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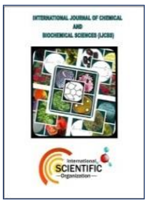

Super Hydrophilicity and Anti-Fouling PVDF Membranes for Dye Removal: A review

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Abstract

Water pollution caused by volatile synthetic dyes. For elimination of pigments from water, membrane-based treatments of wastewater are becoming more and more prominent. Because of its advantageous characteristics, poly (vinylidene fluoride) or (PVDF)-based nanomembranes have become fairly common. The use of these nanomembranes in the cleanup of artificial dyes using various methodologies is discussed in this review. Innovative manufacturing strategies for achieving high performance surface coating and mixing techniques are addressed in relation to PVDF-based nanomembranes. Research on the application of PVDF based nanomembranes in membrane distillation, filtering, adsorption and catalysis (ozonation, oxidant activation, and photocatalysis) has been thoroughly reviewed. In order to boost the performance or working of PVDF membranes, nanomaterials such as metals, metal compounds, (synthetic/bio) polymers, carbon materials, their composites and metal organic frameworks were employed. The benefits and drawbacks of using nanoparticles in the PVDF-based membranes were also discussed. It was explored how nanoparticles affected the PVDF membrane's surface characteristics, mechanical strength, crystallinity, hydrophilicity and catalytic capacity.

Keywords: Poly (vinylidene fluoride), Nanomaterials, Membrane technology, Synthetic dyes, Organic membranes

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1. Introduction

In human society, colors play a vital role [1]. The visible light part of the solar spectrum is absorbed by pigments or dyes, which are employed as colorants. Dyes maybe organic molecules that are very soluble in water, whereas dyes can also be inorganic substances that are not soluble in water. Both natural and synthetic dyes are available, but the tanneries, food, pharmaceutical, plastic, cosmetics, textile, photographic paper/pulp, dye and paint manufacturing industries as well as service contributors (such as hospitals and educational institutions), are the main users of synthetic dyes like xantheneazo, triphenyl methane, phthalein, oxazine, anthraquinone and cyanine dyes [2-3]. According to a survey, the global market for pigments and dyes was valued at roughly 36.4B US dollars in the year 2021, and a higher compound growth rate per year is anticipated between 2022 and 2030 [4]. In particular, the world's dye industries have grown rapidly because of the rising demand for dye. Approximately 7 × 105 metric tons of organic dyes are produced annually [5]. Hazardous dye effluent discharge from the previously mentioned companies has a harmful impact on the environment. For instance, the discharge of wastewater containing synthetic dyes into the environment contaminates ground and surface water supplies and has an impact on humans as well as aquatic life, including bacteria, plants and animals [6]. Several anionic (negatively charged molecules in mixture) and cationic (positively charged molecules in mixture) synthetic/artificial dyes' toxicity to ecosystems have been well studied [7].

Toxic colors therefore need to be eliminated from the wastewater. Thus far, a variety of physical (adsorption, filtration and ion exchange process), chemical (coagulation/flocculation, advanced oxidation processes like photocatalysis, electrochemical method) and biological (including bacteria, enzymes, algal aided biodegradation) methods have been developed to purify or clear the dye-contaminated/polluted water [6-8-9]. Among these instances, the utilization of membrane technology to eliminate dyes from water has received increased attention. Membrane technology has several benefits for wastewater treatment, including easy procedure, reduced equipment size, excellent removal ability, no requirement for special chemicals, low energy usage, selective separation as well as low cost of capital [10]. It must be stated that membrane methodology has extensive applications in treatment of water as well as in a variety of other fields such as hydrogen production, oil water separation, rainwater purification, gasification power plants, space engineering, resource recovery, CO_2 capture and biofuel production [11-12]. A membrane serves as a barrier which separates two phases from one another by limiting the flow of components via it selectively. Depending on its chemical composition, membranes can be classified as either inorganic (that includes ceramic) or organic (like polymers). Inorganic membranes are made of three layers: selective, intermediate, as well as support layer with various sizes of pores.

Inorganic compounds include metal oxides (such as Al_2O_3, ZrO_2, SiC, and metals, and inorganic wastes [13]. Inorganic membranes are resilient, extremely resistant to chemicals, and able to function efficiently across a wide variety of pH levels and temperatures. Additionally, composites made of inorganic materials were created to improve wastewater treatment efficacy [14]. However, organic polymers including polyethylene, polyacrylonitrile, polytetrafluoroethylene, polycarbonate, polypropylene, polyimide, polyvinylidene fluoride (PVDF), polyamides & polyethersulfones make up the majority of organic membranes [10]. Organic membranes have several benefits, including low cost, high reusability, flexibility, and ease of production. Additionally, organic polymer membranes with a greater filtering capacity are available in a variety of forms, including spiral and sheet PVDF membrane is one of the most widely used organic polymer membranes, Because of its exceptional qualities [15]. However, because of its hydrophobic nature, this membrane has unsatisfactory permeation and lessen removal capacity during the separation phenomenon [16]. Typically, membranes that have hydrophobic nature are extremely receptive to fouling phenomena, which is caused by the hydrophobic connection within solutes, microbes and membranes.

The resulting precipitation of insoluble inorganic as well as organic materials causes fouling, that reduces the effectiveness of PVDF membranes in the treatment of dyes [17]. The researchers were inspired by such challenges to enhance PVDF membrane performance in the treatment of wastewater stained with dyes. As a result, PVDF membranes coated with nanoparticles were created for dye treatment [18]. Multiple studies were conducted on PVDF membranes coated with nanoparticles in order to remove various dyes from water. Critical reviews of certain membrane materials with greater implications, membranes with unique applications, or membrane production techniques have been published. The advancement of PVDF membranes for water filtration has been critically reviewed [15]. This review's main objectives are to: (a) provide a report of recent developments in nanomaterial-modified or altered PVDF membranes used for elimination of synthetic/artificial organic dyes using various treatment methodologies; (b) present an overview of the manufacturing processes for these membranes & (c) evaluate impact of nanomaterials on PVDF membrane performance in dye removal techniques. Possible advancements of modified nanomaterial PVDF membranes in treatment of wastewater has also been prophesied based on literature review.

2. Modified PVDF membrane fabrication techniques

Surface coating & blending are the two main fabrication techniques that are the focus of recent developments in PVDF membrane manufacture. PVDF powder, cross-linking agents, organic solvents, and water as a non-solvent are used in the synthesis of these membranes, regardless of the particular method. Organic polymers and nanoparticles are added in process to enhance performance and provide certain features such as superhydrophilicity.

2.1. Surface Coating Techniques

A coating on the surfaces of the PVDF membrane is done to ensure that the nanomaterials are spread uniformly as well as avoid aggregation of the nanoparticles and to ensure complete removal of the dye.

a) Vacuum Filtration

This is only a basic process that involves pouring a mixture of modifying components over the PVDF membrane, followed by its filtering. They are modified and enhanced PVDF with the Graphene Oxide/Ag2CO3/UiO-66-NH2(Metal Organic Framework) using the ultrasonic technique to prepare an aqueous suspension, filter it through the membrane, and dry it at 45'C [19]. Cu-MOF-74 was also used on the PVDF pretreated with polydopamine [20]. The retention of the components was enhanced by using glutaraldehyde or past treatments like wiping the membranes with ethanol and deionized water prior to self-polydopamine polymerization.

b) Grafting method

Membranes made of PVDF may be coated by dipping them directly in modifying solutions. Graphitic carbon nitrate g-C3N4/PVDF membranes are obtained by immersion of pure PVDF in deionized water and immersion in a graphitic carbon nitrate g-C3N4 solution at 55°C throughout the entire night [21]. Afterwards, the membrane was washed with deionized water. Stiffier coating is often acquired through pretreatment or activation. Oxygen plasma is also employed to activate PVDF membranes, after which the samples were put directly into 1H, 1H, 2H, or 2H-perfluorodecyltriethoxysilane or methyltrichloroalkylsilane [22]. The plasma activation on the membrane surface forms functional groups or ions; the solvents are rinsed over and it is dried. The ability of these organosilane-grafted membranes to remove dye is dependent on their concentration and the length of time of grafting.

c) Chemical Modifications

In the coating process, one of the chemical processes such as reduction or solvothermal processes is conducted using PVDF membranes. Membrane surface is first nucleated then development of modifying layers ensues. As an example, Al-Cu layered doubly hydroxide- modified PVDF membrane was produced through a hydrothermal process wherein the membrane was dipped in a mixture of urea, aluminum nitrate and copper nitrate at temperatures of 100C over six hours [23]. Another way of coating Fe-0 nanoparticles involves the binding of Fe-2 + ions to the membrane and reducing Fe'2 + into Fe-0 by dipping in sodium borohydride [24]. One should know that as nanomaterials released off the PVDF, secondary contamination can occur during the dye removal.

d) Synthesis- through contra-diffusion

This technique is based on the simple principle of diffusion of solution across the membrane. All ingredient solutions are kept in different separate beakers and were put in a PVDF membrane to enable the development of the modifying layer. As an example, 2-methylimidazole and zinc nitrate aqueous solutions were kept apart to create the zeolitic imidazolate framework-8 (ZIF-8) layer/PVDF membrane

[25]. Then put a PVDF membrane disc (diameter of 12 cm) into the above-mentioned solutions, which in turn lead to the diffusion of the solutions and the development of a layer of ZIF-8.

2.2. Blending Techniques

Once modifying substances have been mixed with solution of PVDF, the membrane is molded. These membranes can be known as the mixed membranes of the matrix. The process may increase hydrophilicity, avoid fouling, and control material leaching during water treatment.

e) Phase Inversion Strategy

This method is associated with the utilization of organic solvent to create a solution of PVDF and additives and pour them on the substrate (like glass). Additive-loaded PVDF is left as a thin film on the substrate as the solution is evaporated (this gives the material its sheet form). This is then turned into a sheet of membrane by immersion in a water bath. The lithium chloride (LiCl)/graphene oxide/PVDF membrane was created by phase inversion or immersed precipitation, i.e. LiCl, graphene oxide and PVDF powder were put into N,N-dimethylacetamide solvent sequentially until a casting solution was formed [26]. After deaeration, the casting solution poured on a glass substrate and membrane solidified by immersing it in water horizontally. Following phase inversion, produced membrane is immersed in water to remove any solvents that may still be trapped in it. Titanium dioxide nanotubes/Polyaniline/PVDF membranes were also manufactured through phase-inversion approach [27].

f) Spinning Method

PVDF membranes enhanced with nanoparticles were created using electrospinning technique. Electrospinning works by electrifying a liquid to create a jet that is then stretched and elongated to generate fibers on a substrate. PVDF & Chitin nano whiskers were combined in N,N-dimethylacetamide to create the chitin/PVDF membrane [16]. A fraction of the aforementioned fluid is electro spun onto aluminium sheets using a plastic syringe. The polyamide-coated PVDF membrane was prepared using a wet spinning process [28]. A spinneret is used to instantly propel the bore fluid as well as polymer dope solution into a water bath; the inversion of phase process created the membrane. Water and PVDF were used as a bore liquid (extrusion rates = 2.0 mL per minute) and polymer dope (4.0 mL per minute).

3. Applications of dye treatment

PVDF membranes coated with nanomaterials are employed in a number of ways to remove dye contaminants from water (Fig. 1). This section has addressed important membrane-related treatment approaches.

3.1. Adsorption

Adsorption is a very important technique for removing dyes present in water because of its advantages, which include ease of use, high removal efficiency, and affordability [29]. However, it is difficult to recover adsorbent particles from water that has been treated once the adsorption process is over [30]. When membrane technology and adsorption are combined, the problem of adsorbent recovery is resolved and the efficiency of pollutant removal further increased (Fig. 2). For instance, chitin nano whiskers integrated into PVDF membranes to produce an adsorbent membrane. Because it contains polar functional groups, chitin is a hydrophilic biopolymer. Because chitin is hydrophilic, modified PVDF layer had much lower interaction angle of 22.72◦ than original PVDF membrane (contact angle = 93.1◦). An adsorption experiment conducted by dipping a 1 cm2 chitin and PVDF membrane in 40 mL of 200 mg per liter of indigo-carmine solution [16]. Both pure & chitin/PVDF membranes' adsorption capacities were calculated to be 12.5 and 72.6 mg g−1, correspondingly. Both pure as well as chitin-modified PVDF membranes have drainage efficiencies of 22.3% and 88.9%, respectively. Chitin polymer's functional groups made enough space for indigo-based carmine dye to physisorb. Other potential attraction methods include ionic interactions and hydrogen bonding. From first to the third adsorption cycle, removal potency of chitin/PVDF membrane gradually decreased (88.9%–36.1%) [26].

3.2. Filtration

Wastewater containing dyes can be treated by filtering using PVDF membranes. One advantage of adsorptive (nanomaterials enhanced) PVDF membrane filters is their excellent filtering capacity, which prevents dyes by chemical or physical adsorption. Dyes are often separated from water using dead-end and cross-flow filtering systems (Fig. 3). PVDF membranes have been modified to create adsorptive membranes using biopolymers, metal organic frameworks and materials composed of carbon. PVDF membranes have also included the surface-functionalized nanoparticles. For example, it has been reported that dopamine (0.36 weight percent) functionalized halloysite nanotubes/PVDF membrane show good removal effectiveness for direct red 28 (86.5 percent), direct blue 14 (93.7%) and direct yellow 4 (85%) [31]. PVDF membranes with nanocomposite integration have been proven to increase the dye removal rate. When nanocomposites are introduced to PVDF membranes, dye rejection probability significantly increased. Ideal concentration of modifying nanomaterials & structure of molecules of dyes determine how well nanocomposite-modified PVDF membranes filter [32-33]. pH of solution may also affect degree to modified PVDF membrane removes dye. Additionally, nanocomposites enhance PVDF membranes' electrical properties, reusability. For instance, PVDF loaded with Ni–Ag nanoparticles to create an electrically conductive membrane that employed in an electric field- assisted filtering process [34].

3.3. Catalysis

The membrane does not break down or totally mineralise the dyes; it only separates them. By mixing membranes with various catalysts, this problem was resolved. Different types of nanomaterials show catalytic potential in multiple ways. The catalytic PVDF membranes created for dye treatment are described in this section.

3.3.1. Oxidants' Activation

When oxidants like peroxymonosulfate (HSO5-) are activated through catalytic PVDF membranes, some active radicals are produced that may rapidly break down dyes. For example, various quantities of zeolitic imidazolate frameworks-67 (ZIF-67) loaded PVDF membranes have been developed to eliminate methylene blue, rhodamine B and orange II dyes by catalytic degradation caused by sulphate radicals.

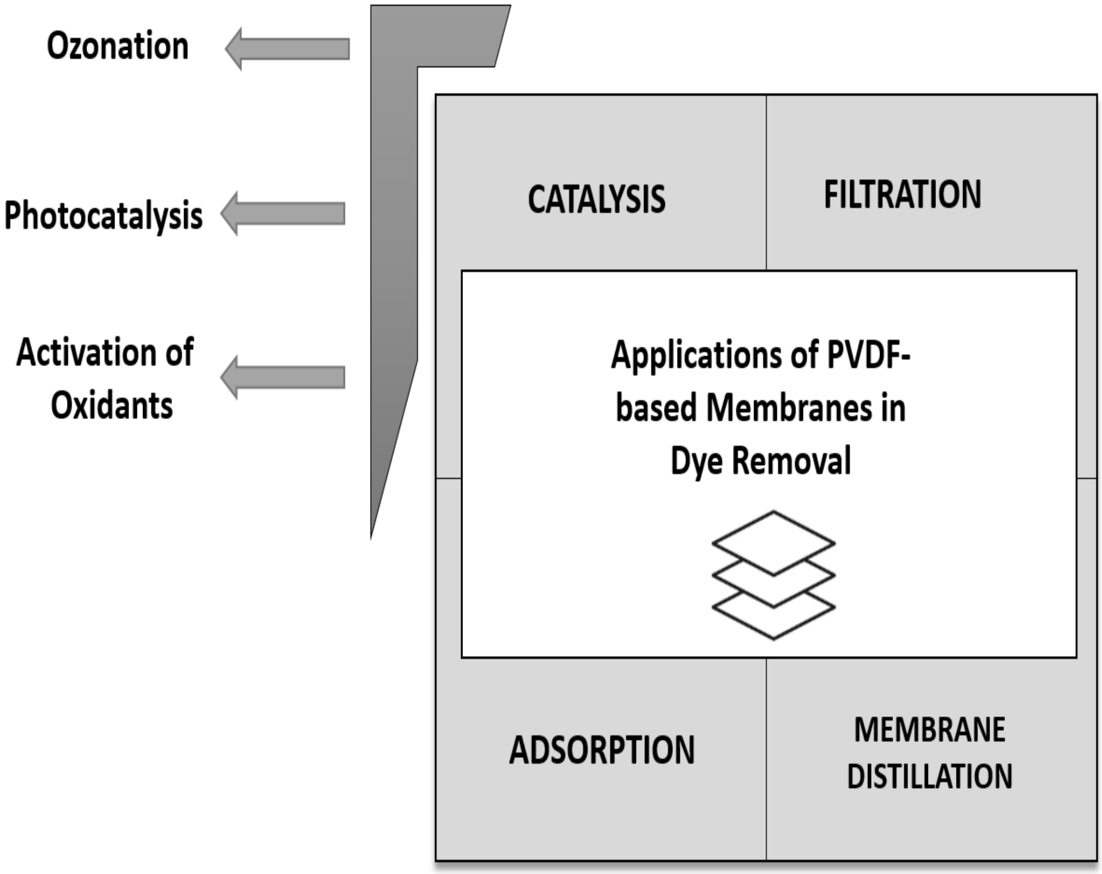

Fig. 1. Employing PVDF membranes modified with nanomaterials for dye treatments

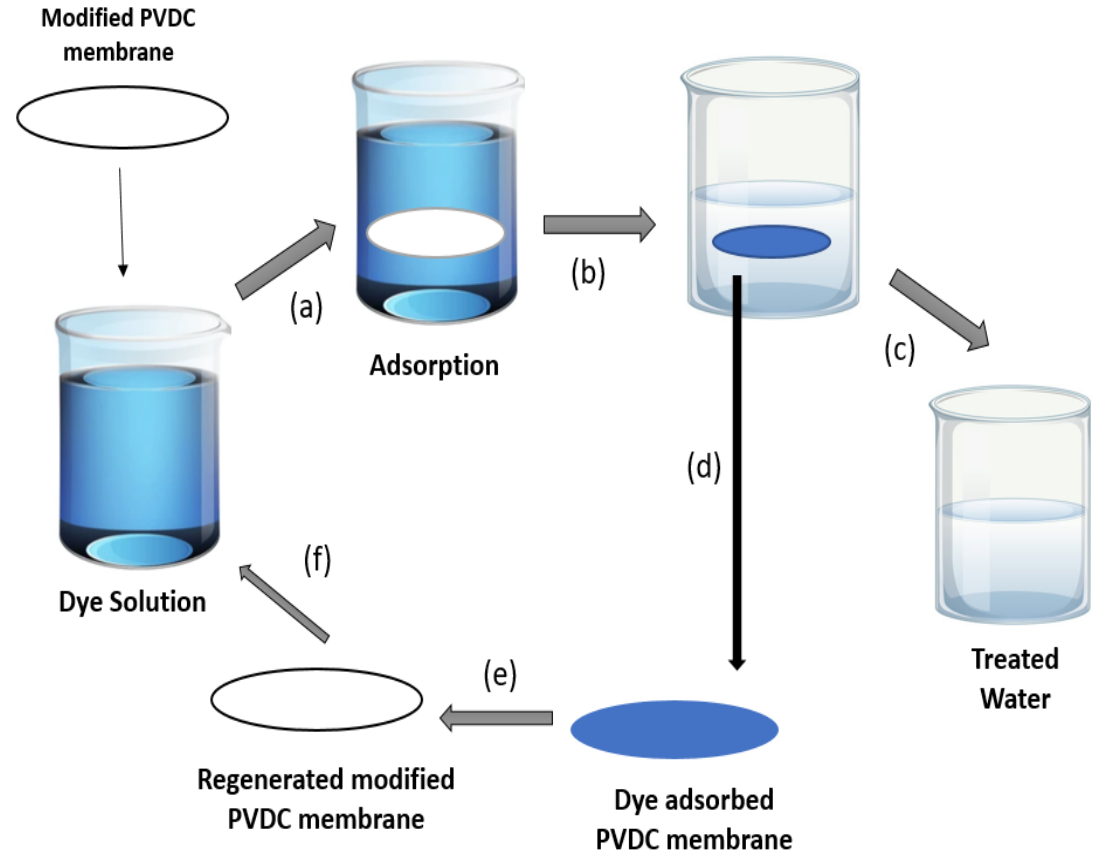

Fig. 2. PVDF membrane use for adsorption-based dye removal

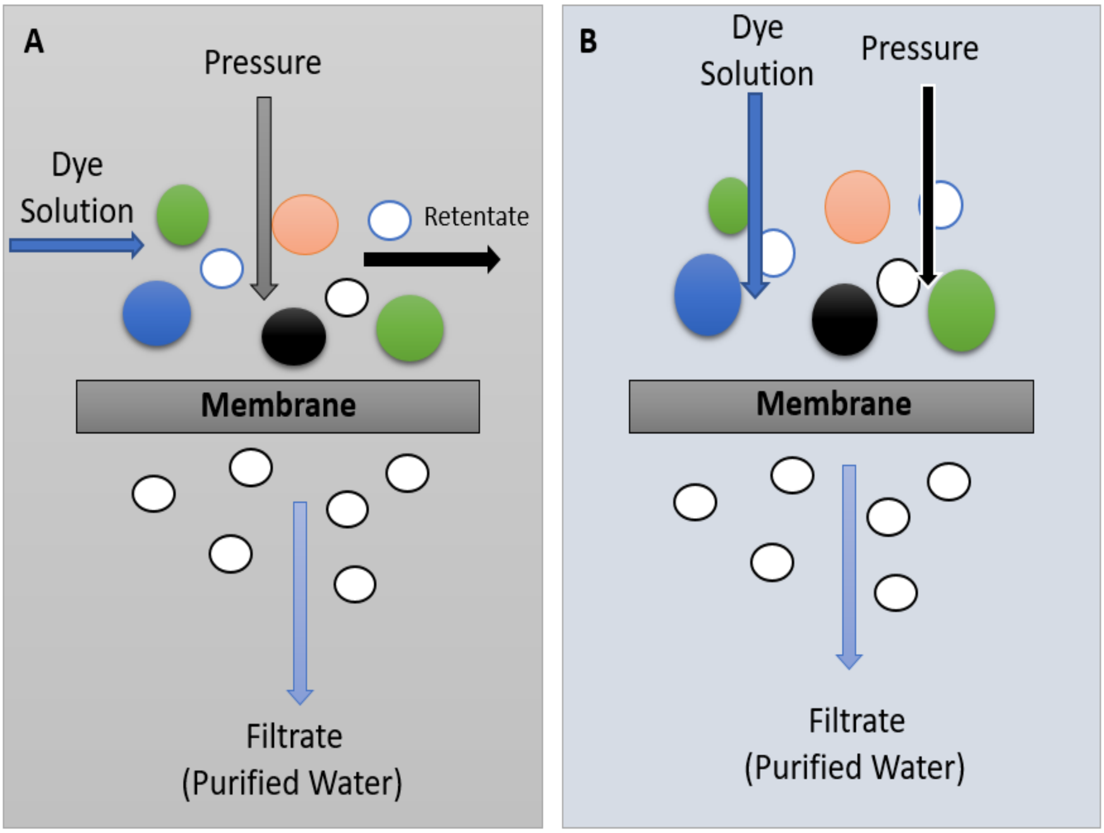

Fig.3. (A) Cross flow and (B) Dead-end

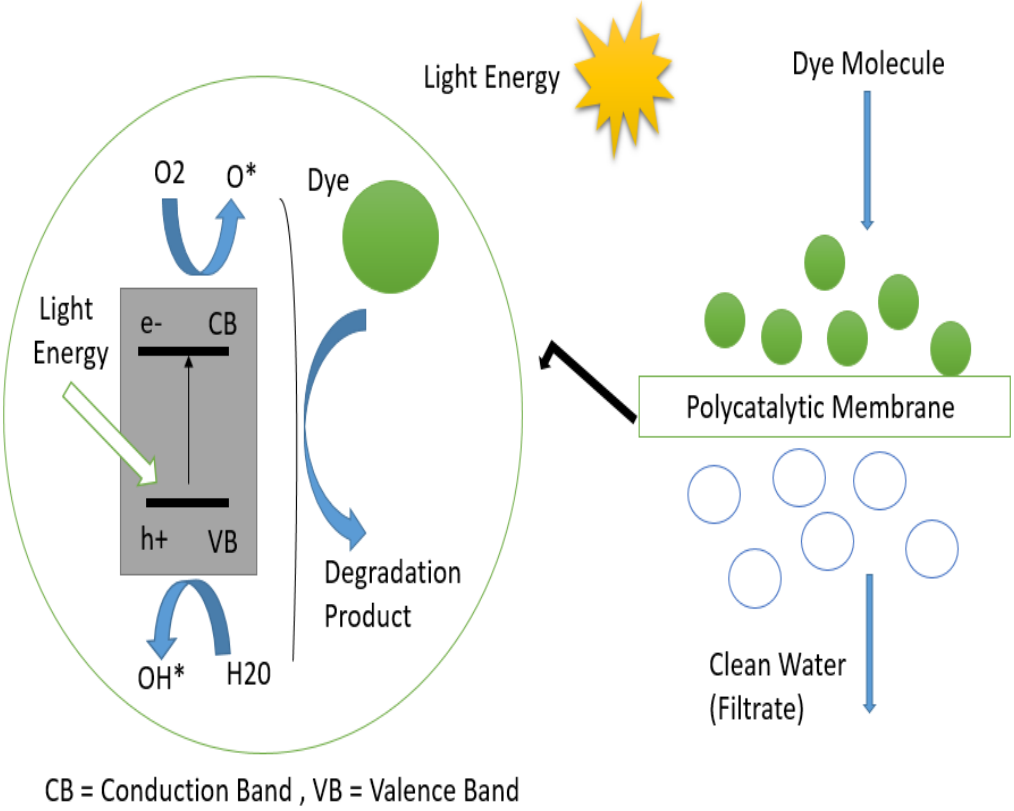

Fig 4: Diagrammatic representation of the role of photocatalytic membranes in dye removal

Table 1. Comparison of modified photocatalyst PVDF membranes' dye decomposition capabilities.

Photocatalyst-modified PVDF membrane	Dye	Source of Light	Decomposition time (min)	Removal Efficiency (%)	References
Polyaniline/TiO2/PVDF	Methyl orange	UV Light	420	90	[35]
C–ZnO/PVDF	Methylene Blue	250 W light Source	4.5h	95.02	[36]
Carbon sphere-TiO2/PVDF	Methyl Orange	Xenon Lamp	100	93	[37]
Graphene/ZnO/PVDF	Methylene blue	300 W Xenon Lamp	100	86.84	[38]
Polyvinyl alcohol-TiO2/PVDF	Reactive blue, Methyl orange, Rhodamine B	UV-C lamp	150	44.4, 47.8, 45.8	[39]

In this study, peroxymonosulfate (1.0 g L^{-1}) was employed as an oxidant. For methylene blue, rhodamine B and orange II, the ZIF-67/PVDF membrane's catalyzed purification efficiency was 98.2%, 90.5% and 97.3% respectively (time=30min). Using electron spin resonance spectroscopy, the synthesis of sulphate radical with the help of this catalytic membrane was proved [40].

3.3.2. Ozonation

Ozonation is a form of water purification where ozone (O3) may break down organic contaminants [41]. Indeed, PVDF membranes may be employed for dye ozonation since they are not harmed by ozone. PVDF membranes should be kept fully dry, especially when it comes to ozone oxidation. Therefore, methyltrichlorosilane as well as trimethylchlorosilane were used to modify pure PVDF membranes [22]. Researchers used pulse inductively associated plasma (argon and oxygen) to treat the unmodified PVDF membrane prior to activation. The membrane's hydrophobicity (water contact angle = 125.3◦) was improved by treatment with plasma (oxygen plasma), kind of chlorosilane (methyltrichlorosilane) as well as timing (4 hours). In just ninety minutes, our enhanced PVDF membrane totally eliminates the dye, Direct Blue 71 [42].

3.3.3. Photocatalysis

Since photocatalytic technique (oxidation of organics) does not produce harmful byproducts, it is regarded as an environmentally friendly method of eliminating organic contaminants [43]. However, there is an important problem with the photocatalyst's simple recovery from the water that has been treated. This problem was prevented by merging photocatalysts with membrane technology that is, coating or blending relevant photocatalysts like semiconductor nanoparticles with the PVDF membrane. Figure 4 shows the wider photocatalytic dye decomposition pathway. In addition to increasing the membrane's removal effectiveness, the photocatalyst prevents it from fouling. For dye treatment, various ultraviolet (UV) and visible light sensitive photocatalytic PVDF membranes have explored. For instance, UV light-active Remazol turquoise blue was eliminated by a TiO_2/PVDF photocatalytic membrane. TiO2/PVDF membrane reached around 99% of remazol turquoise blue removal accuracy. This study assessed the impact of photocatalyst content in the PVDF membrane, H_2O_2, starting pH, feed temperature, as well as the concentration of dye on the photocatalytic process (Table 1) [44].

4. Nanomaterials' function in PVDF membrane applications:

In comparison with PVDF membranes, nanomaterial-blended or coated PVDF membranes show improved adsorption capacity, flux rate, stain resistance, self-cleaning characteristic, stability, catalytic activity, and selectivity. Below is a discussion of the main reasons behind these enhanced functions.

a) Surface roughness

The degree of roughness of a membrane used in the procedure of filtration should be low. This is because contaminants, such as dyes, may stack up on a rough layer and become challenging to remove them by washing with water. As a result, a membrane will eventually exhibit a decrease in flux, rejection rate, and durability. The fouling process is also triggered by high surface roughness. In order to lessen the surface roughness, researchers incorporated nanomaterials with PVDF. For instance, surface roughness in PVDF membranes has been reduced by adding small amounts of chitosan and multiwalled carbon nanotubes [45].

b) Tensile power

For a successful membrane filter, tensile strength is a crucial characteristic. Nanoparticles may contribute to the PVDF membrane's increased strength. A report was provided on the impact of tensile strength by the incorporation of ZIF-8, hydrogen boron nitride nanoparticles, and molybdenum disulphide (MoS2) in silver phosphate (Ag3PO4) improved PVDF membranes. The strength of the membranes raises with the type of nanoparticles. ZIF-8/Ag3PO4/PVDF, MoS2/Ag3PO4/PVDF, and hydrogen boron nitride/Ag3PO4/PVDF membranes found to have tensile strengths of 15.87, 14.53, and 13.67 MPa, respectively [46].

c) Hydrophilicity

Due to its hydrophobic nature, the PVDF membrane typically exhibits lower wettability. However, in order to improve filtration, the membrane needs to be enough hydrophilic to engage with the water flow. Therefore, researchers tried to give PVDF a hydrophilic quality by adding suitable nanomaterials. Contact angle measurements can be used to evaluate a membrane's wettability or hydrophilicity [47]. It was found that introducing a mixture of polyvinyl pyrrolidone and carbon nanotubes could improve the hydrophilic property (contact angle = 20◦) of PVDF membrane after conducting a water contact angle study. Since carbon nanotubes are hydrophobic, polyvinyl pyrrolidone was the only substance found to increase the hydrophilicity of this membrane [48].

d) Photocatalytic activity

Due to its organic nature, natural PVDF membranes are unable to exhibit photocatalytic activity. However, by incorporating semi-conducting nanoparticles, it may achieve photocatalytic activity. Lately, MoS2, ZIF-8 and hydrogen boron nitride were individually linked to a PVDF membrane modified with Ag_3PO_4 [46].

5. Conclusions

PVDF membranes are essential methods of treating wastewater. These membranes are modified using a variety of materials, including as metal oxides, carbon materials and MOFs, in addition to functionalized surface nanomaterials and composites. Surface coating and blending strategies are used in production of modified membranes. These nanoparticles serve the combined purposes of catalytic breakdown and dye separation. For the treatment of different dyes, modified membranes are used in adsorption, filtration, separation, and catalytic treatments. To improve membrane performance, these materials' kind, concentration, and composition are crucial. Surface permeability, hydrophilicity, and anti-fouling ability are all enhanced by their integration. In addition to the separation, modified membranes can use catalytic processes like photocatalysis and ozonation to break down harmful dyes.

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