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Methylene Blue Dye Wastewater Treatment Based On Tertiary Stage of Industrial Wastewater Treatment Process: A Review

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Abstract: Methylene blue (MB) is frequently used in the textile, rubber, plastics, leather, pharmaceutical, cosmetic, and food industries. As a colouring and staining agent, MB is reported as one of the most studied dyes involving degradation and photocatalytic activity. Unfortunately, the discharge of waste from the mentioned industries contains residues of dyes, whereby the presence of a very low concentration of dyes can be highly visible. Discharging the waste without suitable treatment can cause many complications for the environment and human health. Due to that reason, the control of dye wastewater has become the most challenging task. Therefore, this review article will be focused on the MB treatment process, especially membrane filtration technology involving a pressure-driven membrane process, which is reported to be the most efficient method to treat MB dye wastewater.

Keywords: Methylene blue, wastewater treatment, adsorption, advance oxidation process, membrane technology

1. Introduction

In the current worldwide situation, most of the available freshwater within the earth's crust is used for industrial, agricultural, and domestic purposes, which leads to the pollution of water with several geogenic and synthetic materials such as fertilizers, heavy metals, pesticides, and dyes [1], [2]. Among these contaminations, dye water pollution has the most impact on nature since dyes can change the overall natural appearance of water even at a minimum dose of concentration. Dyes are well-known as coloured substances that are used in colouring substrates like cloth, paper, and other materials in which the colour cannot be altered by washing, heat, light, or other factors [3]. Generally, dyes are dissolved in aqueous solutions to improve the fastness of the dye on the substrate. According to the Ecological and Toxicological Association of Dyes and Organic Pigments (ETAD), dyes are defined as intensely coloured or fluorescent organic materials that impart colour to a substrate by selective absorption of light [4]. Dyes are soluble and undergo an application process that temporarily terminates any crystal structure by absorption, solution, mechanical retention, or by covalent or ionic chemical bonds [5]. Some of the dyes' characteristics are that they are soluble, smaller in size, have low resistivity, are compatible with all substrates, and have a short life span [4]. Basically, dyes are used in food to make it look and taste better and to make the final product more appealing.

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There are many methods to classify dyes. Dyes can be classified according to many parameters in the literature, including chemical structure, application, color, fastness, synthesis route, manufacturer, and date invented [4], [6], [7]. However, due to the complexities of the nomenclature based on the chemical structure and other parameters, the classification based on the materials' sources is considered simpler and more advantageous. Common classification of dyestuff is usually based on the source from which it is made. Most original dyes come from organic molecules and are very complex in nature. Dyes based on the sources of materials can be divided into two types, which are natural dyes and synthetic dyes. Natural dye colourant is usually obtained from plants and animals. For example, various plants such as henna, lichens, bugloss *(Alkanna tinctoria)* asafetida, saffron, and madder can be used to make red dyes, and for animals, insects are the most popular animal resource to produce dyes [8], [9]. An insect called Cochineal *(Dactylopius coccus)* found in Central and North America can produce the crimson-colored carmine dye [10]. Other examples of natural dyes are shown in Table 1.

Table 1 - Natural dyes based on their species and specified color

Sources	Species	Colors	Ref.
Plants	Madder *(Rubia Tinctoria)*	Red	[11]
	Mignonette *(Reseda Luteola)*	Yellow	[12]
	True indigo *(Indigofera Tinctoria)*	Blue	[13]
	Orcein/Archil *(Roccella Tinctoria)*	Purple	[14]
	Cutch *(Acacia Catechu)* + (iron and copper mordant)	Brown	[15]
Animals	Muricid *(Muricidae)*	Purple	[16]
	Murex *(Bolinus Brandaris)*	Red	[17]
	Kermes *(Kermes Vermilio or Kermococcus Vermilio)*	Red	[18]
	Cochineal *(Dactylopius Coccus)*	Red	[10]
	Cow urine	Yellow	[19]
	Lac *(Kerria Lacca)*	Red	[20]
	Octopus/cuttlefish	Brown/black	[21]

Synthetic dyes are manufactured from inorganic and organic molecules. Unlike a group of natural dyes, synthetic dyes can be mass-produced consistently [22]. These dyes are made from synthetic resources like petroleum by-products, chemicals, minerals, or earth [23]. Mauvenie was the first human-made organic aniline dye and was found coincidentally as a result of a failed attempt of the quinine synthesis by William Henry Perkin in 1956 [24]. Since then, various dyes have been produced. These dyes are stable in common reduction and oxidation processes but difficult to remove from industrial waste. Most industries use large quantities of dyes and produce large scales of dye waste, which is eventually released directly into the ecosystem and causes serious problems due to the poisonous and disagreeable nature of dyes [25], [26]. Synthetic dyes have been divided into many other classes, such as direct dyes, Sulphur dyes, reactive dyes, vat dyes, mordant dyes, disperse dyes, azoic dyes, acid dyes and basic dyes. A summary of synthetic dyes is shown in Table 2.

The most popular dye, MB, is a thiazine cationic dye in nature and is frequently used by the textile, rubber, plastics, leather, pharmaceutical, cosmetic, and food industries [27]. MB can be classified as a basic dye and is known as a heterocyclic compound with the chemical formula $C_{16}H_{18}N_3SCI$ [28]. It was synthesized in 1876 by Heinrich Caro of Badische Anilin und Soda Fabrik (BASF) as an aniline-based synthetic dye for the discoloration of cotton used in the textile industry [29]. Its utility to colour and inactive microbial species was realized after the synthesized process, and it was later discovered in 1932 to be an antidote to carbon monoxide and cyanide poisoning [29], [30]. MB is reported as one of the most thoroughly studied dyes in many works, especially in degradation of dyes and photocatalytic activity. As a colouring and staining agent, MB has wide and extensive applications in the pharmaceutical and textile industries. However, waste discharge from the involved industries contains residues of dyes, whereby the presence of a very low concentration of dyes in waste can be highly visible. Discharging dye wastewater without proper treatment can result in a variety of complications, including chemical oxygen demand (COD) by the water body, increased toxicity, and adverse effects on human health [27]. For human health, general problems related to this dye are irritation of the skin with itching and redness, high blood pressure, irritation of mouth, throat, stomach and esophagus, nauseam gastrointestinal pain, dizziness, diarrhea, headache and fever [31]. Thus, the control of dye wastewater has become the most strenuous task. The lack of high-quality clean water supplies for human use and the concern for human health have driven the enactment of in-process water-saving techniques and advanced treatment methods for the recycling of wastewater

Table 2 - A summary of synthetic dye classes [32]

Dye	Applying method	Solubility and nature	Applications	Environmental issue
Direct dyes	Dye bath that is neutral or slightly alkaline, or that is nearly boiling with the addition of NaCl, Na_2SO_4, and Na_2CO_3	Water soluble (Anionic)	Wool, paper, cotton, silk, nylon, pH indicators, biological stains	C.I. Direct Orange 62 is toxic dye.
Sulphur dyes	Heating the substance where an organic compound is dissolved and reacts with the sulphide source	Insoluble in water (Anionic)	Cotton, cellulose,	Low biodegradable dispersant
Reactive dyes	Reactive group that reacts with the fibre substrate	Soluble (Anionic)	Cellulosic fiber, fabric, cotton	Presents of heavy metals as impurities from the production process
Vat dyes	The dye is applied to the fabric in stable dispersion, frequently in micro-dispersion	Insoluble in water (Non-ionic)	Cotton, cellulose, blended fibers	Presents of heavy metals as impurities from the production process
Mordant dyes	Pre-mordanting (onchrome) Meta-mordanting (metachrome) Post-mordanting (afterchrome)	Soluble (Amphoteric)	Wool, silk (restricted)	Human toxicity during handling and the pollutant of the effluent by toxic chromium salts used after dyeing.
Disperse dyes	Successive treatments of two chemical component solutions that react with the surface of the fiber's dye	Insoluble in water (Cationic)	Synthetic fibers, cellulose acetate, vilene, velvets, nylon, PVC, polyesters	Conventional dispersant (formaldehyde condensation compounds, ligno-sulphonates, etc.) are poorly biodegradable.
Azoic dyes	Produced by in situ on the textiles in which the dye reacts with coupling compound (naphthol)	Insoluble in water (Cationic and anionic)	Printing inks, pigment, cotton, cellulosic fibers, nylon	Used of amines as developing agents like p-nitroaniline, p-chloroaniline and β-naphthylamine are on the priority list of harmful chemicals and forbidden
Acid dyes	Neutral to acid dye baths	Water soluble (Anionic)	Nylon, silk, wool, leather, synthetic fiber, modified acrylic fibers	ETAD has designated C.I. Acid Orange 150 and 165 as toxic substances. CI Acid Violet 17 might have an allergic reaction
Basic dyes	Neutral to dye baths except for fibers where acetic acid is needed to be added in the dye baths	Water soluble (Cationic)	Acrylic fibers, wool, silk, paper	Causes high aquatic toxicity but with properly handling, the dye fixation can be closed to 100%. C.I. Basic Blue 3,7, 81, Basic Red 12, Basic Violet Basic Yellow 21 are classified toxic by ETAD.

2. Treatment Process for MB Dye Wastewater

Nowadays, people have started to show more interest in preserving natural clean water and treating wastewater for the sake of human health and preserving environmental conditions caused by dye wastewater and other contaminants. Generally, the industrial wastewater treatment process involves four stages, which are preliminary treatment, primary treatment, secondary treatment, and tertiary treatment [33]. Preliminary treatment includes neutralization and equalization; at this stage, wastewater is treated and its characteristics such as pollution load, temperature, and pH are confirmed [34]. Next, primary treatment deals with the omission of solids that froth by skimming and the elimination of settleable organic and inorganic materials by flotation and sedimentation [35]. During secondary treatment, colloidal and dissolved organic matter are subjected to microbial degradation, which stabilizes the waste constituents [36]. Last but not least, tertiary treatment is applied to remove particular pollutants, such as organic colour compounds, by adsorption, treatment with ozone (O_3) or other oxidizing agent and also dissolved solids by membrane filtration techniques [34]. A summary of the classification of industrial wastewater treatment processes is shown in Fig. 1. The treatment of MB dye wastewater will be focused on tertiary treatment.

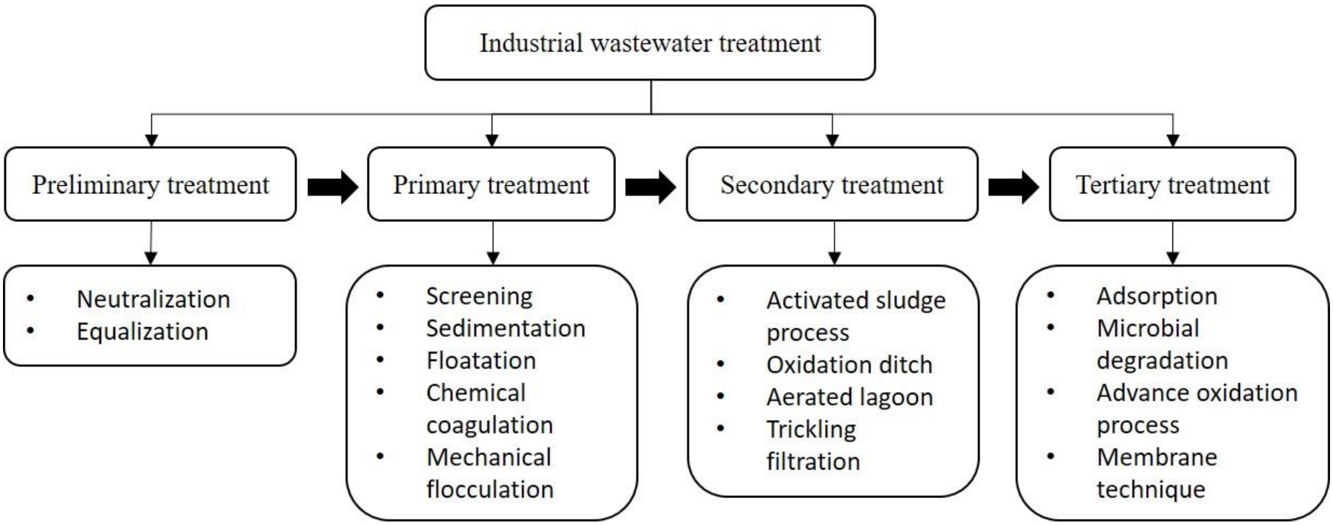

Fig. 1 - Summarize on classification of industrial wastewater treatment process [33]

2.1 Adsorption

Adsorption is a promising technique that offers numerous benefits such as ease of operation, simplicity, broad applicability, and admirable ability to remove a wide range of contaminants [35]. Generally, the adsorption process involves atoms or molecules accumulating in a liquid solution or gas on the surface of a liquid or solid material, thus forming a molecular or atomic thin layer. Adsorbent is used as the material on which the adsorption process occurs, and adsorbate is the material that sits on the surface after being absorbed by the adsorbent [37]. From the data taken from *Santoso et. al* in his review on recent advances in carbon-based adsorbent for methylene blue removal from wastewater, the trend of publication shows the adsorption is among the most investigated methods for the removal of MB [38].

Saafie *et al.*, for example, investigated the use of commercial activated carbon (AC) with enhanced adsorption capacity via alkali-acid treatment (deionized water (AC-DI), potassium hydroxide (AC-KOH), and nitric acid (AC-HNO_3)).The result reveals that AC-DI had excellent adsorption performance, removing 99 % of the MB within 8 hours [39]. Also, the enhanced adsorption of MB dye ions on the AC modified by different anionic and cationic surfactants (sodium lauryl sulphate (SLS), sodium dodecyl sulfonate (SDS), and hexadecyl trimethyl ammonium bromide (CTAB)) in liquid solution was researched by Kuang *et. al*. In the research, the results showed that the AC modified by the anionic surfactants (SLS and SDS) was effective for the adsorption of MB dye [40]. However, the adsorption method has its own disadvantages, such as the high cost of the adsorbents, the difficulty of separating the adsorbent from the dye, and the low surface area capability. Also, the efficiency of the adsorption process depends on the nature of the adsorbents. In some studies, the absorbent itself must be changed to improve adsorption quality and capacity. Some adsorbent modifications are very expensive and involve many chemicals and substances throughout the adsorbent's synthesis [41]. The schematic illustration of adsorption is present in Fig. 2.

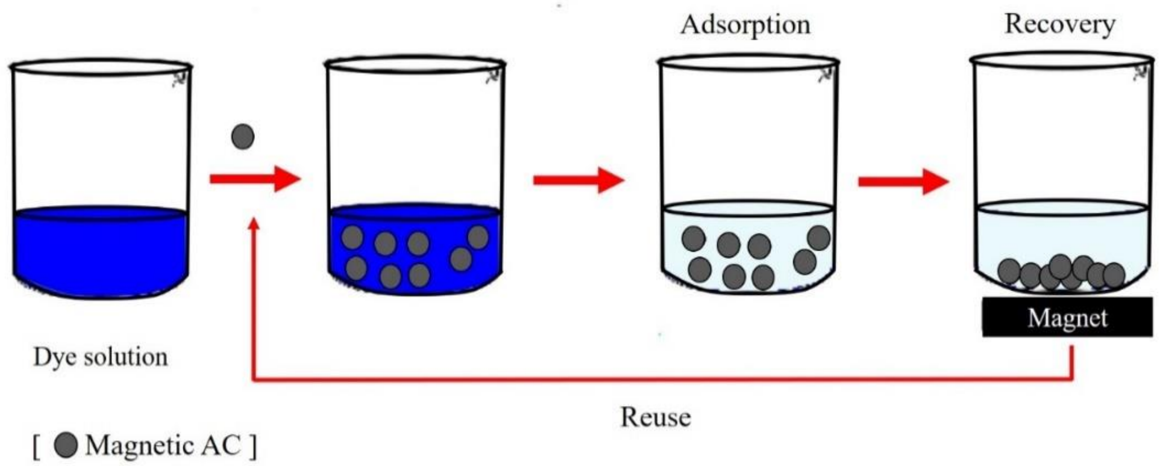

Fig. 2 - Schematic illustration of adsorption by magnetic activated carbon and its recovery [41]

2.2 Microbial Degradation

Microbial degradation is an alternative method to degrade dyes. It is the most reasonable process compared to other methods with the following features: eco-friendly, cost-effective, non-hazardous end product, and it results in complete mineralization. Bacteria, algae, fungi, and yeast are reported as microorganisms that can be used for the degradation of dyes [3]. For bacterial biodegradation, many of the bacterial strains are in aerobic and anaerobic conditions [42]. Eslami *et. al* evaluated the biodegradation of MB from aqueous solutions using bacteria *(Psedomonas aeruginosa)* collected and isolated from contaminated soil. From the result, the degradation efficiency of bacteria was attained from 82.25 to 97.82 % with the MB concentration from 20 to 200 mg/l, respectively, but was reduced to 43.08 % when the MB concentration was increased to 1000 mg/l [43]. Algae, on the other hand, are widely distributed photosynthetic microbes in a variety of environments. Algae like *Chlorella* and *Oscillatoria* can break down harmful aromatic amines into straightforward metabolic byproducts like water [44]. In their study on the remediation of MB from algae, Lebron *et al.* reported 98.20 % and 94.19 % removal efficiencies using *Chlorella Pyrenoidosa* and *Spirulina Maxima*, respectively [45].

For fungi, the microorganism itself can grow in almost any environment because it can quickly adapt and use different carbon and nitrogen sources for its metabolism. Fungi have the ability to produce large amounts of extracellular and intracellular enzymes that have the ability to break down a variety of contaminants, including organic waste and dye effluents, including MB [33]. For example, Mass *et. al* have reported a reduction of 80 % using MB was observed using different species of fungi (*Pleurotus* sp.) [46]. In other reports, a number of yeasts have been informed of their ability to degrade dyes through metabolic activity [47]. Contreras *et al.* in their article on bio-removal of MB from aqueous solution by *Galactomyces geotrichum* KL20A yeast reported that the removal percentages of MB using the yeast microorganism was greater than 70% under the best operating conditions [48]. Despite numerous studies involving various types of microorganisms for dye degradation, this biological method still has its own drawbacks, including the need for a large land area, less biodegradability of the dyes, a high reliance on reaction conditions, a longer time for decolorization, and inflexibility in design and operation [49].

2.3 Advance Oxidation Processes (AOPs)

In wastewater treatment, AOPs are considered one of the most effective methods for removing refractory, low-biodegradability, and chemical pollutants [50]. A highly reactive oxidant species called the hydroxyl radical (*OH), which is formed in-situ, is essential to the mechanisms of AOP. The hydroxyl radical functions as a potent oxidant to oxidize (destroy) substances that are resistant to oxidation by common oxidants like ozone, gaseous oxygen, and chlorine, and mineralize them into carbon dioxide (CO_2), water (H_2O), and inorganic salts [51]. AOPs offer several advantages, like being eco-friendly in nature, low cost, complete degradation, reducing pollutant load and improving the quality of treated water [52]. Different processes of AOP have been studied and developed for pollutant degradation (organic and inorganic compounds) existing in wastewater. Some of the processes are Fenton chemistry, electrochemical oxidation, hybrid AOPs, and photocatalytic.

The Fenton process involves hydrogen peroxide (H_2O_2) as an oxidant and ferrous ions (Fe^{2+}) as a catalyst. The catalytic decomposition of H_2O_2 in Fe^{2+} involves a complex reaction sequence that eventually generates a highly reactive hydroxyl radical and ultimately leads to the organic removal of the wastewater [53], [54]. Cotto-Maldonado *et. al* in their study on the Fenton process for MB degradation using Iron oxide nanowires (Fe_2O_3NWs) and commercial catalyst ($FeCl_2$) reported that both catalysts were able to degrade the MB and could be used to remove contaminants from the water [55]. Researchers confirm that the Fenton process comprises more than 20 chemical reactions and its generally accepted core reaction, which is shown in Equation 1 [53], [56].

$$Fe^{2+} + H_2O_2 + H^+ = Fe^{3+} + H_2O + {}^*OH \tag{1}$$

The efficiency of organic pollutant degradation in this process depends on operation parameters such as Fenton reagent concentration, initial organic pollutant concentration, and wastewater pH. The pH of wastewater is significant because iron species catalyst deactivation prevents the effective treatment of organic pollutants in wastewater at both high and low pH levels [53]. The reaction mechanism for the Fenton process is presented in Fig. 3.

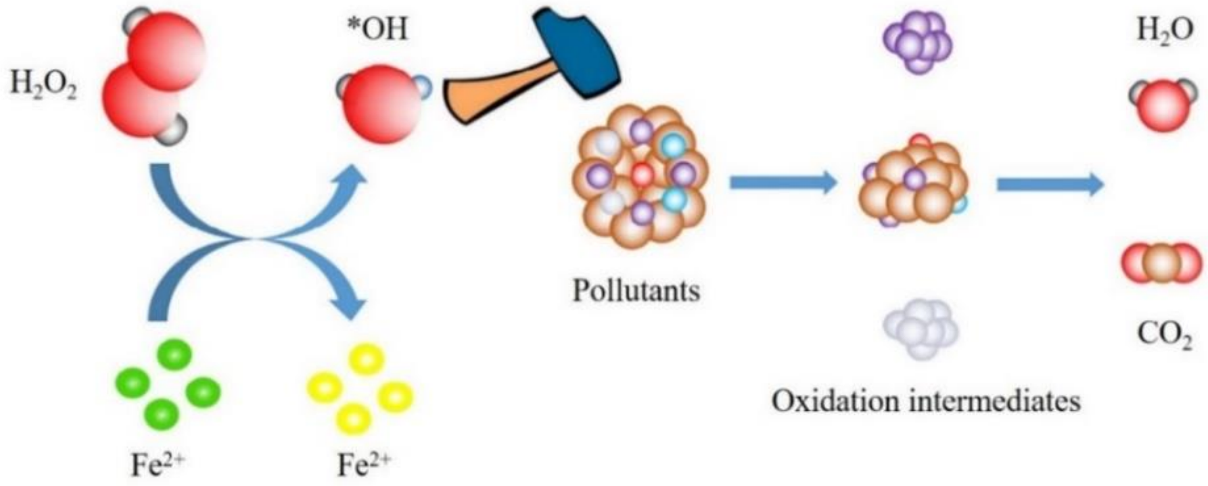

Fig. 3 - Reaction mechanism for the Fenton process [53]

Over the years, newly developed electrochemical advanced oxidation processes (EAOPs) have gained much interest and have been developed by many researchers. EAOPs are broadly used in distillery effluent to treat the present contaminates [57]. Generally, EAOPs use electrons as the main reagent and electrolytes as supporting materials. In the electrochemical process, the pollutants or contaminants are eliminated through either direct or indirect oxidation [58]. In direct oxidation, contaminants adhere to the anode surface, where they are absorbed and later destroyed by the anodic electron transfer reaction. Meanwhile, cathodic or anodic electrochemical processes are used to produce strong oxidants, such as chlorine or hydrogen peroxide, for indirect oxidation. The generated oxidant's oxidation reaction will then destroy the organic contaminants in the bulk solution [50]. Teng *et. al* investigated the comparison between electrochemical processes, electro-fenton processes, and electro-coagulation processes. The outcomes demonstrate that electrochemical processes, in which an iron rod serves as an anode, graphite serves as a cathode, and fly ash red mud particles serve as electrode particles, had more advantages in the degradation of MB. Electrochemical oxidation processes have good performance in the pH range of 3-11 and the electric energy consumption of, electro-fenton processes, electro-coagulation, and electrochemical processes is 81.51, 36.55 and 21.35 $kWhm^{-3}$, respectively, suggesting electrochemical oxidation is more cost-effective [59]. In the meantime, bio-electrochemical systems (BES) are the latest technology in EAOPs, where the combination of bio-electrocatalytic reactions and extracellular electron transfer is able to drive several procedures such as producing electricity from wastewater, synthesizing chemicals, desalinating seawater, and removing pollutants [60].

Other AOPs for degrading dyes are combined oxidation processes or hybrid AOPs. Hybrid AOPs provide economical and flexible options and have been examined by many researchers to increase the efficiency of wastewater treatment [50]. AOPs as individual techniques have their own drawbacks that can be covered by combining other AOPs to create hybrid AOPs, thus giving a better result. For example, photocatalytic processes have problems like slow mass transfer and solids that stick to the catalyst. However, by incorporating an additional AOP, such as ultrasonic irradiation, not only is the production of hydroxyl radicals increased, but the acoustic streaming and turbulence produced by ultrasonic irradiation, as well as mass transfer resistance, are also eliminated [51]. Kumar *et. al*, degraded MB in an aqueous solution using hydrodynamic cavitation combined with H_2O_2 and Bi-doped TiO_2 photocatalyst [61]. The result shows that the degradation of MB was at 94.64 % over 60 min, where the hybrid AOPs-hydrodynamic cavitation/H_2O_2 showed a higher synergetic effect compared to the hybrid AOPs-hydrodynamic cavitation/Bi-doped TiO_2 photocatalytic.

Photocatalytic or photochemical degradation has become the most attractive, efficient, and low-cost AOP treatment technology for enhancing the biodegradability of hazardous and non-biodegradable pollutants like persistent organic pollutants [62]. Usually, photocatalytic degradation of organic pollutants uses semiconductor nanoparticles. According to previous research, many oxides, such as zinc (Zn), vanadium (V), chromium (Cr), tin (Sn), cerium (Ce), and titanium (Ti), can be used for photocatalytic reactions [63]. TiO2, or titania, is the most applied and popular oxide in photocatalytic degradation due to its chemical stability, environmental compatibility, and non-toxicity [64]. Furthermore, titania is also known as a versatile and proficient AOP due to its ability to easily produce reactive oxidative species like hydroxyl radical and super oxide radical, which are needed for photocatalytic degradation of dyes and organic pollutants [65]. For photocatalytic reactions, UV radiation is needed for the activation of the photocatalyst, and, for that reason, titania photocatalyst has a low photocatalytic reaction under visible light. However, the reaction can be improved with certain synthesis methods or by doping with other metals [66], [67]. Cabir *et. al*, synthesized new copper phthalocyanine modified TiO_2 Nano powders (CuPc-TiO_2) via wet chemical impregnation with the purpose of improving the photocatalytic activity that can be used under visible light. The results showed that the CuPc-TiO_2 photocatalyst produced excellent activity, with a complete 100% photocatalytic degradation of MB when exposed to visible light (150 W) [68]. Many factors can influence photocatalytic reaction efficiency, including irradiation source, light power, cation nature, material phases, and particle surface area [69]. However, all factors can be improved with some modification to the material itself.

2.4 Membrane Filtration Technique

Membrane filtration is widely used for wastewater treatment. Using membrane filtration, wastewater permeates through a porous membrane. Any unwanted particles will be trapped while the filtered solution passes through the membrane, depending on the pore size of the fabricated membrane [70]. Membrane technology is the most applied method for removing dye from wastewater due to its efficient removal performance, space-saving procedure, and low operating cost [71]. Membrane processes can be divided by their driving force. A pressure-driven process is commonly used to separate dye wastewater. There are four classes of pressure-driven processes, which are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [72], [73]. MF possess larger pore size around 0.1 to 10 µm thus the membrane is not suitable to be used for dye wastewater treatment. In fact, MF is not commonly used as the main treatment for dye removal but as the pre-treatment unit in a whole filtered system [74]. However, there is also some reported work on the modified MF for the removal of dye. For example, Daraei *et. al* reported on the removal of MB dye using a commercial PVDF membrane modified with organoclay/chitosan nanocomposite to produce a novel thin film composite (TFC) membrane. The results show that the water flux is decreased for all modified MF membranes but is good at removing MB dye. The work also reported that the chemical structure (functional groups) of prepared TFC membranes offered adsorption as a dominant dye removal mechanism [75]. It shows that the modified MF membrane can act as a double treatment medium as separation and adsorption are provided by the adsorbent organoclay/chitosan, thus being able to treat the MB dye wastewater.

Another membrane separation technique is the UF membrane. The UF technique needs lower pressure than RO and NF, thus making it more efficient filtration. However, the molecular weight of dyes is lower than the UF membrane MWCO [76], [77]. For that reason, the application of dye removal using UF membrane is limited and restricted. In industries, UF membranes are usually used as a pre-treatment process before NF and RO. Otherwise, similar to MF, some modification is also needed to improve the efficiency of dye removal. Ngang *et. al* fabricated the modified PVDF-TiO_2 UF MMM in order to evaluate the dye absorption and UV-cleaning properties of MB. The result shows excellent removal of MB with almost 99% efficiency and outstanding UV-cleaning properties, as proved by the 100% flux recovery ratios (FRRs), thus showing that the modified photocatalytic membrane was able to absorb and degrade MB solution [78].

Compared to other separation processes, NF membrane has been widely used for dye effluent treatment and recycling since the process is the most practical method that can effectively eliminate organic compounds with a low molecular weight of around 200 to 1000 g/mol at certain operational pressure [79]. With a membrane pore size ranging from 0.5 to 2 nm, the NF process has major drawbacks, including the fact that the membrane needs to be operated at higher pressure (high operation cost) and the reduction of filtrate water due to membrane fouling during the filtration process. Fouling refers to the unwanted particles or solutes that stick to the membrane surface layer and cause membrane pore blockage. This reduces the flux rate during NF filtration [80]. Because of the high concentration of contaminants, it is not recommended that industrial dye wastewater be treated directly by NF membrane. The use of pre-treatment on the dye effluents or the membrane is needed to avoid membrane fouling and degradation [71]. From Fig. 4, Abdelhamid *et. al* produced a composite-NF membrane using PSf and functionalized graphene oxide (f-GO) for the removal of MB from aqueous media. The modified membrane gave excellent efficiency in MB rejection with 99 % dye removal compared to the control membrane [81].

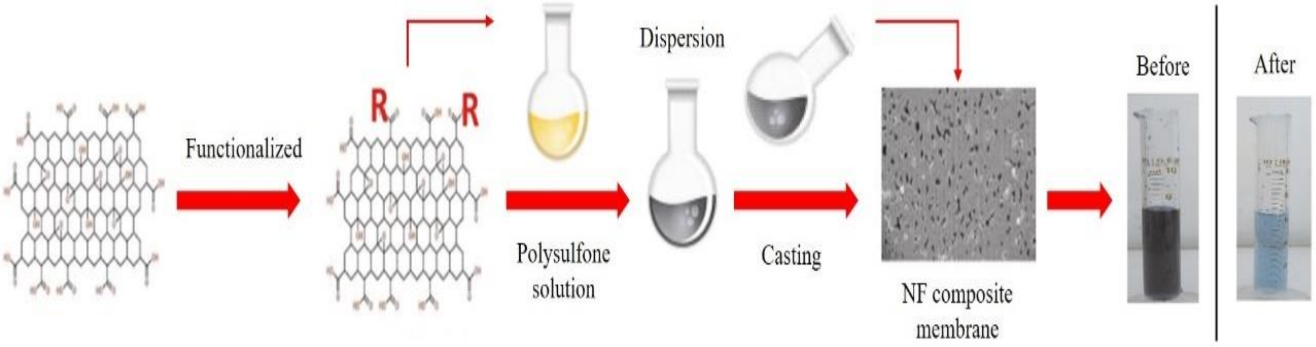

Fig. 4 - Schematic diagram of the fabricated NF membrane for MB removal [81]

Meanwhile, RO filtration, also known as hyperfiltration, has a membrane pore size in the range of 0.5 nm and a high operating pressure of between 7 and 100 bars. RO processes can effectively remove hardness, bacteria, color, and organic contaminant [106]. The implementation of RO membranes is commonly used to treat wastewater with a high concentration of salt. With a smaller membrane pore size, the probability of the membrane being affected by fouling is pretty high when it involves industrial dye wastewater. Thus, to reduce membrane fouling and get an acceptable permeation flux, the operating pressure should be greater than 20 bar, which eventually causes high operation and maintenance cost [71]. Because of this disadvantage, there are not many projects that involve dye removal from wastewater using an RO membrane. The mechanism and characterization of each type of membrane process will be discussed in detail in the coming section. Table 3 summarizes the findings from previous studies involving MF, UF, NF, and RO membranes in treating MB dye wastewater.

As a conclusion, among the four types of treatments, membrane filtration technology shows some promising advantages for treating the MB dye wastewater. The removal efficiency of dye can be improved significantly by making some changes to the standard membrane. The presence of additives inside the membrane structure enhances the properties and functionality of the manufactured membrane. Some additives have their own properties, such as photocatalytic and adsorption. The properties will be included in the membrane by incorporating the unique additive, resulting in a membrane with multiple treatment properties of filtration, adsorption, and photocatalytic reaction. Such a membrane helps in reducing costs by a single treatment, saving operational time, and improving the dye removal efficiency.

Table 3 - A summary of studies involving pressure-driven membranes in treating MB dye wastewater

Type of membrane	Membrane details	Type of MB removal process	Test condition	Efficiency	Ref
MF	Polyethersulfone-microfiltration/Graphene oxide (PES-MF/GO)	Filtration	Dye concentration = 7.5 mg/L Feed flow rate/pressure = 0.02 bar	92.1%	[82]
	Nitrated cellulose acetate (NCA) microfiltration	Filtration adsorption	Dye concentration = 0.5 mg/L & 1 mg/L Feed flow rate/pressure = 50 kPa	<90% & <80%	[83]
	Magnesium silicate fiber membrane (MgSiFM)	Filtration adsorption	Dye concentration = 10 mg/L Feed flow rate/pressure = 2 mL/min	99%	[84]
	Ceramic nanoclay/zeolite	Filtration	Dye concentration = 35.76 mg/L Feed flow rate/pressure = 1.5 bar	90.23%	[85]
	Hydrophilic	Immersion	Dye concentration =	70%, 100%,	[86]

	titania/polyethersulfone (TiO_2/PES), Hydrophilic titania/polyvinylidene fluoride (TiO_2/PVDF), Hydrophobic titania/polyvinylidene fluoride (TiO_2/PVDF)	adsorption	10 µmol/L Feed flow rate/pressure = NA	& 15%	
UF	Micellar enhanced ultrafiltration (MEUF) organic regenerated cellulose membrane	Filtration	Dye concentration = 1 mM Feed flow rate/pressure = 1.4 bar	95-99%	[87]
	Polyethersulfone (PES)	Immersion adsorption	Dye concentration = 4 mg/L Feed flow rate/pressure = NA	98%	[88]
	Cellulose-based cross-linked polymer dicarboxymethyl cellulose (DCMC)	Immersion filtration	Dye concentration = 4 mg/L Feed flow rate/pressure = 5 bar	<85%	[89]
	Micellar enhanced ultrafiltration (MEUF) regenerated cellulose	Filtration	Dye concentration = 4-8 µM Feed flow rate/pressure = 1.5 bar	75% to 99%	[90]
	Polyethersulfone/Barium chloride (PES/$BaCl_2$)	Filtration	Dye concentration = 100 ppm Feed flow rate/pressure = 4 atm	85% to 91%	[91]
	Polyvinylidene fluoride/titanium dioxide (PVDF/TiO_2)	Filtration adsorption photocatalyst oxidation	Dye concentration = 10 mg/L Feed flow rate/pressure = 0.5 bar	<99%	[78]
	Materials institute Lavoiserr (MIL)-53(Fe)/polyvinylidene fluoride (PVDF) composite membrane	Filtration adsorption photocatalyst oxidation	Dye concentration = 20 mg/L Feed flow rate/pressure = 0.5 bar	<75%	[92]
	Cellulose acetate/*pristine*-multiwalled carbon nanotubes (CA/p-(MWCNTs))	Filtration adsorption	Dye concentration = 10 mg/L Feed flow rate/pressure = 100 kPa	>40%	[93]
	Polysulfone/N-titanium dioxide/graphene oxide (PSf/N-TiO_2/GO)	Filtration and photocatalyst oxidation	Dye concentration = 50 mg/L Feed flow rate/pressure = 0.04 MPa	-	[94]
	Polyethersulfone/cellulose nanofibrils/polyvinylpyrrolidone (PES/CNF/PVP)	Filtration	Dye concentration = 25 ppm Feed flow rate/pressure = 0.6-1.4 bar	69.17%	[95]
	Polysulfone/molecular imprinted polymer-titanium dioxide (PSf/MIP-TiO_2)	Filtration and photocatalyst oxidation	Dye concentration = 5 ppm Feed flow	<80%	[96]

			rate/pressure = 2 bar		
	Polysulfone/polydopamine (PSf/PDA)	Filtration	Dye concentration = 0.5 g/L Feed flow rate/pressure = 10 psi	-	[97]
	Polyethersulfone/petrol soot nanoparticles (PES/PSN)	Filtration	Dye concentration = 30 mg/L Feed flow rate/pressure = 100 kPa	42-96%	[98]
NF	Self-assembled positively charged NF membrane (PA6DT-C)	Filtration	Dye concentration = 0.4-10.5 mol/m^3 Feed flow rate/pressure = 200-500 kPa	98%	[99]
	Polysulfone/functionalized-graphene oxide (PSf/f-GO) composite NF (ad gmbr)	Filtration	Dye concentration = 25 ppm Feed flow rate/pressure = 4 bar	99%	[81]
	Commercial ceramic NF/Zinc oxide	Filtration and photocatalyst oxidation	Dye concentration = 500 mg/L Feed flow rate/pressure = 4 bar	94% and 33%	[100]
	Polyethersulfone/chitosan/non-ionic NF	Filtration	Dye concentration = 100 ppm Feed flow rate/pressure = 4.5 bar	<91.7%	[101]
	Covalent organic framework/graphene oxide (COF-TpPa/GO)	Filtration	Dye concentration = 10 ppm Feed flow rate/pressure = 0.1 MPa	97.05%	[102]
	Positive charged NF diallyl dimethyl ammonium (DADMAC)/polysulfone (PSf)	Filtration	Dye concentration = NA Feed flow rate/pressure = 0.5 MPa	99.4%	[103]
	Polysulfone/polyethylene glycol (PSf/PEG)	Filtration	Dye concentration = 15 ppm Feed flow rate/pressure = 400k kPa	99.8%	[104]
	Molecular layer deposition titanium dioxide (MLD TiO_2) NF	filtration	Dye concentration = 64 mg/L Feed flow rate/pressure = 0-7 bar	96%	[105]

3. Conclusion

There is an unending list of works involving the treatment of MB. However, membrane technology is reported to be the most efficient method of filtering MB. In fact, with some modifications to the membrane fabrication, additional properties can be added, and the membrane might possess multiple treatments such as adsorption and photocatalysis. This review attempted to summarize all the work that has been done involving the treatment of MB by citing all the examples from other works. Hopefully, this paper is useful in providing available knowledge about related work that can be used for further research works.

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