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Research Paper

Surface Modification and Integration of Organic/Inorganic Additives Into The Matrix of The Membrane: The Governing Interaction Mechanisms of Dye Adsorption on Adsorptive Membranes

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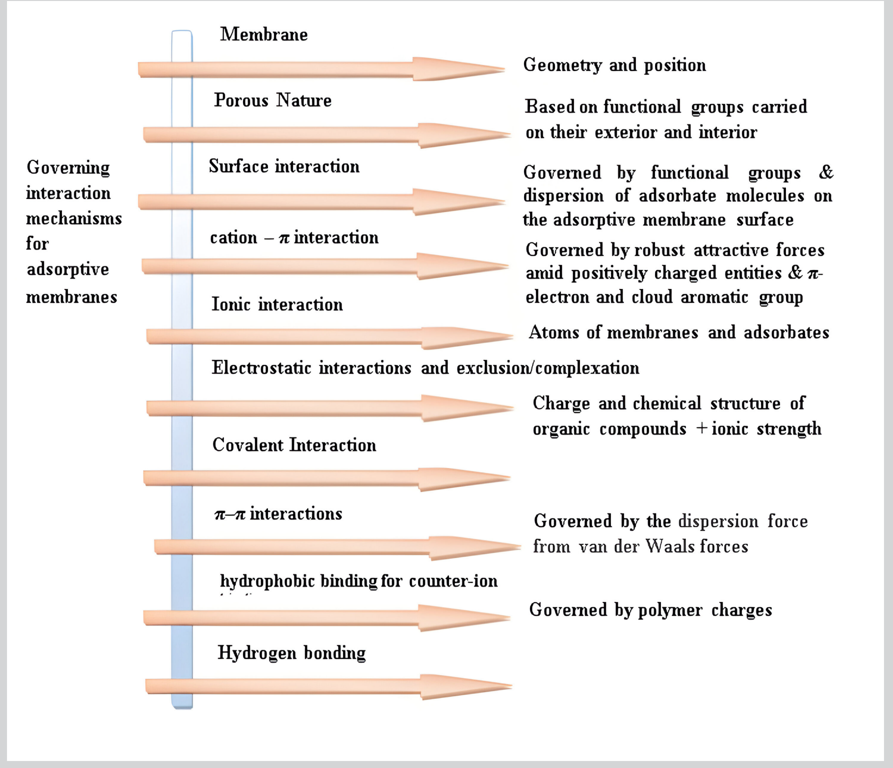
Highlights

- Governing interaction mechanisms between dye and membrane in the membrane separation process
- Adsorptive membranes-based separation mechanism as a prospective technology
- Drawbacks of adsorptive membranes
- Future direction

Graphical abstract



Abstract

The contamination of water from dye industries is considered one of the most global urgent concerns as it compromises the esthetic feature of water bodies, inhibits plant growth, and might stimulate toxicity and carcinogenicity. Adsorptive membranes are highly viewed as one of the prospective technologies that have demonstrated competency in wastewater treatment due to their capacity to make wastewater clean enough for reuse. The adsorption mechanism that lies between adsorptive membranes and dye molecules depends on the individual properties and characteristics. Novel hybrid composite membranes with organic/inorganic additives have been considered for adsorptive membranes as they are expected to advance the effectual removal of dyes from wastewater. The impact of organic/inorganic additives on hybrid adsorptive membranes is highlighted based on the bulk polymer properties like mechanical and chemical resistance together with the structural configurations of the membrane. As such, it is important to understand the interaction mechanisms between adsorbents and dyes for effectual removal of dyes from wastewater. Here, we review the governing interaction mechanisms between dye and adsorptive membrane in the membrane separation process together with the modified adsorptive membranes. Despite the fact that adsorptive membranes possess outstanding effectiveness and capability in wastewater treatment for reuse which provides them a great chance to be employed as prospective technologies for dye adsorption; adsorptive membranes are still racked with some drawbacks. Hence, we present different modification methods used in combating these drawbacks which will subsequently improve the performance of adsorptive membranes.

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Contents

1. Introduction

Numerous sorts of toxic dyes, like basic dyes, acid dyes, and azo dyes originated from different manufacturing sources, and end users are tracked down to the inland surface water [1]. Dye-polluted water discharge into the water bodies, mostly from food processing, textile, leather, cosmetic industries, etc., causes great damage to humans, lives under the water, and the environment as a whole. Hence, they linger in the milieu for an elongated span of time, thereby dropping the appealing worth of water bodies [2]. Due to the negative impact of dye-polluted water, several technological methods have been employed to eliminate these toxic dyes before discharging the wastewater into other water bodies such as streams and rivers. Techniques such as coagulation/flocculation [3,4], oxidation [5], advanced oxidation process [6], ozonization technique [7], use of Fenton's reagent [8,9], adsorption process with activated carbon, [10-13], adsorption process with membrane [14-16], etc. Among all these dye treatment methods, the adsorption process using adsorptive membranes is worth mentioning due to their unique properties. The unique properties that make adsorptive membranes outstanding are low functioning costs, simplicity, reliability, high separation efficiency, and their environmentally friendly nature than conventional water treatment technologies [17].

Adsorption on adsorptive membrane is the buildup of adsorbates at an interface sandwiched between the surface of the membrane [18]. Adsorption at an interface takes place in the course of interactions with the substrate, devoid of forming covalent bonds when instituted by solute molecules. Adsorption on membranes could ensue from a liquid phase or gas phase. The key parameters that control the process are respectively, the concentration of solution, pollutant interactions [19], or partial pressure. However, the temperature has an imperative role in governing the adsorptive membrane filtration process on the account of average kinetic energy of the molecules and their frequency of collision resulting in the surface upsurge with the temperature [20]. Hence the extent of adsorption on adsorptive membranes is dependent on the under-listed factors.

(i) The chemical nature of the adsorbent and the adsorbate. The reason that adsorption ensues from an adhesive process is that, where there is interaction between two types of molecules, the nature of these molecules will ascertain their attractive interactions [21].
(ii) Parameters such as acidity, color, pH temperature, etc., of the system, have an influence on the extent of adsorption on adsorptive membranes, and for the ionic dyes, the exclusion of charge together with size exclusion parameters have an imperative role to play in stabilizing the retention because it is hinged on the size of the dye and membrane pore size [22, 23].
(iii) The upsurge in the temperature leads to greater kinetic energy, subsequently, the faster the movement and diffusion of dye molecules. This will increase the flux [24].
(iv) A high concentration of dye has the potential to reduce the potential of membranes owing to either their buildup on the membrane surface with time or owing to their amassment within the pores, triggering the tightening of the membrane pores [23].

With the grasp knowledge of the factors that influence the extent of adsorption on adsorptive membranes, there is a need to look into the governing interaction mechanisms of dye adsorption on the adsorptive membranes and the modification of adsorptive membranes. Hence, it is the objective of this review to present insights into the governing interaction mechanisms between dye and adsorptive membrane in the membrane separation process. In addition, the novelty of this review aimed to outline the drawbacks of adsorptive membranes and present different types of adsorptive membranes and modification processes for future direction.

2. Governing interaction mechanisms: The role of the membrane as adsorbent

Synthetic dyes are characterized as a reasonably large group of organic chemical compounds with multiple aromatic rings. Synthetic dyes are classified into cationic (malachite green and methylene blue [24-26]) and anionic dyes (Congo red and tartrazine [27]). It is very important to carefully classify dyes as it will enable a precise section of a positively charged adsorptive membrane for effectual removal of negatively charged dyes on account of different governing interactions [27]. Additionally, the efficiency of dye removal is mainly influenced by chemical and physical factors such as the electrostatic interactions between dye and membranes [28, 29], the pore size of the membrane [30], temperature [31], solution pH [32] and dye-substrate contact time [30]. As a result of opposite surface charges, an electrostatic attraction takes place amid the dye and the surface of the adsorbent [26]. Electrostatic interaction primarily speaks of the electrostatic attraction in the midst of positively charged/negatively charged membranes and negatively charged/positively charged dyes, connected to the characteristics of membranes and dyes; and is expressively influenced by the solution's pH [33, 34]. Additionally, dye molecules possess amino, hydroxyl, and sulfonic groups, as groups of atoms taking the place of another atom or group, bonded to the aromatic rings. These functional groups can interact with the membrane of the same functional groups such as carboxyl, amino, and amide [35-38]. Hence, the adsorption mechanism could revolve around numerous interactions like electrostatic, hydrophobic, and hydrogen bonds [26]. All these mechanisms could consequently impact the rate of membrane performance.

The functional group interactions as one of the mechanisms are dependent on the solution pH. Hence, the pH exerts an effect on the membrane surface charge and governs the filtration process. Therefore, membrane surface charge governs the rejection of dyes. Most polymeric membranes evince a negative charge in an extensive range of feed water pH which allows to preferably repel anionic dyes. It is imperative to note that anionic and acidic comprise functional groups that possess a negative charge and these dyes are repelled by the negatively charged polymeric membranes in an extensive range of feed water pH. However, membrane charge can be altered with a suitable surface modification to favor the rejection of specific dyes. Additionally, the zeta potential is also influenced by the pH of feed water. Hence, the zeta potential is contingent on the properties of the liquid phase and the surface of the membrane sample.

Zeta potential is accountable for the electrostatic interaction mechanisms present between the material surface and dye solution. The electrical interaction is a very imperative mechanism between polymeric membranes and dye molecules, especially at the interface of the membrane surface. Furthermore, it is imperative to take cognizance that these interactions are connected with electrical dipole or "double stratum" charge configurations at the nanoscale [39]. Additionally, the zeta potential is employed as an indicator for membrane surface charge. Having a good knowledge of the zeta potential can assist in correlating surface charge to membrane performance and to have a good comprehension of controlling membrane fouling [40]. Studies have shown that to accurately attain zeta potentials, and especially at the isoelectric point; it is imperative to measure the streaming potential on the membrane [41].

At its isoelectric point, the membranes are most expected to possess basic groups and non-ionized acids; otherwise, the absorption of anionic dyes will be less. However the literature has shown that experiments done close to the isoelectric point (pH 6-6.5) [35, 37] resulted in the reduced location of electrostatic forces, and subsequently, other kinds of forces can ensue. Examples of such forces are those responsible for the aggregation of dye (e.g., van der Waals forces, found in dye AO7 adsorption on the membrane), which subsequently improve the efficiency of the adsorption process. Additionally, aggregation upsurges with the concentration of dye and ionic strength [38, 42]. The upsurge in NaCl concentration can stimulate dye molecule adsorption onto the polymeric membrane surface via hydrophobic interactions. Besides, the hydrophobic interactions amid immobilized dye molecules have the capacity to become strong. This is confirmed in the study by Çimen and Yılmaz [43], where the inclusion of salt in a dye solution triggered the heaping of the free dye molecules. Hence, the ionic strength can influence the electrostatic and hydrophobic interactions. Furthermore, investigations have been extensively carried out on dye removal and it was noticed that the magnitude of dye uptake was intensely subjected to the concentration and nature of the electrolyte ionic species in the dye bath [44]. Additionally, studies have shown that the ionic strength could influence the hydrophobic interactions in the course of dye adsorption. For example, at constant pH, an upsurge in ionic strength will upsurge the membrane surface group; however, hydrophobicity will upsurge with the decrease in pH [45]. It has also been found that an upsurge in ionic strength led to a decline in the electrical double-layer thickness of the membrane. This characteristic interprets that H^+ or OH^- concentrations at the surface are nearer to the ones in the bulk. Therefore, for material such as inorganic membranes, at constant pH, the upsurge in ionic strength will result in the ionization of the surface groups [46]. Therefore, the mechanisms through which dye-induced permeability changes are dependent on dye concentration, membrane surface-charge density, dye structure, and ionic strength [47].

The mechanism of dye adsorption can also be understood from the effect of porosity; which is influenced by specific surface area, micropores, and mesopores volume together with the surface chemistry [48]. Hence, porous materials as emergent high-efficiency adsorbents have recently attracted growing attention, as a result of large specific surface area, favorable adsorption capacity to dyes, and fast adsorption rate. Samad et al. [49] examined how heat treatment impacted porous glass microspheres synthesized through an innovative flame spheroidization process for the removal of dye. Their study depicted that the microspheres attained higher dye adsorption efficiency. In addition, the removal of dye was attained through hydrogen bonding, electrostatic interaction, and Lewis acid–base interaction, devoid of functionalization (external or internal) of the microspheres. Fu et al. [50] synthesized a sequence of inventive imidazolium-based cationic organic polymers having diverse ionic capacities and porosity of organic dye adsorption. The synergistic effect between ionic capacities and inherent porosity was confirmed and it was further observed that electrostatic interactions that occur between charges and dyes controlled the removal efficacy. Lakshmi et al. [51] investigated the performance of dye adsorption on 1-butyl-3-methylimidazolium hexafluorophosphate loaded polyethersulfone (PES) polymer inclusion membranes (PIMs). The study arraigned hydrophilic porous PIMs under a well-ordered structure and their performance was examined for dye removal. The PES membranes exhibited high porosity in the span of 79% to 86%. The addition of 2wt% of Poly-N-vinyl pyrrolidone resulted in an upsurge in pore size and the permeability of water. Gorgieva et al. [52] fabricated and evaluated water-stable membranes using carboxymethyl cellulose (CMC) as an ionic adsorbent and cellulose nanofibrils (CNFs) as the stabilizing and citric acid (CA) as structural filler for cationic dyes removal. The resultant, membranes demonstrated anisotropic to isotropic characteristics that are extremely greater than 90% porous structures having pore sizes ranging from a couple of nm to 200 µm. These characteristics subsequently afford reasonably high and stable flux rates in the range of 150-190 L/m^2 h MPa, having close to cationic dye adsorption of 100%, and adsorption capacity in the range of 1828-1398 g/kg.

3. Adsorptive membranes-based separation mechanism as a prospective technology

Adsorptive membranes are highly known to be one of the prospective technologies that have demonstrated efficacy and competency in wastewater treatment for reuse. They are capable of eliminating diverse kinds of emergent contaminants from wastewater that cannot be eliminated by using conventional approaches [53]. Adsorptive membranes possess the same capability of adsorption as adsorbents due to their porosity; hence, they are also regarded as membrane-based adsorbents [54, 55]. Usually, substances that possess adsorption capacity comprise hydroxyl, carboxyl, and sulfonic groups. For example, chitosan can be directly employed as a material of an adsorptive membrane because it contains amino and hydroxyl groups [54, 56]. Hence, adsorptive membranes are considered porous membranes because they externally and internally have functional groups that can bind with dyes by surface complexation mechanism [57].

The adsorption of adsorptive membranes is considered a pressure-driven dynamic membrane filtration-adsorption process because it makes use of the benefits of the adsorption process and membrane separation process [58, 59]. These membranes are described via their strong affinity for ions and molecules, by way of combining ions through ion exchange, chelation bonding, and complexion [55]. Over and above, the high surface area and idleness of the sites of adsorption are significant elements responsible for the efficacy of adsorption for contaminants elimination from wastewater. This membrane technology is essentially governed by three ideologies: adsorption, sieving, and electrostatic phenomenon [53]. In this separation process, the substances separated such as dye are adsorbed on the surface of the membranes, and the solvent infiltrates across membranes' pores (see Fig. 1) [60, 61], with the demonstration of upsurge flow rates, high permeability, low operating pressure, little inner diffusion resistance, swift adsorption/desorption efficiencies together with different adsorption mechanisms. [62, 63].

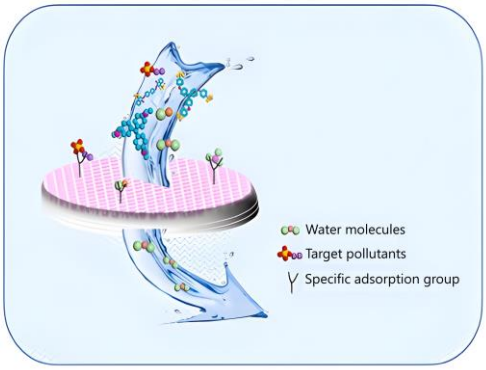

Fig. 1. Separation process of adsorptive membranes (Ref. [55]; with permission from Elsevier to reuse).

Usually, the mechanism of adsorption with regards to the membrane is dependent on rejection and adsorption [64], the hydrophobic interactions of the adsorbent and the adsorbate [65]. Once the solutes (dye molecules) in the wastewater move to the spry functioning stratum of the membrane, the solutes with sizes larger than the membrane's pore are rejected by the membrane. The solutes with reduced sizes will be transported across the active stratum of the membrane and get to the support stratum which functions as an adsorbent; hence the mechanism of the combination of territorial binding and site binding [66]. Subsequently, there will be a reaction/attachment between the membrane and the solute to build a constricted interior spherical complex and then bring out a permeate of sieved water from the membrane [67]. However, the efficacy erasure of dye from wastewater is reliant on surface interactions that exist between the adsorbate and adsorbent governed by the functional groups on the surface of the adsorbent.

The surface interaction characteristics function serves as an essential operating task responsible for the determination of the capability, efficacy, selectivity, and reusability of the adsorbent [68]. Therefore, the mechanisms of adsorptive membranes specifically ensued from membrane-solute interactions such as covalent interaction [69], ionic interaction [69], cation – π interaction [70], π–π interactions, electrostatic interactions and exclusion [71], electrostatic interactions and complexation [72], ion exchange and complexation [55, 61, 73], electro-viscous effects [74], hydrophobic binding for counter-ion binding with immobile charges on polymer chains [66, 75], hydrogen bonding [69] and van der Waals forces [72] to attain highly discerning and swift separation of small organic molecules, such as dye molecules.

Another important factor that will govern the mechanism of adsorptive membranes for dye adsorption is the membrane configuration and its porous nature. Membrane configuration is interrelated to the geometry of the membrane and its location in space relative to the flow of the feed fluid and of the permeate [76]. The coordination of geometry permits a direct interaction of the cation with the quadrupole moment of the adsorptive membrane [77, 78]. Adsorptive membranes are porous and they possess functional groups on their exterior and interior surfaces. These functional groups have the capability of binding with dye molecules via an ion exchange mechanism [79]. Such a mechanism arises on the provision that the adsorbent has energetic locations with free electrons after an electrostatic interaction is present amid the adsorbent and the substance ensues [80]. Barredo-Damas et al. [14] evaluated the operational efficiency of tubular ultrafiltration membranes for treating textile mill raw effluent under diverse working conditions. The flux was increased with an upsurge in pressure which was further enhanced at the least tested pH value. In addition, the upsurge in pressure resulted in the rejection of constricted particles on the membrane. This outcome was boosted via the minor electrostatic repulsive forces. Babu and Murthy [15] employed poly PES/PVA (Polyvinyl alcohol) nanofiltration membranes for the treatment of textile dye wastewater. Outstanding flux was attained with the membrane that contained 1 wt% PVA and an outstanding dye rejection was attained for dispersed dyes. Cao et al. [16] tailored two different membranes for an active elimination of dye intermediates in multifaceted dye wastewater. The two membranes attained a considerable extended stability and over 95% rejection of RB-5 dye was also attained. Ağtas et al. [24] evaluated the influence of membrane properties on membrane efficiency by employing a comprehensive analysis and concluded that the main significant parameter for the removal of dye was the zeta potential. Cseri et al. [79], proposed a single-step preparation of electro-spun nano-fibrous porous polyimides improved with ion exchange characteristics as adsorptive membranes for the adsorption of dye from textile wastewater. The proposed method offers a novel means for the transformation of carboxylic acid groups in polymers to be similar to the functionalities of anion and cation exchange. The ionic sites, together with the carboxylic groups were anticipated to result in an upsurge of robust interactions present between the polymer and ionic organic compounds, like dyes. It was suggested from the results obtained that nano-fibrous polyimide membranes boosted with ion exchange characteristics are auspicious adsorption membranes for the removal of dye from wastewater. Fig. 2 represents all the governing interaction mechanisms for the adsorption dye discussed in this section.

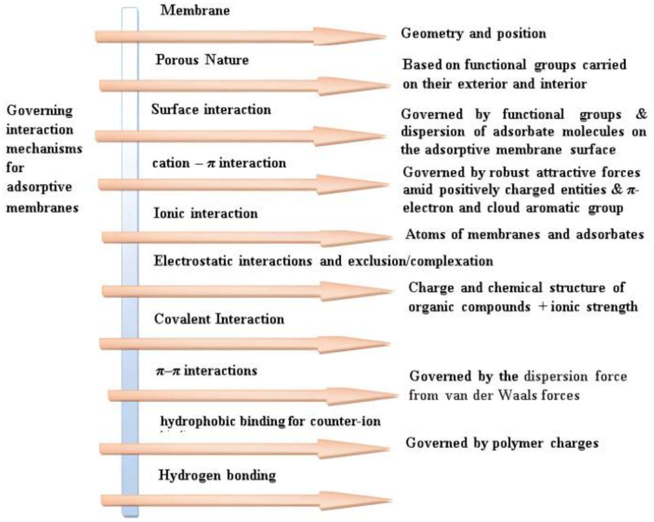

Fig. 2. Representation of all the governing interaction mechanisms for adsorptive membrane dye removal

Literature has established that functional groups on the outward and the interior surface of the adsorbent are principally used to control adsorption because they govern the mechanism together with the selectivity. Sulfonic, carboxyl and phosphonic groups adsorb contaminants through ion exchange, while nitrogen groups such as amine cations and anionic adsorb through electrostatic interactions [81]. Studies have shown that amine groups are more effective for the adsorption of dyes [82]. The utilization of these kinds of membranes for the adsorption of dyes has been lengthily studied and investigated for governing interaction mechanisms. Cheng et al. [83] synthesized deacetylated cellulose acetate (DA)@polydopamine (PDA) composite nanofiber membrane and systematically studied the mechanism of adsorption for the adsorption of methylene blue. The authors construed that the adsorption mechanism of the synthesized material was ascribed to the following points: (1) In the course of the process of adsorption, a huge quantity of phenolic O-H on the surface of the membrane was negatively charged; hence, the membrane can be utilized as effectual adsorbing sites for cationic dye on the account of the build-up of electrostatic interactions that occur between the membrane and dye molecules. (2) π–π stacking interactions ensued amid the membrane and the dye molecules because they both accommodated ample aromatic rings, which was established by the alteration of the peak at 1630 cm^{-1}. Vo et al. [84] provided a perception of adsorption mechanisms by employing a graphene oxide-chitosan interactive membrane for the erasure of several organic dyes from wastewater based on the study of adsorption isotherm and kinetic studies. An outstanding operational effectiveness of the membrane was attained for the elimination of dyes. Furthermore, the adsorptive membrane displayed an excellent recycling performance. Additionally, the dye adsorption and desorption capacities were considerably improved on account of the connecting bond creations and interactions that occur between the surface of the graphene oxide and the active hybrid network via functional groups. Zhao et al. [85] developed adsorptive membranes by employing polyvinylidene fluoride (PVDF), chitosan (CS), and carboxylated carbon nanotubes (CNTs-COOH) (PVDF-CS@CNTs-COOH) and investigated if these membranes can afford to successfully remove anionic and cationic dye from wastewater, effectively. The membranes are highly performed concerning the rejection of the dyes. Furthermore, a reasonable efficiency in recycling, with a flux above 94% was attained. The summary of some of the interactions is shown in Fig. 3 using a Congo dye molecular structure.

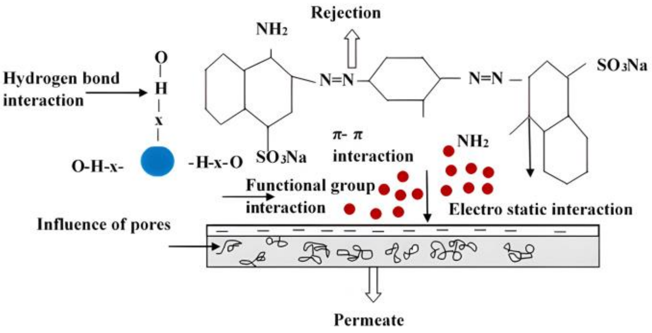

Fig. 3. Graphical demonstration of governing interaction

Additionally, a composite membrane could be utilized as an extremely functional adsorbent for dyes from wastewater owing to the benefits of nanometer scale, high porosity, and excellent hydrophilicity [83]. Lie et al. [86] examined the mechanism and practicability of prepared modified fiber adsorbents using waste Polyacrylonitrile fiber ($PANA_MF$) to remove anionic dyes. The fiber adsorbents exhibited excellent adsorption performance. The mechanism of adsorption studied shows that the adsorption was instigated primarily via the electrostatic force. The dye continually diffuses from the exterior to the interior and; hence swiftly adheres to the adsorption site. Dispersing force, hydrogen bonding, and van der Waals force still pointed out that the dye diffused into the fiber, even when the adsorption site was filled. The dye in the interior stratum of the fiber possesses the capacity to slacken the polymer chain which upsurges the diffusion rate. A graphical demonstration that depicts the process of diffusion even when the adsorption site was occupied is depicted in Fig. 4. Shin et al. [87] studied the separation performance of synthesized nanocomposite membrane for the erasure of Evans blue (EB) dye, and the same functional membrane was subsequently used for treating aqueous solutions having different anionic dyes. The adsorption mechanism was elucidated through the electrostatic interaction between chitosan and dye. All this literature depicted that the removal of dye by adsorptive membranes ensued as a result of the chemical interactions involving the functional group and electron transfer between the membranes and dye molecules. In addition, the sulfate group attraction in dye molecules also had a strong impact on the adsorption of dye molecules. Liu et al. [88] developed a diethanolamine (DEA) - ---modified polyamide composite membrane for effective dye elimination. The integration of DEA molecules on the membrane led to a substantial enhancement of membrane hydrophilicity. This can successfully improve the hydrophobic interactions taking place between dye molecules and membrane surface in return; hence extenuating the adsorption of dye molecules on the membrane. Alardhi et al.

[89] studied the appropriateness of the hybrid MCM41-UF membrane for the removal of dye. The interaction that exists between dye and the adsorptive membrane turned out to be better specifically as the roughness upsurges on the surface where the porous structure characteristics accelerated the spread of adsorbed dye.

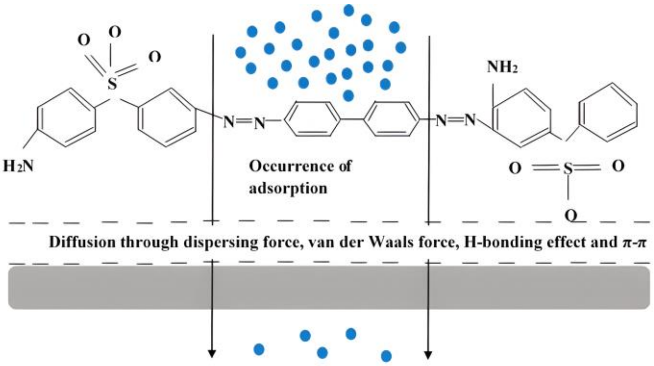

Fig. 4. Graphical demonstration of the process of diffusion even when the adsorption site is occupied.

4. Types of adsorptive membranes

Theoretically, adsorption is considered a transfer movement of substances from the flowing fluid phase to the surface of the material via physical and/or chemical interactions [90]; hence, the supreme advantage of adsorptive membranes over other technology such as the utilization of adsorbent made from activated carbon. Thus, adsorptive membranes possess a twofold function filtration and adsorption [81]. There are different kinds of adsorptive membranes. These membranes are categorized based on the type of membranes and adsorbents incorporated into the membranes during synthesis. A schematic representation of adsorptive membranes that could be used for treating dye wastewater is shown in Fig. 5. These membranes follow the same pattern of operation. They are hinged on the process of mass transfer where pollutants will be adsorbed on the membranes.

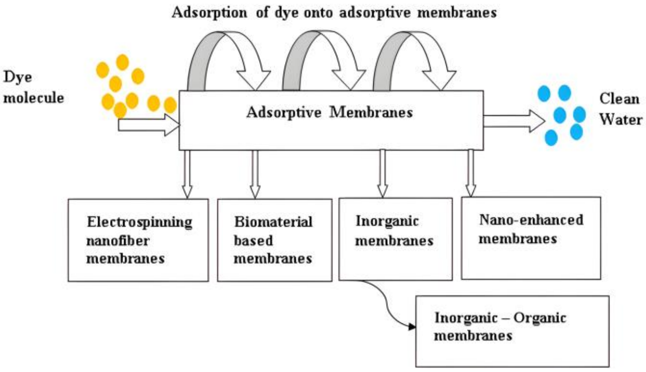

Fig. 5. Classification of adsorptive membranes concerning membrane materials and adsorbents

4.1. Electrospinning nanofiber membranes (ENMs)

Electrospun nanofiber membranes (EMN) are adsorptive membranes that possess an excellent catalyst-supporting material as a result of their benefits (manageable distribution of pore size, large specific surface area, and the volume of the pore, together with insignificant mass transfer resistance in the inner part of the nanofibers) [91]. They possess the capacity to homogenously integrate functional particles in the fiber matrices. They are employed to make fibrous membranes that are of nano and sub-micron diameters [92]. The mechanism of ENM adsorption on dyes could be electrostatic attraction or chelation, which offers outstanding filtration and adsorption ability dyes, and it gives room for improving the capacity of adsorption of ENMs for dyes, via the upsurge of chelation sites [93]. Furthermore, they possess a great number of adsorption sites which subsequently results in an outstanding adsorption capacity; which makes them excellent materials for the elimination of dyes from wastewater. Quite a number of natural and synthetic polymers that could be transformed into electrospun nanofibers are in abundance. This is attained through the process of liquefying these polymers in proper solvents and then spinning them. Concerning their mechanism, electrostatic attraction exists between the anionic dye molecules and the positively charged nanofiber [93]. Moreover, many forms of membrane composites which are also considered polymer membranes comprising diverse structural configurations of nanofibers, such as nanofiltration, ultrafiltration, microfiltration, etc., have been employed for the erasure of dyes in wastewater. Hence, scientists have synthesized ENMs for the erasure of dyes from wastewater with excellent results.

Akduman et al. [94] synthesized nanofiber membranes via the electrospinning method and the membranes possessed exceptional properties like high porosity with controlled pores. The nanofiber membranes demonstrated outstanding potential for the erasure of dye having an excellent sorption capacity of 88.31 mg/g. Ma et al. [95] synthesized a membrane by employing poly(vinylidene fluoride) (PVDF)/graphene oxide (GO) for the erasure of organic dye. The electrospun PVDF fibrous membrane depicted an excellent performance which may be ascribed to the pseudo second-order model with the methylene blue erasure having an excellent adsorption capacity of 621.1 mg g^{-1}. Additionally, the membrane demonstrated outstanding regeneration capability. Chen et al. [96] synthesized a protein-functionalized nanofiber membrane for the erasure of dyes. The potential of the nanofiber membrane for the erasure of dyes was assessed. At the end of five successive adsorption-desorption cycles, the effectiveness of dye erasure utilizing this nanofiber membrane was sustained beyond 97%. Sun et al. [97] designed leaf-like metal-organic frameworks (MOF)-decorated electrospun nanofiber membrane (using polyacrylonitrile nanofibers (NF) and zeolitic imidazolate framework (ZIF)) for the erasure of dye from aqueous solutions. The synthesized membrane effectively adsorbed malachite green having an excellent adsorption capacity of 5103 mg g^{-1}. Nady et al. [98] conveyed the utilization of waste obtained from polystyrene for the synthesis of an electrospun membrane for the erasure of dye from wastewater. The membrane exhibited an excellent dye uptake with excellent regeneration efficiency.

All the excellent work in the literature has made known that, the high adsorption capacity of membranes prepared from nanofiber ensued on account of the manageable distribution of pore size, large specific surface area, and the volume of pore. Hence, nanofibers could be regarded as an innovative group of materials that can provide substantial benefits for applications in removing contaminants such as dye from wastewater [93]. Polymeric nanofiber membranes attained from the electrospinning process have been instituted and established as ecologically and sustainable friendly potential material for the removal of dye from contaminated water. The treated dye wastewater can be subsequently re-claimed in the dyeing process; hence, combating water scarceness as it is not being passed out into the milieu [93].

4.2. Biomaterials based membranes

The utilization of biological materials in adsorptive membranes is well-thought-out to be a recent technology with favorable results from the literature on account of their excellent antifouling property, high permeability, and mechanical reinforcement effect [53]. Recently, there an intensifying need for biopolymers, which are made from renewable source materials; they are economical, and biodegradable without any environmental problems such as toxicity. Membrane-based biomaterials could be attained with varied properties on account of the operational functions to be performed. These membranes are characterized by being porous or dense, hydrophobic or hydrophilic, synthetic or natural, biodegradable or not biodegradable, and stiff or elastic [99]. Biomaterials-based membranes make better filters for the treatment of contaminated water than traditional petrochemical membranes [100]. Hence, the use of biomaterials has been more successful concerning membrane filtration possessing excellent water flux and effective decontamination and decoloring of dyes in wastewater. Dassanayake et al. [101] synthesized manganese dioxide (MnO_2)-chitin-hybrid material for the erasure of methylene blue (MB) from the liquid solution. The hybrid material displayed excellent effectiveness for oxidative decolorization and effective erasure of MB. The hybrid bio-membrane showed outstanding recyclability and sturdiness having 99% degradation for methylene blue at the end of ten successive cycles. Song et al. [102] synthesized an innovative phosphorylated chitosan (PCS) membrane by using surface functionalization with a suitable quantity of graphene oxide (GO) nanosheets via the formation of covalent bonds. The membrane showed a little higher efficiency for the erasure of anionic dyes and significantly superior efficiency for the erasure of salt. Hasanuddin et al. [103] synthesized Poly(Lactic Acid) - poly(ethylene glycol) with magnesium silicate (PLA- PEG/ $MgSiO_3$) to access its dual performance as a filter and adsorbent for the erasure of methylene blue dye. The membrane had 86.36% removal activity of MB dye and established the reusable character of about six cycles. These studies have shown that biomaterial-based membranes are green adsorbates and on account of their cost effectiveness, biodegradability, and easy access. Biomaterials, especially biopolymer membranes, are synthesized from renewable sources that serve as alternative membrane materials to conventional polymers used in synthesizing membranes. More published works on adsorptive membranes and their functions are tabulated in Table 1.

4.3. Inorganic membranes

Inorganic membranes are prepared from oxides or metals, and they can be present in multi-stratum supporting structures, or in the form of self-supporting structures, where they can be made as tubes or standalone sheets, should they be sufficiently permeable. Should that fail to be the situation, they can be synthesized as thin films on multi-stratum with structural supports. The structural supports need to be adequately sturdy and permeable. There is need for a step-wise or graded alterations in porosity to make a well-ordered deposition surface for the membrane [104]. However, the utilization of supports such as inorganic membranes for adsorptive membranes, is restricted on account of the expensive cost of production [55]. Though when likened to membranes made from polymeric materials, inorganic membranes can be more selective and permeable, and they can tolerate highly extreme conditions [104]. Inorganic adsorptive membranes are frequently synthesized through coating and deposition of adsorbents on the inorganic support [55]. In addition, the utilization of inorganic solid particles is considered to be the common technique to make membrane pores. However, their drawback is that the dispersion of these solid particles in the course of membrane synthesis would result in creating an asymmetric pore configuration of the prepared membranes. Thus, to circumvent such shortcomings, the viscosity of the casting solution must be carefully guided to control it via polymer concentration to deter or delay the dispersion of these particles [90]. On account of their adequate sturdiness, high resistance, permeability, and ability to function at extreme temperatures and broad pH, researchers have used membrane composites made of inorganic materials to treat wastewater.

Hebbar et al [105] synthesized polyetherimide nanocomposite membranes with amine-functionalized halloysite nanotubes (MHNTs) for the potential erasure of dye from effluents. By adding inorganic additives or fillers into the casting solution, the morphological configuration, porosity, and hydrophilicity were significantly changed. Their study depicted that the addition of additives into the casting solution exhibited a significant alteration in the effectiveness of the subsequent membrane. The permeation experiments depicted that the flux was enhanced to approximately 195 L/m^{2}h with the inclusion of 4 wt% additive dosage. The membrane demonstrated rejection of 97% at pH 8 and 94% at pH 7 for methylene blue and rhodamine B dyes. Shin et al. [86] synthesized organic-inorganic composite membranes for the erasure of anionic dye from wastewater. The membrane depicted an excellent erasure capacity for Evans blue (EB) and the adsorption capacity was considerably improved to about 434.78 mg/g under acidic circumstances. Alarcón et al. [106] evaluated inorganic membranes for the treatment of textile wastewater at optimized conditions. Using optimized circumstances, over 92% of the dye was attained in less than 30 min. It can be observed that membrane composites made of inorganic materials demonstrated a stimulating case of adsorptive membranes.

It is imperative to take into consideration that the utilization of organic-inorganic materials for the synthesis of membranes for the erasure of dye from wastewater is very attractive. This is because the membrane obtained from these two materials will offer the likelihood of attaining coadjutant effects on the mechanical and thermal stability which will subsequently have an impact on membrane permeability and selectivity. More published works on adsorptive membranes concerning organic-inorganic membranes and their functions are shown in Table 1.

4.4. Nano-enhanced membranes

The distinctive structural configuration and surface properties of nanomaterials (such as carbonaceous materials, nano-metal or nano-metal oxides, etc.) is the enablement of the materials to be employed as adsorbents [90]. Carbonaceous materials (like active carbon (AC), carbon nanotubes (CNTs), and graphene) are well recognized as prospective equivalents to polymer-based composites on account of their excellent mechanical strength, high aspect ratio, compatibility of the carbon matrix with the polymeric structure, and sturdy interactions and adhesion [90]. Nano-enhanced adsorptive membrane is synthesized by dispersing nano-sized adsorbents like metal oxides and zeolites etc., all through the continuous polymeric matrix [107]. Nano-enhanced membranes - adsorptive membranes are usually synthesized by dispersing nano-adsorbent into a polymeric solution before casting and subsequently evaporating the solvent within a well-monitored circumstance [108]. Manoukian et al. [109] synthesized polysulfone-highly even activated carbon sphere mixed-matrix membrane for the removal of dye from wastewater. The study demonstrated enhanced water flux and rejection efficacy of 99.9% was attained, which was attributed to size exclusion and adsorption mechanisms. Kalaivizhi et al. [110] examined the degradation of dye by using synthesized polysulfone (PSF) and polyurethane (PU) membranes blended with AC and green synthesis of ZnO nanoparticles. The synthesized membrane could be utilized as a substrate for dye erasure. Essate et al. [111] synthesized an ultrafiltration composite membrane using a mixture of PSF and polystyrene (PS) on ceramic pozzolan support. The study proved that the inclusion of PS considerably improves the rejection of the membrane.

4.5. Drawbacks of adsorptive membranes

Membrane separation techniques are well-thought-out to be the outstanding technological separation technique employed for combating the challenges encountered by other techniques used for treating dye wastewater. As a result of their effectiveness and capability in wastewater treatment for reuse, adsorptive membranes are very much considered among the prospective technologies used for the adsorption of dyes. Be it as it may, adsorptive membranes are still racked with some drawbacks, like the moderately low adsorption capacity resulting from the inadequate quantity of the adsorbents that are integrated into the membrane matrix. In addition, the issue of fouling and agglomeration are other drawbacks that the membrane industry is experiencing [81]. Furthermore, the trade-off that exists between nanometer-level selectivity and permeability is another intrinsic drawback of membrane technology and most extremely porous materials that offer excellent adsorption capacity come short of the processability of solution and steadiness for attaining adsorption-based molecule separation [59]. Researchers are recently focusing on the modification of membranes to improve dye rejection capacity, reduce the rate of fouling, and overcome other drawbacks, which will subsequently aid in the treatment of textile wastewater in an economical manner. The modification methods that will aid in improving the performance of adsorptive membranes are the employment of conductive polymers, employment of polymer blends [126], use of various additives, such as water-soluble materials [127], nanoparticles, and nanomaterials [128, 129], and surface modification such as free radical graft copolymerization [130].

5. Modifications of adsorptive membranes

This section will discuss different types of modification processes for the enhancement of adsorptive membranes. These modifications will combat the drawbacks of adsorptive membranes.

5.1. Conductive Polymers

In order to overcome these drawbacks, an effective polymeric membrane material and the appropriate integrated adsorbent must be wisely chosen. Due to the outstanding properties of conductive polymers (polyaniline, polypyrrole, and polythiophene) that prevail over traditional membrane materials, conductive polymers should be considered for synthesizing adsorptive membranes. Properties such as ease of functionalization, mechanical stability, chemical versatility, easy synthesis, high electrical conductivity, and environmental sustainability make conductive polymers prevail over traditional membrane materials [81]. Conducting polymers are incapable of being dissolved in aqueous media. It is documented in the literature that the decrease in the optical absorption showing the reduction of the concentration of dye ensued when conductive polymers are included in a solution of an organic dye [131], due to conjugated π-bonds and the exceptional optical, electrical, and physical properties.

The interactions the exit between the dyes and conductive polymers aid the removal of dye from wastewater [131, 132]. The mechanism was founded on dye adsorption or its photocatalytic decomposition, or both could at the same time ensue or in sequence in several proportions [133]. Hence, the interaction emanated on account of physical van der Waals forces or accredited to physicochemical interactions. According to Stejskal [131], one likely interaction that exists between conductive polymers and dyes is the π-π interaction. Again, the electrostatic interactions should be taken into consideration should an insoluble salt be produced from a soluble anionic dye with the conducting polymers. The presence of hydrogen bonding of the atoms of hydrogen in conducting polymers in relation to the nitrogen atoms present in dyes conversely could most likely be one of the strongest interactions that can be accounted for [133]. Mohammad and Atassi [134] reported the synthesis of membranes from polyacrylonitrile (PAN), polylactic acid, and their conforming membranes coated with polyaniline (PANI) for the erasure of MB. The membranes coated with PANI exhibited improved adsorption effectiveness and their DC- conductivities were interrelated to methylene blue concentration. The polyaniline-coated membrane stimulated electrostatic interactions that exist between the adsorbent and the adsorbate.

The interaction mechanism using the conductive polymers for the synthesis of adsorptive membranes for dye erasure is based on the fact that conductive polymers interact with dyes on account of the similarity that the membranes and dye possess in the associated molecular arrangement and layout of their moieties [131]. Furthermore, the interaction mechanism is the adsorbent-adsorbate interactions together with the ionic bonding [135]. In addition, the mechanism is also hinged on hydrogen bonding, π-π interactions, and/or hydrophobic interactions between the adsorbent and the dye. The improvement of the interaction of the mechanism can be attained by making the conductive membrane highly porous through the use of pore enhancers; such as integrating inorganic porous materials into the conductive polymeric membranes.

Table 1
Different types of adsorptive membranes have, a range of performances for the erasure of dye from wastewater

Types of adsorptive membranes	Materials	Method of synthesis	Types of dye removed	Performance parameters	References
Electrospun nanofiber membrane	Functionalized PAN nanofiber membrane with chitosan and proteins	PAN yarn was electrospinning into nanofibers	Anionic dye and cationic dye	After five successive adsorption-desorption cycles, the efficacy of dye erasure was sustained beyond 97%.	[94]
Nano-enhanced membrane	Introduction of ZIF-8 into PSF and CS blend	Phase inversion process	Rhodamine Blue (Rh. B), MB, Acid Blue (AB) and Congo Red (CR)	The efficacy of dye erasure was sustained beyond 85% for all the dyes.	[112]
Nano-enhanced membrane	Activated carbon/ZnO nanoparticles integrated into a PSF/PU membrane	The phase inversion method	CR and MB	The membrane attains 97% erasure within 100 min.	[110]
Nano-enhanced membrane	Integration of iron in titanium oxide nanotubes and integration of silver titanium oxide nanotubes are inserted in the PES matrix	The casting solution method	Rhodamine B dye	97% maximum efficiency removal of rhodamine B was attained.	[72]
Inorganic membrane	2-chloro-1-methyliodopyridine served as an energetic agent for grafting polyimide polymer onto the membrane	Covalent bonding with surface carboxylic groups.	Tropaeolin O, Victoria blue B, and Semixylenol orange	Displayed an excellent erasure efficacy of 98.3%, 99.2%, and 99% respectively for Tropaeolin O, Victoria blue B, and Semixylenol orange.	[113]
Nano-enhanced membrane	Covalent functionalized graphene oxide sheets added to magnetic nanoparticles were incorporated into PES	Phase inversion induced via immersion precipitation method.	-	The hybrid membrane attained about the same percentage of rejection (99%) while the ordinary PES membrane attained 91% rejection.	[114]
Electrospun nanofiber membrane	Poly (arylene ether nitrile) (PEN) nanofibrous substrate used in conjunction with bioinspired polydopamine coated GO barrier stratum	Electrospinning technique	Anionic dyes	An outstanding flux of 99.7 L/m2 h at the operating condition of 0.1 MPa, 25 °C, and pH=3.0, and an outstanding rejection of 99.8% was exhibited.	[115]
Nano-enhanced membrane	Chemically modified halloysite nanotubes uniformly integrated into polyetherimide membrane	Diffusion-induced phase inversion (DIPS) means	Cationic dye	There was an exhibited rejection of 94% at pH 7 and 97% at pH 8.	[105]
Organic-inorganic membrane	Chitosan (CS) and porous geopolymer (PG)	Electrostatic self-assembly method.	Crystal violet (CV)	The erasure efficacy of CV by the PG membrane was just 68.54. However, CS/PG displayed an excellent and steady erasure efficacy of 94.84.	[116]
Organic-inorganic membrane	N-[3-(trimethoxysilyl)propyl] ethylene diamine, 3-(triethoxysilyl)propyl isocyanate and porous glass fiber membranes	Elaborate steps (see Ref. 107)	Anionic dye	There was a recovery of over 92% of dye molecules.	[117]
Nano-enhanced membrane	Poly(ether sulfone) and cerium oxide nanoparticles	A non-solvent phase-induced procedure.	Direct Red 23, Congo Red, and Direct Red	The membrane exhibited rejection of 98.43 and 99.36% for Congo red dye and 99.78% for Direct red dye.	[118]
Inorganic membrane	Si-doped TiO2, Al2O3 membrane	Sol-gel technique	Dye Reactive Red ED-2B (RR ED-2B	The erasure efficiency of the membrane was based on the abroad absorption band in a span of 200 to 350 nm	[119]
Nano-enhanced membrane	Titania nanotubes (TNTs) and poly(vinylidene fluoride)	Phase inversion method	Brilliant green (BG) dye	There was an upsurge in the antifouling properties of the membrane. Excellent Water permeability and excellent antifouling recovery ratio were attained.	[120]
Organic-inorganic membrane	Coating of Polydopamine (PDA) with poly (vinylidene fluoride) modified with Fe2O3@SiO2 cubes	Solvothermal method followed by covalent immobilization of laccase	Azo dye	The membrane displayed an excellent stability and an outstanding reusability.	[121]
Nano-enhanced membrane	Polyvinylidene fluoride (PVDF) membranes coated with ultra-thin zeolitic imidazolate framework-8.	Phase inversion method	Rhodamine B, Direct Black 38) and Reactive Green 19	Rejection of 74%, 98 and 82% were attained respectively for Rhodamine B, Direct Black 38) and Reactive Green 19	[122]
Electrospun nanofiber membrane	PEI Polyethyleneimine(PEI)/Polyacrylonitrile (PAN) and Keratin	Keratin was functionalized over electrospun PEI/PAN using electrostatic interaction	Anionic dye	An improved rejection capacity of 93.25% was attained	[123]
Electrospun nanofiber membrane	Integration of (CNF)/TiO2 nanoparticles in PAN	Electrospun technique	Methylene blue dye	84% rejection was attained for methylene blue dye	[124]
Nano-enhanced membrane	Titanium aluminum nitride (Ti2AlN) as an inorganic additive and cellulose acetate	Reactive sintering method	reactive black 5, reactive red 120, and bovine serum albumin	Excellent percentage erasure efficacy was attained for all the dyes.	[125]

5.2. Use of polymer blends

To overcome the challenges of using adsorptive membranes in treating dye wastewater, the design of a novel material for the synthesizing adsorptive membranes might seem difficult; hence the blending of polymers to synthesize adsorptive membranes gives room for a number of gains such as its easy preparation, ability of replicating the membrane to obtain a consistency and commercial sustainability, resulting to an exceptional polymeric membrane with interdependent properties [136, 137]. The use of polymer blends for synthesizing membranes can be regarded as miscible or immiscible (homogeneous or heterogeneous) [138, 139]. Though, at a molecular level, the mixture of polymers is commonly immiscible as a result of their huge molecular mass; however, the miscibility of the polymers is imperative as the enhancement of membrane properties is usually sought after [137]. Hence, finding the means of how two polymers are mixed to obtain a miscible polymer blend for membrane preparation is of great importance. Polymer blends possess properties that are most likely governed by the structure attained in the course of preparation, and the means of studying this structure is very significant in materials science [140]. In polymer blends, the molecular interaction and miscibility that exist between the small molecule acceptor and the polymer donor usually exercise control over the morphological configuration of the donor–acceptor (D-A) blend [141]. Hence, the miscibility of polymers is imperative for synthesizing polymer mixtures with a precise phase transition.

Organic polymers have proven that they possess good capability for the erasure of colors using the adsorption mechanism [142, 143]. These polymers have been used and they are still in use for membrane preparation. Diverse polymers; be the natural or synthetic modified with some novel materials possess enhanced effectiveness of membranes devoid of permeation and flux alteration [142]. A unique coating with steady and outstanding wettability was constructively created on the polyphenylene sulfide membrane surface through co-deposition with PEI and mussel-inspired polydopamine, which possess imperative impact for polyphenylene sulfide membrane offering outstanding effectiveness and long-standing duration. The resultant membrane gave a higher dye rejection (> 99.0%) when compared with an unmodified membrane on account of the influence of size sieving, and Donnan exclusion. [144]. In the synthesis of polypropylene composite hollow fiber membranes embedded with acrylic monomers, results attained have shown excellent dye retention, respectively having a 99.5% and 98.7% elimination of Congo Red and methylthionine chloride [145]. The cross-linking of these membranes has permitted the formation of polymeric networks with pores and the developing acid-base resistance property, which assisted the retention of dyes via the functional groups in the polymer chains [142]. For example, in the study of Xu et al [144], the membrane improved through modification demonstrated an enhanced acid-base resistance characteristic, as a result of the crosslinking reaction that occurs between catechol and amino.

In addition, organic polymers such as synthetic hypercross-linked polymers usually possess enhanced pores with controlled size and slight working conditions ready to be utilized for the elimination of dyes [146]; hence, considered prospective polymer-based adsorbents. Hyper cross-linked polymers also possess ample pore structures, ease of preparation, high chemical stability, and low cost and they can be produced by employing Friedel Craft reaction, one-step acetal reaction, and a rapid low-temperature crosslinking reaction, etc., employed for an aromatic network with developed high surface area and micro-porosity. Qui et al. [147] prepared different types of cross-linked composite membranes on commercial Polyvinyl chloride hollow fiber substrates through hyper-branched polyester as the reactive macro-monomer and glutaraldehyde in the aqueous emulsion. The membranes were prepared to investigate the impacts of the concentration of glutaraldehyde and the time of heat treatment on membrane performance. Owing to the excellent stability and the viability of backwashing treatment, the membrane composite offers a capable perception for the utilization of dye removal. Lu et al. [148] fabricated a negatively charged aromatic polystyrene fiber cross-linked membrane through a rapid low-temperature crosslinking reaction of polystyrene fiber membrane attained by the means of co-axial electrospinning, then via sulfonation. The membrane displayed an excellent performance for cationic dye removal; like excellent adsorption efficiency with a high flow rate devoid of exterior pressure,100% rejection, notable stability, and sustainability.

The cross-linking of multiple polymers in the synthesis of adsorptive polymers can afford the creation of polymeric linkages with controlled pores which subsequently support the rejection of dye aided through the functional groups in the chains of the membrane. Hence, the interaction mechanism using polymer blends for the synthesis of membranes for the erasure of dye is functional group interaction. Since the atoms in the functional group are connected with the molecules by covalent bonds, the mechanism is also hinged on covalent interaction and hydrogen bonding. The improvement of the interaction of the mechanism for polymer blends can also be attained by making the conductive membrane highly porous through the use of pore enhancers; such as integrating inorganic porous materials into the mixed polymeric blends membranes.

5.3. Use of nanoparticles and nanomaterials

A nanoparticle or ultrafine particle is a spherical particle of matter that has a diameter in the range between 1 and 100 nm. Nano-sized particles, either present in nature or the ones synthesized from diverse products, like carbon or minerals such as silver; are materials at the nanoscale that possess a large surface area to volume ratio because of the small size they possess, enhancing the adsorption efficiency of membranes [149]. These nanoparticles display exceptional physical and chemical features like the prospect of having catalytic properties and great chemical reactivity, optical, conductivity, and magnetic properties [150]. In addition, carbon nanomaterials like graphene, CNT, CS, and several other functionalized materials have demonstrated prospective adsorption capacities towards the removal of dyes, especially when incorporated into membranes [151]. Nonetheless, nanomaterials must possess at the minimum one dimension that is not up to about 100 nanometers [152]. All the properties listed above are what makes nanoparticles better at adsorption as they have an appreciable number of energetic sites for suitable interaction with other chemical species [153].

Kadhim et al. [17] employed graphene oxide nanoparticles for the modification of PES membranes and synthesized mixed matrix membranes (MMMs) for the erasure of acid black and rose Bengal dyes. The membrane performance showed a higher rejection rate of more than 99% for the two dyes. An exhibited excellent antifouling property with the integration of 0.5 wt.% graphene oxide and higher flux was preserved after an extended period of the experiment of the synthesized membrane when embedded with 0.5 wt.% graphene oxide. Koriem et al. [154] examined the impregnation of nano-activated carbon (NAC) prepared from farming waste into acetate-based membranes for dye adsorption from contaminated water. The adsorptive membrane depicted a satisfactory performance in selectively removing methylene blue dye. Zhang et al. [64] modified an adsorptive membrane by incorporating iron (III) oxide (Fe_3O_4) nanoparticles in the membrane-supportive stratum. The authors observed that the membrane maintained excellent adsorption efficacy even at the end of regeneration. This showed that the adsorption capacity of the adsorptive membrane can be upsurged by using nanoparticles to upsurge the functional adsorption sites. Shin et al. [87] reported the modification of functionalized polyvinylidene fluoride membranes by using chitosan-coated iron oxide nanomaterials (Fe-PVDF) for the effective adsorption of anionic dye from polluted water. When the conditions are neutral, Fe-PVDF revealed an excellent erasure efficacy of dye while the adsorption capacity was expressively improved when the conditions were acidic.

Furthermore, the electrostatic interaction that occurs between the positively charged chitosan and the negatively charged dye was used to explain the adsorption mechanism. Additionally, the mechanism was also explained by the attraction of the sulfate groups that the dye molecules possess, which aided the adsorption of dye on the surface of the iron oxide nanoparticles. Cellulose acetate as a biopolymer has been extensively used for the treatment of wastewater on account of its biodegradable characteristics, inexpensive, easily tailored, nontoxic, and ease of access [83, 155]. In addition, it is heat-resistant; hence, chemically and thermally stable. Inspite of all these unique properties, its relatively low adsorption efficiency hinders its employment for the erasure of dye from wastewater [156]. Hence, infusing adsorptive particles within cellulose acetate could improve membrane adsorptive capacity [157].

Additionally, graphene oxide as an adsorptive particle has been incorporated into an adsorptive membrane for the erasure of dye on account of its outstanding adsorption capacity. It is regarded as a prospective adsorbent on account of its large surface area that comprises a huge number of oxygen-functional groups. Vo et al. [84] modified an inorganic membrane by employing GO functional groups for the erasure of dye from wastewater. An exceptional performance in the recycling approach was attained. Some metal oxides have been used as adsorbents for the treatment of contaminated water; however, they possess a very low propensity to adsorb organic molecules such as dye. Hence, they can be mixed with other adsorbents and integrated into adsorptive membranes for treating dye wastewater. Wang et al. [158] introduced titanium dioxide nanowire (TiO_2Nw) and layered double hydroxide (LDH) into a reduced graphene oxide (RGO) to create a unique material. This composite material was integrated into the CA membrane to form a photocatalytic membrane dye removal from wastewater. More than 99% rejection of dye was attained. The interaction mechanism in this study would be hydrogen bonding and functional group interaction due to the presence of layered double hydroxide in the membrane.

The interaction mechanism in the synthesis of adsorptive membranes with the integrating nanomaterials for dye removal ensued from the functional group of the membrane by electrostatic interaction [81]. Hence, the adsorption mechanism of dye by adsorptive membranes synthesized with the integration of nanoparticles is hinged on hydrogen bonding interactions, electrostatic and π-π stacking interactions. The improvement of these interaction mechanisms for adsorptive membranes synthesized with the integrating nanomaterials can also be attained by making the membrane surface negatively charged such as introducing cellulose nanocrystals or the introduction of an anionic group to the membrane.

5.4 Surface modifications

The surface properties of the membrane captiously have an impact on the performance operation of the membrane because the membrane surface makes contact with the feed. Any type of modification must have the capacity to bestow the membranes with unique properties and characteristics that will convert them into ready-to-use products that are more valuable [159]. Fouling of different sorts of unwanted contaminants on the surface of any membrane has a great impact on its properties and subsequently results in an arbitrated performance. Subsequently, a pronounced effort has been dedicated to reducing the undesirable buildup of molecules on the membrane surface [160]. Hence, surface modification is an effective procedure for upsurging the membrane’s hydrophilicity. The modification of the membrane surface is usually carried out to create a hydrophilic stratum on the membrane surface that will subsequently avert the membrane surface from getting in touch with the pollutants; and consequently result in the reduction of fouling [161]. Surface modifications can be grouped into chemical and physical modifications [161].

Chemical modification is a very good approach to give a positive impact on required surface properties at the same time maintain the anticipated bulk properties of the polymer, like mechanical and chemical resistance together with the membrane morphological configuration [160]. Chemical surface modifications bestow the material with novel surface features that are not dependent on the bulk materials [162]. Usually, the principal reason for chemical modification is to convey cationic groups to the surface of the material, and this will aid the adsorption of dyes [163]. Usually, the activation of the surface base polymer chain will first take place through a chemical reaction preceding the institution of new-fangled functionalities [164]. This modification hence, offers a considerable influence on the scaffolding and features of the surfaces of materials owing to the chemical interactions that occur between the modifying agent and the surface of the material. Surface impurities could be removed from this method, producing some chemical changes. In addition, chemical modification enhances membrane wettability, and water permeability and provides resistance to fouling.

Diverse sorts of chemical agents have been employed for the modification of the surface of a membrane which changes the surface through covalent bonding that connects the required chemical moieties through carboxylation, sulfonation, amination, and epoxidation [162, 165]. Zhang et al. [166] reported an excellent super-wettable PVDF/chitosan/dopamine (PVDF/CS&DA) membrane synthesized through a facile one-step co-deposition proposed design using the catalysis of tyrosinase. The composite membrane attained an exceptional separation; with an improved permeation flux and an outstanding performance for the erasure of anionic dyes. The result of this study depicts a good high adsorption efficiency of over 90%; with an excellent anti-fouling performance. Wang et al. [59] reported a hydrophilic amidoxime-modified polymer of inherent micro-porosity (AOPIM-1) membrane for the erasure of dye from wastewater. An excellent adsorption capacity was attained. In addition, the membrane attains >99.9% elimination of several nano-sized organic molecules with an improved flux. Zhao et al. [167] designed positively charged composite ceramic ultrafiltration (UF) membranes through the grafting of aminosilane onto constricted UF membranes. The authors integrated amino functional groups into the ceramic membrane surface via covalent bonds. This resulted into making restitution for the ceramic membranes surface charge weakness. The modified membranes were employed for the erasure of four dyes and the membrane demonstrated steady separation performance.

Another type of chemical modification of adsorptive membrane is free radical graft copolymerization. Grafting is one of the polymer modification techniques by which a polymer is connected to the backbone of a parent polymer, the substrate, through chemical connections which enables surface modification [168]. Grafting enhances the morphology, physical, and chemical properties of the polymer. The membrane's physicochemical properties could be further modified through the creation of a copolymer blended with one more polymer or through grafting. In free radical graft copolymerization of modifying adsorptive membranes, free radical generations occur, then moved to the substrate for monomers to react for the creation of the grafted copolymers [169]. Zhou et al. [170] incorporated zwitterionic carboxy betaine methacrylate into the surface and pore of the PVDF membrane through free radical polymerization proposed procedure to enhance its hydrophilicity and antifouling. The ultrafiltration experiments put forward that the antifouling of the grafted membranes was significantly enhanced. Liu et al. [171] applied an innovative free radical graft and self-cross-linking copolymerization approach to synthesize a catechin-modified chitosan nanofiltration membrane for the removal of dye in wastewater. Their result attained higher rejection of dyes and an excellent dye antifouling ability was also attained.

Chen et al. [172] prepared polyacrylic acid grafted polyethersulfone composite membrane through in-situ grafting and consequently, in-situ deposition of CdS nanoparticles by submerging the membrane in S and Cd^{2+} solutions. The study observed that the membrane degradation was principally ensued from the superoxide anion free radicals. In addition, the membrane demonstrated outstanding rhodamine B dye rejection and the effectiveness of self-cleaning with the aid of visible light was observed to be excellent. Kuar et al. [173] reported the preparation of a green PVA-co-poly(MAA) adsorbent through free radical polymerization. Though the adsorbent is not a membrane, it, however, attained excellent regeneration efficiency for methylene blue removal from wastewater. In all, free radical graft copolymerization of monomers to the surface of the membrane or other adsorbent is a vital route to the erasure of dye from wastewater. However, the nature of the initiator is important as the initiator concentration affects the rate of grafting. In addition, the solubility of the monomer in the solvent is an imperative parameter because the swelling of the polymer backbone in the solvent aids in enhancing the grafting effectiveness, and the swelling of the backbone eases the access of the monomer to the active locations of the membrane for adsorption of dyes. Finally, the reactivity of the monomer is imperative in grafting as the concentration of the monomer influences the grafting efficiency. Apart from chemical modification, physical modification has been considered an excellent approach to give a functional enhancement to the operational effectiveness of membranes.

Physical modification is a common method that aids in enhancing the antifouling characteristics of membranes; this method encompasses coating the surface of the membrane with a sacrificial stratum that will serve as a shielding liner that suppresses the buildup and adsorption attraction of the foulants on the membrane surface [174]. This type of modification can be attained by directly coating the membrane with a hydrophilic polymer or by coating the membrane via the solution of chemically active monomers [161]. In addition, coating the membranes with the utilization of materials such as nanofillers and polymers via weak van der Waals or hydrogen forces gives an instance of modification through physical means [175, 176]. The main purpose of putting a coat on membranes on the top of the interfacially polymerized stratum, such as the method of grafting, is to synthesize membranes that will be more hydrophilic together with a reduction in the surface roughness. Coating could be absorbed on the surface, covalently connected to the membrane surface, in addition, it could also be cross-linked with itself [176]. The coating also makes the surface of the membrane either have positive or negative charges. This method also upsurges the ineradicable properties of membranes. Furthermore, changeable coating can also protect the membrane physically, in the course of fabrication and improve its antifouling properties [176].

Physical modification by directly coating the membrane with polymer is particularly a surface adsorption process that could be swayed via the polymer morphology on the interface [177]. In addition, the modification has shown that hydrophilic modifiers are present on the membrane surface through physical interaction. This approach is adaptable and it is a convenient process done under mild conditions to improve the hydrophilicity and membranes’ performance [178]. Hence, surface alteration can be accomplished through physical modifications that do not need any chemical interactions [179]. Xi et al. [180] reported the coating and polymerization of 3,4-dihydroxyphenylalanine and dopamine for surface modification for polyethylene, polyvinylidene fluoride, and polytetrafluoroethylene porous membranes. The membrane's surface hydrophilicities were assessed. It was established that there was a remarkable decrease in the water contact angle of the modified membranes, signifying that the membrane hydrophilicity was considerably enhanced. Qin et al. [177] reported the impacts of stationary and pore-flowing techniques for modifying polyacrylonitrile ultrafiltration. The capacities of some polymers to create a coating stratum were initially assessed and the polymers exhibited an excellent antifouling property. A compact and well-leveled stratum through a slack and arbitrary stratum was created, which altered the flux and antifouling properties of the membrane. However, physical modification application is restricted because they are recommended to flat sheet membranes on the account of their suitability, rather than hollow fiber membranes [177]; it is however done with ease if the exterior surface of the hollow fiber membranes is coated by using dilute solution coating to improve membrane hydrophobicity. Li et al. [181] reported a slack and porous hydrophobic zeolitic imidazolate frameworks-71/PVDF coating stratum deposited on the external surface of PVDF hollow membrane using the means of diluting solution coating to improve membrane hydrophobicity for the erasure of Congo red using vacuum membrane distillation (VMD). The rejection of the dye depicts a minor alteration; however, remains above 99.9%. At the end of the modification process, surface area, roughness, surface charge, surface energy, hydrophobicity, and functional groups, reactivity will be enhanced [182].

For coating of the membrane via the solution of chemically active monomers, a solution of chemically active monomers will be firstly submerged by the polymeric membrane, or a coat can equally be applied using the membrane. Subsequently, they are fixed on the membrane surface via crosslink or polymerization reaction, devoid of upsetting the chemical configuration of the membrane sub-structure [162]. Monomers in active stratum have an impact on the cross-linking density, thickness, pore structure, and surface properties of the membranes which in turn impact the performance of membrane filtration [183]. The existence of hydrophilic monomers in membranes could also affect the membrane durability. Hence, introducing hydrophilic materials on the membrane surface would give the membrane the capability to hinder fouling; thus, upsurging the membrane service life [184]. The most commonly used reactive monomers are water-soluble diamines like piperazine, tannic acid, p-phenylenediamine; and acid chloride monomers like isophthaloyl chloride and trimesoyl chloride [185]. However, tannic acid is usually chosen owing to its oxidation resistance, antienzymatic, astringent, anti-bacterial with outstanding anti-fouling properties, these properties avert the degradation of membranes. In addition, trimesoyl chloride is employed as an acyl chloride monomer for the creation of thin films on the porous support coating [186]. Chen et al. [187] reported the synthesis of negatively charged polyether sulfone membranes for the erasure of dye, employing two types of vinyl monomers (acrylic acid and sodium styrene sulfonate) for the functionalization of PES through in-situ cross-linking copolymerization. The results showed that membranes depict effective adsorption capacity for methylene blue. Zhang et al. [188] designed an interfacial polymerization (IP) reaction scheme to make the Polyamide stratum on the polysulfone support. An innovative acyl chloride, trimellitic anhydride chloride (TAC) together with an anhydride group, were then instituted into the piperazine/1,3,5-benzenetricarbonyl trichloride (PIP/TMC)-based IP system, to prepare a TFC NF membrane with enhanced permeability. The resulting membrane showed an outstanding water-salt separation. Kumar et al. [186] used monomers (tannic acid (TA) and m-phenylenediamine (MPD) to synthesize a novel polyether sulfone (PES) based nanofiltration membrane to treat common effluent in textile wastewater. The membrane depicts excellent water permeability and rejection of salt, and the water can be further reused in the industry of agricultural irrigation. In addition, the modified membrane possesses a boosted anti-fouling property and outstanding hydrophilicity.

The interaction mechanism in modifying the surface of adsorptive membranes for dye removal with the utilization of the carboxylate group yields negative interfacial charges and offers colloidal steadiness to take out cationic dyes through electrostatic interactions. While the use of epoxide and hydroxyl functional groups in the plane could interrelate via noncovalent interactions. Hence, the interactions between them could be of pronounced significance to the resultant properties of the efficient hybrid adsorptive membrane linkages [84]. The improvement of the interaction of the mechanism for polymer blends can also be attained by making the conductive membrane highly porous through the use of pore enhancers; such as integrating inorganic porous materials into the modified membranes.

Literature has hence, proven that surface modification of adsorptive membranes has resulted in desirable surface properties that provide the membranes with excellent fouling resistances which will subsequently improve the membrane performance and durability. A summary of recent works published in the year 2023 concerning mechanisms, separation efficiency, types of modification methods, and types of additives are depicted in Table 2.

Table 2
Summary of recent works, mechanisms, types of modification methods, types of additives, and separation efficiency

Recent works	Mechanisms	Types of modification methods	Type of additives	Separation efficiency	References
Self-assembled silk nanofibrils doped with different ratios of palygorskite to synthesize composite membranes using Genipin as a crosslinking agent.	Adsorption-assisted pore size filtration.	Functional modification	-	Excellent flux and high percentage rejection were attained for anionic dyes.	[189]
The modification of GO via granules of blast furnace slag derived from geopolymer was used to coat stainless-steel mesh for the preparation of an inorganic geopolymer membrane.	Participating in activities of physical retaining and electrostatic adsorption	Nanomaterial modification (Graphene oxide)	Graphene oxide	The adsorption rate of Methylene blue (MB) surpassed 91% in 4 hours.	[190]
Development of an inventive membrane made of blends of PVC and nanocellulose reinforced with titanium aluminate ($TiAl_2O_4$) nanoparticles for dye removal.	Adsorption mechanism	-	$TiAl_2O_4$ nanoparticles	The nanocomposite membrane exhibited 98.6% rejection of MB dye.	[191]
The utilization of a byproduct in the manufacturing of carpets (acrylic fibers waste), for the synthesis of membrane for the erasure of dyes	Adsorption mechanism	Chemical modification	Polyvinyl pyrrolidone (PVP)	The addition of 5% PVP raised its water flux to 320 $Lm^{-2}h^{-1}$ and attained 90% rejection of MB	[192]
Development of chitosan, polyvinyl alcohol, and cornstarch integrating nanocellulose (CPCN) film as an alternative method for the removal of methylene blue dye.	Adsorption mechanism	Modification by use of nanomaterials	Nanocellulose	Excellent adsorption of MB was attained at pH 10.	[193]
Synthesis of Polyvinyl chloride PVC/PbO-graphite membrane for photocatalytic dye-degradation activity	Adsorption mechanism	Modification by use of nanomaterials	Lead(II) oxide (PbO)-graphite	The ACM was employed for the photodegradation of Congo red dye degraded about 71 % dye in 50 min.	[194]
Designed of spinnable highly protonated montmorillonite composites.	Adsorption mechanism via electrostatic attraction present between dyes active groups and the energetic locations on the surface of nanofiber	-	Montmorillonite	The adsorption capacity of the membranes was remarkably improved when the content of Montmorillonite was raised.	[195]
Development of composite films for effective investigation in removing dyes under diverse experimental conditions.	Surface adsorption mechanism	-	-	The two films prepared demonstrated excellent adsorption capacities.	[196]
Synthesis of graphene oxide (GO) incorporated into nanostructured polyethersulfone (PES) membranes for dye removal.	Adsorption mechanism	Physical modification	GO	The membrane demonstrated 85% performance at the end of 2 h contact with both microorganisms and an enhancement in colorant retention at the end of two and four cycles of filtration.	[197]

6. Proposed future direction of adsorptive membranes

In view of the drawbacks of adsorptive membranes. Several modification methods have been discussed. However, researchers need to look in the direction of making well-stable membranes, which will also possess less fouling properties and extended cleaning operations concerning dye wastewater treatment in the future. One way of achieving this is the use of ceramic adsorptive membranes. Ceramic membrane offers the benefits of narrow pore size distribution, thermal and chemical stability, and excellent mechanical strength which will in turn allow back flushing, high porosity and consequently, high flux; and extended life span [198]. Hence, the use of solid waste in synthesizing ceramic adsorptive membranes is proposed because ceramic membranes synthesized from solid wastes could possess catalytic properties devoid of the need to load additional catalysts in the course of synthesis. In addition, the waste-synthesized ceramic adsorptive membrane could possess multiple functions such as catalytic and/or twofold separation-adsorption properties [199]. All these properties will result in high efficiency of the erasure of dyes from wastewater.

It is therefore imperative to find more efficient solid waste treatment approaches to have reduced particle sizes, have well-ordered distributions of particles, and work on improving the purity of the solid waste; this will combat the drawback of the trade-off that lies between nanometer-level selectivity and permeability. This will also aid in achieving excellent ceramic membranes with well-controlled pores which will in turn magnify the utilization of waste-derived adsorptive membranes for diverse applications. In addition, researchers should also look into investigating the sustainability of solid wastes used in synthesizing adsorptive membranes; hence, increased efforts ought to be dedicated to the transformation of waste-derived ceramic adsorptive membranes. Nanoparticles that will aid the filtration-adsorption characteristic of adsorptive membranes can be synthesized from agricultural wastes. It is also important to study the interaction mechanisms between dye and waste-derived adsorptive membranes. Though, one would believe that the ceramic adsorptive membranes synthesized from waste would be cost-effective; however, it is important to do the overall cost breakdown and life cycle assessment of these adsorptive membranes. Finally, it is also imperative to find out if the waste materials are hazardous as this will in turn affect the adsorptive membrane in the course of its long-term operation [199]. Hence, to an increasing extent, future research should focus on the sustainability of waste-derived ceramic adsorptive membranes.

7. Summary and conclusion

This review made available a readable fusion of the up-to-date resources available in the literature on the surface modification and incorporation of organic/inorganic additives into the membrane matrix. Up-to-date resources were also used to give a rundown on the governing interaction mechanisms of dye adsorption on adsorptive membranes. The summaries of findings are given in Table 3.

Adsorptive membranes have shown their effectiveness in the adsorption of different dyes from wastewater. Hence, adsorptive membranes are considered prospective technology as they make use of the adsorption process and separation process. The governing interaction mechanisms that aid their performance for the erasure of dyes from wastewater are found to be electrostatic interaction, functional group interaction, hydrophobic interaction, ionic and cation-π interactions, π-π interaction, and covalent interaction. Different types of adsorptive membranes and their performances have been discussed; however, adsorptive membranes are racked with some drawbacks. Though researchers have come up with different modification techniques to prevail over the downsides accompanied by employing adsorptive membranes for dye erasure; it is important to select an appropriate modification method that will help in optimizing the membranes for the proposed application. This technology, however, requires lots of investigation to make it sustainable and scale it up for industrial applications. The future direction of adsorptive membranes was also discussed. Thus, future research should focus on the sustainability of adsorptive membrane applications for the erasure of dye from wastewater.

Table 3
The summary of the findings

Purpose	Type of resources	Summary points
To Identify interaction mechanisms between dye molecules and membranes	Journal articles	The interaction mechanisms are electrostatic interaction, functional group interaction, hydrophobic interaction, ionic and cation-π interactions, π-π interaction, and covalent interaction.
To put forward adsorptive membranes as prospective technology for dye wastewater treatment	Journal articles	Adsorptive membranes are described via their strong attraction for molecules and ions, by way of combining ions through ion exchange, chelation bonding, and complexion. Hence, the reaction between the membrane and the solute constructs an interior spherical complex bound which will subsequently bring out a permeate of sieved water from the membrane.
To identify kinds of adsorptive membranes	Journal articles	The different types of adsorptive membranes are characterized by performance efficiency such as high removal efficiency, high water flux, high adsorption rate, and suitable reusability.
To identify the shortcomings of adsorptive membranes.	Journal articles	Different modifications have been used to combat the drawbacks of adsorptive membranes.
To propose the future direction of adsorptive membranes	Journal articles	The future direction of adsorptive membranes was proposed.

Data availability

Not applicable.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRedit authorship contribution statement

F.A. Akinyemi: Investigation; Methodology; Resources; Writing – original draft; Writing – review & editing.
O. Agboola: Conceptualization; Formal analysis; Funding acquisition; Project administration; Resources; Supervision; Writing – review & editing.
O. Oladokun: Resources; Supervision.
E.E. Alagbe: Resources; Supervision.
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