques such as in-situ/interfacial polymerization [114–116] and dip coating methods [117–119] have been reported for the fabrication of TFN membranes. In order to prepare novel TFN membranes with specific characteristic, nanoparticles in the range of 20–200 nm have been incorporated within the ultrathin active layer or support layer during the fabrication process [120].

Plate and frame module, tubular membrane module, spiral wound module, and hollow fiber membrane module are four configurations of NF membrane elements [49].

2.5. Fouling and Control

Membrane fouling is one of the important inevitable challenges in NF process that can be because of blockage of the membrane surface and pores by colloidal, microbiological, and chemical (organic and inorganic) components [26,27,45,47,49,50,121]. It may be reversible or irreversible [56]. Generally, solutes adsorption on the membrane surface or in pores, blockage of pores by solutes, cake layer formation and gel layer formation are forms of fouling [50,122]. Fouling leads to reduction in NF process performance (e.g., flux decline) and cost efficiency [26,50].

Physical, chemical, and hydrodynamical techniques may be used to control membrane fouling [26]. Using pre-treatment processes (e.g., coagulation, flocculation, ozonation, adsorption, MF, UF) upstream of NF, operating the system with high cross-flow velocity, using a cleaning cycle, backwashing or backflushing, and changing the operating temperature are some strategies that may be considered to prevent and mitigate the fouling [27,50,56,123,124]. Note that chemical cleaning may damage the membrane structure [26,125] and suitable cleaning agent and conditions of the cleaning process should be selected to maintain membrane performance [49,126].

Kim et al. [127] studied coagulation-flocculation-sedimentation with and without coagulant and coagulant aids as pre-treatment methods of NF process to treat oil sands process-affected water (OSPW), and concluded that the strategy improves the desalination of OSPW using NF membrane [127]. Additionally, Moser et al. [23] used direct UV and hydrogen peroxide-assisted (UV/H$_2$O$_2$) photolysis as pre-treatment methods for NF treating membrane bioreactor (MBR) permeate of a petroleum refinery to mitigate fouling. High quality water was produced using a MBR-H$_2$O$_2$/UV-NF system that could be reused in the refinery process (e.g., in cooling systems) [23].

3. Literature Review of Petroleum Industry Wastewater Treatment by Nanofiltration

The required discharge standards from the petroleum industry cannot be reached by common treatment methods. In addition, the need for water reuse in the petroleum industry drive attention to use effective technologies like membrane separation processes (e.g., NF) for better performance and optimized cost [128,129]. However, membrane fouling by oil, sulfides, or bacteria and generation of hazardous reject streams can be drawbacks of these processes [17,129]. In general, several studies have revealed that enhanced flux, minimized membrane fouling, simple cleaning strategy, and chemical and
thermal stability of membranes are major issues/barriers for utilization of membrane separation technologies in the petroleum industry wastewater treatment [1,17,130].

Over the last 30 years, NF technology has been used to treat various wastewater streams from the petroleum industry [1,16,23,36,37,44,47,52,53,59,93,127,129–145]. A summary of recent NF treatment studies on various petroleum wastewaters and performance evaluation is listed in Table 1. Various wastewater streams including produced water, OSPW, desalter effluent wastewater from a refinery, MBR permeate from refinery plant, refinery’s clarifier effluent, oily wastewater from washing of gasoline reserving tanks, etc. have been successfully treated by NF. Both polymeric and ceramic NF membranes have been applied. In other words, polymeric membranes, such as polyamide (PA) TFC NF membrane (NF-90), piperazine-based semi-aromatic PA TFC NF membrane (NF-270) [136], unmodified and poly(N-isopropylacrylamide) (PNIPAAm) and PNIPAAm-block-poly(ethylene glycol methacrylate) (PPEGMA) modified PA TFC NF (NF-270) membranes [137], nano-porous membrane (polyacrylonitrile) [36], PA-SiO$_2$ nanocomposite NF membrane [44], PSF-penta-block copolymer (PBC) composite NF membrane [37], PAN NF membrane [52], NF membrane with graphene oxide (GO)/aminated GO (NGO)-incorporated substrate [140], PES-poly acrylic acid (PAA)-ZrO$_2$ NF membrane [142], etc., and ceramic NF membranes [130,141,143] have been used to treat wastewater streams from the petroleum industry. As presented in Table 1, for instance, almost 100% removal of total suspended solids (TSS), 44.4% removal of TDS, 99.9% removal of oil and grease content, 80.3% removal of COD, 76.9% removal of biological oxygen demand (BOD$_5$) [36], 72–89% rejection of soluble organics [53], 6–43.7% retention of benzene, 19–89.2% retention of toluene, 48.5–98.5% retention of p-xylene, 48.5–98.7% retention of m-xylene, 30.7–98.7% retention of o-xylene, 21 ≥ 99.9% retention of 2-isopropyl phenol, 19.6–99.5% retention of 4- or 3-isopropyl phenol [132], higher than 95% rejection of total organic carbon (TOC), higher than 95% rejection of naphthenic acids (NAs), 62–66% rejection of sodium, higher than 92% rejection of calcium, higher than 90% rejection of magnesium, 95–98% rejection of sulfate, 20–39% rejection of chloride, 58–81% rejection of bicarbonate, and permeate flux of greater than 15 L/m$^2$/h [133] have been reported in different research studies using various NF membranes.

Peng et al. [133] investigated the performance of three commercially available TFC NF membranes (Deasol-5 from Osmonics/Desal; NF-45 and NF-90 from Dow Chemical) for removal of TOC, NAs, and different ions from OSPW to improve water management in oil sands operation. Among these membranes, Desal-5 was reported to be a suitable membrane for this purpose. Incomplete rejection of monovalent ions of sodium, chloride and bicarbonate (20–80%), higher than 95% rejection of divalent ions (calcium, magnesium, and sulfate), TOC, and NAs were reported for Desal-5. Permeate flux decline of Desal-5 due to fouling was tested in experiments for about 18 h and flux maintaining at 15 L/m$^2$/h or higher at a pressure of 10.3 bar was reported [133]. In other work, the average efficiency of salt removal from raw OSPW using PA TFC NF membrane (GE Osmonics) was reported to be about 68.9%. The study revealed that OSPW components could bound to the NF membrane surface; and chemical cleaning using both HCl (1 mM) and NaOH (1 mM) showed similar flux recovery ratio (HCl had slightly higher recovery ratio) [127]. These studies [127,133] did not investigate the fouling mechanisms of OSPW desalination by NF; however, they addressed fouling.
Table 1. Summary of recent NF treatment studies on various petroleum wastewaters and performance evaluation.

| Membrane | Wastewater | Studied Parameters | Influent Concentration | Major Findings | Reference |
|----------|------------|--------------------|------------------------|-----------------|-----------|
| NF       | Offshore produced water | Soluble organics | 176 mg/L | 72–89% rejection of soluble organics and 15–20% removal of salts | [53] |
| NF       | Produced water | Oil, sodium, calcium, magnesium, potassium, ammonium, chloride, and sulfate | <1 ppm oil, 9610 ppm sodium, 715 ppm calcium, 412 ppm magnesium, 174 ppm potassium, 110 ppm ammonium, 8010 ppm chloride, and 1090 ppm sulfate. | Concentrations in NF permeate were: non-detectable oil, 5250 ppm sodium, 163 ppm calcium, 115 ppm magnesium, 77 ppm potassium, 68 ppm ammonium, 4710 ppm chloride, and non-detectable sulfate. Recovery was 90–95%. | [16,131,145] |
| Membranes: UTC-60 (aromatic polyamides) from Toray (Tokyo, Japan); NRT-729HF (polyvinyl alcohol/polyamides), ES-10C (polyamides), and LF-10 (polyvinyl alcohol/polyamides) from Nitto Denko (Osaka, Japan) | Soluble organic pollutants | Benzene, toluene, p-xylene, m-xylene, o-xylene, 2-isopropyl phenol, 4- or 3-isopropyl phenol, etc. | Benzene, toluene, p-xylene, m-xylene, and o-xylene concentrations were 1.25 mg/L; whereas 2-isopropyl phenol and 4- or 3-isopropyl phenol concentrations were 0.05 mg/L. Retention rates for organic compounds at 0.3 MPa varied among membranes: Benzene, 6–43.7%; toluene, 19–89.2%; p-xylene, 48.5–98.5%; m-xylene, 48.5–98.5%; o-xylene, 30.7–98.7%; 2-isopropyl phenol, 21 –> 99.9%; 4- or 3-isopropyl phenol, 19.6–99.5%, etc. Approximately, retention rates for UTC-60 < NTR-729HF < ES-10C < LF-10. | [132] |
| TFC NF membranes (Deasl-5 from Osmonics/Desal; NF-45 and NF-90 from Dow Chemical (Midland, MI, USA)) | OSPW | TOC, NAs, sodium, calcium, magnesium, sulfate, chloride, and bicarbonate | 44 mg/L TOC, 30–57 mg/L NAs, 434–1,170 mg/L sodium, 23.4–46 mg/L calcium, 13–33 mg/L magnesium, 94–1300 mg/L sulfate, 225–760 mg/L chloride, and 545–1040 mg/L bicarbonate + carbonate | >95% rejection of TOC, >95% rejection of NAs, 62–66% rejection of sodium, >92% rejection of calcium, >90% rejection of magnesium, 95–98% rejection of sulfate, 20–39% rejection of chloride, and 58–81% rejection of bicarbonate. Permeate flux was 15 L/m²/h or higher at a pressure of 10.3 bar. | [133] |
Table 1. Cont.

| Membrane                                                                 | Wastewater                             | Studied Parameters          | Influent Concentration                                                                 | Major Findings                                                                 | Reference |
|--------------------------------------------------------------------------|----------------------------------------|----------------------------|----------------------------------------------------------------------------------------|--------------------------------------------------------------------------------|-----------|
| NF-90 (Dow/Filmtec), TFC-S (Koch (MA, USA)), and ESNA (Hydranautics (Oceanside, CA, USA)) | Methane produced water                | TOC, conductivity, and iodide | TOC, conductivity, and iodide concentrations were 5243 ± 561, 9647 ± 652 µs/cm, and 55.6 ± 10.8 mg/L, respectively. | TOC, conductivity, and iodide rejection efficiencies of NF-90 > TFC-S > ESNA. TOC, conductivity, and iodide rejection efficiencies of NF-90 were 87.6 ± 0.6, 72.7 ± 5.4, and 78.3 ± 1.3, respectively. | [134]     |
| NF-90 (Dow/Filmtec)                                                     | Produced water from a natural gas production site in Eastern Montana | TDS, TOC, barium, boron, bromide, chloride, and iodide | 5520 ± 718 mg/L TDS, 2 ± 0.5 mg/L barium, 3.8 ± 0.3 mg/L boron, 51 ± 7 mg/L bromide, 3306 ± 854 mg/L chloride, and 50 ± 8 mg/L iodide | Salt rejection was 85.3–94.9%. Concentrations in the NF final product water were 566 mg/L TDS, 0.08 mg/L TOC, 0.02 mg/L barium, 2.6 mg/L boron, 14.0 mg/L bromide, 372 mg/L chloride, and 22.9 mg/L iodide. | [135]     |
| Piperazine-based semi-aromatic polyamide TFC membrane (NF-270) and polyamide TFC membrane (NF-90) from Filmtec (MN, USA) | Produced water from Colorado, USA     | TDS and TOC                | TDS and TOC were 722–2090 ppm and 68.8–136.4 mg/L, respectively.                      | NF 270 had the largest membrane pore size; the conductivity, TDS, and TOC of the permeate were the highest. | [136]     |
| NF-200 (Polyamide TFC from Filmtech (MN, USA))                          | Vakiflar oil produced water           | COD, TDS, sodium, chloride, and salinity | 1483 mg/L COD, 6510 mg/L TDS, 5169 mg/L sodium, 2949 mg/L chloride, and 6.7% salinity | Effluent concentrations were: 137 mg/L COD, 2240 mg/L TDS, 1059 mg/L sodium, 1200 mg/L chloride, and 2.3% salinity | [1]       |
| Unmodified and poly(N-isopropylacrylamide) (PNIPAAm) and PNIPAAm-block-poly(ethylene glycol methacrylate) (PPEGMA) modified NF-270 polyamide TFC membranes | Coal bed methane produced water       | TDS and conductivity       | TDS and conductivity were 722 ppm and 1448 µs, respectively.                          | Effluent TDS and conductivity for unmodified membrane were 648 ppm and 1297 µs, respectively. Whereas effluent TDS and conductivity for one of the modified membrane were 342 ppm and 694 µs, respectively. | [137]     |
| Ceramic NF membrane                                                     | Oilfield produced water               | Oil and TOC                | Oil and TOC were 113 and 94 ppm, respectively.                                         | Oil and TOC removals were 80% and 13%, respectively.                            | [130]     |
| Polyamide TFC NF membrane from GE Osmonics (Fairfield, CT, USA)        | OSPW                                   | Salts                      |                                                                                       | The average efficiency of salt removal from raw OSPW was about 68.9%             | [127]     |
| Membrane  | Wastewater                          | Studied Parameters | Influent Concentration                                                                 | Major Findings                                                                                                                                  | Reference |
|-----------|-------------------------------------|--------------------|----------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------|-----------|
| Polyamide TFC NF commercial membrane (NE2540-90, SAEHAN Corp., Korea) and self-made TFC NF membrane | Oily wastewater from washing of gasoline reserving tanks | COD and EC | The COD and EC of pre-treated wastewater were 2940 ppm and 73 μs/cm, respectively.    | The COD and EC removals were 84% and 88% for commercial membrane and 79% and 93% for self-made membrane, respectively. | [47]      |
| NF-90 (Dow Filmtec) | The MBR permeate from REGAP-Gabriel Passos Refinery Plant, Brazil | Ammonia, chloride, calcium, nitrite, COD, TOC, and TDS | 30 mg/L ammonia, 573 mg/L chloride, 34 mg/L calcium, 0.66 mg/L nitrite, 440 mg/L COD, 91 mg/L TOC, and 1575 mg/L TDS | 98.60% removal of ammonia, 98.75% removal of chloride, 100% removal of calcium, 100% removal of COD, 99.36% removal of TOC, and 98.35% removal of TDS | [23]      |
| Self-made polyacrylonitrile (PAN) NF membrane | Synthetic produced water | oil and salts | 10 ppm oil and 6000 ppm of salts | Water flux and overall rejection were 78.8 (L/m²·h) and 46.2%, respectively. | [52]      |
| Self-made NF membrane with graphene oxide (GO)/aminated GO (NGO)-incorporated substrate | Petrochemical wastewater and shale gas produced water | Ions | Generally, better performance of TFC_{NGO} than TFC_{GO}; remarkable increase of water flux (higher than 24.8%) and similar divalent ion rejection for petrochemical wastewater; better performance in permeability and divalent ion rejections (approximately 6% higher than pristine membrane) for shale gas produced water | | [140]    |
| A commercial titania ceramic NF membrane | Recycle water from a Canadian oil sands mine | Ions, TSS, and TOC | High rejection of divalent cations, 75-90% TOC rejection, and almost 100% TSS rejection | | [141]    |
| NF (GE Osmonics) | Whiting refinery’s clarifier effluent | Mercury | Effluent mercury concentration of <1.3 ppt | | [138]    |
| Self-made PA-SiO₂ nanocomposite NF membrane | Oily wastewater from Daqing oilfield | Salts | Nearly 50% salts removal | | [44]      |
| Nano-porous membrane (polyacrylonitrile) | Desalter effluent wastewater from Tehran refinery | TSS, TDS, oil, and grease content, COD and BOD₅ | 250 mg/L TSS, 8200 mg/L TDS, 196 mg/L oil and grease, 456 mg/L COD and 321 mg/L BOD₅ | 100% removal of TSS, 44.4% removal of TDS, 99.9% removal of oil and grease, 80.3% removal of COD and 76.9% removal of BOD₅ | [36]      |
| Membrane | Wastewater | Studied Parameters | Influent Concentration | Major Findings | Reference |
|-----------|------------|--------------------|------------------------|----------------|-----------|
| **NF1 from Amfor Inc. (Amei Ande Membrane Technology Ltd., Beijing, China)** | Produced water | TDS, oil and grease, TSS, COD, and TOC | 854 mg/L TDS, 2 mg/L oil and grease, 10 mg/L TSS, 96 mg/L COD, and 26.3 mg/L TOC | Effluent concentrations were 520 mg/L TDS, <1 mg/L oil and grease, <1 mg/L TSS, 60 mg/L COD, and 22.9 mg/L TOC | [139] |
| **TFC NF membrane (Sepro Membrane Inc., Oceanside, CA, USA)** | Oily wastewater | Oil and magnesium | Oil and magnesium concentrations were 200–2000 and 40–403 ppm, respectively. | 95–98% oil rejection and 56–99.8% magnesium rejection | [59] |
| **TFC NF membranes (HL4040F) of polyamide chemistry (GE/Osmonics)** | Oilfield produced water | TDS, hydrocarbons, oil droplets, sulfate, silica, boron, and SS | Concentrations of TDS, organics including hydrocarbons, oil droplets, sulfate, silica, boron, and SS were 96,472.6, 268.2, 120.4, 7087.5, 134.4, 29.3, and 20.2 ppm, respectively. | Intermittent chlorination/coagulation/NF combined unit efficiently rejected sulfate, uranium, and other metal cations and polished the removal of SS, bacteria, and organics. | [129] |
| **Self-made polysulphone (PSF)-penta-block copolymer (PBC) composite NF membrane** | Engine oil in water emulsion | Oil | 500–1000 ppm engine oil in water emulsion | 95.5–99.5% oil rejection; and flux recovery of 89–95% | [37] |
| **NF (Polyamide, JCM-1812-50N, USA)** | Produced wastewater from dewatering unit of an oil and gas well drilling industry | Ba, Ni, Cr, NaCl and TDS | 209 mg/L Ba, 6.2 mg/L Ni, 5.3 mg/L Cr, 14,180 mg/L NaCl and 61,500 mg/L TDS | 85.3% removal of Ba, 77.4% removal of Ni, 58.5% removal of Cr, 79.6% removal of NaCl and 56.3% removal of TDS | [93] |
| **Polyethersulfone (PES)-poly acrylic acid (PAA)-ZrO$_2$ NF membrane** | Synthetic wastewater | Polycyclic aromatic hydrocarbon (PAH) | More than 90% PAH rejection rate | | [142] |
| **Ceramic NF membranes with γ-Al$_2$O$_3$ support and ZrO$_2$, Al$_2$O$_3$ and TiO$_2$ selective layers (Rauschert Inopor, Veilsdorf, Germany)** | Produced water from different SAGD operations in Alberta, Canada | Residual organic matter | Complete removal of non-polar oil components including saturated and aromatic hydrocarbons, approximately 80% removal of polar components, and 95.0–98.3% removal of total solvent extracted material | | [143] |

BOD$_5$—biochemical oxygen demand, COD—chemical oxygen demand, EC—electric conductivity, NF—nanofiltration, NAs—naphthenic acids, OSPW—oil sands process-affected water, PAH—polycyclic aromatic hydrocarbon, SAGD—steam assisted gravity drainage, SS—suspended solids, TFC—thin film composite, TOC—total organic carbon, TDS—total dissolved solids, TSS—total suspended solids.
Several studies have been reported for produced water treatment using polymeric [1,16,53,93,129,131,134–136,139,140,145] and ceramic [130,143] NF membranes. In general, application under extreme operating conditions beyond the operating range of typical polymeric membranes, cleaning with aggressive reagents such as organic solvents or hot water steam and long lifespan can be advantages of ceramic membranes [143]. Xu et al. [135] used PA TFC NF membrane (NF-90) to treat produced water from a natural gas production site in Eastern Montana for beneficial use of it by meeting potable and irrigation water quality standards. Salt rejection was 85.3–94.9%. TDS, TOC, barium, boron, bromide, chloride, and iodide concentrations in the NF-90 final product water were 566, 0.08, 0.02, 2.6, 14.0, 372, 22.9 mg/L, respectively. Effluent water from the NF-90 could not meet US Environmental Protection Agency secondary drinking water standards with regard to chloride and TDS [135]. Mondal and Wickramasinghe [136] reported that effluent conductivity, TDS, and TOC of piperazine-based semi-aromatic PA TFC NF membrane (NF-270) were higher than those of the PA TFC NF membrane (NF-90) in treating produced water [136]. In a study [130], oilfield produced water was treated using a ceramic NF membrane and oil and TOC removals were reported to be 80% and 13%, respectively [130]. In other work [143], produced water from different steam assisted gravity drainage (SAGD) operations in Alberta, Canada was treated using ceramic NF membranes with γ-Al2O3 support and ZrO2, Al2O3 and TiO2 selective layers (Rauschert Inopor, Veilsdorf, Germany); and complete removal of non-polar oil components including saturated and aromatic hydrocarbons, approximately 80% removal of polar components, and 95.0–98.3% removal of total solvent extracted material were reported [143].

NF has been used as an effective process for sulfate removal in the petroleum industry especially in offshore oilfields [3,144]. In sulfate removal using NF process, cartridge filters are utilized upstream of the system to reduce pre-treatment upsets [3]. A pilot study (Figure 1) including membrane separation processes (cartridge filter, UF, NF, and two RO units) was conducted by Osmonics Inc. (MN, USA) in 2001 to treat produced water in northern California. Oil, sodium, calcium, magnesium, potassium, ammonium, chloride, and sulfate concentration in influent feed were 10–50, 9610, 715, 412, 174, 110, 8010, and 1090 ppm, respectively. Whereas, oil, sodium, calcium, magnesium, potassium, ammonium, chloride, sulfate concentrations in NF permeate and recovery percent were non-detectable, 5250, 163, 115, 77, 68, 4710 ppm, non-detectable, 90–95%, respectively. More than 80% was reported as the overall system recovery [16,131,145].

![Figure 1](image-url) Figure 1. Schematic flow diagram of the GE pilot scale produced water treatment system (modified after [16,131,145]).

Kim et al. [127] used PA TFC NF membrane (GE Osmonics) for desalination of OSPW and reported that coagulation-flocculation-sedimentation pretreatment of OSPW before filtration with NF is an efficient technology to manage water in oil sands operation [127]. Moser et al. [23] studied the effect of AOP (UV/H2O2) pretreatment on NF (NF-90, Dow Filmtec) process performance treating MBR permeate of REGAP-Gabriel Passos Refinery Plant, Brazil. The pretreatment mitigated the flux decline because of membrane fouling and improved membrane cleanability. Ammonia, chloride, calcium, COD, TOC, and TDS removal efficiencies were reported to be 99.07%, 98.74%, 100%, 100%, 98.95%, and 98.22%, respectively. They concluded that water produced using the MBR-H2O2/UV-NF system could be reused in the refinery process [23]. Khedr [129] proposed the “intermittent chlori-
nation/coagulation/NF” process using PA TFC NF membranes (HL4040F, GE/Osmonics) to filter oilfield-produced water for re-injection; and reported that the process could effectively reject sulfate, uranium, and other metal cations and polish the removal of suspended solids, bacteria, and organics. In addition, the process could prevent the formation of scales and biofilm as well as the related unwanted phenomena [129]. Thus, it seems that NF process should be used in combination with the other separation processes to manage petroleum industry wastewaters [2,129]. Depending on the pretreated oily wastewater quality, this process may provide effluent water for reuse in the petroleum industry applications [19,23,144]. Although the separation efficiency of RO process is better than NF process [19,127,145], the NF process can be cost-effective to reuse water in the petroleum industry [19,144].

Commercially available NF membranes have widely been used for treating wastewater streams from the petroleum industry. However, some studies have been carried out to prepare self-made NF membrane for treatment of petroleum industry wastewaters: PA-SiO$_2$ nanocomposite NF membrane for desalination of oily wastewater from Daqing oilfield [45]; PSF-PBC composite NF membrane for separation of oil-water emulsion [37]; PAN NF membrane for synthetic produced water treatment [52]; NF membrane with GO/NGO-incorporated substrate for desalination of petrochemical wastewater and shale gas produced water [140]; and PES-PAA-ZrO$_2$ NF membrane for removal of polycyclic aromatic hydrocarbon (PAH) from synthetic wastewater [142]. Limited information can be found in the literature for modification of commercially available NF membranes for petroleum industry wastewater treatment. In a study carried out by Tomer et al. [137], PNIPAAm and PPEGMA nanolayers were grafted from NF-270 (Filmtec) via surface-initiated atom transfer radical polymerization in order to mitigate fouling in treatment of coal bed methane produced water. Improved permeate water quality and constant flux for modified NF-270 were reported as compared to those of unmodified NF-270 during filtration of produced water [137].

4. Conclusions and Future Perspectives

In this study, recent NF treatment studies on various petroleum wastewaters were reviewed. Key findings of this review are:

- Approximately 100% removal of TSS, 44.4% removal of TDS, 99.9% removal of oil and grease content, 80.3% removal of COD, 76.9% removal of BOD$_5$ [36], higher than 95% rejection of TOC, higher than 95% rejection of NAs, 62–66% rejection of sodium, higher than 92% rejection of calcium, higher than 90% rejection of magnesium, 95–98% rejection of sulfate, 20–39% rejection of chloride, 58–81% rejection of bicarbonate [133], etc. have been reported in different research studies for treating petroleum wastewaters using various NF membranes.
- NF has the potential to replace RO membranes because of lower operating pressure and/or energy consumption, relatively lower investment, and more economical operation and maintenance costs.
- NF process should be used in combination with other separation processes (e.g., pre-treatment processes) to manage petroleum industry wastewaters [2,129]. Depending on the pretreated oily wastewater quality, this process may provide effluent water for reuse in the petroleum industry applications [19,23,144]
- The mitigation of membrane fouling; selection of appropriate pre-treatment technique; and selection of a suitable, cost-effective, non-hazardous cleaning strategy are the vital items in designing of NF process [17].

Further investigations on the enhanced flux, minimized membrane fouling, simple cleaning strategy, and chemical and thermal stability of membranes for long-term operations are still desirable to extensively/efficiently apply NF process for petroleum industry wastewater treatment at full-scale. In particular, further studies on fouling mechanisms of petroleum industry wastewaters (e.g., OSPW) desalination by NF [127] can be beneficial. In addition, studies on the effect of membrane properties such as membrane molecular
Separations 2021, 8, 206 12 of 17

weight cut-off and surface properties [142, 143] on its performance (e.g., NF separation efficiency) in treating oily wastewater are of interest. For instance, incorporating stable and inexpensive nanoparticles in NF membrane manufacturing technology [142] can result in developing/fabricating high performance (e.g., enhanced surface hydrophilicity and fouling resistance) membranes.

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