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REVIEW **Open Access**

A comprehensive review on textile wastewater treatment by coupling TiO_2 with PVDF membrane

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Abstract

Background The textile industry represents a great portion of the global industry due to the increase in population and demand for sustainable products. Tons of textile wastewater contains predominantly synthetic complex organic dyes like direct dyes, processing dyes, reactive dyes, …etc. making discharge of colored effluents challenging.

Main body of the abstract Textile wastewater treatment is essential to maintain the environmental balance and reduce public health threats. Conventional wastewater treatment methods cannot overcome and decompose these toxic wastes; therefore, numerous modern approaches have been studied and implemented for pollutant degradation to be suitable for environmental disposal. Membranes and photocatalysis have proven their significant effect on the photodegradation of different dyes and the production of pure water for further use in industrial purposes.

Short conclusion This review paper aims to represent a comprehensive review of textile dyeing wastewater treatment by integrating polyvinylidene fluoride (PVDF) and titanium dioxide (TiO_2) in a hybrid system named "Photocatalytic membrane reactor, PMR".

Keywords Textile wastewater treatment, Methylene blue (MB), Polyvinylidene fluoride (PVDF), Photocatalysis, Titanium dioxide (TiO_2)

Background

Wastewater is a critical problem that threatens the environmental balance (Yang et al. 2020; Zhao et al. 2022; Azanaw et al. 2022; Mavlanova et al. 2023). It is particularly important to work hard to decrease water pollutants and issue strict regulations for waste disposal to keep our environment clean and healthy (Maity et al. 2020; Arutselvan et al. 2022; Mahboob et al. 2023). Finding new wastewater treatment methods should be one of the important concerns in scientific research. Countries should cooperate to recycle and reuse wastewater due to the decrease in water resources (Zahmatkesh et al. 2023; Stefanakis 2020; Bouwer 2000). Several aspects produce large volumes of wastewater (e.g., the industrial sector (Rajoria et al. 2022)), the agricultural sector (Khan et al. 2022), and the domestic sector (Ghani and Mahmood 2023), all of which should collaborate to treat wastewater to be disposed of according to the environmental regulations (Paredes et al. 2019; Partyka and Bond 2022).

The textile industry is one of the largest sources yielding tons of waste dyeing effluents that, even at low concentrations, reduce wastewater transparency, and oxygen solubility, and are toxic (Mondal et al. 2018; Castillo-Suárez et al. 2023; Islam et al. 2023). Dyeing and finishing processes are one of the main reasons for producing textile wastewater. A large variety of inputs, as chemicals

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and dyestuff, are used in these processes and unfortunately, not all the inputs are contained in the final product, therefore, they become waste and discharged to the environment (Jahan et al. 2022; Yaseen and Scholz 2019). The main reason for the obstacle to textile wastewater treatment is the difficulty in dealing with the chemical structure of the textile chemicals (Azanaw et al. 2022). Pollutants exist as suspended solids, heat, color, acidity, and chemical oxygen demand (Uddin 2021; Palani et al. 2021). Moreover, the pH can be changed over a wide range from 2 to 12 which increases challenges in front of treatment methods. (Gadow et al. 2022; de Araújo et al. 2020). Some characteristics of textile wastewater are shown in Table 1 (Pal 2017b).

Therefore, the treatment of textile wastewater is a serious challenge due to the presence of strong color and chemical composition of liquid waste. Significant concern was directed to investigate different methods to treat textile wastes before being discharged into the environment to meet the limitations imposed by legislation (Castillo-Suárez et al. 2023; Sharkey et al. 2020; Pervez et al. 2021). Over the years, wastewater treatment methods were advanced beyond conventional methods (i.e., coagulation, filtration, adsorption, etc.) to overcome the complexity and diversity of pollutants existing in domestic, industrial, and agro-industrial waste streams and to provide clean drinking water to confront the population growth and water scarcity issues (Abuhasel et al. 2021; Rahman et al. 2023).

To date, several studies have reported the remarkable effect of coupling membranes and photocatalysis for dye photodegradation (Li et al. 2023; Farouq 2022; Khan et al. 2023). Nevertheless, the fabrication of polyvinylidene fluoride (PVDF) membrane and integration with titanium dioxide (TiO_2) photocatalyst is still challenging. This leads to the current review to highlight the developments of PVDF membranes and TiO_2 photocatalyst for methylene blue degradation in a hybrid system named "Photocatalytic Membrane Reactor, PMR".

Table 1 Industrial textile wastewater characteristics (Pal 2017b; Katal et al. 2014; Kapdan and Alparslan 2005)

Parameters	Values based on literature
Dye concentration (mg/L)	700
pH	9–10
Chloride (mg/L)	17,750–34,000
Sulfate (mg/L)	1400
Total nitrogen (mg/L)	23
Biological oxygen demand (mg/L)	363
Chemical oxygen demand (mg/L)	1781
Dye (mg/L)	15–8000
Total dissolved solids (mg/L)	1950–2925
Color	50–2500

Main text

Wastewater treatment

Different technologies are involved to improve wastewater quality to be disposed of according to environmental regulations. Owing to the large volumes of wastewater, treatment is carried out in continuous open systems. Wastewater treatment can be classified according to the nature of the treatment process to physical, chemical, and biological processes (Bera et al. 2022; Donkadokula et al. 2020; Crini and Lichtfouse 2019).

Physical treatment is the removal of material ready to be settled out by gravity or floating in the water where no change in chemical or biological composition occurs (Darra et al. 2023). In domestic wastewater treatment, physical treatment will approximately lead to the removal of a few of the organic and non-organic loads (Al-Mawla et al. 2023). Physical treatment is carried out by comminution, screening, sedimentation, grit removal, aeration, pH control, and flotation. Chemical treatment involves chemical reactions to improve water quality. It consists of different processes like coagulation, neutralization, disinfection, and ion exchange (Akbar et al. 2023). Some processes represent a combination of physical and chemical treatment as adsorption by activated carbon.

The biological treatment method is defined as the decomposition of dissolved organic matter by microorganisms under aerobic or anaerobic conditions (Ilmasari et al. 2022). It is widely used for domestic sewage treatment. Wastewater is introduced into a bioreactor where microorganisms start to feed on the dissolved organic matter in wastewater. Microorganisms responsible for decomposition are bacteria (aerobically or anaerobically), algae, and fungi (aerobically). Due to the presence of food and oxygen, biological oxidation occurs to end up with stable thick bacterial biomass, therefore, it is necessary to separate the produced biomass using sedimentation. This sludge contains bacterial cells in contrast to the sludge from the physical treatment process which is fecal solid. Several methods are operated under aerobic conditions such as oxidation ponds, aerobic digestion, and trickling filtration. On the other hand, septic tanks and anaerobic digestion utilize the anaerobic conditions by holding the wastewater in tanks for 1–2 days to reduce the biochemical oxygen demand (BOD) by about 35–40% (Kurniawan et al. 2020; Arlyapov et al. 2022).

Photocatalysis for wastewater treatment

Photocatalysis is a physicochemical wastewater treatment process in which a catalyst is induced by light for activation and accelerating the rate of reaction (Ali et al. 2023). The catalysts used in this reaction are semiconductors (Hong et al. 2022). Each semiconductor has its bandgap for activation (Table 2). Thus, by illuminating the catalyst with light having energy higher or equal to the bandgap energy, electrons transfer from the valence band to the conductive band creating a hole in the valence band. Oxidation and reduction photodegradation reactions take place by the generated hole and electron, respectively (Chaves et al. 2020; Zhu and Zhou 2019).

Photocatalysis is widely studied and investigated in different aspects (Patial et al. 2022; Yu et al. 2022). Illumination of the catalyst can be carried out by visible light or UV light according to the required bandgap energy. Photocatalytic degradation of organic water pollutants by a photocatalyst present as a slurry can be described as (Baruah et al. 2019):

a. Dispersing the photocatalyst in wastewater.
b. Illuminating the catalyst by the proper wavelength.
c. Recording water concentration.
d. Separating the dispersed photocatalyst from the treated water.

Photocatalysis is divided into two categories, homogeneous photocatalysis, and heterogeneous photocatalysis. The main difference between the two categories is the phase of the catalyst and reaction medium. If the catalyst and the reaction medium are in phase (same phases), then the photocatalysis process is said to be homogenous photocatalysis. A simple example of that is the photodegradation of dye using water-soluble carbon dots. On the other hand, heterogeneous photocatalysis is termed when the catalyst and the reaction medium are out of phase (different phases). Usually, heterogeneous photocatalysis takes place between a solid-phase catalyst and a water-soluble organic pollutant compound. Separation of the catalyst from the reaction medium in homogenous photocatalysis is difficult in contrast to heterogeneous photocatalysis. Heterogeneous photocatalysis is the most common process for wastewater treatment and degradation of MB by the TiO_2 solid particles represent this process (Zeitoun et al. 2020; Riaz and Park 2020; Antonopoulou 2022).

Table 2 Bandgap energies for various semiconductors (Serpone and Pelizzetti 1989; Schiavello and Sclafani 1989; Sakthivel et al. 2000)

Semiconductor	Bandgap energy (eV)	Semiconductor	Bandgap energy (eV)
Diamond	5.4	WO_3	2.76
CdS	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
SnO_2	3.54	ZrO_2	3.87
CdSe	1.7	Cu_2O	2.172

Semiconductors suitable for photocatalysis

Different semiconducting materials, such as oxides (TiO_2, ZnO, CeO_2, ZrO_2, WO_3, V_2O_5, Fe_2O_3, etc.) and sulfides (CdS, ZnS, etc.) have been used as photocatalysts (Hong et al. 2022; Tahir et al. 2020). A photocatalyst should have the following properties (Weldegebrieal 2020; Xiao et al. 2020):

- Significantly active.
- Non-poisoning and stable for long-term operation at high temperatures.
- Resistant to attribution and mechanically stable.
- Physically and chemically stable under various conditions.
- Activated by visible light and UV light.
- Cost-effective and eco-friend.
- High surface area, mobility, and lifetime.

Titanium dioxide (TiO_2) has proven its outstanding performance in many applications over other semiconductors (Ijaz and Zafar 2021; Kang et al. 2019). Binary metal sulfides such as CdS and PbS are toxic and unstable for catalysis. ZnO has been reported to have the same energy characteristics as TiO_2, however, it suffers instability in illuminated aqueous solution leading to photocorrosion affecting its activity after long-term operation (Barnes et al. 2013; Štrbac et al. 2018). WO_3 has been studied by different researchers and it was found that it is less active than TiO_2. Combining two or more different photocatalysts can improve the photocatalytic activity and stability significantly (Ferreira et al. 2023; Al-Mamun et al. 2019; Pasini et al. 2021).

Titanium dioxide structure, characteristics, and activation mechanism

Titanium dioxide has three well-known natural polymorphs which are anatase (tetragonal), brookite (orthorhombic), and rutile (tetragonal) (Elamin et al. 2023; Lavrov et al. 2022). The unit cells of these structures are shown in Fig. 1 where grey and dark red spheres represent titanium and oxygen, respectively (Hiroi 2022; Eddy et al. 2023; Bai et al. 2014; Hu et al. 2003).

Rutile is the thermodynamically stable phase whereas anatase and brookite are metastable phases that transform into rutile upon heating (Hu et al. 2003; Hanaor and

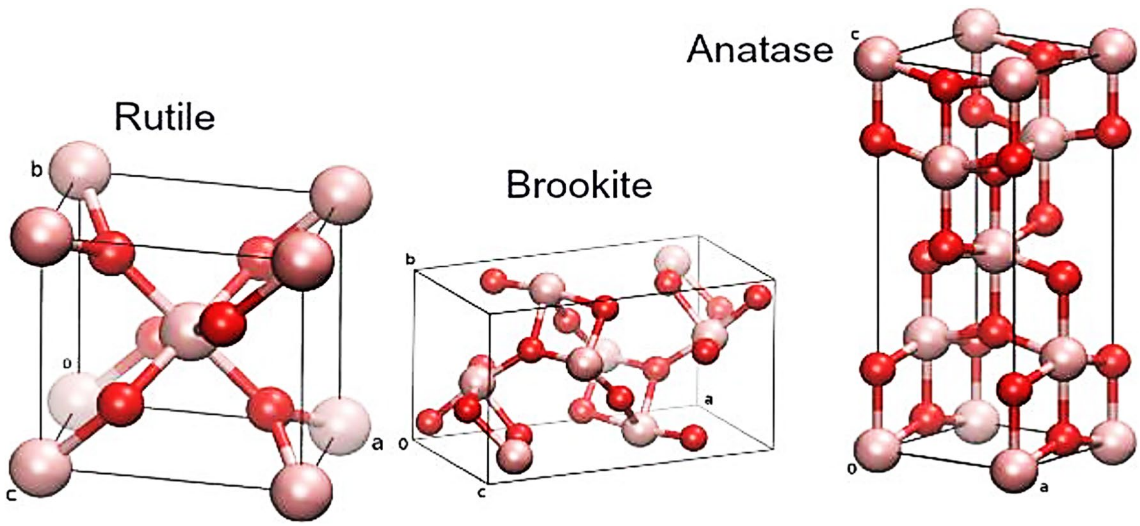

Fig. 1 Titanium dioxide polymorphic structures

Sorrell 2011; Janczarek et al. 2022). The different octahedral arrangement for each polymorph brings about their diverse physical and electronic properties. In particular, the energy values of their band gaps vary from 3.0 eV ($\lambda = 413$ nm) in rutile to 3.2 eV ($\lambda = 387$ nm) in anatase and 3.2–3.4 eV ($\lambda = 387–365$ nm) in brookite (Ibhadon 2008). The photocatalytic activity of TiO_2 depends on many factors including crystalline phase, crystal size, lattice or surface defects, specific surface area, particle size, duration of light irradiation, charge carrier lifetimes, or efficiency for charge transfer to molecules adsorbed on the semiconductor surface (Riaz and Park 2020; Yamazaki et al. 2020; Navidpour et al. 2023). Regarding the pure phases' evaluation, it is well established that anatase is the polymorph that shows the highest photocatalytic activity in water treatment applications (Žerjav et al. 2022).

Owing to the wide bandgap energy of anatase TiO_2 (3.2 eV), various studies were concerned about improving TiO_2 activity to be activated by visible light. These modifications were presented in numerous studies (Saianand et al. 2022; Tang et al. 2022; Arora et al. 2022; Kanakaraju et al. 2022) and include:

- Coupling of TiO_2 with dye sensitization (Behera et al. 2022), polymer sensitization (Enesca and Cazan 2022), and semiconductors to improve surface properties (Thambiliyagodage 2022; Cui et al. 2022).
- Creation of oxygen vacancies and oxygen sub-stoichiometry for bandgap modification (Khatibnezhad et al. 2022, 2021).
- Doping with non-metals (N Asahi and Morikawa 2007; Di Valentin et al. 2007; Shen et al. 2007; Feng et al. 2008; Li et al. 2008), S (Sakai et al. 2008; Wang et al. 2008; Zhou et al. 2008; Wei et al. 2008), C (Xu et al. 2006; Ren et al. 2007), B (In et al. 2007; Zhang and Liu 2008), F (Todorova et al. 2008; Wu and Chen 2008), Cl (Long et al. 2007)) and co-doping with two different non-metals as (N and S (Wei et al. 2008), N and F (Xie et al. 2007), N and B (Xue-li and Wei 2022), etc.
- Doping with metals as (Fe, V, Cr, Mn, Co, Ni, Cu, Pt, etc.) (Kanakaraju et al. 2022; Khatibnezhad et al. 2021).

Although the only disadvantage of TiO_2 is inactivity by visible or sunlight and a UV light source must be used for activation (Binjhade et al. 2022). Moreover, it has superior characteristics as photo-stability, chemically and mechanically inert, non-toxic, efficient photoactivity, resistance to photo-corrosion and microbe, and lower cost (Nasrollahi et al. 2021).

Titanium dioxide is widely used in wastewater treatment as shown later. Several studies investigated the mechanism of organic pollutant (i.e., methylene blue) degradation, and as shown below it is based on activation by the suitable wavelength to transfer the electron from the valence band to the conductive band (Konstantinou and Albanis 2004; Mohammad Jafri et al. 2021; Bullo et al. 2022).

$$TiO_2 + h\nu \rightarrow TiO_2\left(e^- + h^+\right) \quad (1)$$

$$h^+ + H_2O \rightarrow H^+ + HO^{\cdot} \quad (2)$$

$$h^+ + HO^- \rightarrow HO^{\cdot} \quad (3)$$

$$e^- + O_2 \rightarrow O_2^{\cdot -} \quad (4)$$

$$O_2^{\cdot -} + H^+ \rightarrow HO_2^{\cdot} \quad (5)$$

$$HO_2^{\cdot} + HO_2^{\cdot} \rightarrow H_2O_2 + O_2 \quad (6)$$

$$H_2O_2 + O_2^{\cdot -} \rightarrow HO^{\cdot} + HO^- + O_2 \quad (7)$$

$$e^- + H_2O_2 \rightarrow HO^{\cdot} + HO^- \quad (8)$$

$$H_2O_2 + h\nu \rightarrow 2HO^{\cdot} \quad (9)$$

$$\text{Pollutant} + \left(h^+, HO^{\cdot}, e^-, HO_2^{\cdot}\right) \rightarrow \text{degradation products} \quad (10)$$

Membranes

A membrane can be defined as a selective barrier or interphase between two bulk phases Membranes are essentially used for separation processes, and they have been widely adopted over the last 30 years for various industrial applications (Vasishta et al. 2023; Suresh et al. 2023b). Separation processes based on membranes are more capital and energy-efficient than conventional separation processes (Suresh et al. 2023b). A membrane can be solid, liquid, or gel and symmetric or asymmetric and homogenous or heterogenous and it can be neutrally, positively, or negatively charged. Membrane thickness ranges from less than 1 nm to more than 1 cm. The driving force for mass transport through the membranes can be gradients in concentration, temperature, or pressure (Al-Najar et al. 2020). Membranes include various fabrication materials, structures, morphology, and configurations. Despite the existence of different types of membranes, all membranes have the same function which is the prevention of undesired species from the passage to the permeate side, Fig. 2 (Qamar et al. 2023; Graham and Higgins 2020; Costa et al. 2019).

Membrane distillation (MD) is a favorable technique for desalination and water/wastewater treatment, and it provides several advantages over conventional treatment methods (Yang et al. 2022; Nishad and Rajput 2023). It is a thermally driven process where vapor molecules of the solvent are only capable to pass through a hydrophobic porous membrane (Meng et al. 2023; Julian et al. 2023). The existence of a vapor pressure difference between the two sides of the membrane improves the passage of vapor molecules from the feed compartment to the permeate compartment. MD has many advantages (Gude 2018; Pal 2017a; Zhong et al. 2021; Kebria and Rahimpour 2020), such as:

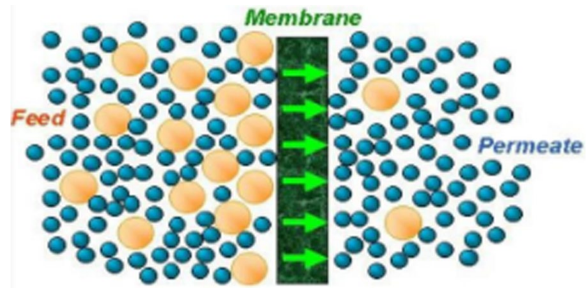

Fig. 2 Membrane separation mechanism (Sani 2015)

- Low operating temperatures where it is not required to heat the feed solution to its boiling point.
- The hydrostatic pressure in MD is lower than all other membrane processes as reverse osmosis.
- Low cost.
- Membranes can be made from cheap materials such as plastic therefore there will be no corrosion problems.
- High rejection factor and complete separation process.
- Less fouling in comparison to other membrane processes due to the large membrane pore size.
- MD can be combined with other separation processes to form an integrated system producing outstanding products and high efficiency.
- Renewable energy sources can be utilized to heat the feed.

However, there are some limitations for MD (Nishad and Rajput 2023; Reddy et al. 2022; Yang et al. 2022):

- Low permeated flux due to concentration and temperature polarization.
- The presence of trapped air inside the membrane cell increases the resistance to mass transfer, hence permeating flux decreases.
- Heat loss by conduction.
- Membrane fouling and wetting.

Contribution of PVDF membrane in wastewater treatment

A popular polymeric membrane material is polyvinylidene fluoride (PVDF), it is a semi-crystalline polymer containing 59.4 wt% fluorine and 3 wt% hydrogen (Dohany 2000; Rajeevan et al. 2021; Ismail et al. 2021). A PVDF monomer structure is ($-CH_2-CF_2-$) and the spatial arrangement of the atoms can be verified by

Fourier transform infrared spectroscopy (FTIR) characterization (Van Tran et al. 2019). Moreover, ease of PVDF dissolution by different organic solvents such as dimethylacetamide (DMAC), dimethylformamide (DMF), and *n*-Methyl-2-Pyrrolidone (NMP) encourages the evaluation of PVDF performance in different applications and purposes (Saxena and Shukla 2021; Fei et al. 2019).

Recently, PVDF has gained remarkable attention due to its exceptional characteristics in comparison to other polymers such as polysulfone (PS), polyethersulfone (PES), and polyimide (PI). High mechanical properties, thermal stability, chemical resistance, and high hydrophobicity have provided PVDF a notable center of research for various operations (Mohammadpourfazeli et al. 2023; Dallaev et al. 2022; Suresh et al. 2023a, b) from 2010 to 2023 as shown in Fig. 3.

This review article focuses on the role of PVDF membrane in wastewater treatment. Figure 4 shows that the number of publications increased from 2010 to 2023 revealing the outstanding performance and high removal efficiency of pollutants from wastewater to be suitable for environmental disposal.

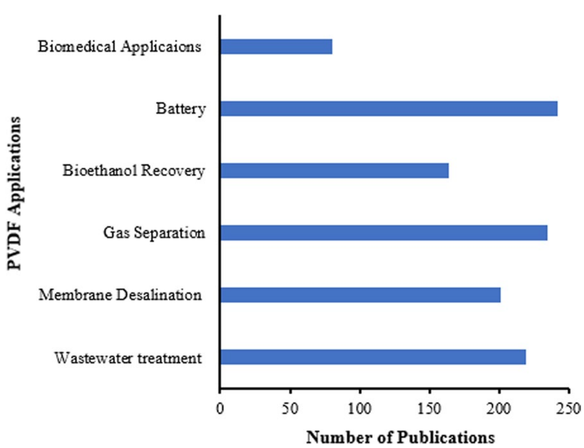

Fig. 3 Contribution of PVDF in various processes

Integrating PVDF and TiO_2 for methylene blue removal

The limitations accompanied by membrane separation processes and photocatalytic degradation of contaminants can be solved by coupling both methods to form a hybrid system named photocatalytic membrane reactors (PMRs) (Samuel et al. 2022). As a result, PMRs have gained a noticeable focus for evaluation in the wastewater treatment aspect (Chen et al. 2022) as shown in Fig. 5. The performance of different PMRs combinations of membrane fabrication materials and photocatalysts have been assessed in the open literature (Mozia 2010; Chakachaka et al. 2022; Chen et al. 2022), and it was found that an integrated system of PVDF membrane along with TiO_2 photocatalyst seems to be a promising system for methylene blue photocatalytic degradation owing to the previously mentioned advantages of the PVDF, as well as TiO_2.

Jia et al. (2009) compared the performance of PVDF pure membrane versus PVDF/TiO_2 composite membrane on the removal of two different dyes (i.e., MB and Congo red (CR) dyes). Pure PVDF membrane and PVDF/TiO_2 hybrid membranes were prepared by phase inversion. Membrane properties were examined by a series of analytical methods. It was found that rejection of dye increased by increasing TiO_2 content to 21%. The retention of methylene blue was more than CR due to the ability of the negatively charged membranes to retain positively charged methylene blue components. Moreover, the anti-fouling performance was examined by using a protein solution (BSA) and the results showed that the relative flux of the blend membranes are higher than the pure membrane and reached more than 80%.

Likewise, Ngang et al. (2012) synthesized polyvinylidene fluoride (PVDF)-Titanium dioxide (TiO_2) mixed-matrix membranes by phase inversion and proved that the mixed matrix membrane is photo-catalytically active as it shows better MB degradation compared to the neat membrane with $\sim$ 100% pure water flux recovery

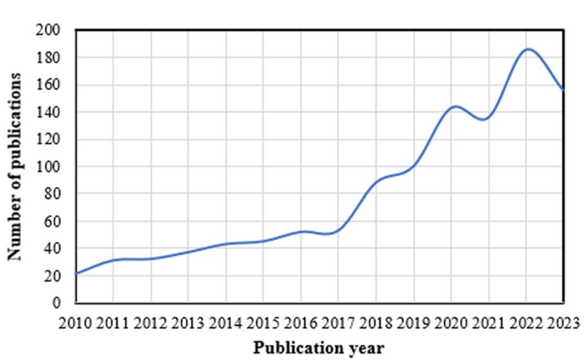

Fig. 4 Contribution of PVDF studies in wastewater treatment annually

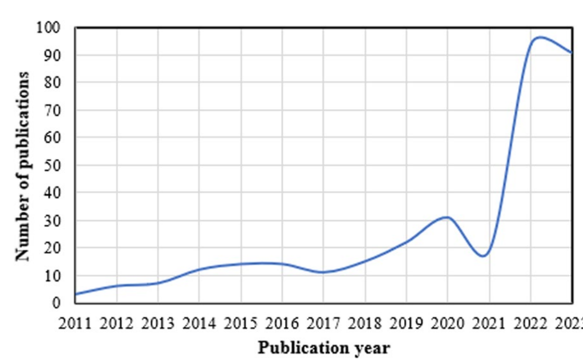

Fig. 5 Contribution of PMRs in wastewater treatment annually

under 1 h of UV light irradiation. Membrane characterization was measured, and photocatalytic performance was tested against the degradation of methylene blue (MB). The hydrophilicity of the mixed-matrix membranes $\left(392.81 \pm 10.93 \frac{l}{m^2} \cdot bar\right)$ showed significant enhancement in comparison with the pure PVDF membrane $\left(76.99 \pm 4.87 \frac{l}{m^2} \cdot bar\right)$.

Li et al. (2015) evaluated the performance of $Ag/TiO_2/PVDF$ composite membrane in wastewater treatment. The composite membrane was prepared via the blending/photo reduction combined method. Membrane characterization was performed and showed high hydrophilicity owing to a large amount of hydroxyl group (−OH) formed on the PVDF membrane. The composite membrane showed excellent activity in the degradation of MB and the inactivation of bacteria under visible-light illumination. Correspondingly, (He et al. 2016) utilized the electrospinning technique to fabricate nanofibrous PVDF membranes containing TiO_2 nanoparticles and Ag nanoparticles as well. Mechanical strength was improved due to the presence of Ag nanoparticles. Degradation of MB was recorded to be 39.5%, 50.9%, 78.9% and 92.3% for pure PVDF membrane, Ag/PVDF membrane, $TiO_2/PVDF$ membrane and $Ag–TiO_2/PVDF$ membrane respectively.

Martins et al. (2014) evaluated the performance of a copolymer (PVDF/TrFE) membrane with immobilized TiO_2 nanoparticles doped with erbium (Er) and codoped with Er and praseodymium (Pr). High porosity (75%) coupled with a low bandgap (2.63 eV of these TiO_2-modified nanoparticles) and high surface area (273 m^2/g) achieved 98% MB degradation after exposure to UV for 100 min.

Li et al. (2014) prepared a novel hollow fiber composite membrane of PVA/PVDF with nano TiO_2 particles by dip coating. Crosslinking between the polymers was done by glutaraldehyde which improved the mechanical, chemical, and thermal stabilities as well. Characterization was performed to examine the surface morphology and chemical structures of the modified membranes. Membrane separation efficiency was highly influenced by dye concentration, salt concentration, pH, and operating temperature. The results showed that the optimum amount of TiO_2 is 1 g/L by which rejection to CR, Methylene Orange (MO) and MB reached $94 \pm 2.57\%$, $52.1 \pm 2.45\%$, and $92 \pm 2.20\%$, respectively. The modified membranes with TiO_2 proved higher antifouling, stability, and separation efficiency in comparison to neat PVA/PVDF membranes.

Fischer et al. (2015) studied the performance of three membranes (hydrophilic PES and PVDF, hydrophobic PVDF) coated with TiO_2 nanoparticles. The coating was carried out via hydrolysis of titanium tetraisopropoxide. All the tested membranes degraded MB; however, the hydrophilic $TiO_2/PVDF$ membrane achieved the highest degradation rate. Moreover, Hydrophilic (TiO_2/PES and $TiO_2/PVDF$) membranes degraded two non-inflammatory drugs (diclofenac and ibuprofen) in contrast to hydrophobic $TiO_2/PVDF$ membrane.

Ramasundaram et al. (2016) thermally fixed TiO_2 nanoparticles on PVDF at different temperatures to study their photocatalytic activity. TiO_2 nanoparticles dispersed in methanol were electrosprayed on a steel mesh (SM) coated with PVDF followed by thermal fixation to improve the mechanical strength. When the electrosprayed volumes were 10, 20, 30, 40, 50, and 60 mL, the TiO_2 loading on both sides of the PVDF-coated SM was $0.20, 0.43, 0.73, 0.97, 1.10,$ to 1.60 mg respectively. The SM sample with $1.1mg$ TiO_2 proved to be the optimum for MB degradation under UV irradiation due to its high stability for 20 photocatalytic runs. Thermal fixation at 160 °C showed higher MB degradation than those fixed at 180 °C and 200 °C because of PVDF melting (165–172 °C) leading to entrapping the TiO_2 nanoparticles and decreasing their photocatalytic activity. The rate constant for MB removal (100% removal efficiency) was found to be 0.0251 min^{-1} for the optimum SM-TiO_2.

Dadvar et al. (2017) investigated the characteristics and performance of different polymeric membranes such as a perfluorinated polymer (Nafion), cellulose acetate, polycarbonate (PC), polysulfone fluoride (PSF), and polyvinylidene fluoride (PVDF). The examined membranes were supported with semi-conductor TiO_2 and graphene oxide (GO) to improve the antifouling property, photocatalytic activity for removal of Azo dyes as methylene blue, and these membranes proved their ability to be used in the treatment of wastewater from polycyclic aromatic hydrocarbons (PAHs).

The blended PVDF with different dosages of $Ag/TiO_2/APTES$ for dyeing wastewater treatment (Peng et al. 2018). Membrane hydrophilicity was tested by comparing the contact angle of the composite membrane (61.4°) with the original PVDF membrane (81.8°). Rejection of MB increased to 90.1% after the addition of $Ag/TiO_2/APTES$ in comparison with the neat PVDF membrane which showed rejection rate of only 74.3%. Moreover, the composite membrane with a concentration of 0.5% $Ag/TiO_2/APTES$ showed a significant effect on inhibition of bacteria preventing it from reproducing or causing serious membrane fouling.

Li et al. (2017) coated PVDF membrane with three dimensional (3D) TiO_2/ZnO photocatalyst and detected its stability, reusability, and photocatalytic oxidation.

Coating of the PVDF membrane surface and the pore walls were carried out by atomic layer deposition (ALD). Excellent stability, reusability, and photocatalytic oxidation were realized during MB degradation. The photo-induced super-hydrophilicity was realized with a decline of 82.6% for the water contact angle and an increase of 33.5% for the pure water flux. The antifouling property was enhanced especially for the composite membrane ($TiO_2 : ZnO = 1 : 3$) to reach 73% humic acid (HA) removal.

Galiano et al. (2018) tested the photocatalytic activity of ultrafiltration $TiO_2/PVDF$ hollow fiber membranes (HFs) for MB degradation in water and salty seawater. These hollow fibers proved high hydrophilicity, stability, and catalytic activity under UV-A irradiation. Despite the low amount of catalyst added (0.5%), degradation of MB was found to be 97% under UV-A irradiation. Moreover, HFs achieved 97% MB reduction which reveals their excellent catalytic activity for degradation of MB in synthetic seawater.

Cheng and Pu (2018) improved the photodegradation of MB by blending multi-walled carbon nanotubes (MWCNTs) with TiO_2. A novel PVDF composite nanofibers were fabricated using nanolayer coextrusion. TiO_2 and multi-walled carbon nanotubes (MWCNTs) were blended with the PVDF to develop a highly active PVDF photocatalyst membrane. It was observed that MWCNTs played a vital role in MB degradation as they acted as bridges between TiO_2 particles which resulted in an improvement in transfer of electrons. The photocatalytic reaction rate for PVDF/(30 wt% TiO_2/MWCNTs) and PVDF/(40 wt% TiO_2/MWCNTs) reached 0.412 h^{-1} and 0.531 h^{-1}, respectively, which exceeded that of pure TiO_2 (0.375 h^{-1}).

Abdullah et al. (2018) prepared $PVDF/TiO_2$ hollow fibers membranes and evaluated their performance by photodegradation of MB. The results showed that the undoped PVDF as well as TiO_2 doped VDF membrane was capable of degrading MB. The composite membrane (9 wt% $TiO_2/PVDF$) showed the highest performance while the neat PVDF membrane showed the lowest performance which reveals the significant effect of incorporating the TiO_2 photocatalyst into the PVDF membrane. The rate of degradation of the MB fitted well into first order kinetic data with apparent kinetic constants of 0.0591, 0.0295, 0.0188, and 0.0100 obtained using pure membrane, undoped PVDF, 3 wt% $TiO_2/PVDF$, 6 wt% $TiO_2/PVDF$, and 9 wt% $TiO_2/PVDF$, respectively.

Lee et al. (2018) studied the effect of changing the ratios of PVDF and PVP on performance of the composite membranes. P25-TiO_2 was entrapped with a bi-polymer system of electrospun fibers of PVDF and PVP in different ratios. Concentration of TiO_2 in the fabricated membranes ($PVDF : PVP = 2 : 1$, $PVDF : PVP = 1 : 1$, $PVDF : PVP = 1 : 2$) was maintained to be 4%. Methylene blue (MB) degradation was tested to visualize contaminant removal, assess the sorption capacity (5.93 ± 0.23 mg/g) and demonstrate stable removal kinetics ($k_{MB} > 0.045\ min^{-17}$) under UV-A irradiation (3.64×10^{-9} einstein/cm^2/s) over 10 cycles. The membrane ($PVDF : PVP$ weight ratio of $2 : 1$) proved to have the highest photocatalytic activity.

Liu et al. (2018) fabricated a hollow fiber membrane of ($TiO_2/PVDF$) using single orifice spinneret. The fabricated membrane was used for degradation of dye, i.e., MB and Congo red (CR), and Na_2SO_4 resulting from textile wastewater. The results revealed outstanding properties such as good hydrophilicity, high stability for long operations, and good flux recovery ratio. Additionally, the hollow fiber ($TiO_2/PVDF$) membrane rejected with excellence the CR and MB dye and less retention extent to Na_2SO_4.

Suriani et al. (2019) synthesized a composite nanofiltration membrane of ($PVDF/SDS - GO/TiO_2$) by phase inversion technique. Synthesis of dimethylacetamide-based graphene oxide (GO) was carried out by electrochemical exfoliation in combination with single-tail sodium dodecyl sulfate (SDS) surfactant. Comparison between neat PVDF membrane and the composite membrane was performed in a dead-end cell at the pressure of 2 bar. Results showed the excellent performance of the composite membrane in MB rejection (92.76%) in comparison with the pure PVDF membrane. The composite membrane achieved the highest flux (7.770 L/m^2 h) due to the presence of GO and TiO_2 which affected the membrane morphology and properties as the increase in hydrophilicity, an increase in porosity (57.46%), and a decrease in contact angle ($64.0 \pm 0.11°$). Moreover, the fabricated membrane attained the highest water permeability (4.187 L/m^2 h bar).

Martins et al. (2019) prepared by solvent casting a microporous membrane based on polyvinylidene difluoride-co-trifluoro ethylene (PVDF/TrFE) with immobilized TiO_2 nanoparticles. Characterization was conducted by scanning electron microscopy, energy dispersive X-ray spectroscopy, Fourier-transform infrared spectroscopy, porosimeter, and contact angle goniometry. Performance of the fabricated membrane exhibit high degradation for MB (99%), ciprofloxacin (95%), and ibuprofen (48%). Moreover, long-term stability and high efficiency were observed after 20 h of UV irradiation, corresponding to four use cycles.

Venkatesh et al. (2020) modified electrospun PVDF membrane by graphitic carbon nitride (G-C_3N_4) decorated on reduced graphene oxide (RGO) with titanium dioxide (TiO_2). The novel ternary composite membrane exhibited outstanding thermal stability, antifouling performance, and increased contact angle. Moreover, performance was tested against the rejection of oil–water emulsion and MB which reached $95.4 \pm 0.1\%$ and $94.2 \pm 0.5\%$, respectively. These results are attributed to the reasonable hierarchical structure and formation of the hydration layer which chemically and physically helps in the separation of oil–water emulsion and dye rejection.

Abdelmaksoud et al. (2021) examined the effect of a composite of nano black (NB–TiO_2), graphene oxide (GO), and PVDF on photodegradation of MB and malachite green dyes. At first NB-TiO_2 was prepared by simple hydrogenation of anatase TiO_2 while modified Hummer's method was utilized for the preparation of graphene oxide. Secondly, GO–PVDF membrane was synthesized by electrospinning technique followed by submerging the electrospun GO–PVDF into a crosslinking medium with 2.5 wt% glutaraldehyde (GA). Finally, a solution of NB–TiO_2 was added to the PVDF–GO nanofibers resulting in the required composite PVDF–GO/NB–TiO_2. Evaluation of the composite nanofibers was carried out by the photodegradation of MB and malachite green. It was found that the optimum degradation efficiency of malachite green and MB dyes occurred at pH values of 8 and 10, respectively, while the maximum photocatalytic degradation efficiencies of malachite green and MB were found to be 74 and 39%, respectively, under visible light after 30 min.

Zeitoun et al. (2020) developed a new hybrid photocatalytic membrane reactor for treating industrial waste (e.g., organic dye MB waste). The experimental setup was designed in a flexible way to enable both separate and integrated investigations of the photocatalytic reactor and the membrane, separately and simultaneously. A membrane cell containing electrospun PVDF membrane and a photocatalytic reactor with TiO_2 as a slurry were implemented for MB photodegradation. Different amounts of TiO_2 and dye concentrations were investigated, and results showed that 100% removal is achieved at certain conditions. Additionally, kinetic analysis was carried out to indicate that degradation of MB follows pseudo-first-order reaction.

Yadav et al. (2021) prepared photocatalytic TiO_2 sheets (PTS) incorporated PVDF-co-hexafluoropropylene (HFP) UV-cleaning mixed matrix membranes with various PTS compositions (0–5 wt%). This prepared membrane was utilized to eliminate Congo red (CR) and MB from synthetic textile industry wastewater, with equal CR and MB concentrations $\left(100\ \mathrm{mg/l}\right)$ and 4% NaCl via direct contact membrane distillation (DCMD). The results showed that the prepared membrane with 3 wt% PTS has a dye removal efficiency close to 100% for both MB and CR and 6.1 $\mathrm{kg/m^2}$ h vapor flux. Moreover, after UV cleaning, the flux recovery ratio (FRR) for long-run DCMD studies (5 days) was more than 91%.

Since membranes are the most effective dye wastewater treatment approach, (Azhar et al. 2022) listed all prior research in their review article relating to membrane technology used to recover MB from wastewater. The Fischer et al. study in 2015 (Fischer et al. 2015) is included in this review article, where the results showed that after 2 h, the MB recovery percentage was 100% when using the hydrophilic $TiO_2/PVDF$.

A glass hollow membrane fabricated using glass waste, PVDF, and *N*, *N*-dimethylacetamide DMAC, coated with TiO_2 via dip-coating and calcination by Zhang et al. (2022). In this study, the photocatalytic removal of MB from wastewater using the fabricated membrane was examined and the results showed that the TiO_2-coated membrane calcined at 550°Chas good photocatalytic and antifouling properties. Also, the photocatalytic removal of MB was higher than 97.2% and could be recycled multiple times by a simple treatment. Moreover, the MB removal percentage was in the range of 92.3–93.6% after five recycling operations.

Ma et al. (2023) studied MB recovery from synthetic wastewater with initial MB concentration equals 0.5 mg/L using PVDF, carbon black (CB), and TiO_2 membrane hybridized via a polyvinylpyrrolidone (PVP)by phase inversion method. The fabricated PVDF–CB–TiO_2 membrane recorded recovery percentage equal to 98.6%.

PVDF–TiO_2–graphene oxide (GO) based hybrid membrane was blended with various polymer additive (poly methyl methacrylate (PMMA), polyvinyl alcohol (PVA), and polyvinylpyrrolidone (PVP)) using phase inversion method in Mohamat et al. (2023a, b). The impact of combining 4 wt% from the different polymer additives was evaluated based on MB dye recovery percentage and antifouling performance. The Experimental outcomes showed that adding PVP to the membrane matrix resulted in high recovered water flux 635.897 $\mathrm{L/m^2}$ h and permeability 55.833 $\mathrm{L/m^2}$h Pa together with 57.56% MB recovery percentage. Whereas, adding PMMA to membrane enhanced the MB recovery percentage to 79.18% with 86.46% flux recovery ratio (FRR). Additionally, Mohamat et al. study in 2023 (Mohamat et al. 2023a, b) investigated how various polymer types (such as polyether sulfone (PES) and PVDF) utilized in membrane fabrication affected the membrane performance. Based on the findings of this investigation, the PVDF–GO–TiO_2 demonstrated a higher MB recovery percentage of 92.63% and an FRP that was near to 100%.

All above list of studies involving the treatment of MB from wastewater (illustrated in Table 3) show that membrane technology is the most efficient method to recover MB from wastewater. Specially with some modifications to the membrane fabrication by integrating PVDF and TiO_2 to improve the membrane performance.

Table 3 Previous studies on MB removal by PVDF and TiO_2

PVDF configuration	Dissolution solvents	Fabrication technique	% TiO_2	MB Conc	Percentage removal/photo-degradation	References
Flat sheet	DMAc	Phase inversion	6 vol% 12 vol% 21 vol%	0.01 mM	67.52% 75.89% 82.63%	Jia et al. (2009)
Flat sheet	DMAc	Phase inversion	1.5 wt%	10 ppm 50 ppm 100 ppm	98.84% 93.11% 85.24%	Ngang et al. (2012)
Flat sheet	DMAc	Blending/photo reduction combined method	4 wt%	10 ppm 20 ppm 30 ppm	24% 23% 21%	Li et al. (2015)
Flat sheet	DMF and acetone	Electrospinning	0.4 wt%	0.05 M	78.9%	He et al. (2016)
Flat sheet	DMF	Solvent casting	2 ml of titanium isopropoxide solution	0.00001 M	98%	Martins et al. (2014)
Hollow fiber	–	Purchased	1 g/L	50 mg/L	92 ± 2.20%	Li et al. (2014)
Flat sheet	–	Purchased	2.00 × 10 M titanium isopropoxide solution	10 µm	100%	Fischer et al. (2015)
Flat sheet	DMAc	Dip coating of PVDF solution on steel mesh	10 ml 20 ml 30 ml 40 ml 50 ml 60 ml	10 µM	68.7% 76% 87.3% 91.6% 100% 100%	Ramasundaram et al. (2016)
Flat sheet	DMAc	Immersion phase inversion	Amount of (Ag–TiO_2–APTES): 0.1 g 0.2 g 0.5 g	3 mg/L	80.3% 86.7% 90.1%	Peng et al. (2018)
Flat sheet	–	Purchased	Titanium tetraisopropoxide solution	10^{-5} M	27%	Li et al. (2017)
Hollow fibers	N-Methyl-2-pyrrolidone (NMP)	Dry/wet spinning	0.5 wt%	10 µM	97%	Galiano et al. (2018)
Flat sheet	–	Nanolayer coextrusion	10 wt% 20 wt% 30 wt% 40 wt%	10 ppm	22% 32% 57% 64%	Cheng and Pu (2018)
Hollow fibers	DMAc	Electrospinning	3 wt% 6 wt% 9 wt%	10 ml/L	85% 90% 100%	Abdullah et al. (2018)
Flat sheet	DMAc and acetone	Electrospinning	4 wt%	3.2 mg/L 6.4 mg/L	93.75% 81.25%	Lee et al. (2018)
Flat sheet	DMAc	Solvent casting	1 wt%	10 ppm	92.76%	Suriani et al. (2019)
Flat sheet	DMF	Solvent casting	0.087 g	2 mg/L	99%	Martins et al. (2019)
Flat sheet	DMF	Electrospinning	5 ml Titanium butoxide	–	94.2%	Venkatesh et al. (2020)
Flat sheet	DMF	Electrospinning	5 mg 10 mg 20 mg	5 ppm	29% 32% 39%	Abdelmaksoud et al. (2021)

Table 3 (continued)

PVDF configuration	Dissolution solvents	Fabrication technique	% TiO_2	MB Conc	Percentage removal/photo-degradation	References
Flat sheet	DMAc and acetone	Electrospinning	0.1 g/L	4 ppm 7 ppm 11 ppm 15 ppm	100% 100% 100% 99.4%	Zeitoun et al. (2020)
			0.2 g/L	4 ppm 7 ppm 11 ppm 15 ppm	100% 100% 98.1% 100%	
			0.3 g/L	4 ppm 7 ppm 11 ppm 15 ppm	100% 100% 100% 99.3%	
Flat sheet	Methanol	Spray drying followed by calcination	0–5 wt%	100 mg/L	100%	Yadav et al. (2021)
Flat sheet	TTIP/ethanol	Dip coating	0.092 wt%	10 μmol/L	100%	Fischer et al. (2015)
Hollow fiber	Ethanol	Phase inversion calcination/dip coating	0.62 wt%	20 mg/L	97.2%	Zhang et al. (2022)
Flat sheet	Ethanol	Phase inversion	2–5 wt%	0.5 mg/L	98.6%	Ma et al. (2023)
Flat sheet	Dimethylacetamide (DMAC)	Casting solution	1 wt%	10 ppm	79.18%	Mohamat et al. (2023a, b)
Flat sheet	DMAC	Non-solvent induced phase separation (NIPS)	1 wt%	10 ppm	92.63%	Mohamat et al. (2023a, b)

Conclusions

Membrane separation in combination with photocatalysis has a promising effect on textile wastewater treatment. TiO_2 is a stable semiconductor where effective dye removal can be achieved under UV-irradiation. Various modifications for TiO_2 to be activated under visible light are reported. The limitation of membrane processes as organic fouling can be decreased by coupling photocatalysis and membrane in a photocatalytic membrane reactor. Several studies for MB removal by photocatalytic membrane reactors have been reviewed showing higher performance than conventional membrane processes and photocatalysis as well. However, different photocatalytic membrane reactors configurations should be studied to determine the most suitable design for highest dye removal and flux. This review article will provide valuable knowledge for further research work about wastewater treatment by membrane photocatalytic reactors.

Abbreviations

ALD	Atomic layer deposition
APTES	Aminopropyltriethoxysilane
BOD	Biochemical oxygen demand
BSA	Protein solution
CR	Congo red
CB	Carbon black
DMF	Dimethylformamide
DMAC	Dimethylacetamide
DCMD	Direct contact membrane distillation
Er	Erbium
FRR	Flux recovery ratio
FTIR	Fourier transform infrared spectroscopy
GA	Glutaraldehyde
GO	Graphene oxide
$G\text{-}C_3N_4$	Graphitic carbon nitride
HA	Humic acid
HFs	Hollow fiber membranes
MB	Methylene blue
MD	Membrane distillation
MO	Methylene orange
MWCNTs	Multi-walled carbon nanotubes
NMP	*N*-methyl-2-pyrrolidone
PAHs	Polycyclic aromatic hydrocarbons
PC	Polycarbonate
PI	Polyimide
PS	Polysulfone
PES	Polyethersulfone
PMR	Photocatalytic membrane reactor
PMMA	Poly methyl methacrylate
PTS	Photocatalytic TiO_2 sheets
PVA	Polyvinyl alcohol
PVP	Polyvinylpyrrolidone
PVDF	Polyvinylidene fluoride
SDS	Sodium dodecyl sulfate
SM	Steel mesh
TiO_2	Titanium dioxide
TrFE	Trifluoro ethylene

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