Review

Two-Dimensional Nanomaterial (2D-NMs)-Based Polymeric Composite for Oil–Water Separation: Strategies to Improve Oil–Water Separation

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Abstract: Oil leakage and organic solvent industrial accidents harm the ecosystem, especially aquatic and marine life. Oil–water separation is required to combat this issue, which substantially enhances the ecosystem and recovery of oils from water bodies. In this aspect, significant efforts have been made by scientists to develop newer composite materials that efficiently separate oils from water bodies with exceptional recyclability. Membrane filtration is an efficient option for oil–water separation due to its ability to separate oil from water without involving any chemicals. However, relatively less water permeability and a high degree of surface fouling limit their applicability. The advent of two-dimensional nanomaterials (2D-NMs) gives newer insight in developing membranes due to their exceptional characteristics like hydrophobicity/hydrophilicity, selectivity, antifouling ability, flexibility, and stability. Incorporating 2D-NMs within the polymeric membranes makes them exceptional candidates for removing oil from water. Moreover, 2D-NMs offer rapid sorption/desorption rates and boost water transportation. Additionally, 2D-NMs provide roughness that significantly enhances the fouling resistance in the polymeric membrane. This review focuses on properties of 2D-NM-based polymeric membrane and their roles in oil–water separation. We also discussed strategies to improve the oil–water separation efficiency. Finally, we discussed oil–water separation’s outlook and prospects using 2D-NM-based polymeric membranes. This review might provide new insight to the researchers who work on oil–water separation.

Keywords: polymers; 2D-NMs; environmental remediation; oil–water separation; membrane

1. Introduction

Oil pollution is a common pollutant that has severe concern nowadays due to technological advancement and industrialization. Industries like textiles, petrochemicals, mining, and metals produce significant oil–water waste globally. Usually, oil contaminants are mixed with water in the form of grease, fat, and petroleum products, including emulsified oil (<20 µm), dispersed oil (15–145 µm), and free oil (>145 µm) [1–4]. Moreover, oil leakage during production and transportation probably devastates the water bodies and ecosystems. Oil pollutants also increase natural resource loss [5–8]. In this aspect, energy-efficient oil–water separation technology is required to purify water from industrial discharge.

Numerous oil–water separation techniques have been developed, including physical, chemical, and biological techniques. The physical treatment methods are flocculation-coagulation, flotation, and skimming. These processes are simple, economical, and energy-efficient [9–11]. However, these are inefficient for treating wastewater contaminated with...
dissolved elements and small suspended particles. Chemical processes such as ozonation [12], electrochemical [13], photocatalysis [14], and demulsification [15] have been developed for oil–water separation. These techniques are efficient for dissolved elements and small particles. However, the processes are costly and tend to produce hazardous sludge [16]. On the other hand, biochemical methods have been developed in which some enzymes, such as lipase, break down the large molecule into small molecules [17]. The biochemical technique was cost-efficient and environmentally friendly. However, the maintenance and management of the microorganism requires sophisticated instruments and skilled personnel. Therefore, it is necessary to develop newer techniques that resolve such associated issues and efficiently separate oil–water from wastewater.

Membrane filtration is an efficient alternative for oil–water separation because it can separate huge amounts of oil waste without requiring additional chemicals. Also, the membrane separation system is more straightforward than conventional methods [18,19]. The membrane separation technologies are highly efficient, easy to scale up, and do not require additional chemical or phase change. This makes membrane separation processes a highly potential technology for portable oil and gas and offshore platforms [20]. However, it has several drawbacks, such as less water permeability and a high degree of surface fouling. For that, periodic chemical cleaning must be performed, which consumes considerable energy. In this aspect, low pressure requires high permeable membranes for oil–water separation.

Polymeric membranes are extensively used for wastewater treatment due to some unique properties of the membranes, such as easy processing, high density of packing, and cost efficiency. These characteristics make polymeric membranes efficient. However, they also have some drawbacks, such as most polymeric membranes are unstable in strong acid or alkaline environments, high temperatures, and low intrinsic permeability [21,22]. Typically, altering the polymer’s composition and morphology afterwards makes it possible to change its properties, such as hydrophility and hydrophobicity. The hydrophility or hydrophobicity of a designed polymeric composite can also be adjusted by changing the polymeric composite’s surface roughness. The shape and wrapping of another polymer used to create the membrane determine how planned polymeric composites’ hydrophilic/hydrophobic surface roughness is amplified, creating a membrane with an improved surface [23–26]. The hydrophilic/hydrophobic surface roughness of planned polymeric composites is enhanced depending on the shape and wrapping of another polymer employed to create the membrane, resulting in an improved surface texture of the membranes. Hydrophilicity and hydrophobicity are important factors that affect the oil–water separation efficiency.

Researchers are continuing the development of membranes having fouling resistance with high resistivity against the chemical, thermally stable, and selective contaminants of the water without affecting the performance efficiency [27–29]. Therefore, integrating new filler materials into the polymers to develop polymeric membranes might be one of the promising strategies for separating oil–water.

Recently, two-dimensional (2D) materials such as graphene, graphene oxide (GO), carbon nitride (C₃N₄), boron nitride (BN), and transition metal dichalcogenides (TMDs) (WS₂, MoS₂, MXene), layered double hydroxides (LDHs), and black phosphorus (BP) etc. have drawn a key interest among the researchers in this field due to the unique properties of 2D-NMs such as high surface area, amenability to surface modifications, high mechanical strength, facile synthesis, and cost-effectiveness that provide a chance for scaling up the process, thereby effectively using various applications including oil–water separation [30–39]. 2D-NMs provide fast diffusion (sorption–desorption) rate and enhance water transportation. 2D-NMs improved roughness, enhancing the fouling resistance in the polymeric membrane [40–42]. Furthermore, tunable structure, pore volume, and high surface area of the 2D-NMs might improve the sorption ability of the polymeric composite, thereby effectively adsorbed oil from oil–water pollutants [43,44]. This review article focuses on synthesizing the incorporated polymeric membrane of 2D-NMs and its uses in oil–water separation. First, numerous 2D-NMs and their polymeric composites are briefly
discussed. The next part describes the incorporated polymeric membrane of 2D-NMs used for the oil–water separation and its properties. Next, the recent progress and impact of these membranes are described. The last section concludes with a perspective of the challenges and opportunities in using 2D NM-based membranes for oil–water separation in future technologies.

2. Improvement of Polymeric Membranes Properties by Incorporating 2D-NMs

Numerous approaches have been used to fabricate polymeric membranes to achieve desired properties of the polymeric membranes like permeability, selectivity, hydrophobicity, hydrophilicity, pH, and temperature responsiveness that depend on the physico-chemical characteristics of the materials. The advent of 2D-NMs offers newer paradigms in membrane technology due to their exceptional characteristics. With the help of 2D-NMs, we can easily tune the various physicochemical characteristics of the polymeric membranes, making them suitable for separating oil–water. 2D-NMs have gained significant attention in polymeric membrane fabrication due to their unique properties like excellent mechanical strength (provides rigidity to the membrane), high surface area (for more absorption of the desirable product), and exceptional transport properties.

Usually, incorporating 2D-NMs within the polymers significantly improves the membrane’s permeability. The innovative polymeric membrane is made of stimuli-responsive polymers that have exceptional characteristics, including self-cleaning, antifouling, and switchable wettability on the surface by changing their physical (like light, temperature, electric, and magnetic field) and chemical (like pH, ions, and solvent) [45–47]. We can develop stimuli-responsive polymeric membranes by incorporating surface functional groups and 2D-NMs at the surface of polymeric membranes. Usually, innovative smart polymeric membranes were synthesized by using various processes like blending of polymers, cross-linking of polymers, surface coating, surface grafting, and plasma treatment. Moreover, the fabrication/synthesis of polymeric membranes might be adopted according to their desired property, thereby significantly improving their applicability. It is important to mention that incorporation of 2D-NMs significantly improved hydrophobicity/hydrophilicity, permeability, stability, reusability, and scalability [40,48]. Incorporating 2D-NMs derivatives makes them more stable and rigid in oil–water separation applications. For instance, 2D-NMs like graphene are rich in oxygen-containing functional groups on their surface. These are used as the starting filter for composite formation that can drastically change the van der Walls interaction between the graphene sheets and allow easy dispersion in water [49]. Therefore, their property could be enhanced by making a composite with several hydrophilic polymers, such as polyvinyl alcohol (PVA), which is a biodegradable and hydrophilic polymer used by Liang et al. The composite of GO/PVA gained the enhanced thermal stability and mechanical strength. Additionally, the tensile strength and modulus of the composite also improved from 22 MPa to 42 MPa and 0.45 GPa to 121 GPa, respectively [50]. Similarly, Chang et al. used a composite of graphene, nano clay, and polyaniline for fitter water vapor. Adding 0.5% clay and graphene reduces the water vapor permeability by 63% and 80%, compared with the bare polymer [51]. Kiran et al. added GO in a polyethersulfone (PES) mixed matrix to enhance the water flow through the membrane during its fabrication process. This creates the nanogaps in between the polymeric matrix and GO nanoparticles that commonly occur in nanoparticle-based nanocomposites and enhances the water flow [52]. The studies mentioned above suggested that incorporating 2D-NMs within the polymeric composite significantly improved the hydrophobicity/hydrophilicity, fouling resistance, tensile strength, thermal resistance, and sweetened water flow. Undeniably, we can say that the 2D-NM-based polymeric membranes achieve exceptional characteristics including reusability that might be beneficial for the oil–water separation. Figure 1 shows the schematic illustration of the fabrication of 2D-NM-dispersed polymeric membrane and its application.
3. 2D-NM-Incorporated Polymeric Membranes for Oil–Water Separation

The advent of 2D-NMs and their incorporation within the polymeric membranes offers ample opportunities to fabricate/design polymeric composite with exceptional characteristics, thereby significantly enhancing their applicability to the end applications. Although 2D-NMs have almost similar geometries, different 2D-NMs have different characteristics like electrical, mechanical, optical, and thermal, thereby offering new avenues for developing multifunctional materials by changing the 2D-NMs. Moreover, different synthesis processes also affect the characteristics of the 2D-NMs. Usually, 2D-NMs provides rigidity to the membrane, high surface area (for more absorption of the desirable product), and exceptional transport properties [53–55]. Therefore, incorporating 2D-NMs within the polymeric matrix might benefit the oil–water separation.

3.1. Graphene and Its Derivative

Graphene, a single layer of carbon atoms arranged in a hexagonal lattice, has exceptional mechanical strength, high gas permeability, and molecular sieving capabilities. The nano-porous structure of graphene exhibits excellent water permeability while rejecting ions and other contaminants, making them promising candidates for desalination and water purification applications [56–58]. Graphene-based membranes have attracted significant interest in oil–water separation applications due to their unique properties, such as high mechanical strength, excellent chemical stability, and selective permeability. While graphene itself is a single layer of carbon atoms arranged in a hexagonal lattice, it can be incorporated into various membrane configurations to achieve effective oil–water separation [58–60]. For instance, Prince et al. synthesized polyethersulfone (PES) modified graphene (PES-graphene)-based hollow membrane for oil–water separation. The data suggested that the PES-graphene shows excellent hydrophilicity, permeability, and selectivity, thereby effectively separating oil–water [61].

Similarly, GO derived from graphene by introducing oxygen functional groups offer excellent oil–water separation capabilities. The hydrophilic nature of GO attracts water...
molecules while repelling oil droplets, allowing for effective separation. GO membranes can be fabricated by methods like vacuum filtration or layer-by-layer assembly. They demonstrate high oil rejection efficiency and can be regenerated for repeated use [62–64]. Numerous graphene and their derivative-based polymeric membranes have been used for oil–water separation. Singhal et al. developed a super-hydrophilic and oleophobic membrane by cross-linking GO into tannic acid (TA). The membrane enhanced with surface and mechanical properties was used for vacuum-driven oil–water separation. Figure 2 shows the schematic illustration of the synthesis and its application of the GO-PVDF membrane [65]. In another study, Abdalla et al. incorporated the aseptic acid (AA) functionalized GO into a polysulfone matrix by the phase inversion method. The composite membrane showed enhanced flux, oil rejection, and fouling resistance [66].

![Figure 2](image_url)

**Figure 2.** A schematic illustration of the synthesis and its application of the GO-PVDF membrane (a) synthesis process, and (b) oil–water separation. The image was taken with permission [65] under a common creative license.

Similarly, Alammar et al. used polybenzimidazole polymeric metric to incorporate GO. The prepared membrane was dipped in polydopamine and then used for oil–water separation. GO and polybenzimidazole polymeric composite showed excellent antifouling properties [67]. Zhang et al. fabricated poly (vinylidene fluoride) (PVDF) and GO nanocomposite-based nanofibrous membranes having super-oleophilic and under oil super-hydrophobic properties. The membrane is capable of separating oil and water from emulsion with excellent efficiency [68]. Similarly, George et al. developed GO-dispersed resorcinol-formaldehyde membrane coated over an activated carbon fiber support. The composite was used as a microfiltration membrane and showed excellent efficiency in oil–water separation from emulsion [69]. Instead of polymeric material, Dong et al. used stainless-steel (SS) mesh coated by GO at different pore diameters. The prepared membrane exhibited super-hydrophobic nature underwater and hydrophilicity in the air, completely
against wettability and allowing gravity-driven oil–water separation [70]. Similarly, many other studies have been performed on the development of GO-polymeric nanocomposite membranes [71–76], some of which are described in Table 1. The studies mentioned above and the comparative data of Table 1 suggested that graphene and its derivative efficiently separate oil–water. Moreover, surface functionalization, like the incorporation of functional groups and the encapsulation of polymers, is one of the decisive factors that significantly improved desired properties of the membranes.

Table 1. Graphene-based polymeric membranes for oil–water separation.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Graphene-Based 2D-NMs</th>
<th>Polymeric Material</th>
<th>Enhanced Properties</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Graphene</td>
<td>Polyethersulfone</td>
<td>Hydrophilicity and permeability</td>
<td>[61]</td>
</tr>
<tr>
<td>2.</td>
<td>GO</td>
<td>TA</td>
<td>Surface texture and mechanical strength</td>
<td>[65]</td>
</tr>
<tr>
<td>3.</td>
<td>AA-GO</td>
<td>Polysulfone</td>
<td>Enhanced flux, oil rejection, and fouling resistance</td>
<td>[66]</td>
</tr>
<tr>
<td>4.</td>
<td>GO</td>
<td>Polybenzimidazole</td>
<td>Antifouling</td>
<td>[67]</td>
</tr>
<tr>
<td>5.</td>
<td>GO</td>
<td>PVDF</td>
<td>Super-oleophilic and under oil super-hydrophobic</td>
<td>[68]</td>
</tr>
<tr>
<td>6.</td>
<td>GO</td>
<td>Resorcinol-formaldehyde</td>
<td>Super-oleophilic and under oil super-hydrophobic</td>
<td>[69]</td>
</tr>
<tr>
<td>7.</td>
<td>GO</td>
<td>Polyethylene</td>
<td>Enhanced surface hydrophobicity and oil–water separation efficiency</td>
<td>[71]</td>
</tr>
<tr>
<td>8.</td>
<td>GO</td>
<td>Melamine sponge</td>
<td>Reserve original wettability in harsh environments (acidic, alkaline, saline, and hot waters)</td>
<td>[72]</td>
</tr>
<tr>
<td>9.</td>
<td>GO</td>
<td>Dicarboxylic cellulose</td>
<td>High oil absorption capacity</td>
<td>[73]</td>
</tr>
<tr>
<td>10.</td>
<td>GO</td>
<td>Sodium alginate</td>
<td>Porosity, hydrophilicity, and high pure water flux</td>
<td>[74]</td>
</tr>
<tr>
<td>11.</td>
<td>rGO</td>
<td>Polyurethane sponge</td>
<td>Enhance the hydrophobicity and mechanical properties</td>
<td>[75]</td>
</tr>
<tr>
<td>12.</td>
<td>Fe-GO</td>
<td>PVDF</td>
<td>Super-hydrophilic and underwater super-oleophobic</td>
<td>[76]</td>
</tr>
</tbody>
</table>

3.2. Molybdenum Disulfide (MoS$_2$)

MoS$_2$ is a layered TMDs material with unique electronic and mechanical properties that gained attention for various applications, including oil–water separation. MoS$_2$ membranes can be fabricated by exfoliating bulk MoS$_2$ into few-layered or single-layered sheets. Due to their atomically thin nanopores, these membranes show selective permeability for molecules and ions. MoS$_2$ membranes have the potential ability for gas separation and water purification applications, offering high permeability and selectivity. However, commercial applications require support for their stability. Therefore, immobilization or incorporation of MoS$_2$ into a polymeric membrane to fabricate the MoS$_2$-polymeric membrane enhances the stability of the composite material. Additionally, it provides some unique characteristics to the material that are useful in oil–water separation [77]. MoS$_2$ within the polymeric membrane significantly improved the wettability, stability, and scalability. Numerous MoS$_2$-based polymeric membranes are effectively used for oil–water separation. For instance, Krasian et al. doped MoS$_2$ into polylactic acid, a fibrous biodegradable polymer. The composite material shows enhancement in oil absorption capacity and good reusability. The membrane can separate the floating oil in water by gravity-driven force [78]. Similarly, Dong et al. used MoS$_2$-incorporated PVDF membranes for oil–water separation. The composite membrane shows dual nature i.e., underwater super-oleophobic and underoil super-hydrophobic properties. With the super-wettability
properties, the membrane shows excellent potential for oil–water separation [79]. In another study, Wan et al. synthesized MoS\(_2\)-coated melamine-formaldehyde membrane. The prepared membrane shows oil–water separation effectively. The prepared MoS\(_2\)-based polymeric membranes could also remove other oil/organic contaminants and water-soluble dyes like methylene blue and methyl orange from the water [80]. Razak et al. developed MoS\(_2\) and silica-doped polyethersulfone (PES) membranes. The developed membrane showed high water flux and hydrophobicity. Excellent performance in oil–water separation was determined by the developed membrane, which was due to the change in surface properties of the composite membrane [81]. Similarly, Ansari et al. fabricated MoS\(_2\) doped cellulose acetate membrane. In this composite, MoS\(_2\) showed hydrophobic characteristics; however, cellulose acetate is hydrophilic. The composite membrane shows enhanced water flux leads to efficient oil–water separation with excellent reusability [82]. Instead of polymeric material, Gaourasi et al., coated MoS\(_2\) and quantum dots mixed material over the stainless-steel mesh to fabricate the membrane. The prepared membrane exhibits hydrophobic and oleophobic properties. The membrane shows high flux with good selectivity and also excellent reusability up to 23 times [83]. Remanan et al. synthesized MoS\(_2\)-PVDF membrane for oil–water separation and antibacterial activity. The data suggested that the prepared membrane shows high separation efficiency (~90%) and recovery (~88%) with six times reusability [84]. Sui et al. synthesized the octadecylamine-MoS\(_2\)-PU sponge for the oil–water separation and degradation of dye. The data suggested that the prepared composite materials have excellent hydrophobicity and outstanding oil adsorption ability with high stability [85]. Figure 3. (a) Wettability of PU sponge, (b) Surface texture of pristine-PU sponge, (c) Surface texture of the ODA-Fe\(_3\)O\(_4\)@PU sponge. The image was taken with permission [85].

<table>
<thead>
<tr>
<th>S. No.</th>
<th>2D-NMs Polymeric Material</th>
<th>Enhanced Properties</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>MoS(_2) Polylactic acid</td>
<td>Oil absorption capacity and reusability [78]</td>
<td></td>
</tr>
<tr>
<td>2.</td>
<td>MoS(_2) PVDF</td>
<td>Amphiphilic nature of the membrane [79]</td>
<td></td>
</tr>
<tr>
<td>3.</td>
<td>MoS(_2) Melamine-formaldehyde</td>
<td>Oil separation efficiency and capability to remove oil/organic contaminants also [80]</td>
<td></td>
</tr>
<tr>
<td>4.</td>
<td>MoS(_2)-Silica PES</td>
<td>Enhanced water flux and hydrophobicity [81]</td>
<td></td>
</tr>
<tr>
<td>5.</td>
<td>MoS(_2) Cellulose acetate</td>
<td>Amphiphilic nature of the membrane [82]</td>
<td></td>
</tr>
<tr>
<td>6.</td>
<td>MoS(_2) PVDF</td>
<td>Efficient separation with reusability and antibacterial activity [84]</td>
<td></td>
</tr>
<tr>
<td>7.</td>
<td>Fe(_2)O(_3)-MoS(_2)</td>
<td>Polyurethane enhanced hydrophobicity and oil–water separation efficiency [85]</td>
<td></td>
</tr>
<tr>
<td>8.</td>
<td>Polydimethylsiloxane-MoS(_2)</td>
<td>Polyethylene enhanced surface hydrophobicity and oil–water separation efficiency [86]</td>
<td></td>
</tr>
<tr>
<td>10.</td>
<td>MoS(_2) PES</td>
<td>Enhanced mechanical strength, water uptake, and dye removal [88]</td>
<td></td>
</tr>
<tr>
<td>11.</td>
<td>rGO-MoS(_2) PVDF</td>
<td>Enhanced separation with efficient dye rejection [89]</td>
<td></td>
</tr>
</tbody>
</table>

Figure 3. (a) Wettability of PU sponge, (b) Surface texture of pristine-PU sponge, (c) Surface texture of the ODA-Fe\(_3\)O\(_4\)@PU sponge. The image was taken with permission [85].
Table 2. Different MoS$_2$-based polymeric membranes for oil–water separation.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>2D-NMs</th>
<th>Polymeric Material</th>
<th>Enhanced Properties</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>MoS$_2$</td>
<td>Polylactic acid</td>
<td>Oil absorption capacity and reusability</td>
<td>[78]</td>
</tr>
<tr>
<td>2.</td>
<td>MoS$_2$</td>
<td>PVDF</td>
<td>Amphiphilic nature of the membrane</td>
<td>[79]</td>
</tr>
<tr>
<td>3.</td>
<td>MoS$_2$</td>
<td>Melamine-formaldehyde</td>
<td>Oil separation efficiency and capability to remove oil/organic contaminants also</td>
<td>[80]</td>
</tr>
<tr>
<td>4.</td>
<td>MoS$_2$-Silica</td>
<td>PES</td>
<td>Enhanced water flux and hydrophobicity.</td>
<td>[81]</td>
</tr>
<tr>
<td>5.</td>
<td>MoS$_2$</td>
<td>Cellulose acetate</td>
<td>Amphiphilic nature of the membrane</td>
<td>[82]</td>
</tr>
<tr>
<td>6.</td>
<td>MoS$_2$</td>
<td>PVDF</td>
<td>Efficient separation with reusability and antibacterial activity</td>
<td>[83]</td>
</tr>
<tr>
<td>7.</td>
<td>Fe$_2$O$_3$-MoS$_2$</td>
<td>Polyurethane</td>
<td>Enhanced hydrophobicity and oil absorption capacity</td>
<td>[85]</td>
</tr>
<tr>
<td>10.</td>
<td>MoS$_2$</td>
<td>PES</td>
<td>Enhanced mechanical strength, water uptake, and dye removal</td>
<td>[88]</td>
</tr>
<tr>
<td>11.</td>
<td>rGO-MoS$_2$</td>
<td>PVDF</td>
<td>Efficient separation with efficient dye rejection</td>
<td>[89]</td>
</tr>
</tbody>
</table>

3.3. Boron Nitride (BN)

BN is another 2D-NMs that has gained attention for membrane synthesis. It possesses excellent chemical stability, high thermal conductivity, and good mechanical strength. BN membranes can be created by exfoliating bulk BN into thin layers or by synthesizing them through chemical vapor deposition (CVD) techniques. These membranes exhibit high water permeability while maintaining excellent selectivity, making them suitable for water desalination and filtration [90–95]. Numerous studies have been conducted to date that suggests the BN and its hybrid membrane are effectively used for oil–water separation. For instance, Lei et al. synthesized a BN-guanidine hydrochloride (BN-GH)-based membrane to separate oil–water. The super-hydrophobicity of the porous BN-GH-based membrane makes them suitable for separating oil–water. The prepared porous BN-GH-based membrane has sturdy oxidation resistance, thereby easily recycling [96]. Hao et al. synthesized h-BN incorporated polycaprolactone (BN-PCL) using co-precipitation to separate oil–water. Figure 4 shows the SEM images of BN-PCL composite materials. The SEM images confirm the porous texture and surface roughness that might be beneficial to the separation of oil–water. The data suggest that the prepared BN-PCL-based composite materials effectively separated oil from the water up to 6.1 times their weight [97]. Han et al. synthesized a salinized BN-infused sponge (SBNS) using a simple dipping process. The prepared SBNS-based composite has super-hydrophobicity, flame retardancy, and chemical stability. The intrinsic hydrophobic nature of the SBNS aided advantages for exceptional sorption ability around 22–99 times with different solvents. Moreover, flame retardancy aided advantages in maintaining surface texture in different conditions like acidic and basic. Therefore, the prepared SBNS might be next-generation materials for oil–water separation [98]. Figure 5 shows the schematic representation of the SBNS for oil–water separation. He et al. synthesized a BN-PAN-based fibrous mat using an electrospinning process to separate oil–water. The data suggested that the prepared BN-PAN-based fibrous mat has high porosity and mechanical strength with 99.99% separation efficiency. Moreover, the prepared fibrous mat efficiently used up to 25 consecutive cycles without any significant separation efficiency changes [99].
Hao et al. synthesized the BN tubes-PAN-based fibrous mat using an electrospinning process and atomic layer deposition. Hao et al. synthesized PAN-based fibrous mats using an electrospinning process and deposited BN tubes using an atomic layer deposition process to produce BN-tube-PAN composite materials. The prepared BN-tube-PAN composite materials have high super-hydrophobicity and effectively separate oil up to five consecutive cycles [100].

Liu et al. synthesized Poly dimethyl diallyl ammonium chloride (PDDA)-BN (BN-PDDA)-based composite materials using layer-by-layer assembly. The data suggested that the prepared BN-PDDA have high porosity and excellent sorption ability ~112 times its weight [101]. Li et al. synthesized a free-standing BN sheet using a template-assisted CVD process and attached on to the porous ceramic. The data suggested that the prepared free-standing BN effectively separates oil–water up with 100% efficiency. The high separation efficiency is mainly due to the super-hydrophobicity and lipophilicity nature of the film [102]. Liu et al. synthesized BN-PVDF-based membranes using a phase inversion process for water cleaning, including oil–water separation. Figure 5 shows the SEM, TEM, photographic images, water permeation vs. PVDF wt, and water permeation vs. pressure. The images confirm the uniformity of pores within the BN-PVDF. The water permeation data confirm the porosity of the composite materials; thereby, the prepared BN-PVDF composite material sufficiently separates oil from the water up to 99.99% efficiency. Moreover, the prepared BN-PVDF composite shows a multifunctional ability to clean water by removing dye as well [103]. Another study synthesized the BN-PVDF using a sol-gel process for oil–water separation. The data suggested that the prepared BN-PVDF-based composite material has super-hydrophobicity and super-oleophilic characteristics, significantly improving the oil–water separation efficiency. Moreover, high chemical stability, anti fouling ability, and durability make them exceptional candidates for oil–water separation [104].

The literature and comparative data of Table 3 suggested that the BN-based polymeric membranes improved the hydrophobic nature of membranes, repelling water molecules due to their low surface energy, high stability and scalability, and high oil–water separation efficiency. Furthermore, researchers continue to focus on the improved performance of oil–water separation by controlling the pore size, improved selectivity, and fabrication process. Moreover, compatibility of BN-based polymeric membranes with different types of oils and water sources is underway to ensure their applicability toward real application.

Table 3. BN-based different composite materials for oil–water separation.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Materials</th>
<th>Fabrication Process</th>
<th>Efficiency (%)</th>
<th>Remarks</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>BN-GH</td>
<td>Thermal process using a templating approach</td>
<td>33 times</td>
<td>Super-hydrophobicity makes them selective.</td>
<td>[96]</td>
</tr>
<tr>
<td>2.</td>
<td>BN-PCL</td>
<td>Co-precipitation</td>
<td>6.1 times</td>
<td>BN enhance porosity and internal roughness.</td>
<td>[97]</td>
</tr>
<tr>
<td>3.</td>
<td>SBNS</td>
<td>Dipping</td>
<td>22–99 times</td>
<td>Super-hydrophobicity and flame retardation efficiency make exceptional sorption ability.</td>
<td>[98]</td>
</tr>
<tr>
<td>4.</td>
<td>BN-PAN</td>
<td>Electrospinning</td>
<td>99.99</td>
<td>Porous membranes have high mechanical adsorption ability and reuse up to 25 cycles.</td>
<td>[99]</td>
</tr>
<tr>
<td>5.</td>
<td>BN-tube-PAN</td>
<td>Electrospinning process and atomic layer deposition.</td>
<td>99.99</td>
<td>Porous membranes have high stability and separation ability up to 5 times.</td>
<td>[100]</td>
</tr>
<tr>
<td>6.</td>
<td>BN-PDDA</td>
<td>Layer-by-layer</td>
<td>112 times</td>
<td>High resistance to oxidation with exceptional sorption ability</td>
<td>[101]</td>
</tr>
</tbody>
</table>
Table 3. Cont.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Materials</th>
<th>Fabrication Process</th>
<th>Efficiency (%)</th>
<th>Remarks</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.</td>
<td>Free-standing BN</td>
<td>Template-assisted CVD</td>
<td>100</td>
<td>Super-hydrophobicity and lipophilicity</td>
<td>[102]</td>
</tr>
<tr>
<td>8.</td>
<td>BN-PVDF</td>
<td>Phase Inversion</td>
<td>99.99</td>
<td>Porous membranes have multifunctional water cleaners.</td>
<td>[103]</td>
</tr>
<tr>
<td>9.</td>
<td>BN-PVDF</td>
<td></td>
<td>99.99</td>
<td>Super-hydrophobicity and super-oleophilic characteristics make it suitable for oil–water separation.</td>
<td>[104]</td>
</tr>
</tbody>
</table>

Figure 4. A schematic illustration of the SBNS-based composite materials for oil–water separation (a) synthesis, and (b) oil/solvent separation process. The image was taken with permission [98].

Figure 5. (a) SEM, (b) TEM, (c) SEM cross-section images of PVDF, (d) surface texture of BN-PVDF, (e,f) cross-section images of BN-PVDF, (g) photographic images, (h) water permeation vs. PVDF wt, and (i) water permeation vs. pressure. The image was taken with permission [103].
3.4. Transitional Metal Carbide/Nitride/Carbonitrides (MXene)

MXene is an emerging 2D-NMs and belongs to the family of metals carbide, carbonitride, and nitride. MXene was first used in membrane fabrication by Professor Gogotsi et al. in 2015, which opened the door to MXene in the membrane world [105]. The general chemical formula of MXene is Mn+1XnTx, where M stands for early transition metal element, X is carbon or nitrogen, and T is the active group attached to the surface like O−, OH−, etc. [106]. MXene has inherent characteristics, mainly negatively charged sorption sites, high surface area, chemically stable, tunable inter-layer spacing, and exceptional hydrophilicity that attract researchers for different end applications, including oil–water separation. The material has various chemical and physical properties that are desirable for oil–water separation applications. However, small interlayer space and poor mechanical strength limit its application [107]. To overcome such problems in past studies, MXene was doped into the polymeric matrix. The polymeric composite enhances the interlayer space of MXene and provides mechanical strength to the material. For instance, Zeng et al. synthesized MXene modified by halloysite nanotubes (Hal) and polydopamine (PDA) material over cellulose acetate for membrane fabrication. Due to modifications, the composite material exhibits excellent hydrophobicity with enhanced antifouling and anti-swelling properties. The composite membrane showed excellent potential in oil–water separation [40]. Similarly, Long et al. synthesized a hydroxyl-rich titanium carbide MXene-doped polyvinylidene fluoride membrane. The composite membrane exhibits excellent underwater wettability and permeation flux, effectively separating oil and other contaminants from water with the reusable nature of the membrane [108]. In another study, Huang et al., synthesized nanoribbons of MXene and doped them into a mixed cellulose matrix. The composite membrane showed super hydrophilicity, and the underwater super-oleophobic nature leads to enhanced oil droplet resistance. The fabricated membrane showed excellent performance in separating four different oils including gasoline, diesel, edible oil, and n-hexane form an oil–water mixture [109]. In a study, Imsong and Purkayastha fabricated the Ti3C2T1X MXene nanoflakes-doped polycrylonitrile (PAN) composite membranes. The membrane showed super-hydrophilic/underwater super-oleophobic nature and excellent performance in vacuum separation of various oil–water emulsions. The material has anti-oil-fouling properties, excellent reusability, and flux recovery of up to 80% [110]. Similarly, Zhang et al. developed a Na-Bentonite-modified MXene membrane over PVDF polymeric support. Na-Bentonite made a good adhesion between the layers of MXene, which enhances the water holding capacity and stability of the membrane [42]. Instead of polymeric matrix Saththasivam et al., used MXene as an ink and printed over a paper for fabrication of membrane. The paper-based membrane exhibits high oleophobic and hydrophilic properties with excellent robustness and flexibility and shows 99% separation efficiency in oil–water emulsion [111]. Figure 6 shows the schematic illustration, SEM, TEM, and photographic images of MXene-paper based composite. The photographic images confirm the flexibility of the MXene-paper-based composite, whereas the SEM images conform to the deposition of MXene onto the surface of the paper. The inter-layer spacing of MXene might be beneficial for the oil–water separation (Figure 6e,f). Numerous other studies have been conducted on the applications of MXene in membrane fabrication and oil–water separation application in recent years [112–117], and some of them are briefly described in Table 4. The aforementioned studies suggested that the MXene-based polymeric membranes were efficiently used in oil–water separation. The exceptional inherent characteristics of the MXene mainly negatively charged the sorption sites, exceptional surface area, stability, changing inter-layer spacing, and high hydrophilicity. Moreover, the high oleophobic, hydrophilicity, excellent robustness, and flexibility due to the incorporation of MXene within the polymeric composite. In general, MXene significantly improved the oil–water separation’s mainly tuneable inter-layer spacing, which significantly improved the reactive sites. Moreover, optimizing the inter-layer spacing might be beneficial to oil–water rejection, and thereby to high oil–water separation efficiency.
6. MXene nanosheets PVDF Self-cleaning capability, fouling resistance, and durability [112]

7. MOF-MXene Cellulose acetate Enhanced flux recovery with stability in the acidic, alkaline, and salty medium [113]

8. A-APTES-MXene nanosheets Cellulose acetate Enhanced hydrophobicity and separation performance [114]

9. Ni on MXene PES Enhanced flux, fouling resistance, and oil rejection efficiency [115]

10. Titanium carbide MXene PES Enhance stability and separation efficiency for oil–water emulsion [116]

11. Ti3C2Tx-MXene PVDF Enhanced separation efficiency and permeation flux [117]

Figure 6. (a) A schematic representation of MXene-paper based composite, (b) TEM images of MXene, (c) photographic images of MXene-paper based composite, (d) SEM images of paper, (e,f) Edge of MXene-paper based composite. The image was reproduced with permission [111] under a common creative license.

Table 4. Different MXene-based polymeric composite for oil–water separation.

<table>
<thead>
<tr>
<th>S. No.</th>
<th>2D-NMs</th>
<th>Polymeric Material</th>
<th>Enhanced Properties</th>
<th>Refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Hal-PDA-MXene</td>
<td>Cellulose acetate</td>
<td>Enhanced hydrophobicity, antifouling, and anti-swelling.</td>
<td>[40]</td>
</tr>
<tr>
<td>2.</td>
<td>hydroxyl-MXene</td>
<td>PVDF</td>
<td>Enhanced wettability and permeation flux</td>
<td>[108]</td>
</tr>
<tr>
<td>3.</td>
<td>Nanoribbons of MXene</td>
<td>Mixed cellulose</td>
<td>Enhanced oil droplet resistance and capacity to separate variety of oil form oil–water emulsion</td>
<td>[109]</td>
</tr>
<tr>
<td>4.</td>
<td>Ti3C2TX MXene nanoflakes</td>
<td>PAN</td>
<td>Enhanced antifouling reusability and flux recovery</td>
<td>[110]</td>
</tr>
<tr>
<td>5.</td>
<td>Na-Bentonite modified MXene</td>
<td>PVDF</td>
<td>Enhanced water holding capacity and mechanical stability</td>
<td>[42]</td>
</tr>
<tr>
<td>6.</td>
<td>MXene nanosheets</td>
<td>PVDF</td>
<td>Self-cleaning capability, fouling resistance, and durability</td>
<td>[112]</td>
</tr>
<tr>
<td>7.</td>
<td>MOF-MXene</td>
<td>Cellulose acetate</td>
<td>Enhanced flux recovery with stability in the acidic, alkaline, and salty medium</td>
<td>[113]</td>
</tr>
<tr>
<td>8.</td>
<td>A-APTES-MXene nanosheets</td>
<td>Cellulose acetate</td>
<td>Enhanced hydrophobicity and separation performance</td>
<td>[114]</td>
</tr>
<tr>
<td>9.</td>
<td>Ni on MXene</td>
<td>PES</td>
<td>Enhanced flux, fouling resistance, and oil rejection efficiency</td>
<td>[115]</td>
</tr>
<tr>
<td>10.</td>
<td>Titanium carbide MXene</td>
<td>PES</td>
<td>Enhance stability and separation efficiency for oil–water emulsion</td>
<td>[116]</td>
</tr>
<tr>
<td>11.</td>
<td>Ti3C2Tx-MXene</td>
<td>PVDF</td>
<td>Enhanced separation efficiency and permeation flux</td>
<td>[117]</td>
</tr>
</tbody>
</table>
3.5. Tungsten Disulfide (WS\textsubscript{2})

WS\textsubscript{2} possesses a hexagonal structure and belongs to the spatial group P6\textsubscript{3}/mmc. WS\textsubscript{2} consists of layers in which tungsten atoms form covalent bonds with sulfur atoms. The connections between adjacent layers, specifically the S-W-S joints, are relatively weaker due to the presence of van-der Waals forces. The adjacent layers are stacked in a way that each layer aligns with the accumulation axis of tungsten atoms and rotates after the sulfur atoms, leading to a complete deviation from a flat state\cite{118,119}. WS\textsubscript{2} gives exceptional properties to the composite material that are useful in oil–water separation applications. However, the polymeric matrix provides stability and rigidity to the WS\textsubscript{2} leading to exploring their applicability in commercial applications. Numerous studies have been conducted on the fabrication and application of WS\textsubscript{2}-based polymeric membranes for oil–water separation applications. For instance, Krasian et al. used a combination of two-dimensional materials (MoS\textsubscript{2} and WS\textsubscript{2}) to enhance the performance of PLA fibrous mats in terms of oil adsorption and oil–water separation. The results showed that this hybrid material proved effective in separating surfactant-stabilized oil–water emulsion, with approximately 70% flux recovery achieved across multiple separation cycles\cite{78}. Similarly, Zhou et al. focused on the electroplating process of non-fluorinated super-hydrophobic Ni-WC-WS\textsubscript{2} composite coatings that exhibit excellent resistance to abrasion. These coatings, which are strengthened by WC and lubricated by WS\textsubscript{2} inclusions, demonstrated exceptional durability against abrasion, with a remarkable bearing capacity of at least 10,000 mm of abrasion length\cite{120}. In another study, Ajibade et al. synthesized a polydopamine-modified WS\textsubscript{2} membrane over PLA polymer. The composite material exhibits outstanding antifouling and oil-repellency properties. Also, the composite material is able to separate the oil and dyes from the water in saline environments\cite{121}. Similarly, Zhai et al. fabricated a composite membrane of SiO\textsubscript{2} nanoparticles and WS\textsubscript{2} nanosheets over PDMS. The membrane showed higher oil adsorption than the conventional composite membranes. The prepared membrane is stable in saline, thermal, acidic, insolation environments, and strong wind waves by maintaining its hydrophobicity\cite{122}. Cheng et al. synthesized hybrid WS\textsubscript{2}/GO nanofiltration membranes to achieve precise molecular sieving. One particular membrane, GO15WS\textsubscript{2}, consisting of 15% GO in weight, displayed excellent rejection rates (>90%) for dyes and ions with a hydrated radius greater than 4.9 Å. In this research, a super-hydrophilic composite of nano-porous AC-WS\textsubscript{2}-QDs was prepared using a one-step hydrothermal method and coated onto a mesh for easy collection. The composite was then investigated for its ability to separate oil–water mixtures, considering factors such as pH, solvent type, and varying water-to-solvent ratios. It is important to highlight that the stabilization of AC/WS\textsubscript{2}-QDs nanoparticles on a metal mesh and their application in oil–water separation present a remarkable research innovation\cite{123}. The aforementioned studies suggested that the WS\textsubscript{2} significantly improved the oil–water separation efficiency by improving the hydrophilicity/hydrophobicity.

4. Recyclability of the 2D-NM-Incorporated Polymeric Membranes

The reusability of the 2D-NM-based polymeric composite is a very important aspect in end applications including oil–water separation. The reusability depends on numerous factors like composition of the materials (types of 2D-NMs and polymers), surface properties, and recycling process. 2D-NMs, mainly graphene and GO, exhibit unique characteristics like exceptional mechanical strength, stability, and permeability that makes them ideal candidates for the development of polymeric membranes for oil–water separation. Numerous polymeric composite materials for oil–water separation have shown excellent reusability. For instances, Rong et al. synthesized 3D-composite decorated with carbonized pollen grain for oil–water separation. The prepared hydrophobic 3D-composite effectively separate oil–water with exceptional stability. Moreover, the prepared hydrophobic composite material is efficiently reusable up to five consecutive cycles due to excellent chemical stability\cite{124}. Jiang et al. synthesized superhydrophobic and superhydrophilic sponges for the removal of oil from water. The prepared sponges efficiently separate oil from water surface up to
three consecutive cycles without any significant changes in the separation efficiency [125]. Zhu et al. synthesized super-hydrophobic polyurethane sponges for oil–water separation. The data suggested that the prepared sponges have remarkable reusability up to less than a 400 cycle [126]. Similarly, Alammar et al. synthesized graphene-based membrane for oil–water separation. The data suggested that the prepared graphene-based membrane shows excellent oil separation ability from water surface with reusability up to four consecutive cycles [67]. The aforementioned data suggested that the reusability of the 2D-NM-based polymeric composite depends on their materials composition that directly/indirectly affect their chemical stability. The reusability of the 2D-NM-based polymeric composite have some challenges that might be resolved in future study. Researchers continue to focus on the reusability of the composite materials and their recyclability methods, as most of the process decrease/degrade the materials. It is important to mention here that the effective recycling process should be developed to improve the shelf life of the materials.

5. Strategies to Improve Oil–Water Separation Efficiency

Researchers are actively exploring the optimization of polymeric membranes by incorporating 2D-NMs, surface modification, and their synthesis process for high oil–water separation efficiency [55,127,128]. Usually, various factors affect oil–water separation efficiency. (1) Selection of appropriate membrane, some basic properties such as hydrophobicity/hydrophilicity, porous texture, and exceptional mechanical strength, are required for oil–water separation. We must choose polymers with hydrophobicity/hydrophilicity and chemical resistance. (2) Modification of polymeric membranes, surface modification is a decisive factor affecting the oil–water separation efficiency. Numerous processes, such as surface coating, grafting, and plasma treatment, have produced hydrophobic layers that prevent oil droplets from wetting. (3) Surface texture can be optimized by the efficacy of the polymeric membrane by tuning the pore size and surface morphology of the membrane using various processes like the electrospinning process, self-assembly, and phase inversion. With the help of controlling pore size, we can easily improve the oil rejection ability of the membranes. (4) Stimuli-responsive polymeric composites provide a newer avenue by tuning/modification of surface properties like by changing the pH and temperature. The wetting behavior of the polymeric membranes is directly related to the liquid/solid interfaces. The hydrophobic/hydrophilic and oleophilic/oleophobic characteristics of polymeric composite might be beneficial for oil separation from water surfaces. The polymeric composite having hydrophobic or oleophilic allows oil droplets to pass through, whereas hydrophilic or oleophobic allows water molecules and blocks oil droplets. Therefore, designing the polymeric membranes with super-wettability significantly improved the separation efficiency. Interestingly, such types of materials have reversible behavior that easily switches the super-hydrophilicity and super-hydrophobicity, thereby making them potential candidates for oil–water separation. (5) Development of hybrid membranes: hybrid membranes are an important aspect of synthesizing desired polymeric membranes, as according to desired properties we can choose materials like polymers and 2D-NMs that significantly enhance the oil–water separation efficiency. It is important to mention that the specific strategies required to enhance the polymeric membranes for oil–water separation efficiency depend on the desired separation efficacy, mixture of oil–water, and operating conditions [129–131]. Moreover, selecting 2D-NMs is also one of the decisive factors for separating oil–water. Additionally, stability, cost-effectiveness, and scalability are considered for developing 2D-NM-based polymeric membranes.

6. Conclusions

The integration of 2D-NMs within the other materials, including polymers, produces a hybrid polymeric composite that significantly enhances the oil–water separation efficiency. With the help of 2D-NMs, mechanical strength, selectivity, permeability, hydrophilicity/hydrophobicity, and pore size are controlled. For oil–water separation, a hybrid membrane may comprise layers of hydrophilic materials for water permeation and
hydrophobic materials for oil rejection. Combining 2D-NMs with polymers creates a multifunctional membrane with improved separation performance. Ongoing research focuses on optimizing membrane structures, surface modifications, and fabrication techniques to enhance their oil–water separation performance further and address practical implementation challenges. It is important to mention that developing sustainable and cost-effective membranes/adsorbents remains a challenge. Most of the prepared membranes/adsorbents with a high capacity for the separation of oil–water have some drawbacks, such as being environmentally unfriendly, having a complex synthesis route, and being costly. Furthermore, these materials show exceptional oil adsorption in a laboratory. However, oil pollution has higher density and viscosity than oil used in laboratory experiments. Therefore, more research is required that needs to be conducted on crude oil, which has high density and viscosity. It is important to mention that the 2D-NMs alone have some issues for the development of membranes, mainly stability. Incorporating 2D-NMs within the polymers gives better results in end applications. Further research is ongoing to optimize their performance, scalability, stability, reusability, and their waste management. Scientists are exploring strategies to enhance their selectivity, improve membrane fabrication methods, and investigate their compatibility with different oils and water sources. Therefore, strategically combining 2D-NMs within the polymers can offer improved performance and address the challenges associated with oil–water separation.

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