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염료제거용 효소고정화막 생물반응기: 리뷰

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Enzyme Immobilized Membrane Bioreactor for Removal of Dye: A Review

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요 약: 효소 고정화 막 생물반응기(EMBRs)는 폐수 내의 염료를 처리하는 새로운 방법입니다. 이 분야는 효소의 효능과 환경에 대한 높은 저항성 때문에 많은 양의 연구가 진행되었습니다. 효소 자체와 해당 효소의 구조를 모두 포함하는 다양한 방법이 EMBR에 접근할 수 있습니다. 생물반응기 자체는 염료 제거의 필요에 맞게 변형될 수 있습니다. 효소적 생물반응기부터 산화 그래핀 또는 탄소 나노튜브와 같은 나노구조를 사용하는 것까지 다양합니다. 또한 TiO_2와 같은 나노입자는 EMBR을 더욱 향상시키기 위해 사용될 수 있습니다. 폴리머 기반의 막 지지 구조는 또한 효능 증가의 문제에 접근하는 다양한 방법을 포함합니다. 본 바와 같이, 지난 수십 년 동안 EMBR을 사용하는 이 문제에 대한 다양한 접근법이 수행되었습니다. 이 검토는 방법론을 요약하고 EMBR에 대한 다양한 개선 사항을 설명하는 것을 목표로 합니다.

Abstract: Enzyme Immobilized Membrane Bioreactors (EMBRs) are a novel method to treat dyes within wastewater. Due to their efficacy and high resistance to the environment, there has been a large amount of research being done in this area. There are a variety of ways to approach EMBRs that include both the enzyme itself and the structure of said enzymes. The bioreactor itself can be modified to suit the needs of the dye removal. Ranging from Enzymatic bioreactors to utilizing nanostructures such as graphene oxide or carbon nanotubes. Furthermore, nanoparticles such as TiO_2 can be used to enhance the EMBR further as well. The polymer-based membrane supporting structure also includes a variety of different ways to approach the problem of increasing efficacy. As seen, during the past decades, different approaches to this issue that utilize EMBRs have been done. This review aims to summarize the methodologies and describe the various improvements to EMBRs that have been made.

Keywords: *EMBR, membrane, enzyme, dye*

1. Introduction

Recently there have been many advancements in the textile industry. With the global textile industry being a large market an equally large need for dyes has emerged. However, the usage of said dyes is also problematic to the environment when not treated properly. It is estimated that around 800,000 tons of dyes are produced worldwide, of which 200,000 tons are produced in the textile industry[1]. These dyes are useful for the industry

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itself, but when released into water bodies, but are resistant to processing and discoloration while remaining on the water surface. This in turn hinders natural sunlight penetration in the water which affects plant growth and thus increases the toxicity of the water[2]. Additionally, most dyes themselves are toxic due to their phenolic rings and functional groups, negatively impacting local ecosystems and damaging natural resources[3].

To combat these issues various different methods are being used in the present day. Despite this though, conventional treatments for wastewater that contain dyes are inefficient in the removal of these pollutants and the new technologies in research are currently either costly, not environmentally friendly, or not applicable on a larger scale[4]. One answer for these issues is enzyme-immobilized membrane bioreactors. This membrane-based filtration method can purify wastewater and remove color in practical applications, and is resistant to microbial attacks, temperature fluctuations, and the toxicity of the chemicals themselves[5]. While the membrane itself shows good efficiency; the high operation costs, the chance of the membrane clogging, and a huge sum of sludge forming has restricted the ability of using only the membrane[1]. Thus, a need for a more sustainable and more environmentally friendly approach is needed. This is where the enzyme immobilization enhances the membrane-based technology. These biocatalysts in the form of enzymes can be substituted for the traditional chemical-based catalysts and help the degradation of both natural and synthetic dyes. Various enzymes have already been tested for these applications, including azoreductase, phenoloxidase, peroxidase, and laccase[6]. Each has its own characteristics in degrading said dyes. Azoreductase only degrades azo dyes and requires additional cofactors such as $NADH_2$ and $FADH_2$. Peroxidases can only catalyze reactions when there is hydrogen peroxide and its applications are limited to synthetic dyes. Phenol oxidase can degrade most synthetic dyes without the need for cofactors. Laccase is a common polyphenol oxidase that breaks down many dyes, releasing only water as a byproduct.

Regarding the immobilization of the enzyme itself, there have been a variety of methods to do so. Some practical ways of immobilization itself are adsorption, encapsulation, entrapment, covalent binding, and cross-linking. Each presents a myriad of advantages and drawbacks. Adsorption is relatively cheap but has a weak binding force between the membrane and the enzyme[7]. Encapsulation involves trapping the molecules into different matrices and maintaining the biological systems in a fine film to avert contact with the environment itself. Allowing the enzyme to be stable for long periods of time[8]. However, it has severe diffusion problems and rupturing of the membrane is an issue if products gather too quickly[9]. Entrapment cages the enzyme in a network of fibers. Thus, allowing a high load capacity and low fabrication costs[10]. However, enzyme leakage due to a large pore size is a concern. Covalent binding is a well-established method that produces durable enzymes and a low probability of enzyme leakage[11]. However, the costly production and restrictions that lead to enzyme modification and thus productivity loss are an issue[12]. Cross-linking is a method where the enzymes are interconnected and intermolecular cross-linkage is achieved. It boasts strong binding and high stability. But as a high economic cost[13]. This review aims to cover various methods and usages of enzyme-immobilized membrane bioreactors in the field of dye removal in wastewater. Covering recent developments in the field and their mechanisms.

2. Enzymatic Membrane Bioreactor

This study aimed to investigate the effectiveness of laccase treatment and membrane filtration in removing azo dyes from wastewater[14]. The researchers fabricated new biosystems made of nanofiltration or ultrafiltration membranes combined with laccase entrapped between polystyrene electrospun fibers. The biosystems were then used to decolorize aqueous solutions of three azo dyes: C.I. Acid Yellow 23 (AY23), C.I. Direct

Blue 71 (DB71), and C.I. Reactive Black 5 (RB5). The synergistic action of laccase treatment and membrane filtration was effective in removing azo dyes from wastewater. This method was successful in decolorizing dyes over a wide range of pH and pressure conditions, resulting in a significant decrease in toxicity. The use of electrospun fibers in the reactor further enhanced its efficiency. The decolorization efficiency of the dyes was dependent on factors such as the type of dye, the concentration of laccase, and the pH of the solution. The highest removal efficiencies of 97% for AY23 and 100% for both DB71 and RB5 from permeate solutions were achieved at pH 5 and pressure of 2 bar. The decolorization of retentate for RB5 reached 65% under these conditions. The study also analyzed the mass contribution of the selected elements for biosystems before and after dye decolorization using energy dispersive X-ray spectroscopy (EDS). The amount of total organic carbon (TOC), total carbon (TC), and total nitrogen (TN) in samples of permeates after the dye decolorization processes were investigated using a TC/TN 3100 Analytic Jena analyzer. The decolorization efficiency of dye was calculated from spectrophotometric measurements of absorbance of permeates and retentates at wavelength 257 nm, 587 nm, and 595 nm for AY23, DB71, and RB5, respectively. The concentrations of the azo dyes in permeates and retentates, separately, were determined using calibration curves for each of the compounds.

This paper presents a study on the use of spore surface displayed small laccase (SLAC) for the decolorization of textile dye[15]. More than 90% of Indigo carmine was successfully decomposed using recombinant SLAC displaying Bacillus spore. The surface localization of SLAC on the spores was confirmed by flow cytometry. It was also found that the recombinant SLAC displayed on spores retained over 70% of laccase activity even after heat treatment and eight rounds of repeated decomposition. The CotE-SLAC on spore display platform showed excellent performance in thermal stability and is robust enough for repeated usage for the decomposition of synthetic dye, Indigo carmine and alizarin. Overall, the study provides a promising approach for the remediation of industrial dye contaminated water using spore surface displayed SLAC.

This paper is a study on the production of an ultrafine fibrous membrane from the waste culture of Ganoderma lucidum, a type of medicinal mushroom[16]. It was found that this membrane was effective in decolorizing dyes such as methyl violet and malachite green, due to the presence of laccase, an enzyme that can break down these compounds. The membrane was characterized by SEM, FT-IR, and TGA, and was shown to have good thermal resistivity and biocompatibility. It is suggested that this membrane has potential applications in bioremediation and environmental protection. In addition to its environmental applications, the study found that the membrane could be used to extract bioactive compounds from the culture of G. lucidum, which could have potential applications in the pharmaceutical industry. Overall, this study presents a promising approach to recycling the waste culture of G. lucidum and producing a useful material for environmental protection and other applications.

This study covers Dopamine-assisted deposition technology[17]. Which is a widely employed method for enhancing the functionality of polymer membranes. The technique itself offers several advantages, including ease of use, gentle reaction conditions, and strong material adhesion. However, it has faced challenges such as spontaneous polymerization in alkaline environments and non-selective adhesion, making it difficult to simultaneously achieve directional deposition, stability, and rapid deposition rates. Inspiration was drawn from the directional deposition of melanin induced by tyrosinase on skin surfaces. Here, a biomimetic catalysis approach for the targeted deposition of dopamine on specific surfaces was introduced. This method involves the immobilization of laccase on the target surface, which inhibits the oxidative self-polymerization of dopamine and enables biomimetic-induced directional deposition on polymer membranes. The resulting biomimetic catalytic membrane, coated with a

uniform and stable laccase-dopamine layer, proves highly effective for degrading azo printing and dyeing wastewater. Furthermore, the catalytic membrane exhibits impressive performance, with a degradation efficiency of 19.8 μg/min for methyl orange and maintaining approximately 81% relative activity even after six cycles, demonstrating both high catalytic activity and stability.

The study presents the development of a unique dye decolorization system termed VRMs-Bt-WlacD, which involves a triplicate combination of volcanic rock matrix, Bacillus thuringiensis, and laccase WlacD[18]. To achieve this, WlacD was strategically anchored onto the surface of Bacillus thuringiensis MB174 cells, creating a whole-cell laccase biocatalyst. This anchoring process employed two repeat N-terminal domains of autolysin Mbg, denoted as $(Mbgn)_2$. Immunofluorescence microscopy was employed to confirm the successful attachment of the fusion protein (Mbgn)2-WlacD onto the recombinant B. thuringiensis MB174 cell surface. Following this initial step, a series of optimization methods, including single-factor tests, L 9(34)-orthogonal testing, Plackett-Burman tests, the steepest ascent method, and Box-Behnken response surface methodology, were utilized. These optimizations led to a significant enhancement in the whole-cell specific laccase activity of B. thuringiensis MB174, reaching 555.2 U L-1, which represents a 2.25-fold improvement compared to the primary culture conditions. Subsequently, the optimized B. thuringiensis MB174 cells were immobilized onto volcanic rock matrices (VRMs), resulting in the creation of VRMs-Bt-WlacD—an immobilized whole-cell laccase biocatalyst. When tested for decolorization capacity, VRMs-Bt-WlacD exhibited promising results, achieving a 72.36% decolorization rate for a textile dye, reactive blue 19 (RB19), at an initial concentration of 500 mg L-1 in an aqueous solution. This performance was achieved with a solid-to-liquid ratio of 10 g-100 mL. Furthermore, the study explored the potential for large-scale or continuous operations by conducting repeated decolorization-activation cycles, highlighting the high decolorization capacity of VRMs-Bt-WlacD.

In this study, the researchers successfully achieved the display of Laccase CotA from Bacillus subtilis 168 on the membrane of Escherichia coli cells, utilizing poly-γ-glutamate synthetase A protein (PgsA) from B. subtilis as an anchoring matrix[19]. The resulting fusion protein, PgsA/CotA, demonstrated efficient translocation to the cell surface of E. coli, exhibiting an enzymatic activity of 65 U/108 cells. The surface-displayed CotA exhibited notable improvements in its enzymatic properties when compared to the wild-type CotA. These enhancements included higher thermal stability, with over 90% activity retained at 70°C and nearly 40% activity maintained at 90°C after a 5-hour incubation period. Additionally, the displayed CotA displayed stronger tolerance to inhibitors, retaining approximately 80% and 65% activity when incubated with 200 mM and 400 mM NaCl, respectively. The whole-cell system demonstrated remarkable enzymatic activity against various dyes, including anthraquinone dye Acid Blue 62, triphenylmethane dye Malachite Green, and azo dye Methyl Orange. After a 5-hour incubation period, the decolorization percentages for these dyes were 91%, 45%, and 75%, respectively. These findings underscore the potential of this system for efficient dye decolorization applications.

2.1. Graphene oxide

This study explores the promising application of graphene oxide (GO) nanosheets as a support for the immobilization of laccase, an enzyme used for the eco-friendly degradation of organic compounds in water[20]. The immobilized laccase on GO-coated polyethersulfone (GO-PES) membranes exhibited outstanding stability under varying temperature and pH conditions, along with improved water permeability. It focuses on the treatment of synthetic dyes, which are a major contributor to water pollution, especially from the textile industry. Synthetic dyes are challenging to remove due to their colorfastness, resistance to photolysis, and resistance to microbial degradation. Furthermore, they often contain toxic and carcinogenic components, exacerbat-

ing environmental issues. Conventional dye removal methods are expensive, inefficient, and can generate hazardous by-products. In light of this, Enzymatic treatments have emerged as a more efficient and eco-friendly alternative for dye removal. Laccase, a multi-copper oxidase enzyme, has shown promise in degrading various aromatic and phenolic compounds without producing toxic by-products. However, the sensitivity of laccase limits its industrial applications, leading to poor long-term stability and degradation capacity. Immobilization of laccase onto suitable matrices, such as functionalized membranes, has addressed these limitations. Various supports for enzyme immobilization, such as porous glass, chitosan, and nanoparticles, have been explored. Still, they often suffer from extended reaction times and low surface areas, leading to limited degradation efficiency, especially for azo dyes. To overcome these challenges, GO nanosheets with their large surface area have been utilized. However, the development of a biocatalytic GO membrane for dye degradation has not been widely studied. Researchers created a novel biocatalytic GO membrane for dye degradation. GO nanosheets were deposited onto a polyethersulfone support using a layer-by-layer approach and then immobilized laccase onto the GO coating via physical adsorption. The membrane demonstrated potential as a high-performance carrier for efficient enzyme immobilization, particularly for laccase. The study systematically investigated various parameters related to laccase immobilization, including activity recovery rate, optimum temperature, optimum pH value, and operational stability. Four representative dyes with different molecular sizes and structures were selected for degradation tests, with the biocatalytic membrane showing significant improvements in dye degradation compared to GO-PES without laccase immobilization.

In conclusion, this research showcases the potential of GO-coated membranes as a support for laccase immobilizatuvion, offering a green and efficient approach to the removal of synthetic dyes from wastewater, thereby addressing a significant environmental challenge.

2.2. Carbon nanotube

Myceliophthora thermophila laccase was covalently immobilized on functionalized multiwalled carbon nanotubes (MWNT) arranged over a supporting membrane to obtain a permeable bio-barrier that could be applied in multibatch or continuous processes[21].

Several supporting materials showed promise, namely cellulose nitrate, agarose and polyvinyl alcohol, and were evaluated for the MWNT. The highest enzyme loading, reaching 0.286 U mg^{-1} of MWNT, was achieved when cellulose nitrate support was supplemented with 0.21 mg of MWNT. The immobilized laccase demonstrated robust operational stability, maintaining over 95% of its initial activity even after 10 reaction cycles. Additionally, it exhibited enhanced resistance to temperature, pH variations, and acetone exposure compared to its free counterpart. For instance, when incubated in the presence of 20% acetone (v/v) for 6 hours, free laccase retained only 21% of its activity, while immobilized laccase retained 49%. The immobilized laccase was effectively employed for decolorizing Reactive Black 5 (RB5). The highest RB5 decolorization rates were achieved in the presence of 1-hydroxybenzotriazole as a mediator and at a pH of 5.0. After 6 hours, 68.09% of RB5 was decolorized, and after 24 hours, an even higher decolorization rate of 84.26% was observed.

In this study, the authors conducted experiments involving laccase-rich enzymatic extract pretreatment using either granulated activated carbon or microfiltration with a polyimide hollow fiber membrane (0.4 μm pore size) before proceeding to ultrafiltration with a polyether sulfone spiral membrane (10 kDa cut-off)[22]. They found that ultrafiltration without any pretreatment yielded the highest recovery percentage (141%) but required longer process times and had a lower permeate flow rate (20 L m^{-2} h^{-1}). Conversely, when both pretreatments were employed, the recovery rate was moderately lower (109%), but the permeate flux was higher. To concentrate approximately 110 L of laccase crude broth effectively, only the microfiltration pretreatment was used due to its advantages in reducing

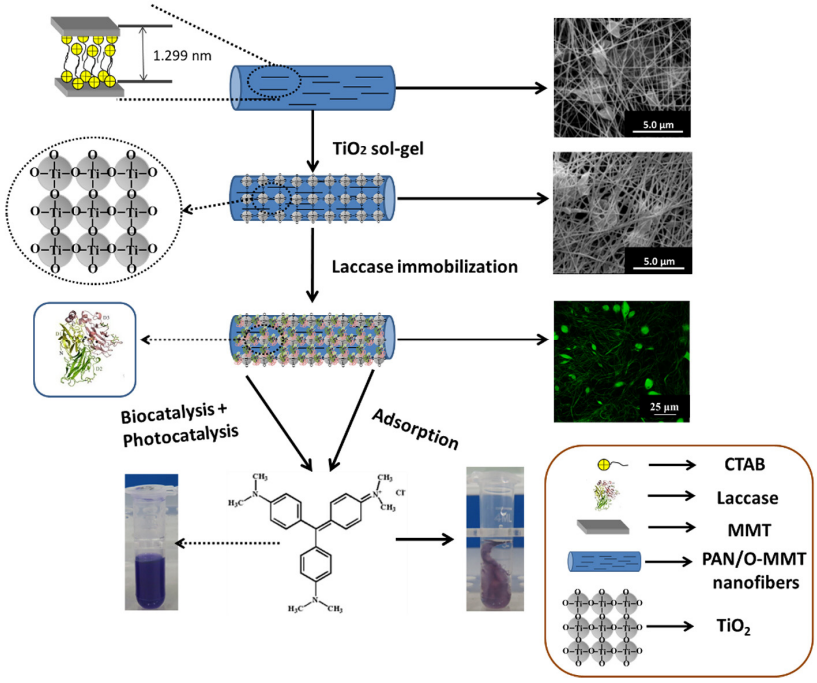

Fig. 1. Schematic illustration of polyacrylonitrile/organically modified montmorillonite (PAN/O-MMT) composite nanofibers functionalized by sol-gel coating of TiO_2 and then used as support for laccase and its application in crystal violet (CV) treatment (Reproduced with permission from Wang *et al.*[23], Copyright 2020, MDPI).

process time and cost. This approach achieved a volumetric concentration factor of 60 times, resulting in an enzyme recovery percentage of 183% and an activity concentration factor of 102. The resulting extract had a laccase activity of approximately 3033 U mL^{-1}. These concentrated enzymatic extracts were both utilized, containing 150 U mL^{-1} laccase activity, along with 0.3 μmol L^{-1} mediator syringaldazine, to conduct decolorization experiments. Decolorization percentages of approximately 75% for brilliant green, 68% for acid blue 80, and 52% for reactive red 198 were achieved.

2.3. TiO_2

This study explores a novel approach for the removal of crystal violet (CV), a triphenylmethane dye, through a synergistic combination of enzymatic, photocatalytic, and adsorption processes[23].

The researchers achieved this by immobilizing laccase (Lac) onto the surface of electrospun polyacrylonitrile/organically modified montmorillonite (PAN/O-MMT) nanofibers coated with TiO_2 sol-gel. This immobilization process enhanced the stability and performance of Lac, particularly under various pH levels, temperatures, and operational conditions. Crystal violet (CV) itself is a challenging dye to remove in wastewater due to its persistence and toxicity and traditional treatments can be costly and produce secondary pollutants. In light of this, biodegradation is a technique that has been used which is cost-effective and environmentally friendly, making it a preferred choice. Laccase (Lac), a versatile enzyme, can degrade a wide range of substrates without generating harmful by-products. However, it's sen-

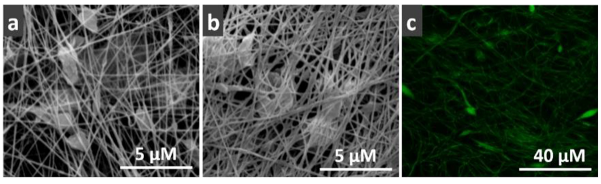

Fig. 2. SEM images of (a) PAN/O-MMT, and (b) PAN/O-MMT/TiO_2 nanofibers. (c) Confocal laser scanning microscope (CLSM) micrograph of Lac-immobilized PAN/O-MMT/TiO_2 nanofibers (Reproduced with permission from Wang *et al.*[23], Copyright 2020, MDPI).

sitive to industrial conditions, necessitating a reduction in reaction time for practical use. To enhance degradation efficiency, titanium dioxide (TiO_2) is combined with Lac due to its effective photocatalytic properties in dye removal. The simultaneous action of Lac and TiO_2 can improve dye degradation and reduce energy consumption. The researchers in this paper developed a functional composite nanofibrous material by electrospinning PAN/O-MMT nanofibers, coating them with TiO_2 sol-gel, and immobilizing Lac. This innovative approach combines the adsorption capabilities of O-MMT and electrospun nanofibers with the enzymatic and photocatalytic degradation abilities of Lac and TiO_2. The resulting material shows promising potential for dye degradation. The paper includes detailed characterizations, kinetic analyses, pH and temperature optimization for Lac, as well as assessments of thermal and operational stability. The effect of initial dye concentration, pH, and temperature on CV degradation was investigated as well. Furthermore, the researchers compared the removal efficiency with and without UV light, revealing the synergistic effect of simultaneous enzymatic-adsorption or enzymatic-photocatalytic-adsorption treatment. In summary, this research demonstrates the effectiveness of immobilizing Lac onto TiO_2-coated PAN/O-MMT nanofibers for the removal of CV dye. This approach offers potential applications in industrial dye degradation and addresses the challenges of wastewater pollution.

It is important to have efficient and sustainable methods for dye degradation in wastewater treatment. In this study, a specific approach of using bacterial cellulose as a support system for immobilizing laccase and TiO_2 nanoparticles was introduced[24]. Bacterial cellulose is a highly pure and biocompatible material that can be easily modified and functionalized, and has a high surface area and porosity that can facilitate the immobilization of enzymes and nanoparticles. The bacterial cellulose was prepared by the fermentation of Komagataeibacter xylinus, and then oxidized to create aldehyde groups that served as anchors for covalent immobilization of laccase and TiO_2 nanoparticles. Atomic Force Microscopy (AFM) and Scanning Electron Microscope (SEM) confirmed the installation of both TiO_2 and laccase on the surface of the bacterial cellulose nanofiber membrane. The results indicate that the immobilized laccase and TiO_2 nanoparticles on the bacterial cellulose support system exhibit excellent catalytic activity for dye degradation under UV irradiation. All in all, composite membranes demonstrate good stability and reusability, making them promising candidates for practical applications in wastewater treatment. It was suggested that further optimization of the composite membrane structure and catalytic properties could enhance their performance and expand their potential applications.

2.4. Polymer

This study aimed to investigate the effectiveness of a new type of electrospun fiber membrane made from polycaprolactone and polyethyleneimine, with immobilized laccase enzymes, for biodegrading textile dyes and phenolic compounds[25]. It was found that the membrane had high decolorization and removal efficiency for various pollutants, including Acid Orange 7 and Reactive Black 5 dyes, and 2,6-dichlorophenol. The immobilized laccases were also found to be operationally stable over time. The membrane had a higher decolorization efficiency for Acid Orange 7 and Reactive Black 5 dyes compared to other studies using immobilized laccase. Additionally, the removal efficiency of 2,6-dichlorophenol was found to be 98.5% after 24 h of treatment. Overall it was concluded that this innovative material shows promise for addressing the issue of pollutants released by the textile and other industries, and that further collaborative research is needed to explore its potential.

While water pollution due to dyes have become a significant problem, enzymatic treatments such as laccase catalysts are considered solutions[26]. Laccase, in particular, is capable of degrading aromatic compounds commonly found in industrial wastewater. However, its sensitivity and limited stability in practical water environments have hindered its widespread use. To over-

come these challenges, enzyme immobilization has been employed to enhance laccase activity, stability, and recyclability. Porous membranes have recently emerged as a practical solution, simplifying enzyme recovery and reuse. Additionally, membranes with biocatalytic properties are well-suited for the removal of aromatic compounds like azo dyes.

Polymeric membranes, particularly PVDF membranes, are highly attractive as supports for enzyme immobilization due to their excellent mechanical strength and chemical stability. Incorporating inorganic materials onto the membrane surface has been shown to enhance its chemical stability. In this study, a poly(vinylidene fluoride) (PVDF) membrane was modified to enhance its chemical stability and mechanical strength, making it suitable for laccase immobilization via covalent bonding. Creating a hybrid bio-inorganic structure on the surface of a polydopamine (PDA)-coated PVDF membrane was one of the key goals. This structure is achieved by using the PDA layer as a secondary platform for attaching Fe_2O_3@SiO_2 cubes (FS@cubes) modified with 3-triethoxysilylpropylamine (APTES) through a solvothermal process. Laccase is then immobilized onto this modified membrane (Lac-FS@cubes-PDA@PVDF) through glutaraldehyde (GA) crosslinking. The resulting biocatalytic membrane exhibits excellent stability, especially in terms of temperature and pH. It demonstrated a remarkable ability to remove Congo Red, achieving a removal efficiency of 97.1% under optimized conditions (pH 7.0 and temperature 35°C). This performance surpasses that of free laccase. Importantly, the immobilized laccase on this membrane maintained its stability during low-temperature storage and showed outstanding reusability.

3. Conclusions

Enzyme-Immobilized Membrane Bioreactors (EMBRs) emerge as an innovative methodology for wastewater treatment, displaying versatility in many diverse approaches. This innovative technique is a response to the challenges of environmental pollution. Most notably wastewater pollution by dyes that are used in various industries. EMBRs boast notable advantages, including heightened stability, reusability, and refined control over enzymatic reactions. Furthermore, the versatility of EMBRs are shown in the literature above. It can encompass the utilization of existing enzymes like laccase, or compounds such as dopamine, and natural ingredients like medicinal mushrooms or E. coli cells. The application of EMBRs does not only include enzymatic options but also extends to the nano-scale, where the supporting structure can be made up of nanomaterials such as carbon nanotubes or graphene oxide. Additionally, TiO_2 nanoparticles also present a valid option in supporting and enhancing the efficacy of EMBRs. Polymers based membranes are also valuable in removing dyes that are normally difficult to handle such as Congo Red. As this combination of biological and nanotechnological advancements continues the field forward, EMBRs stand as an important asset in the pursuit of sustainable and efficient wastewater treatment solutions. Current research focused on increasing the efficacy of the EMBRs itself. However, there are still concerns about the cost effectiveness and economic issues which makes its industrial feasibility questionable. Considering these issues these gaps require further research to compete with existing wastewater dye treatments to provide a sustainable method.

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