Membrane Bioreactor for Removal of Dyes

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Access to clean water is crucial for human health and the advancement of society. However, the decline in water quality has become a serious global issue due to human activities. The United Nations introduced 17 Sustainable Development Goals (SDGs) with the aim of creating a sustainable future for all humankind. One of the most significant of these goals is "Clean Water and Sanitation for All". However, the discharge of various contaminants into aquatic environments impedes progress towards achieving SDG6. Among industrial effluents, the textile and dye industries are considered to be major contributors to wastewater production. Dyestuffs, which are synthetic, complex aromatic compounds, and ionizing agents, are widely used as coloring agents in industries such as paper, textiles, food, dyeing, and cooking. Following the dyeing process, approximately 15% of the used dyes remain in the wastewater stream, making the colored wastewater effluent a major concern. Conventional wastewater treatment plants have difficulty in removing such chemicals, resulting in over 200,000 tons of dyes being discharged each year in the environment. The release of dyes results in water pollution with resistant compounds that are not easily broken down by natural degradation processes. Several methods have been established for treating dyes from water bodies, including physicochemical and biological approaches. Physicochemical methods, such as membrane filtration, adsorption, ion exchange, advanced oxidation processes (AOPs), and coagulation, have limitations in the removal of dyes due to high cost, inefficiency, and the potential for secondary pollution. In contrast, biological treatment methods, such as membrane bioreactors (MBRs), are cost-effective, safe, environmentally friendly, and efficient for removing dyes. Among the various biological treatment methods, MBRs are regarded as one of the most effective methods for treating wastewater. MBRs are a combination of units for biological degradation and physical filtration

Keywords: dyes ; AnMBR ; wastewater

1. Introduction

The integration of membrane separation (mostly microfiltration—MF and ultrafiltration—UF) and biological degradation processes in various conditions (aerobic, anoxic, and anaerobic) has demonstrated promising potential in treating contaminated wastewaters over the past 20 years [1]. Membrane bioreactor (MBR) systems use membrane filtration units with different pore sizes, either within biological tanks or installed separately, to effectively separate clear wastewater from suspended solids, which are kept within the wastewater treatment plant (WWTP) [2]. Membrane materials can be grouped into three main categories: polymer, ceramic, and metallic [3].

Polyvinylidene fluoride (PVDF), polystyrene (PS), polysulfone (PSF), polyetherimide (PEI), polyacrylonitrile (PAN), cellulose acetate (CA), polyethylene (PE), and polyethersulfone (PES) are commonly used in polymeric membranes [4]. The main benefit of polymeric MBR is that it is cost-effective compared to ceramic and metallic options; however, polymeric membranes have some limitations, such as excessive fouling and lower stability under challenging operating conditions such as salt [5].

Ceramic membranes have seen wide use in water treatment, gas purification, and product concentration. However, their high fabrication cost makes them a challenge to implement on a large commercial scale [6]. Ceramic membranes have certain advantages over polymeric membranes because of their inorganic matrix and specific micro- and nanostructural properties. They offer long-term stability at high temperatures, exceptional chemical stability against acids and solvents, mechanical stability under high pressure, and a long service life, making them suitable for use in harsh environments [7].

Metallic membranes can be manufactured using different methods, including electroplating, chemical vapor deposition (CVD), physical vapor deposition (PVD), electroless plating, and sintering. The resulting metallic membranes come in different shapes, such as disks and tubes, and can be customized in terms of size to meet specific needs [8]. Typically, metallic membranes are preferred for applications in harsh environments (e.g., high temperatures, high chemical concentrations).

The treatment of most dyes, such as azo dyes, is challenging through aerobic methods due to their recalcitrant and toxic nature to microorganisms. However, under anaerobic conditions, azo dyes serve as electron acceptors and can be easily broken down into aromatic amines, making anaerobic bioreactors more effective for removing color [9]. Therefore, the AnMBR has been considered in this research. Anaerobic digestion (AD) is a process where facultative anaerobic bacteria break down biodegradable organic matter in anaerobic conditions, resulting in the production of biogas. This method is more energy-efficient, degrades organic matter effectively, and produces less biosolids compared to methods that require oxygen [10].

According to Ji et al. [11], anaerobic biological treatment can eliminate emerging contaminants (ECs) from wastewater. However, conventional anaerobic digestion technology has limitations in removing hydrophilic and toxic ECs due to low biomass levels. Furthermore, the stability and functional microbial community in the conventional AD system are vulnerable to the toxicity of ECs [11]. The AnMBR system, compared to conventional anaerobic digestion process technology, has a higher biomass and a longer sludge retention time (SRT) because of the membrane module's role [12].

As described by Ji et al. [11], AnMBR technology can be categorized into three forms based on how the membrane module is incorporated into the bioreactor: side-stream AnMBR (SS-AnMBR), submerged AnMBR (S-AnMBR), and external submerged AnMBR (ES-AnMBR). SS-AnMBR involves placing the membrane module outside the bioreactor and pumping the treated wastewater at a high flow rate to the outer membrane module for separation. S-AnMBR has the membrane module submerged in the bioreactor, and the clean water is separated from the reactor through vacuum, which is commonly used in industrial wastewater treatment, such as in the textile industry. ES-AnMBR is mainly employed in the treatment of municipal sewage and has the membrane module submerged in an external chamber. Apart from that, some novel AnMBRs (such as the dynamic AnMBR, the AnDMBR) have been applied.

As mentioned above, novel AnMBRs are frequently applied in the treatment of industrial wastewater. Most reported novel AnMBRs are anaerobic electrochemical membrane bioreactors (AnEMBRs), anaerobic dynamic membrane bioreactors (AnDMBRs), anaerobic membrane distillation bioreactors (AnMDIBRs), and anaerobic biofilm membrane bioreactor (ABMBR) [13].

AnDMBR is a technology designed to improve performance and is more affordable than AnMBRs. It uses a low-cost support structure, such as coarse cloth or mesh with a 10–100 μm pore size, on which a biofilm layer develops. Wastewater is filtered through this dynamic membrane, retaining suspended solids in the bioreactor. AnDMBRs use less energy-intensive fouling mitigation strategies such as backwashing and relaxation rather than sparging [14].

Researchers are developing the AnEMBR to improve the performance of AnMBRs by combining membrane separation, anaerobic microbiology, and microbial electrolysis cell technology for better control of membrane fouling and increased contaminants removal. The membrane reduces sludge loss when an electrical bias is applied to the membrane surface, creating electrochemical conditions that help alleviate membrane fouling through electrostatic forces, electro-flocculation, electrochemical reactions, and the scouring effect of gas produced by the electrode [13].

Anaerobic membrane distillation bioreactors use hydrophobic membranes and temperature differences for efficient wastewater treatment and reuse. The mass transfer in AnMDIBRs is driven by temperature differences, allowing for water vapor to evaporate through the membrane and improve effluent quality [13].

The anaerobic biofilm membrane bioreactor (AnBMBR) combines the advantages of a conventional MBR process, such as a minimal physical footprint, efficient solid-liquid separation, low sludge generation, and more, with the added benefit of utilizing biofilm to enhance pollutant removal. This is due to the high density of biomass and diverse biological environment within the biofilm, as well as the favorable conditions it creates for microorganisms to thrive [15]. In AnBMBR, certain carriers are utilized for the growth of microorganisms. These carriers enhance mass transfer and foster the growth of biofilms [16].

Several studies [17][18] have indicated that AnMBRs are highly effective in eliminating nutrients and organic contaminants from water. Ramadan et al. [19] found that AnMBRs were able to remove more than 95% of the chemical oxygen demand (COD) when operated at a temperature of 35 °C. The study also showed that the hydraulic retention time (HRT) and solid retention time (SRT) were 9–16 days and 365 days, respectively. An anaerobic biofilm membrane bioreactor (ABMBR) was utilized to effectively eliminate 95% of the chemical oxygen demand (COD) from wastewater. The hydraulic retention time (HRT) was set at 1 day, and the system was operated at a temperature of 35 °C [20]. Keerthi et al. [21] used a hybrid membrane bioreactor (HMBR) with integrated electrocoagulation to remove 90% of the COD and 93% of color from tannery wastewater, while the membrane bioreactor alone was able to remove 73% of the COD and 76% of the color from tannery wastewater. In another study, a dynamic membrane bioreactor (DMBR) was used to remove 91% of the COD and over 97% of dye from textile wastewater when HRT and dye concentration were 1–2 d and <200 mg/L, respectively, at temperature 32–34 °C [22]. According to Sari Erkan et al. [23], a moving bed membrane bioreactor (MBMBR) was found to be highly effective in removing contaminants, achieving a 98% removal of chemical oxygen demand (COD) and 89.5% removal of color. The STR was set at 30 days, and the HRT was between 15.2 and 16.5 days. **Table 3** summarizes the literature on the removal of dyes with AnMBR. AnDMBR was performed, using nylon mesh support material with pore sizes ranging from 20 μm to 100 μm, to remove 99% of the color [24]. Katuri et al. [25] used AnEMBR, with nickel-based hollow-fiber membranes, for treating organic aqueous solutions. They found that more than 97% of COD was removed when the voltage was between 0.5 and 0.9 V and the current was below 25 mA at a temperature of 25 °C. According to Li et al. [20], an AnBMBR was able to reduce 95% of COD from wastewater.

2. Removal Mechanisms and Vital Factors

In AnMBRs, there are two primary methods for removing dyes: physical treatment through membrane filtration and biological treatment through the use of microbial communities [26].

The membrane's performance is dependent on various factors, such as pore size, material composition, wastewater characteristics, solubility, and retention time. Retention occurs as a result of the concentration difference between the retentate (the portion of the solution that cannot pass through the membrane) and the permeate (the solution that has been filtered) [27]. The molecular weight of dyes varies widely, ranging from 265 to 1419 g/mol for Acridine Orange and Reactive Green 19, respectively. This characteristic is of utmost importance for wastewater treatment technologies, as it can significantly impact the process performance in a positive or negative manner [28]. For instance, membrane rejection mechanisms may occur through size exclusion, where the membrane's ability to retain a molecule of a particular compound is determined by the fraction of membrane pores that are smaller than the molecule's size [29]. As a result, dyes with a higher molecular weight are more readily retained by the sieving effect of the membranes [30]. Additionally, other removal mechanisms such as electrostatic repulsion/attraction or adsorption can influence the membrane's efficiency. Therefore, the efficacy of the filtration system is also determined by the physicochemical properties of the dyes and the membrane characteristics, such as the average pore size, molecular weight cut-off (MWCO), and surface charges [28]. A research study employed a self-prepared thin-film composite NF membrane, which was created through interfacial polymerization on the surface of a dual-layer hollow fiber membrane, to remove dyes from textile wastewater. The results of the study revealed that the membrane surface's physical and chemical properties led to a rejection rate of more than 99% as it interacted with the positive and negative groups of reactive dyes [31]. According to Ding et al. [32], during dye filtration, the permeate flux of membranes was found to be lower than the permeability of the membranes for pure water. This phenomenon is attributed to the adsorption of dye on the surfaces and pore walls of the membrane [32]. Zhong et al. [33] conducted a study in which they utilized an NF membrane to eliminate dye from textile wastewater. The NF membranes had a mean effective pore diameter ranging from 1.13 to 1.20 nm, a molecular weight cutoff (MWCO) of 1627–1674 Da, and a high pure water permeability (PWP) of 9–14 LMH bar^{-1}, which demonstrated effective performance in removing dye [33].

In terms of the biological process of dye removal, several studies have demonstrated the potential usefulness of anaerobic reductive cleavage of the dyes bonds by microbes. Some of these studies have utilized anaerobic activated sludge, while others have employed mixed bacterial cultures [34]. As stated by Türgay et al. [35], due to their unique properties, anaerobic microorganisms are often the preferred choice for decolorizing certain dyes present in textile wastewaters. One notable example is their ability to generate electrons that can effectively cleave the azo bond. Khalili and Bonakdarpour utilized a bacterial consortium (activated sludge) to remove dye under anaerobic conditions [36]. Fang et al. [37] conducted a study in which a membrane bioreactor inoculated with activated sludge (a bacterial consortium) successfully removed a significant amount of dye. One key factor that contributes to the effectiveness of AnMBRs is the unique bacterial communities that are present in these systems. These bacteria play an essential role in breaking down the dye molecules and converting them into less harmful substances.

In one study conducted by Zhou et al. [38], the top three dominating phyla in AnMBRs were found to be *Chloroflexi*, *Euryarchaeota*, and *Firmicutes*, with other phyla such as *Proteobacteria*, *Nitrospirae*, *Bacteroidetes*, and *Chlorobi* also commonly detected under diverse conditions. According to a different study, the dominant phyla in an AnMBR were *Bacteroidetes* and *Firmicutes*, which are known for their ability to degrade organics [39]. According to Liao et al., *Firmicutes*, *Chloroflexi*, *Bacteroidetes*, and *Proteobacteria* are commonly identified as the dominant bacterial phyla in anaerobic systems [40]. Chaudhari et al. [41] confirmed the prevalence of the *Firmicutes*, *Proteobacteria*, and *Bacteroidetes* phyla during different stages of dye wastewater treatment.

Maintaining a stable granular structure is crucial in anaerobic systems, and *Chloroflexi*, a strictly anaerobic bacterium, plays a vital role in this process. Studies have reported that *Chloroflexi* can degrade starch into acetate and other short-chain fatty acids, which can then be utilized by methanogens [42]. The presence of electrochemically active bacteria that can transfer electrons, such as *Firmicutes*, *Acidobacteria*, and *Actinobacteria*, is crucial in the removal of dye under anaerobic conditions [43]. Chaudhari et al. [41] stated that with continuous exposure to the dye, there was a notable increase in the relative abundance of *Firmicutes*. This suggests that *Firmicutes* may have played a significant role in the decolorization of reactive blue HERD dye [41]. In addition, Qiu et al. [44] stated that bacteria classified under the *Firmicutes* phylum are accountable for breaking down aromatic amines into CO_2 and alkenes. Additionally, a majority of *Firmicutes* bacteria demonstrate heterotrophy and can facilitate the production of electron equivalents, suggesting that they can enhance the decolorization of azo dyes by breaking down aromatic intermediates [44].

The significant role that some *Bacteroidetes* play in breaking down complex molecules into simpler ones within the host intestine implies that these bacteria may also have a crucial function in the bioconversion of complex molecules into simpler ones during anaerobic processes [45]. In addition, *Bacteroidetes* are capable of extracellular electron transfer, which could be linked to the process of methanogenesis in anaerobic systems. Within anaerobic sludge, these bacteria may be involved in the direct transfer of electrons between species during azo dye reduction [40]. The *Proteobacteria* phylum possesses the capability to break down complex carbon sources, and a majority of these bacteria are either obligate or facultative anaerobes that are commonly found in anaerobic reactors [40]. Within the *Proteobacteria*, *Alphaproteobacteria*, *Betaproteobacteria*, *Deltaproteobacteria,* and *Gammaproteobacteria* are mostly reported in dye removal processes under anaerobic conditions [46].

3. Fouling and Cleaning

In AnMBR technology's application to waste and wastewater treatment, membrane fouling is the most significant challenge, encompassing various parameters such as sludge concentration, HRT, soluble metabolic products, and extracellular polymeric substances [47]. The main issue in the MBR process is fouling of membranes, which causes decline in permeate flux, an increase in trans-membrane pressure, and higher operating costs for cleaning. Fouling occurs due to organic, inorganic, particulate, and biofouling, and is influenced by wastewater and biomass composition [26].

External fouling takes place when particles, colloids, and macromolecules larger than the membrane's pore size deposit on the surface. Internal fouling is caused by the presence of small particles, solutes, and undissolved matter that are retained or submerged within the membrane pores [12]. Fouling of the membrane can also be differentiated into two further types, reversible and irreversible, based on the ease of cleaning [3]. According to Maaz et al. [12], irrecoverable fouling occurs over a long-term experiment, where once the membrane is fouled, its original permeability can never be regained. Residual fouling, on the other hand, involves the buildup of fat, protein, and minerals that can be attributed to various fouling mechanisms [48]. Biofouling refers to the interaction of components of the biological treatment broth with the membrane surface [12].

Membrane cleaning methods are mainly categorized as physical and chemical based on the membrane materials, foulant composition, and nature of cleaning reagents used [49]. In addition to physical cleaning (e.g., backwash and in-line ultrasonic), chemical cleaning is a widely used method to clean polymeric membranes. Chemical cleaning agents can be divided into seven categories, including acids (e.g., nitric acid, sulfuric acid), alkalis (e.g., carbonates, hydroxides), caustics (e.g., sodium hydroxide), disinfectants (e.g., hydrogen peroxide, chlorine, sodium hypochlorite, peroxyacetic acid, metabisulfite), enzymes (e.g., proteases, lipases), sequestrants (e.g., ethylenediaminetetraacetic acid), and surfactants (e.g., sodium dodecyl sulfate, alkyl sulfate) [50]. Backwashing is a more effective cleaning method for ceramic membranes, reducing the risk of concentration polarization, cake layer formation, and fouling [51]. Physical cleaning methods are ineffective in treating irreversible fouling, and therefore, the efficiency (flux) of the membrane can only be restored through chemical cleaning [49].

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