

# Two-Dimensional Nanomaterial (2D-NMs)-Based Polymeric Composite for Oil–Water Separation

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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. 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.

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

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## 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  $\mu\text{m}$ ), dispersed oil (15–145  $\mu\text{m}$ ), and free oil (>145  $\mu\text{m}$ ) <sup>[1][2][3][4]</sup>. Moreover, oil leakage during production and transportation probably devastates the water bodies and ecosystems. Oil pollutants also increase natural resource loss <sup>[5][6][7][8]</sup>. 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 <sup>[9][10][11]</sup>. However, these are inefficient for treating wastewater contaminated with dissolved elements and small suspended particles. Chemical processes such as ozonation <sup>[12]</sup>, electrochemical <sup>[13]</sup>, photocatalysis <sup>[14]</sup>, and demulsification <sup>[15]</sup> 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 <sup>[16]</sup>. On the other hand, biochemical methods have been developed in which some enzymes, such as lipase, break down the large molecule into small molecules <sup>[17]</sup>. 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 issue 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 <sup>[18][19]</sup>. 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 <sup>[20]</sup>. 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 <sup>[21][22]</sup>. Typically, altering the polymer's composition and morphology afterwards makes it possible to change its properties, such as hydrophilicity and hydrophobicity. The hydrophilicity 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

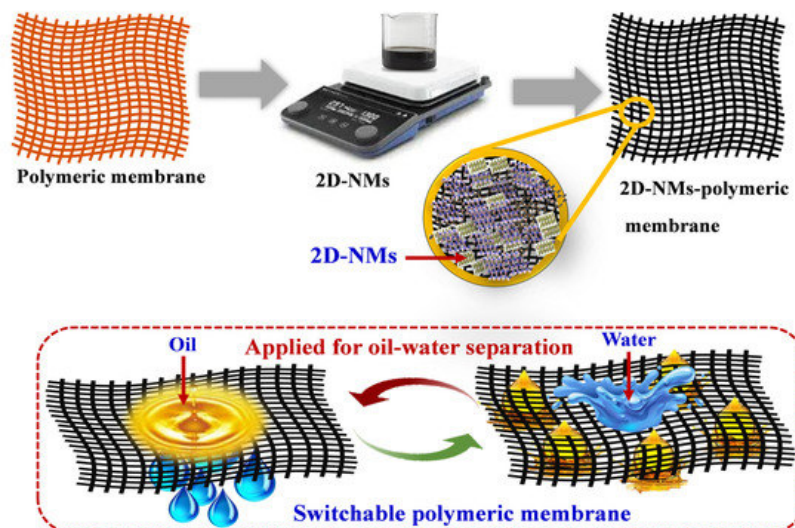
determine how planned polymeric composites' hydrophilic/hydrophobic surface roughness is amplified, creating a membrane with an improved surface [23][24][25][26]. The hydrophilic/hydrophobic surface roughness of planned polymeric composites is enhanced depending on the shape and wrapping of another polymer employed to create the membrane, resulting in an improved surface texture of the membranes. Hydrophilicity and hydrophobicity are important factors that affects the oil–water separation efficiency.

Recently, two-dimensional (2D) materials such as graphene, graphene oxide (GO), carbon nitride ( $C_3N_4$ ), boron nitride (BN), and transition metal dichalcogenides (TMDs) ( $WS_2$ ,  $MoS_2$ , 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 [27][28][29][30][31][32][33][34][35][36]. 2D-NMs provide fast diffusion (sorption–desorption) rate and enhance water transportation. 2D-NMs improved roughness, enhancing the fouling resistance in the polymeric membrane [37][38][39].

## 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 physicochemical characteristics of the materials. The advent of 2D-NMs offers newer paradigms in membrane technology due to their exceptional characteristics.

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) [40][41][42]. Scholars 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 [37][43]. 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 Waals interaction between the graphene sheets and allow easy dispersion in water [44]. 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 [45]. **Figure 1** shows the schematic illustration of the fabrication of 2D-NM-dispersed polymeric membrane and its application.



**Figure 1.** A schematic illustration of the fabrication of 2D-NM-dispersed polymeric membrane and its application.

## 3. 2D-NM-Incorporated Polymeric Membranes for 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 [46][47][48]. 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 [48][49][50]. 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 [51].

### 3.2. Molybdenum Disulfide (MoS<sub>2</sub>)

MoS<sub>2</sub> is a layered TMDs material with unique electronic and mechanical properties that gained attention for various applications, including oil–water separation. MoS<sub>2</sub> membranes can be fabricated by exfoliating bulk MoS<sub>2</sub> into few-layered or single-layered sheets. Due to their atomically thin nanopores, these membranes show selective permeability for molecules and ions. MoS<sub>2</sub> 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<sub>2</sub> into a polymeric membrane to fabricate the MoS<sub>2</sub>-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 [52]. MoS<sub>2</sub> within the polymeric membrane significantly improved the wettability, stability, and scalability. Numerous MoS<sub>2</sub>-based polymeric membranes are effectively used for oil–water separation. For instance, Krasian et al. doped MoS<sub>2</sub> 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 [53].

### 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 [54][55][56][57][58][59]. 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 [60].

### 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 [61]. The general chemical formula of MXene is  $M_n+1X_nT_x$ , 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. [62]. 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 [63]. 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 [37].

### 3.5. Tungsten Disulfide (WS<sub>2</sub>)

WS<sub>2</sub> possesses a hexagonal structure and belongs to the spatial group P6<sub>3</sub>/mmc. WS<sub>2</sub> 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 [64][65]. WS<sub>2</sub> 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<sub>2</sub> leading to exploring their applicability in commercial applications. Numerous studies have been conducted on the fabrication and application of WS<sub>2</sub>-based polymeric membranes for oil–water separation applications. For instance, Krasian et al. used a combination of two-dimensional materials (MoS<sub>2</sub> and WS<sub>2</sub>) 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 [53].

## 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 [66].

## 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 [67][68][69]. 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. Scholars 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, it 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 scholars 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 [70][71][72]. Moreover, selecting 2D-NMs is also one of the decisive factors for separating oil–water.

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