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REVIEW ARTICLE

A Review on Separation Techniques of Graphene Oxide (GO)/ Base on Hybrid Polymer Membranes for Eradication of Dyes and Oil Compounds: Recent Progress in Graphene Oxide (GO)/ Base on Polymer Membranes-Related Nanotechnologies

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Abstract

Nowadays, obtaining drink water for saving health of humans is very important. There have been various investigations about the cost effective and environmental friendly methods of water treatment. The purpose of the current study is reviewing the application of hybrid polymeric membranes Graphene Oxide (GO) as new techniques for separation of pollutants (Figures 1 and Figure 2) [1,2]. One of the methods for separation of containment is filtration. The filtration of pollutant is a useful method because of its limited need to energy and no production of chemical material, so less pollution. The hybrid polymeric membranes have many advantages such as permeation high flux, low antifouling and reducing odor compared to traditional kind. In addition, it can be used for separation of aromatic hydrocarbons which have four or more rings in waste water. Aromatic hydrocarbons are included in the priority list of pollutants of US EPA and European Union, especially those with four or more rings.

Keywords

Recent patents, Graphene oxide (GO), Hybrid polymer membranes, Separation techniques, Eradication, Dyes and oil compounds, Nanotechnologies

Abbreviations

GO: Graphene Oxide; SSA: Specific Surface Area; TiO_2: Titanium Dioxide; MF: Microfiltration; UF: Ultrafiltration; NF: Nanofiltration; RO: Reverse Osmosis; CA: Cellulose Acetate; PC: Polycarbonate; PSF: Polysulfone Fluoride; PES: Poly Ether Sulfone; PVDF: Polyvinylidene Fluoride; MMM: Mixed Matrix Membrane; MB: Methylene Blue; MV: Methyl Violet; MO: Methyl Orange; CS: Chitosan; HM: Hybrid Membrane; AOPs: Advanced Oxidation Processes; rGO: Reduced Graphene Oxide; MWCNTs: Multi-Walled Carbon Nanotubes; TMP: Transmembrane Pressure; TDS: Total Dissolved Solid; RF: Rejection Flux

Introduction

By developing heavy industries during recent decades, availability of healthy drinking water becomes a global concern [3]. Pollutants such as gas petrochemical, pharmaceutical and organic substances, industrial dyes, microorganisms and heavy metals (including Cobalt, Cadmium, Mercury, Chromium and Lead) are typically observed in the water [4-9]. Because of high retention, these compounds possess harm effect like carcinogenetic in animal, humans' bodies and environment [10]. It is difficult to treat water by conventional physical methods such as flotation, aeration, coagulation and gravity settling. These methods have drawbacks such as high cost, using toxic compounds, large space, high energy consumption and producing secondly compounds. In recent decades, applications of clean water were se-

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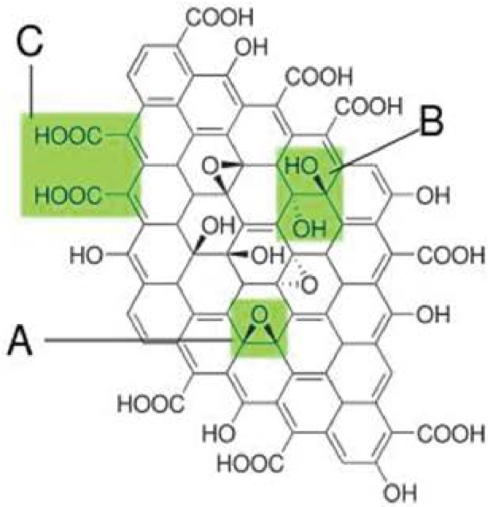

Figure 1: Structure proposed in 1998 with functional groups. A): Epoxy bridges; B): Hydroxyl groups; C): Pairwise carboxyl groups [1].

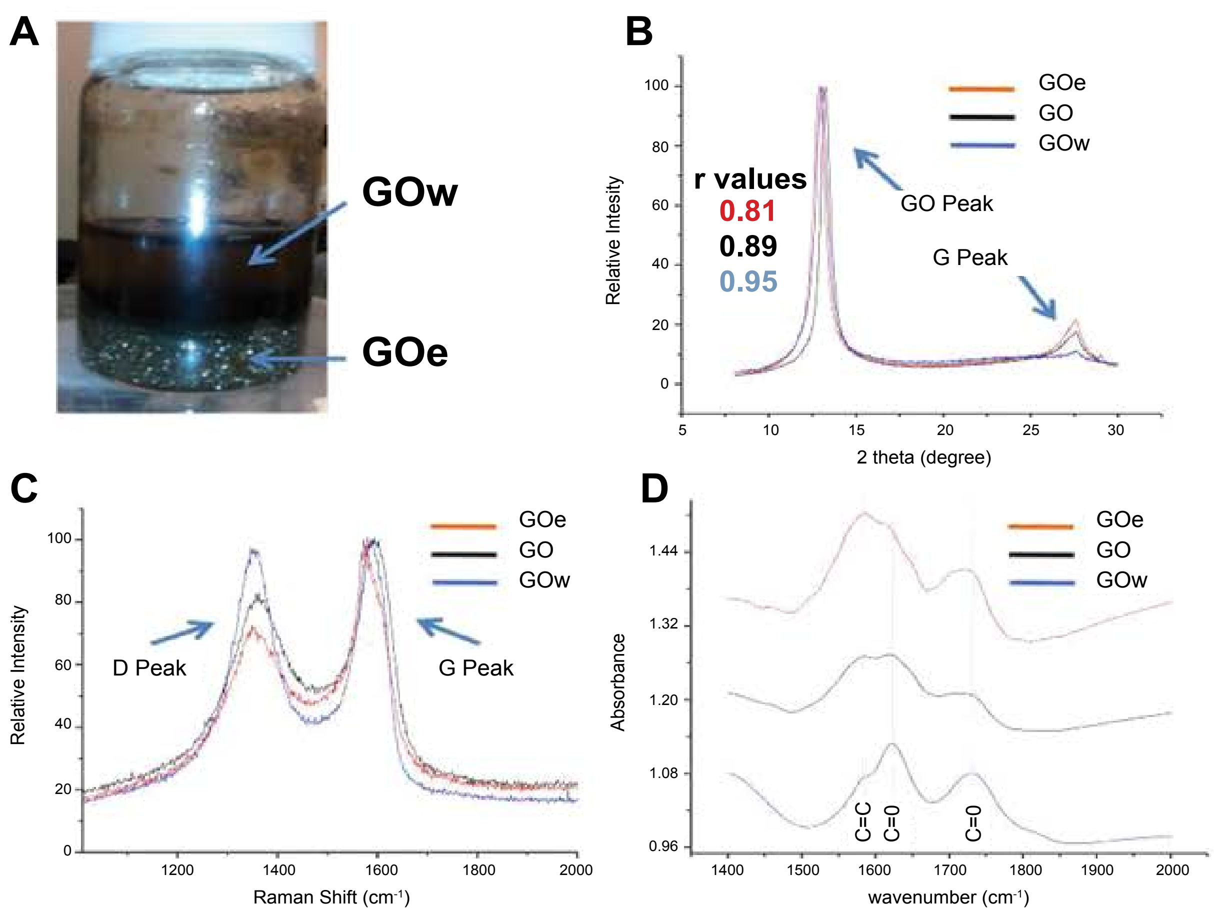

Figure 2: A) Image of fractionated GO. The upper phase contains GO suspended in water (GOw) and the lower region contains GO at the interface of a chloroform-in-water emulsion (GOe); B) Overlaid XRD spectra of the original GO sample (black), the GOw from the water phase (blue), and GOe from the emulsion phase (red); C) Overlaid Raman spectra of GO (black), GOw (blue), and GOe (red) showing the G and D peaks; D) Overlaid FTIR spectra of GO (black), GOw (blue), and GOe (red) highlighting changes in relative intensities of peaks at 1580 cm^{-1} (sp^2-hybridized C = C), 1620 cm^{-1} (ketone C = O), and 1730 cm^{-1} (carboxyl C = O) [2].

verely increased [11-13]. With high efficiency and low energy consumption, filtration is one of the most appropriate technologies for decreasing pollution [14-18]. The formation of a good membrane is an important and practical step toward increasing the efficiency of water treatment; this film can be made from materials such as polymer, fiber, ceramic and Carbon nanotubes [19-22].

Background

Graphene is a single layer or few layers of graphite,

Table 1: Band gap energy for some common semiconductor materials [46].

Semiconductors	Band Gap Energy (eV)	Semiconductors	Band Gap Energy (eV)
Diamond	5.4	WO_3	2.76
SdC	2.42	Si	1.17
ZnS	3.6	Ge	0.744
ZnO	3.436	Fe_2O_3	2.3
TiO_2	3.03	PbS	0.286
CdS	2.582	PbSe	0.165
CnS_2	3.54	ZrO_2	3.87
CdSe	1.7	Cu_2O	2.172

a two-dimensional (2D) one atom thick nanomaterial consisting of sp^2-hybridized Carbon packed in honey combed crystal lattice [23]. Scientists are heavily interested in Graphene due to its unique properties; including high Specific Surface Area (SSA) of 2600 $m^2 g^{-1}$ [24] or 2630 $m^2 g^{-1}$ [25], excellent thermal conductivity of 5000 W m^{-1} K^{-1} [26], high-speed electron mobility of 200000 cm^2 V^{-1} s^{-1} at 250 °C [27] and strong adsorption capacity on poly aromatic hydrocarbon [28].

Many efficient adsorbents [29] and photocatalysts [30] are developed for the removal and photocatalytic degradation of pollutants. Graphene mono layers have an excellent electrical conductivity 103 WmK^{-1} [3]. Many sensitive biosensors, glucose biosensor [31] and fluorescent [32] were used as detector. Finally, chemical properties and chemical modification are also of particular interest for photocatalysis or separation of pollutant such as heavy metal and azo dyes. GO consists of Graphene sheets having functional groups R-O-H hydroxyl, epoxy, COOH and C = O. It causes Graphene has negative charge which can affect cationic compounds [33].

One of the interesting properties of GO is that it is a photocatalyser material in photoluminescence. GO is a p-type semiconductor. The optical band gap of GO is around 3.06 eV. Graphene is a suitable material as an alternative for photoanode due to its specific surface area and good capacity to transport electron.

Titanium dioxide (TiO_2) is the most important semiconductor among all of the semiconductors which are widely used with Graphene in many studies. Titanium dioxide is the oxide of Titanium, with a molecular weight of 79.87 g/mol and chemical formula of TiO_2. One of the important properties of TiO_2 is the photocatalytic activity. Also, the photocatalytic activity of TiO_2 has been applied in a wide range of metal oxides and their sulfides [34,35] including ZnO [36], WO_3 [37], WS_2 [38], Fe_2O_3 [39], V_2O_5 [40], CeO_2 [41], CdS [42] and ZnS [43]. ZnS is an II-VI semiconductor with a wide band gap (3.75 eV). ZnS is used in transistors, LEDs, photocatalysis and solar cells [44,45]. The band gap energy values for some common semiconductor materials are presented in Table 1 [46].

Graphene is one of the Carbon allotropes which possess photocatalic performance. One of the material photocatalytic performances of Graphene is related to the Carbon material. It has low toxicity, excellent electrical conductivity and thermal conductivity as well as other special properties such as electron-acceptor and electron-transport Dye Sensitized Solar Cell (DSSC), quantum-dot sensitized solar cell eradication of organic, ionic and biological pollutant water splitting [47-50].

Preparation of GO

GO was prepared from natural graphite powder according to the Hummers method [48] and many procedures have been described with some modification, previously [51,52]. Briefly, 3 g of graphite was added into 70 mL of sulfuric acid (98%) and the resulted mixture was stringed at room temperature for 24 h. Then, 300 mg of Nitrate Sodium was added into the mixture and stirred for 1 h. The dispersion was cooled by ice bath. Next, 15 g of $KMnO_4$ was slowly added to the mixture during 15 min. Afterwards, the temperature of mixture was kept to 40-50 °C while stirring for another 1 h. The reaction was followed by adding 138 mL of DI water into the dispersion during 30 min. Finally, 30 mL of H_2O_2 was added into the dispersion. The pH of solution was remained at 7. Next, the synthesized GO was dried at 60 °C for 24 h.

Preparation of modified GO

Modifications in the conditions of the Hummer's method can be improved to be efficient in Graphene-based p-semiconductors production (it does not use Na_3NO_3, increasing added amount of $KMnO_4$ and volume ratio of H_2SO_4:H_3PO_4 in a 9:1 mixture), various advantages can be achieved such as fewer defects, higher efficiency, equivalent conductivity and no production of toxic gases. The mixture was stirred at 50 °C for 12 h. Then, the mixture was cooled at 25 °C, under vigorous stirring in an ice bath with adding 3 mL H_2O_2. Therefore, the mixture was washed with DI water several times. This is considered as the most excellent method to prepare GO in large quantities [53].

Characterization of GO by FT-IR and raman spectroscopy

Figure 3 depicts the FT-IR spectrum of GO. Fourier Transform Infrared (FT-IR) spectroscopy was used to detect chemical composition assignment (cm^{-1}): C = O (carbonyl/carboxy) groups at 1736.56; C = C (aromatics) at 1633.56; C-O (carboxy) at 1419.16; (epoxy) C-O group at 1267.74; C-O (alkoxy) at 1037.07 [54,55].

Furthermore, Figure 4 illustrates the Raman spectrum of GO includes D and G peaks where the D peak at ~1350 (cm^{-1}) is the result of defects in the Graphene sheets and the G peak at ~1600 (cm^{-1}) is the result of bond stretching of sp^2 hybridized Carbons, respectively [56,57].

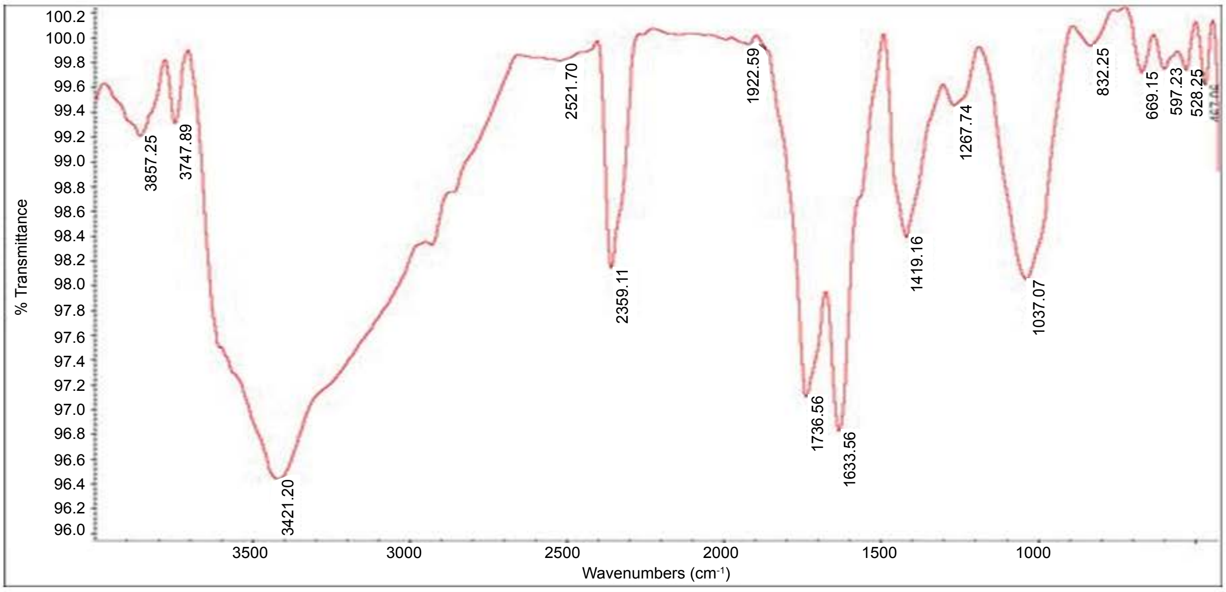

Figure 3: FT-IR spectrum of GO.

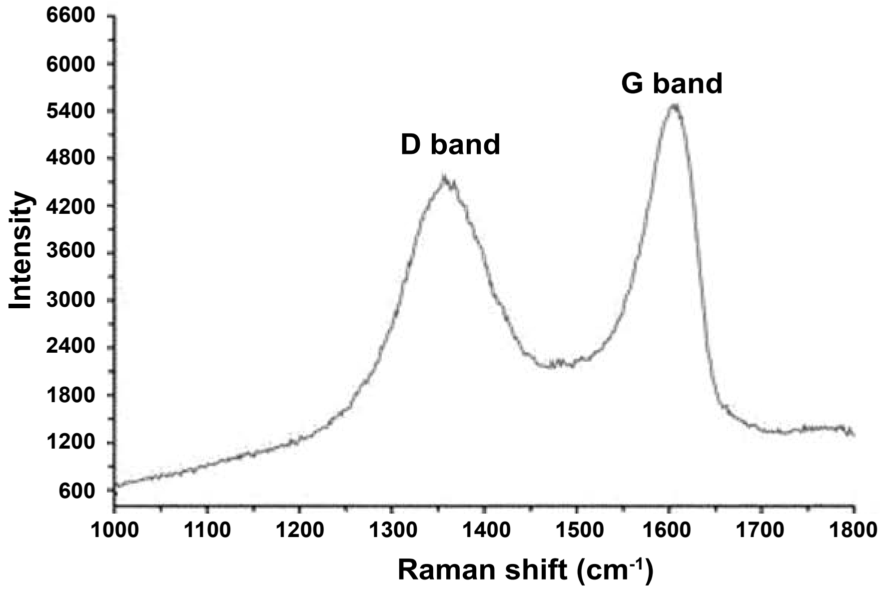

Figure 4: Raman spectrum of GO.

Separation Techniques

Filtration

As shown in Figure 5, membrane separation is based on three principles: Adsorption, sieving and electrostatics [58]. The type of membrane has relation with its pore size: Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF) and Reverse Osmosis (RO) as shown in Table 2. Among the membrane processes, the advantage of Ultrafiltration (UF) with polymeric membrane is that it is able to omit pollutants such as organic pollutants, heavy metal, suspended solid and various dyes.

It should be noted that when the pore size of membrane is more than pollutant molecules' size, deep filtration occurs. In this regard, when the pore size of membrane is smaller than pollutant molecules' size, sieving or adsorption or surface filtration occurs. Furthermore, electrostatic reaction takes place for the reason of membrane and particles' charges. In this regard, when pollution membrane and particle have same charge, adsorption does not occur. But when pollution membrane and particle have opposite charge, adsorption occurs.

Advanced technology in membrane

In recent decades, ceramic and polymeric membranes have been increasingly used due to their strong mechanical characteristics, chemical stability, high efficiency for removing pollutants, high photocatalytic power and odor decline. They are produced in various types such as hollow fiber spiral and tubular structure.

Polymeric membrane: Different polymeric membranes such as Anionic Perfluorinated Polymer (Nafion), Cellulose Acetate (CA), Polycarbonate (PC), Polysulfone Fluoride (PSF), Poly Ether Sulfone (PES) and Polyvi-

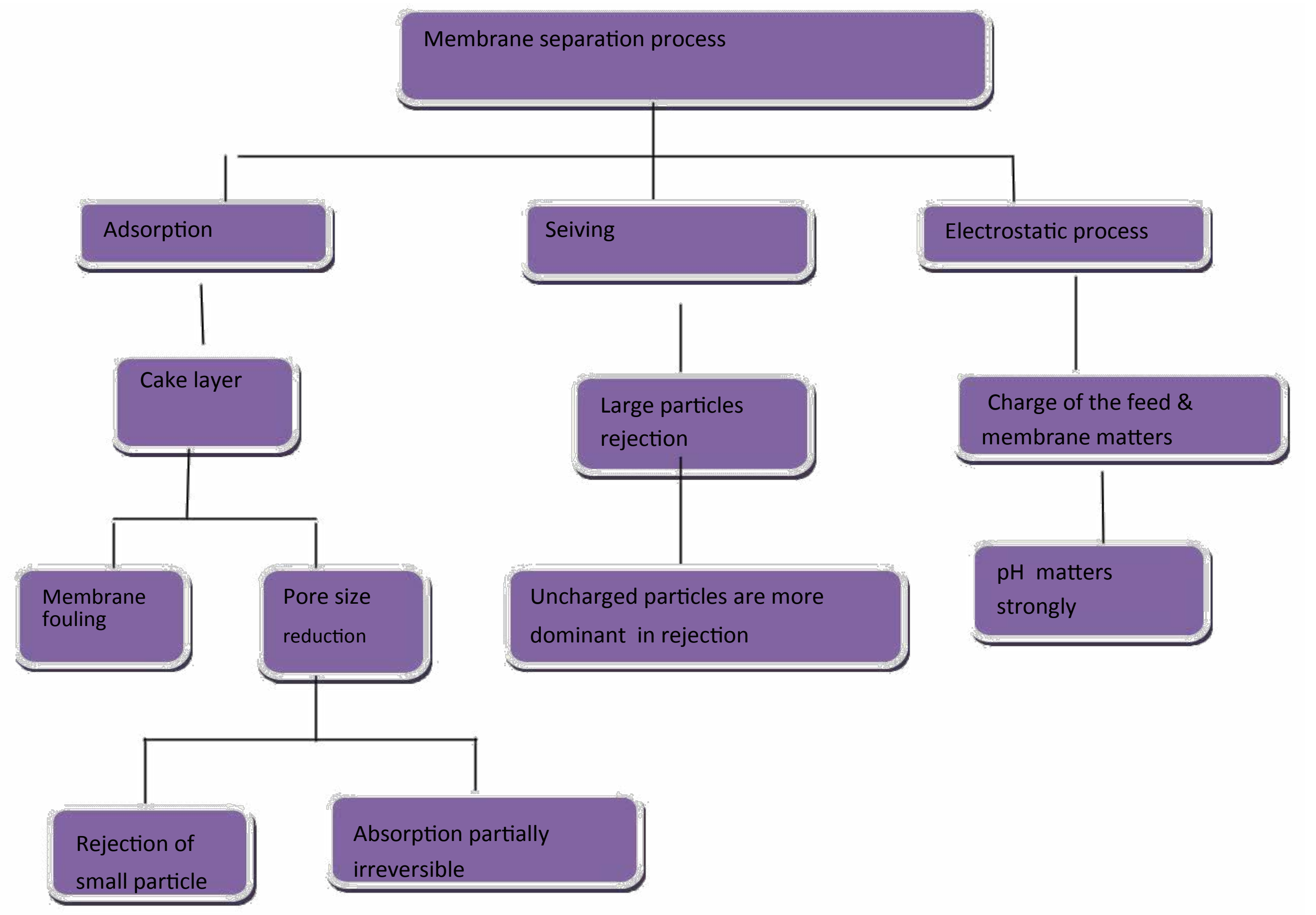

Figure 5: Schematic representing the basic principles involved in membrane separation [58].

Table 2: Membrane separation process [58].

Pore Type	Membrane Type	Species	Dimension (nm)
Macropores	Microfiltration	Yeasts & fungi	1000-10000
> 50	50-500	Bacteria	300-10000
		Oil emulsion	100-10000
Mesopores	Ultrafiltration	Colloidal Solid	100-1000
2-50	2-50	Viruses	30-300
		Protein/Polysaccharide	30-300
		Humic/Nucleic Acids	3-10
			< 3
Micropores	Nanofiltration ≤ 2	Common antibiotics	0.6-1.2
0.2-2	Reverse Osmosis	Organic antibiotics	0.3-0.8
	0.3-0.6	Organic ions	0.2-0.4
		Water	0.2

nylidene Fluoride (PVDF) were used in separation method. The polymer membranes have specific characteristics such as thermal stability, low energy requirements, chemical resistant, high flux and low fouling compared to usual kinds [59]. Although polymers are hydrophobic, many methods have been developed to enhance the properties of polymer membrane in the past decades. Among the methods, the strategy of doping inorganic oxide particles for casting the solution to prepare organic-inorganic composite membranes or Mixed Matrix Membrane (MMM) is promising, owing to its simple operating process. The main reason for the reduction of membrane fouling and increasing water permeability is increasing of membranes hydrophilicity which causes to decline the interpolation of oil and dyes compounds [59]. The asymmetric polymeric membrane is ideal for obtaining high permeability, good hydrophilicity and excellent chemical resistance to the feed solution.

Separation of azo dyes

The discharge of wastewater from textile industries into the environment is regarded as a main source of pollution and cause of eutrophication in rivers and lakes. There have been many studies for aiming to find a better way for separating azo dyes from wastewater. Fei Liu and his group reported a three-dimensional (3D) GO sponge from a GO suspension through a simple centrifugal vacuum evaporation method. They removed both Methylene Blue (MB) and Methyl Violet (MV) by GO sponge with good efficiency. The adsorption process completed with high efficiency of 99.1% for Methylene Blue (MB) and 98.8% for Methyl Violet (MV) in 2 min. The effective parameters investigated on adsorption three-dimensional (3D) GO were temperature, pH and stirring speed. The adsorption mechanism follows the second-order kinetic equation [60].

Lulu Fan, et al. also reported synthesized GO/Magnetic Chitosan (CS) for absorption of Methylene Blue (MB) in aqueous solution. The amount of adsorbent can be reduced using specific surface area of Chitosan (CS). The kinetic study fitted the pseudo second-order model. Although the equilibrium data well-modeled according to the Langmuir and Freundlich isotherm models, the best fitted results was observed in Langmuir model [61].

Tonghao Liua, et al. investigated adsorption capacity of Graphene at Methylene Blue (MB). The kinetic study illustrated that the adsorption of Methylene Blue (MB) onto Graphene fit the pseudo second-order model. The effective parameters investigated on adsorption GO were temperature, pH, contact time and dosage in adsorption [62].

Lulu Fan and his group synthesized the magnetic β-cyclodextrin-chitosan/GO as a nanoadsorbent. β-cyclodextrin-chitosan/GO has good adsorption properties for the reason of magnetic properties of Fe_3O_4 as a functional group in Chitosan (CS) and also hydrophobicity of cyclodextrin. This magnetic nanoadsorbent has low cost and easy operation and can be extracted from aqueous solution. It will be used as other novel biosorbent suggested in future [63].

Hem Raj Pant and coworkers reported the synthesizing of ZnO micro flower nanoparticles. It assembled on GO which have excellent characteristic for removing the pollutants, especially the Methylene Blue (MB). ZnO nanoparticles can be recovered and extracted from reaction system which is useful for environment [64].

Jiaqian Qin, et al. also reported the synthesizing of ZnO microspheres-r GO nanocomposite. It had been used for photocatalytic degradation of Methylene Blue (MB) under UV irradiation. The better adsorption is in higher Brunauer-Emmett-Teller (N_2-BET) Spacific Surface Area (SSA) (16 m^2/g for ZnO, 20 m^2/g for ZRGO80 and 26 m^2/g for ZRGO120). The best photocatalytic activity among of synthesized pure ZnO, P25 TiO_2 in this study was ZnO-rGO (4.06 wt. % rGO) composite [65].

It is essential to determine the factors affecting the reaction rate and the mechanisms that control the adsorption, including adsorption at the surface, chemical reaction and diffusion mechanisms through kinetic evaluation. Pseudo-first-order and pseudo-second-order kinetic models are widely used to characterize adsorption. A pseudo-first-order kinetic model indicates that diffusion takes place from inside a layer and is based on solid capacity. A pseudo-second-order kinetic model indicates that chemical adsorption reduces the rate and controls the adsorption, which is based on solid phase adsorption.

The Langmuir, Freundlich and Tamkin models were used to determine the mechanism, superficial characteristics and adsorption tendency of experimental data at equilibrium. In the Langmuir model, adsorption takes place at a series of sites inside the adsorbent on the homogenous surface, every adsorption site have same adsorption power. The equation form is according to the following:

$$\frac{C_e}{q_e} = \frac{1}{K_L Q_0} + \frac{C_e}{Q_0}$$

where q_e denotes the amount of pollutant adsorption onto the adsorbate mg/g, Ce denotes the final equilibrium concentration of the solution (mg/l), KL denotes the equilibrium constant Langmuir and Q_0 denotes the maximum adsorption capacity at monolayer coverage (mg/l).

In the Freundlich (1906) isotherm, adsorption occurs in response to the heterogeneous surface having a non-uniform distribution of adsorption heat. The Freundlich model is an empirical model. The equation form is according to the below:

$$log q_e = log K_F + \frac{1}{n} log C_e$$

where K_F is the adsorption capacity at unit concentration and 1/n is the intensity of adsorption and indicates the type of isotherm;

$1/n = 0$ is irreversible, $0 < 1/n < 1$ is desirable and $1/n > 0$ is undesirable. The Tamkin isotherm assumes that the adsorption heat of all molecules in a layer declines linearly due to the interaction between the adsorbent and the adsorbate. This model describes the interaction between the adsorbent and adsorbate.

All of previous experiments have been reported that the adsorption of Methylene Blue (MB) on the surface of Graphene were endothermic and spontaneous. The adsorption increases as the contact time increases. It is clear that the percentage removal of dyes increases by increasing the dosage of Graphene. All of above results show that GO plays important roles for π-π interaction between aromatic rings like cationic dyes and GO or negative.

It should be noted that GO are used as heavy metals removal in nanocomposites, nanoadsorbates, etc. For example, Fang Fang and coworkers have functionalized GO as a heavy metals removal using aromatic diazonium salts. A new designed GO-NH_2 has excellent adsorption for removal of Co (II) ions. Ca^{+2} and Mg^{+2} are the usual ions with Co (II) in nature; they are increased the adsorption of Cobalt [66].

Chaohui Ruan and his groups also investigated on novel porphyrin sensitized TiO_2/Graphene. The effects of photocatalytic properties of TiO_2/Graphene and porphyrin synthesis TiO_2/Graphene under visible light were studied. The results showed the improved photocatalytic degradation Methylene Blue (MB) under visible light with the novel composite [67].

According to the Chao Xu, et al. synthesis of GO-TiO_2 composite as filtration membrane to remove of (Methyl Orange (MO) and Rhodamine B) from the water was successful. These attached TiO_2 can shore up GO sheets and enlarge the interlayer distance, which leads to the formation of special pores and channels in the films, making them good treatment of water [68].

Peng Gaoa and coworkers synthesized GO-TiO_2 microsphere as membrane filtration. They used a commercial Cellulose Acetate (CA) membrane as a kind of polymer GO-TiO_2 nanohybrid was uniformly assembled on the surface of Cellulose Acetate (CA) membrane that has extraordinary removal pollutant of water [69].

Yong Gao, et al. found effective of GO-TiO_2 assembled on polysulfone base membrane which show significantly improved photodegration of Methylene Blue (MB) under UV irradiation. The synthesizing method of Hybrid Membrane (HM) in their study was LbL (Layer-by-Layer). The LbL (Layer-by-Layer) method has the ability to form a Hybrid Membrane (HM) [70].

The LbL (Layer-by-Layer) method has the ability to form a Mixed Matrix Membrane (MMM). This experiment used a Gold covered sensor to determine the mass of TiO_2 and GO during the LbL (Layer-by-Layer) procedure. The sensor was coated using a base polymer membrane. The sensor was put in a desiccator for drying. After that, it was put in DI water. A fixed concentration on the surface of the sensor was at 0 ng/cm^2 after 15 min and the DI water was changed with TiO_2 solution. TiO_2 nanoparticles were adsorbed on the base polymer after 2 hr, the mass of the nanoparticle slowed down and became stabilized. After that, TiO_2 solution was replaced with DI water for the removal of nanoparticles. Next, the TiO_2 coated sensor was soaked in the GO solution. In particular, the advantage of LbL (Layer-by-Layer) properties of the membrane can be readily controlled through polymer compositions, concentrations, numbers of applied layers and application conditions/methods [71].

Photocatalytic nanocomposite films of Titanium dioxide (TiO_2)/polyoxometalate ($Na_4W_{10}O_{32}$, abb. $W_{10}O_{32}$) were prepared via electrostatic LbL (Layer-by-Layer) self-assembly method by Ping Niu and coworkers. The influence of operational parameters was studied including bilayer number of films, initial dye concentration and electron acceptor. The UV light catalytic activities were evaluated by the degradation of Methyl Orange (MO) aqueous solution as pollutant. The experimental results show that the degradation rate of Methyl Orange (MO) decreases with an increase of initial dye concentration. Degradation of Methyl Orange (MO) and the photodecolorization follows first-order kinetics according to the Langmiur- Hinshelwood kinetics [72].

Simona Filice and his groups also investigated effectiveness of permeation flux of pollutant water for anatase-type TiO_2 nanoparticles, GO and organo-modified GO (abbreviated as GO_{SULF}). All membranes were prepared by the solvent casting method. The results showed excellent removal of Methyl Orange (MO) by GO_{SULF} during less than three hours UV irradiation [73].

Prototype composite membranes was produced and prepared through partial reduced of GO-TiO_2 as photocatalytic ultrafiltration water treatment under visible light for treating azo dyes such as Methyl Orange (MO) and Methylene Blue (MB) by Chrysoula P, et al. Among all the membranes, GO-TiO_2 (GOT-10) was the best membrane for removing Methyl Orange (MO) under UV irradiation and GO-TiO_2 was the best membrane for removing Methylene Blue (MB). GOT-10 has the higher photocatalytic activity compared to GOT-5. The amounts of Methylene Blue (MB) and Methyl Orange (MO) which were removed by GO-TiO_2 (GOT-10) under UV irradiation are 7.9, 4.9 mg/min, respectively [74].

Song Bai and coworker synthesized RGO-MFe_2O_4 hybrids with slovothermal method. The results of experiment showed the hybrid removed over 92% Rhodamine B (RhB) and 100% Methylene Blue (MB) with a concentration of 5 mg/L within 2 min. The kinetic data were using pseudo-second-order kinetics. The hybrids RGO-MFe_2O_4 was separated by magnate which was one of advantages for adsorbent from pollutant water [75].

Synthesizing of Graphene/Magnetite composite by solvothermal method for removal of Methylene Blue (MB) from aqueous solution accomplished by Lunhong Aia and his worker. The kinetics and isotherm well described by pseudo-second-order kinetic and Langmuir isotherm model, respectively. Additionally, magnetics nanoparticles (NPs) also showed advantages such as low toxicity, low cost and eco-friendliness [76].

Separation of oil compounds

Polycyclic Aromatic Hydrocarbons (PAHs) can be found in all environmental compartments, atmosphere, soil, aqueous, etc. These compounds are usually generated by natural and anthropogenic processes and can be released into the environments through different ways. There are three types of antropogenic sources of Polycyclic Aromatic Hydrocarbons (PAHs): Phytogenic, petrogenic and pyrogenic sources. These sources are associated with the generation of high molecular weight

Polycyclic Aromatic Hydrocarbons (PAHs) which include creosote, coal tar, which is released into the environment in the form of exhaust and solid residues. Phytogenic-hydrocarbon compounds were derived from plants [77-79]. These compounds are 16 and classified as hazardous compounds by World Health Organization (WHO).

Ainhoa Rubio-Clemente, et al. investigated the methods for removing Polycyclic Aromatic Hydrocarbons (PAHs) from aqueous environment by chemical treatments. Advanced Oxidation Processes (AOP_s) coupled with biological treatments seems to be one of the best solutions for the treatment. In recent decades, Advanced Oxidation Processes (AOP_s) have become useful. This process is based on the production of super active species such as hydroxyl (OH), which is capable of oxidizing a wide range of materials, but its action is non-selective [80].

Mahdie Safarpour, et al. used reduced GO-TiO_2 nanocomposite with variant molar ratios on the Polyvinylidene Fluoride (PVDF) ultrafiltration membranes for treatment. Series of Mixed Matrix Membrane (MMM) Polyvinylidene Fluoride (PVDF) ultrafiltration were prepared by assembled GO, TiO_2 and rGO- TiO_2 nanocomposite using the phase inversion way. The membranes modified with 0.05 wt. % rGO-TiO_2 nanocomposite with rGO to TiO_2 ratio of 70/30 had the greatest hydrophilicity. The bovine serum albumin used as pollutant in this study [81].

Based on Na Wang and his group achievement about GO synthesis according to the Hummers-Offeman method, this technique can be used in sorption of soluble diesel oil from water, too. The adsorption isotherm well fitted to the Freundlich isotherm. Soluble oil molecules were strongly adsorbed on the surface of Graphene by hydrophobic interactions, π-π bonds and Van der Waals reaction. This sorption process composed physical and chemisorption. In comparison of equilibrium sorption capacity of soluble diesel oil on Graphene, expanded Graphite and activated Carbon, Graphene has highest equilibrium adsorption 241.88 mg g^{-1} [82].

X.S. Yi, et al. modified Polyvinylidene Fluoride (PVDF) by nano-sized TiO_2/Al_2O_3 to separate oil/water emulsion by phase inversion method. The effective operating parameters such as initial concentration, Trans Membrane Pressure (TMP), Total Dissolved Solid (TDS) and pH were investigated. All the modified Polyvinylidene Fluoride (PVDF) membranes exhibited higher Rejection Flux (RF) and better antifouling property compared to the bare Polyvinylidene Fluoride (PVDF) membrane under the same operational conditions. The Rejection Flux (RF) decreased sharply by increase in oil concentration from 20 to 400 mg/l. The influence of pH between 2-6.8 was not significant. However, after pH 10, the Rejection Flux (RF) was decreased. Total dissolved solid has no significant influence on Rejection Flux (RF) until 8000 mg/l. There was good flux recovery when Mixed Matrix Membrane (MMM) washed by pure water and NaClO [83].

Yuqing Zhang and coworker studied on phosphorylated TiO_2-SiO_2 particles/polysulfone composite membrane for wastewater treatment. At the first step, Phosphorylated TiO_2-SiO_2 (PTS) particles were successfully prepared by sol-gel method and then, composite membrane was prepared by phase inversion. The results showed that composite membrane was a suitable membrane for treating oily waste water. Phosphorylated TiO_2-SiO_2/Polysulfone (PTS/PSF) membrane has good permeation flux and antifouling compared with other prepared membranes. The operating pressure showed when pressure was increased, the membrane pores became blocked and also permeation flux reduced. The operating temperature and the influence of different oil concentration were investigated [84].

R. Jamshidi Gohari and his groups synthesized a novel Polyethersulfone (PES)/Hydrous Manganese Dioxide (HMO_2) as a new hybrid matrix membrane with improved anti-fouling properties for oily wastewater (TOG or Total Oil and Grease) treatment. PES/HMO_2 membrane showed excellent oil rejection and Flux Recovery Rate (FRR) when the concentration was 1000 ppm compared to the other membranes. Inorganic Hydrous Manganese Dioxide (HMO_2) nanoparticles was synthesized by permanganate in accordance to the Parida's method [85].

Vahid Vatanpour and coworkers synthesized novel antibifouling nanofiltration polyethersulfone membrane by embedding TiO_2 coated Multi-Walled Carbon Nanotubes (MWCNTs). The flux for protein solution was measured at 5 bar for 2 h. The pure water flux, contact angle and hydrophilicity of membrane increased by assembled TiO_2 on the surface of Multi-Walled Carbon Nanotubes (MWCNTs). 0.1 Wt% TiO_2 coated on Multi-Walled Carbon Nanotubes (MWCNTs) showed the highest antifouling properties due to lowest surface roughness [86].

Polysulfone is one of the polymers which are used for preparing Mixed Matrix Membrane (MMM), PSF functionalized by SiO_2 nanoparticle, by A. L. Ahmad and coworkers. The pollutant candidate for separation is oil in water. The result showed that the addition of nanoparticles improved antifouling and permeation flux properties by increasing the pore size. The pore size of the SiO_2 filled membrane increased and became interconnected due to the interfacial stresses between the polymer and filler SiO_2 nanoparticles) [87].

GO-TiO_2 nanocomposite was assembled on poly sulfone for treatment of humic acid by Mahendra Kumar, et al. The different loading amount of adding GO-TiO_2 on PES were 0-5 Wt % by the Non-solvent Induced Phase Separation (NIPS) method. Different concentration of Hyaluronic Acid (HA) prepared to test the antifouling and removal efficiency, increasing concentration of Hyaluronic Acid (HA) which decline efficiency removal of membrane containing 5 wt% nanocomposite (MG-5).

The membrane MG-5 had ability to remove 98.7% of Hyaluronic Acid (HA) from its 10ppm solution at pH = 7. Irreversible fouling decreased by increasing GO-TiO_2. The lowest irreversible fouling is for MG-5 which has highest nanoparticles [88].

Huiqing Wuet, et al. synthesized the Polysulfone (PSF)-based Hybrid Membranes (HMs) by doping with SiO_2-GO nanohybrid. 0.3 Wt % GO-SiO_2/PSF among other membrane showed the best antifouling, permeation flux rate protein rejection. The rejection to egg albumin as containment was at a high level (98%). PSF, GO/PSF, SiO_2/PSF were membranes which tested by Huiqing Wu and coworkers [89].

M. Padaki and his workers also investigated about the membrane separation in oil-water. The results showed that membrane separation process is economically effective compared to other commercial methods. They reported several operating parameters influencing on the ceramic and polymeric membranes. The surface modification by particular monomers and nanohybrids enhanced the Mixed Matrix Membrane (MMM) performance and showed better antifouling and hydrophilicity properties [58].

Zonghua Wang, et al. synthesized novel GO-blended Polyvinylidene Fluoride (PVDF) ultrafiltration membranes. They blended various amount of GO (0.1, 0.15, 0.2, 0.25 and 0.3) on the Polyvinylidene Fluoride (PVDF). The results showed that when the content of GO added in the casting solution was 0.20 wt.%, the water flux and porosity were 457.86 l /m^2.h, 70.7%, respectively. The properties of this membrane were better than other ones. The GO blend Polyvinylidene Fluoride (PVDF) membrane displayed higher water flux recovery ratio compared to the pure Polyvinylidene Fluoride (PVDF) membrane [90].

According to the Zhong–Kun Li, *et al.* PVDF/SiO_2@GO Nanohybrid Membranes (NHMs) synthesized via thermally induced phase for separating the pollutants. Different amount of GO-SiO_2 (0, 0.3, 0.6, 0.9 and 1.2) assembled on the Polyvinylidene Fluoride (PVDF). The M-4 presented the highest Bovine Serum Albumin (BSA) rejection and the lowest permeation flux as 91.7% and 182.6 Lm, respectively. However, more SiO_2@GO addition (1.2 wt%) led to the outstanding pure water permeation flux and much lower Bovine Serum Albumin (BSA) rejection. The melting temperature (Tm) increased with the addition of nanohybrid up to 0.9 wt%. More SiO_2@GO addition (1.2 wt%) led to a small reduction [91].

According to the results of all experiments, optimum operating parameter shows that suitable operating temperature should be between 25-30 °C. By increasing oil concentration, the membranes permeate flux declines. One of the reasons for flux declination is concentration polarization on the surface of membrane as the oil concentration increases. Also, when operating temperature increases, the membrane pore will be blocked, tend to decline of water permeate flux. According to the R. Jamshidi Gohari, the permeation flux under different pHs were not only affected membranes properties but influenced oil dropt size [85].

Other Applications for Removal Pollutant

Wenbo Lu, et al. reported synthesizing of GO-SiO_2 by combining sonication with sol-gel technique. After that, GO-SiO_2 functionalized with AgNP. AgNP/F-SiO_2/GO nanocomposites were prepared. The results showed that glucose biosensor can be used for the glucose detection in human blood serum [92].

Another application for GO is used for antibacterial and antifouling in mixed matrix membrane. Preparation and characterization of HPEI-GO/PES ultrafiltration membrane with antifouling and antibacterial properties was studied by Liang Yu and coworkers. The Ultrafiltration (UF) membrane showed effective antibacterial performance against *Escherichia coli (E. coli)* [93].

On the other hand, complications related to infectious diseases have significantly decreased due to the availability and use of a wide variety of antibiotics and antimicrobial, antibacterial and biological agents. However, excessive use of antibiotics and antimicrobial, antibacterial and biological agents over years has increased the number of drug resistant pathogens. Antibiotics and antimicrobial, antibacterial and biological multidrug resistance poses serious risks and consequently research attention has refocused on finding alternatives for antimicrobial treatment. Among the various approaches, the use of engineered nanostructures is currently the most promising strategy to overcome microbial drug resistance by improving the remedial efficiency due to their high surface-to-volume ratio and their intrinsic or chemically incorporated antibacterial activity. GO, a two-dimensional ultra-thin nanomaterial, possesses excellent biocompatibility, putting it in the forefront for different applications in biosensing, drug delivery, biomedical device development, diagnostics and therapeutics. GO-based nanostructures also hold great promise for combating microbial infections. Yet, several questions remain unanswered such as the mechanism of action with the microbial entities, the importance of size and chemical composition in the inhibition of bacterial proliferation and adhesion, cytotoxicity, and other issues when considering future clinical implementation [94-235].

Results and Discussion

As previously mentioned, GO can improve ultrafiltration membrane hydrophilicity, high permeation flux, mechanical properties and salt retention; it has been used in separation field.

Summary of research findings and achievements

Du Chunhui and coworkers prepared Chitosan modified-Graphene Oxide (CS-GO) nano composite positive osmotic membrane. In many polymers, Chitosan (CS) is a biodegradable, non-toxic and bio-compatibility. The natural polysaccharide polymer is reusable, distributed in nature and it has a rich source. It is a very useful, environmentally friendly material widely used in medicine, food, chemical, membrane separation and other fields. As a natural polymer, membrane material has been widely used in the preparation of ultrafiltration, nanofiltration, reverse osmosis membranes and smart responsive film. The various amount of percentage by mass GO add to nanocomposite 0.1%-1.0%, 2%-5% of Chitosan (CS), 0.1-0.3% of a hydrophilic additive. The results of their patent on nanocomposite show that a better salt interception effect can be achieved via a unique sheet structure of GO [236].

He Linghao, et al. synthesized modified polyvinylidene difluoride (PVDF) super-hydrophobic material and preparation method. The result of study show that the modified polyvinylidene difluoride (PVDF) super-hydrophobic material has excellent oil absorption performance and can be used for efficiently separating oil and water where the amount of the (G)/PVDF or (GO)/PVDF is (1 to 5) (100; 3) under a closed condition [237].

According to the Xu Nanping and his groups, who investigated the enhanced GO hollow fiber composite membrane and preparation method thereof. The prepared membrane was strong in hydrophilicity, and the water permeability of the composite membrane was greatly improved. The thickness of the membrane separation layer was 0.01-1 mm and the membrane pore diameter of the enhanced GO hollow fiber composite membrane is 1-500 (nm) [238]. LbL (Layer-by-Layer) assembly of GO membranes via electrostatic interaction and elucidation of water and solute transport mechanisms also reported by Baoxia Mi and coworkers. The purification of the water and removing contaminant was excellent [239].

Peng Wang, et al. also provided thin films of nanoscale materials. A solution for creating micro arrays on thin film material surfaces was provided. It was demonstrated that filtration-based membrane preparation had potential to impact the produced rGO membrane property. A vacuum filtrated GO or rGO membrane has two surfaces, which are formed at different interfaces [240].

Jae Min Hong and his workers described a method for preparing an asymmetric porous membrane by a dry- wet phase inversion method. The results showed higher permeability because of high hidropilicity and excellent rejection ability of the asymmetric membrane for the sake of high porosity and uniform pore size of mixed matrix membrane [241].

Summary of recent patents

Meng Chuan Koh, et al. also investigated the methods for forming GO/polymer composite membranes. Their patent is recorded. The ratio of mass GO embedded to 14 wt% PSF was 0.0, 0.5 and 1.0 wt%. The method for preparation was wet phase inversion [242].

Ho Bum Park and coworkers also prepared composite separation membrane including GO coating layer and method for manufacturing. The composite separation membrane of the invention has high selectivity for Carbon dioxide. The concentration of the GO in the dispersion obtained preferably from 0.5 to 1.5 g/L [243].

Nanocomposite ultrafiltration membrane containing GO or reduced Graphene Oxide (rGO) and preparation method thereof also reported by Saira Bano, *et al.* The polyacrylonitrile (PAN)/GO nanocomposite ultrafiltration membrane has improved mechanical properties; high permeability and high salt rejection ratio and excellent anti-fouling property. Wherein the GO or rGO was present in an amount of 0.1-10 wt % based on the total weight of the nanocomposite ultrafiltration membrane. Meanwhile, an ultrafiltration membrane including only a hydrophobic polymer is susceptible to fouling. Therefore, according to the their exposure, a hydrophobic polymer matrix is impregnated with GO or rGO to enhance hydrophilic property so increasing permeation and to control the roughness of the membrane surface so that the anti-fouling property may be improved [244].

The patent of Feng Kai, et al. reported added go on the polymer Hybrid Membrane (HM) for exchanging proton. The prepared GO was added to the polymer solution. Proton exchange especially at high temperatures and/under low humidity environment has improved. The exchange of proton on the Hybrid Membrane (GO-HM) is better than pure polymeric membrane [245].

Reported no patent of GO prepared by blending method to the Nafion (perfluorosulfonic acid resin) matrix was prepared by Bong Gill Choi and coworkers. The Hybrid Membrane (HM) was proton exchange film and application in fuel cell [246].

The asymmetric composite membrane synthesized by Nouran Ashraf and coworkers. It was containing a polymeric matrix and Carbon nanotubes within a single membrane layer, wherein the Carbon nanotubes were randomly oriented within the polymeric matrix and the composite membrane was formed by phase inversion. The invention also relates to a desalination method using the composite membrane [247].

With increasing of various containment in the water, it is critical to find a way for obtaining drinking water with low consumption energy and no production of chemical material. This invention reported by Rahul Nair, et al. Pervaporation (or pervaporative separation) is a method of separating mixtures of liquids by partial vaporization through a non-porous or porous membrane [248].

Zhengtang Luo and coworkers prepared methods for synthesizing GO with high surface area, electrical conductivity for using in sensor and device [249].

The forward osmosis composite membrane was a polysulfone-sulfonated polysulfone-inorganic filler blend/polyamide composite membrane synthesized by Xu Tongwen and his groups. Since conventional permeable membrane is generally used in the Reverse Osmosis (RO) system the permeate traditional forward osmosis is low than theoretical value, a high-water flux forward osmosis composite membrane and the preparation method was invented by their group. The method used for synthesis was phase inversion [250].

Graphene membrane filtration and its production method for removing alcohol and water, with efficiency close to 100%, synthesized by Hong Wei-song, et al. [251].

The polyelectrolyte membrane is disposed between the positive and the negative electrode and can include a sulfonated tetrafluoroethylene (PTFE) based fluoropolymer-copolymer that can be subjected to ultraviolet/ozone (UV/O_3) exposure. The metallized nanoparticles increase the efficiency of the fuel cell by at least 50% when the feed gas includes at least 1000 ppm Carbon patented by Miriam Rafailovich, et al. [56,57,94-235,252-256].

Conclusions, Perspectives, Useful Suggestions and Future Studies

We have summarized special characteristics of GO and several investigations about adsorption or filtration of oil compounds by polymeric compounds. The results show high permeation flux, reduce antifouling in polymeric compounds compare to traditional kinds at several investigations. Fouling of membrane occurs due to formation of cake layer on the membrane surface or adsorption on to the membrane surface or within the pores. Although many investigations about the fouling of the membrane carried out by scientists, it will be used for reducing the fouling and increasing the flux. Polycyclic Aromatic Hydrocarbons (PAHs) are neutral, non-polar molecules found in charcoal and in tar deposits. These are also produced by the incomplete combustion of organic materials (e.g., in engines and incinerators, when biomass burns in forest fires, etc.). Exposure to Polycyclic Aromatic Hydrocarbons (PAHs) has also been linked with carcinogenetic disease and poor germinal development. Because of specific characteristics of polymeric membrane such as thermal stability, low energy requirements, chemical resistant, high flux and low fouling compared to usual kinds, it will be used to remove Polycyclic Aromatic Hydrocarbons (PAHs) from wastewater at future by different polymeric membranes such as Polyvinylidene Fluoride (PVDF), cellulose acetate poly ether sulfone and polycarbonate.

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