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Review

Recent Advances in Plasmonic Chemically Modified Bioactive Membrane Applications for the Removal of Water Pollution

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Abstract: Population growth has reduced the available freshwater resources and increased water pollution, leading to a severe global freshwater crisis. The decontamination and reuse of wastewater is often proposed as a solution for water scarcity worldwide. Membrane technology is a promising solution to the problems currently facing the water and wastewater treatment industry. However, another problem is the high energy costs required to operate systems which use membranes for water treatment. In addition, membranes need to be replaced frequently due to fouling and biofouling, which negatively affect water flow through the membranes. To address these problems, the researchers proposed membrane modification as a solution. One of the exciting applications of plasmonic nanoparticles (NPs) is that they can be used to modify the surface of membranes to yield various properties. Positive feedback was reported on plasmonic-modified membranes as means of wastewater treatment. However, a fundamental gap exists in studies of plasmonic membranes' performance and applications. Given the importance of membrane technology for water and wastewater treatment, this paper reviews recent advances in the development of plasmonic chemically modified bioactive membranes and provides a perspective for future researchers interested in investigating modified membranes.

Keywords: plasmonic NPs; wastewater treatment; bioactive membranes; membrane

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1. Introduction

Population growth has reduced available freshwater resources and increased water pollution, resulting in a severe global freshwater crisis [1]. Therefore, there is a need to develop alternative freshwater supply methods beyond the hydrologic cycle's capabilities to meet the world's growing freshwater needs [2]. The uncontrolled discharge of various waste materials and drugs such as ibuprofen, acetaminophen, and antibiotics has created unfavorable and dangerous environmental conditions. Pharmaceuticals such as diclofenac and ibuprofen are poorly and slowly mineralized through photocatalysis. Thus, such materials accumulate in the environment and circulate through water resources in the ecosystem [3–5]. Detoxifying and reusing wastewater has been widely proposed to solve water scarcity worldwide. As the global population grows, sustainable water treatment technologies need to be developed to meet the increasing demand for clean water [6]. Nowadays, membrane technology is a key focus of interest among researchers.

A membrane is a solid or liquid between two systems which allows the selective transport of matter and energy. Modern nanostructured membranes are highly efficient and easy to use, exhibit high permeability and selectivity for transporting specific molecules, and require low energy inputs. They are also modular, stable under various operating conditions, and easy to control and scale up. The use of membrane technology in separation processes can provide an alternative to energy-intensive methods for the selective and effective separation of specific components [7]. However, membrane technology still faces critical challenges. Solving the problems of membrane technology requires a multidisciplinary approach and collaboration with disciplines such as solid-state physics. In particular, the use of thermoplasmonics is an exciting option in the development of membrane technology [8,9]. Thermoplasmonics is the thermal heating of metal nanoparticles (NPs) by optically resonant plasmonic excitations and is based on the control of nanoscale thermal hotspots by light irradiation [8,10–17]. Plasmonic NPs can act as ideal confined nano-heat sources within polymer membranes and convert absorbed light radiation into heat [8,18,19]. The energy of plasmons can be converted into heat, increasing the temperature of the surrounding medium [8,20–22]. The evaporation of water using solar energy is a promising and environmentally friendly method for water and wastewater treatment.

The enhancement of steam generation in solar steam generators by a plasmonic heating effect has been shown to be practical. In such systems, redundant heating of the liquid is a limiting factor for efficiency, and the commonly used noble metal nucleotides are not cheap or readily available and require extensive preparation. Studies have shown that simple thermal evaporation is possible using a paper-like plasmonic microporous membrane with fabricated indium NPs. Indium NPs are lightweight, absorb light very well, and are excellent for plasmonic heating. Therefore, indium-based plasmonic membranes can be used for wastewater treatment in places with limited or no electricity access [23,24]. A wide range of desired membrane properties for various applications can be achieved by modifying the surface of textile membranes. Plasma treatment is proposed as a clean alternative to harmful chemical methods of membrane surface modification. Unlike chemical surface modification methods, plasma treatment does not produce hazardous waste products. The electrospinning process can achieve a different level of adhesion between nanofiber layers and the pad. This way, several unique membrane properties can be achieved, such as higher numbers of active sites, ionic capacities, and wear resistance [25–27].

In addition to using modified membranes for wastewater removal, various other water and wastewater treatment methods exist. Some of the most significant water and wastewater treatment methods are osmosis, oxidation, chemical precipitation, solvent extraction, micellar ultrafiltration, organic and inorganic ion exchange, and adsorption. An exciting option for removing contaminants from water is the adsorption method. The simplicity of the processes involved in the adsorption method and the availability of cheap adsorbent materials have attracted the attention of researchers to this method. Gładysz-Płaska et al. [28] investigated the usage of red clay as an adsorbent of uranium(VI) and phosphate ions. Uranium metal is a toxic heavy metal and a potent source of radioactivity. Uranium and its compounds naturally exist in the environment and water resources; however, in recent years, civil and military nuclear activities have increased uranium concentrations in the environment beyond safe levels [29,30]. Gładysz-Płaska et al. [25] demonstrated that the adsorption method is promising for the inexpensive and effective removal of many toxic materials, such as heavy metal ions, from the environment and water resources. In another study by Gładysz-Płaska et al. [31], the usage of an inorganic/organic hybrid uranium(VI) adsorbent consisting of halloysite functionalized with isothiouronium salts was investigated. It was shown that the adsorption capacity could be increased by modifying the halloysite with the isothiouronium salts, especially those with four nitrogen atoms in their structure.

The adsorption of lanthanides by adsorbent materials can be investigated as a model to obtain information about interactions between the adsorbent materials and radioactive materials and the performance of the adsorption method in the removal of such materials, which is due to similar chemical properties of lanthanides and radioactive actinides. Gładysz-Płaska et al. [31] investigated the usage of raw and unmodified red clay as an adsorbent for lanthanides. Based on the studies, thermally modified clay (T-clay) is the most active lanthanide sorbent among the different forms of red clay. The adsorption of lanthanide on red clay was also modeled by Gładysz-Płaska et al. [31] based on the molar fractions of individual lanthanide complexes as a function of pH. However, further experiments are required to refine the provided model. Barakat et al. [32] investigated the usage of bentonite/sawdust interfaces loaded with NPs of magnetite (Fe_3O_4) as adsorbents for the removal of methylene blue (MB). It was found that the composites prepared using NPs of Fe_3O_4 were successful at effectively removing MB through adsorption.

Energy can be harnessed from renewable resources and used for water treatment. Solar desalination and water treatment is an environmentally friendly method for water purification, which various researchers in recent years have studied extensively. Elsheikh et al. [33] reviewed the usage of heat exchangers in solar desalination. Based on studies by various researchers, it was concluded that using heat exchangers as means of waste heat recovery in solar desalination systems improves their performance. Based on another study by Elsheikh et al. [34], it was concluded that using nanofluids in systems which utilize the sun as an energy source increases the heat capacity and the absorption of solar energy. The major problem with solar desalination systems is that they are costly and complex. Increasing the amount of bulk water is a significant challenge in conventional solar desalination systems due to the reductions in their efficiency in such conditions. The efficiency reduction is because the bulk water dissipates a considerable amount of heat. To address these issues, using a thin film of water is suggested in solar steam generation systems instead of vast amounts of water. Elsheikh et al. [35] reviewed the thin film technology for solar steam generation. Investigations have showed that using thin films of water in solar steam generation units can make achieving high efficiencies possible.

Microorganisms can be used in membranes to achieve the bioremediation of wastewater. Bioremediation offers the possibility of destroying or rendering various harmless pollutants through natural biological activity. Relatively inexpensive techniques with low technical complexity are used, which generally have high acceptance among the population and can often be carried out on-site. Due to the importance of membrane technology for water and wastewater treatment, this paper reviews recent advances in developing plasmonic chemically modified bioactive membranes.

2. Overview of Plasmonic Membrane Technology

Textile filters are an essential component of many industrial processes. Their main task is the separation of solids from liquids or gases. In this way, they contribute to the purity of final products, energy consumption, the efficiency of the processes in which they are involved, the recovery of valuable materials, and the controlled release of pollutants into the environment. The methods used to classify membrane materials depend on many factors. Some of these factors include flexibility or hardness, the effect of gravity on the filtration process, the intended use, the regeneration method, the manufacturing method of the membrane itself, and the geometry of elements inside the filter [8,25].

Regarding flexibility, membrane materials and filter media can be divided into solid, bulk, and composite. Solid filters are composed of solid particles such as metals, ceramics, or activated carbon. The solid particles of filter media are bonded together by a specific type of fastener. Bulk filters, such as solid filters, comprise solid particles, but their manufacture does not use binders, pliable metals, non-metallic fabrics, knitted and non-woven surface textile products, or perforated polymer films. Composite filters consist of several elements. Woven fabrics, non-woven fabrics, and solid particles with and without binders can be used to develop composite filters. Composite filters are fascinating because of their versatility and the possibility of using them simultaneously as reinforcing and geotextile materials [8,36].

Filter barriers can also be used for purification [25,37–39]. In the remediation of industrial wastewater, it is necessary to use appropriate water treatment methods which lower the concentration of toxic intermediates [40–43]. Membranes used in water treatment can remove various contaminants, from large colloids, algae, and bacteria with a characteristic size in the order of micrometers, to individual ions with a hydration radius in the order of angstroms [44,45].

One of the earliest examples of the modification of the transport properties of membranes by surface modification can be found in W. Pfeffer's monograph of 1877, which describes the formation of a layer of copper ferrocyanide on the surface of porous porcelain. The membranes in Pfeffer's work were prepared by saturating porous porcelain with a copper sulfate solution (II). One side of the porcelain was then kept in contact with a potassium ferrocyanide solution. These procedures resulted in semipermeable copper ferrocyanide membranes [46,47]. Pfeffer's membrane preparation method is similar to what was reported over 100 years after Pfeffer's work as the preparation method for composite polyamide membranes RO.

Ceramic Pasteur–Chamberland water filters were first modified in 1896 by C. J. Martin using gelatin or silica [48,49]. Advances in membrane manufacturing processes continued, and in 1928, membrane technology became attractive for industrial use; modern membrane manufacturing processes were developed in the early 1980s. Since then, many membrane researchers have focused on improving the properties of basic membrane architectures through surface modification. Currently, there are several surface modification techniques, such as chemical treatment, plasma treatment, and UV irradiation. The membrane surface modification methods can be used alone or in conjunction with other techniques [36,50]. As mentioned above, one of the key membrane surface modification methods is plasma treatment, which is widely used in polymer surface modification. In plasma treatment, the surface of the membrane is exposed to various plasmas to achieve a hydrophilized membrane surface [51–54]. Plasma can also activate the membrane surface, and the desired modification can be carried out by other methods [52,55,56].

Plasma membrane modification has been addressed in several publications [36,51,52]. In some of the published articles, researchers evaluated the performance and properties of plasma-modified membranes [36]. However, little information is available about the performance of membranes modified using microwave-driven plasma-associated processes such as plasma-induced polymerization [57,58]. Case studies published between 2000 and 2010 highlighted the performance and properties of membranes modified using low-pressure plasma processes [59–61]. Plasma-modified polymeric membranes have been reported to exhibit antifouling properties [62]. In addition, some published articles on plasma-modified membranes have highlighted how the surface hydrophilicity of water purification membranes can be improved. Modification processes for low-pressure and atmospheric-pressure membranes are also being studied in the context of pure plasma physics and chemistry [36,60,63,64].

3. Plasmonic Membranes

A plasmon is a quantum of vibration of plasma. The energy of a plasmon can be defined as $E_p = h\omega_p$, where h is Planck's constant and ω_p is the characteristic frequency which depends on the plasma's mass, density, and charge. Exploiting the properties of collective modes in conducting systems has led researchers to investigate the applications of plasmons. Thus, a new field of research, a subfield of nanophotonics, plasmonics, was defined [65,66]. Metals consist of solid positive ion cores and mobile conduction electrons; thus, the study of the properties of metals from the electromagnetic point of view leads to the conclusion that metals are plasmas [67,68]. Thus, NPs of metals can be used as the plasma required for modifying membranes. Membranes modified with metallic NPs are called plasmonic membranes. Plasmonic metallic NPs can convert light from various sources, such as the sun, into heat. Therefore, plasmonic membranes can be used in photothermal water purification systems [8]. The most commonly studied plasmonic metallic NPs are noble metals such as gold (Au) and silver (Ag). Although the use of gold and silver NPs for membrane modification is preferred due to the exceptional plasmonic properties of gold and silver, several problems limit the widespread use of gold- and silver-modified membranes in the industry [69,70]. One main problem is the price and availability of gold and silver metals. In the case of silver-modified membranes, there are safety and environmental issues due to the probability of silver NPs leaching into the water. Leached silver particles can harm humans and other living organisms, especially aquatic life.

Therefore, some researchers have recently investigated alternative plasmonic materials to gold and silver metals. For example, Ren and Yang [71] proposed using NPs made of metal nitrides as substitutes for noble metals. One of the most studied metal nitrides is titanium nitride (TiN) [72]. Naik et al. [73] introduced TiN NPs in 2011, and Patsalas et al. [70] listed the advantages of using TiN NP instead of gold and silver NPs. The main advantage of using TiN NPs is that titanium and its compounds are cheap and abundant compared with Au and Ag and have similar plasmonic resonances as Au NPs. TiN NPs can also withstand high temperatures. A theoretical study by Lalisse et al. [44] found that TiN NPs exhibit better or the same thermoplasmonic properties as Au NPs under the same illumination conditions.

It was observed that membranes modified with plasmonic metallic NPs have antifouling properties. Therefore, the use of different plasmonic materials to achieve antifouling properties has been investigated by researchers. For example, Yang et al. [74] proposed a membrane modified with copper–zinc–tin–selenide (CZTSe) nanocarambolas. The synthesized CZTSe nanocarambolages were deposited on the surface of a hydrophilic membrane in a solar-powered interfacial water evaporation system. The modified membrane could be operated for over 30 days without decay or loss of efficiency. Amoli-Diva et al. [75] proposed two new types of composite membranes with antifouling and anti-biofouling properties. These membranes were prepared by modifying commercial polyamide membranes (PA) with synthetically prepared biplasmonic Au–Ag and Ag–Au photocatalysts. Shen et al. [76] proposed using a plasmonic p–n heterojunction of $Ag/Ag_2S/Ag_2MoO_4$, which exhibits enhanced photocatalytic activity under visible and infrared radiation for the purification of wastewater. The use of Ag/Ag_2S/Ag2MoO4 can remove 99% of rhodamine B (RhB), 100% of methylene blue (MB), 83% of tetracycline (TC), and 77% of hexavalent chromium (Cr (VI)) under visible light irradiation. Up to 62% RhB, 58% MB, 52% TC, and 50% Cr(VI) can be removed by $Ag/Ag_2S/Ag_2MoO_4$ under near-infrared illumination generated with an 808 nm laser light source. The sensitivity of such photocatalysts to visible and infrared radiation has attracted the attention of researchers due to the high utilization efficiency of solar energy. Thus, membranes modified with such materials may provide a sustainable method for purifying wastewater using renewable energy sources such as solar energy. Another attractive property of $Ag/Ag_2S/Ag_2MoO_4$ is that it can be used as an efficient and recyclable photocatalyst.

Plasmonic Chemically Modified Bioactive Membranes

As mentioned above, most of the developed plasmonic materials used to modify membranes are toxic to humans and other life forms. Therefore, it is necessary to develop bioactive plasmonic materials. Fungi, bacteria, and algae can be used to biologically treat wastewater, such as those contaminated with dyes from the textile industry. Therefore, biological treatment methods have attracted the attention of researchers in recent years. Biological treatment is advantageous because it is less energy-intensive and environmentally friendly. The processes associated with biological treatment methods are simple and less sludge is produced [77–79]. Srikanlayanukul et al. [80] studied the application of the fungus Coriolus versicolor for the biological treatment of textile wastewater. Immobilized Coriolus versicolor was applied to the polyurethane foam surface to treat the wastewater in an aerobic bioreactor. The objective was to decolorize the wastewater. It was found that using Coriolus versicolor on the foam removed 67% of the chemical oxygen demand (COD), and 80% of the dye dissolved in the wastewater within 48 h. Selvakumar et al. [81] studied the use of the fungus Ganoderma lucidum in the treatment of textile wastewater. It was found that 81.4% of the dye and 91.3% of COD in the wastewater were removed in 5 days in a batch reactor using Ganoderma lucidum. Kıvanc and Doğruer [82] investigated the application of Aspergillus flavipes. It was found that 92% of the dye and 45.92% of COD were removed from the water within one week. However, the biological wastewater treatment methods could not achieve the discharge standards required for textile wastewater. Therefore, Isik et al. [83] developed a bioactive ultrafiltration membrane from the fungus Aspergillus carbonarius, which is filamentous. The ability of the developed bioactive ultrafiltration membrane to treat wastewater was tested using a sample of industrial textile wastewater. The performance of the developed membrane was then evaluated in terms of the decolorization capacity and COD rejection of fungal biomass in batch reactors. The developed bioactive membrane material achieved 91% decolorization and a 73.2% reduction in COD. Figure 1 shows the process used to prepare the bioactive ultrafiltration membrane, as described in the study by Isik et al.

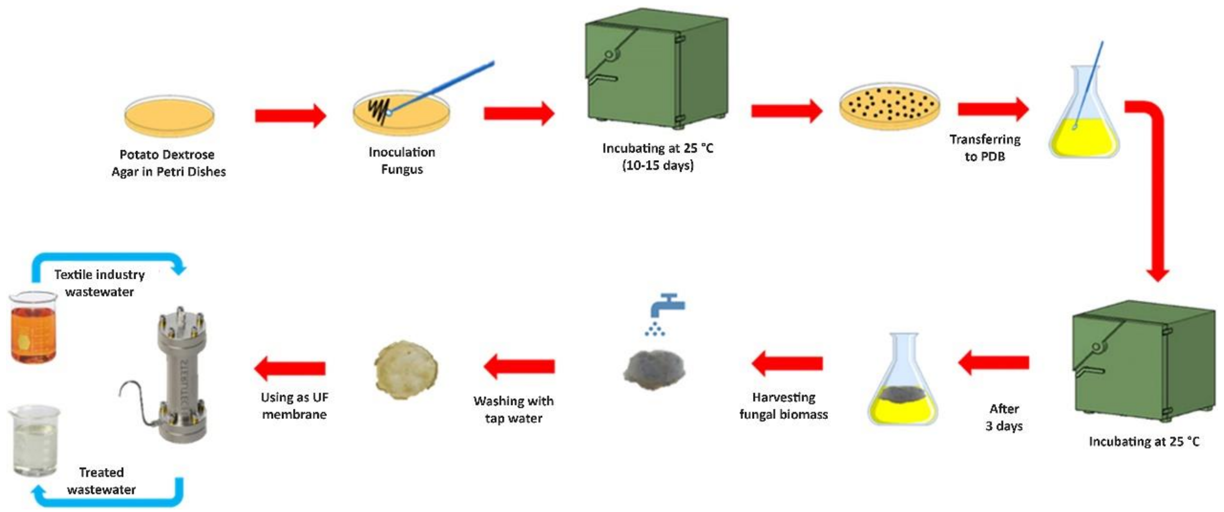

Figure 1. The preparation procedure of a bioactive ultrafiltration membrane. Reprinted with permission from [83]. Copyright © 2019, Elsevier.

The main drawback preventing the use of bioactive membranes made from fungi or other organisms is that most industrial wastewater contains substances which are toxic to all life forms. For example, industrial wastewater is usually not biodegradable due to impurities such as dyes, heavy metals, salts, and nitrogen and phosphorus compounds, e.g., in some detergents [84–86]. Sulfur is an abundant, cheap, and biologically active element [87–89]. Therefore, a candidate for solving the problem of bioactive membranes made from different organisms is the use of different forms of sulfur. Sulfur is in the same group of the periodic table of elements such as oxygen, but is much weaker in terms of its electronegativity. Therefore, the electrical conductivity of sulfur is poor, which is the main problem in using sulfur as a bioactive agent in plasmonic membranes. Several methods have been proposed to solve this problem. Some of the proposed methods include using composite materials composed of sulfur and doping other materials with sulfur. The combination of sulfur or nanosulfur with carbonaceous materials, metal oxides, conductive polymers, polar inorganic materials, and transition metal sulfides is known for its excellent electrochemical performance. It is believed that using complex structures with plasmonic properties is even more desirable than single particles for some applications due to the strong coupling between the plasmons of individual particles [89–93].

Plasmonic NPs used to modify membranes can be prepared from the leaf extracts of some plants containing bioactive substances such as alkaloids and phenolic compounds. The anti-inflammatory, anti-tumor, and antibacterial properties of the extracts of such plants make their use interesting for the preparation of bioactive plasmonic membranes. One of these plants is Crinum latifolium (CL). Vo et al. [56] used the aqueous extract of the leaves of CL to biosynthesize silver and gold NPs. It was found that the synthesized CL–Ag NPs could strongly inhibit the action of four bacterial strains. Figure 2 shows the biosynthesis of CL–Ag NPs and CL–Au NPs and their applications for wastewater treatment [94].

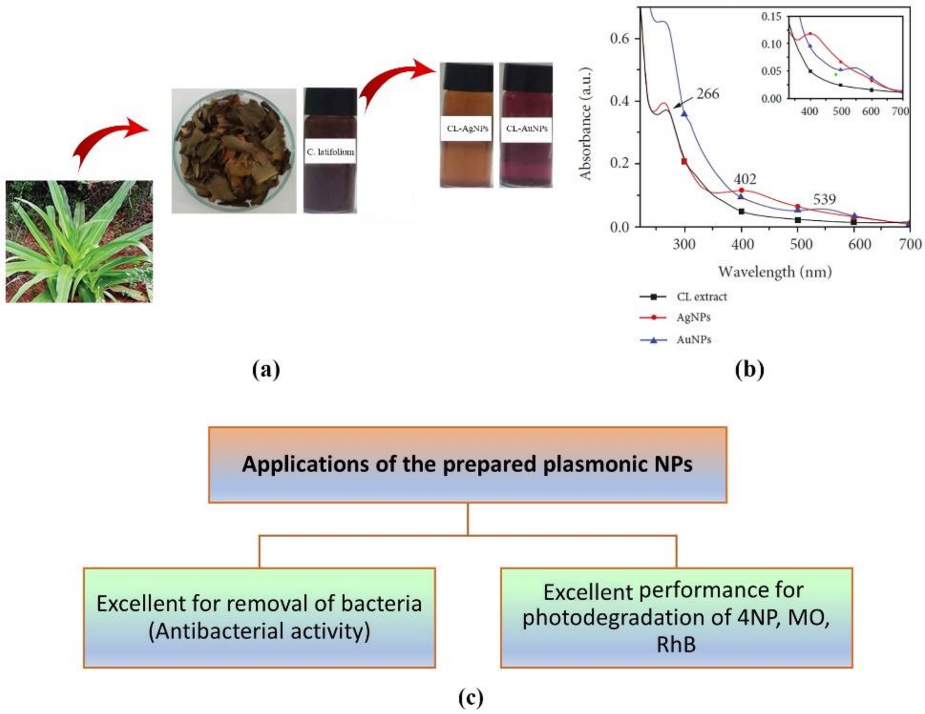

Figure 2. Biosynthesis of CL–AgNPs and CL–AuNPs explained by Vo et al.: (**a**) preparing the extract of oven-dried leaves of CL and modifying it with Ag and Au NPs; (**b**) comparing the light absorbance of different compositions of the modified CL-based NPs; and (**c**) applications of the prepared Plasmonic NPs in wastewater treatment. Reproduced with permission from [94]. Copyright © 2019, Hindawi.

Plant waste can be recycled and converted into plasmonic bioactive hybrids. Barbinta-Patrascu et al. [95] used environmentally friendly approaches to synthesize and develop novel plasmonic biohybrids which can be used as bioactive coatings. The garden herbs Mentha piperita and Amaranthus retroflexus were used to produce Ag NPs, and lemon peels were used to obtain pectin. The Ag NP and the extracted pectin were then used to prepare a pectin-coated bio-nanosilver. The prepared nano-biohybrids were found to be spherical nanoscale particles. Studies on the biological performance of the prepared plasmonic materials in terms of antibacterial properties showed that the proposed novel pectin-based hybrid coatings were impressively effective against Escherichia coli bacteria. The studies of the antioxidant properties of the novel pectin-based plasmonic hybrid coating showed that the developed coating was capable of removing 96.1% to 98.7% of short-lived and 39.1% to 91% of long-lived free radicals. The obtained results were promising, and the bioapplication of such coatings in situations where potent antioxidant and antimicrobial properties are important has been proposed [95–97]. Thus, such materials can be used as coatings for membranes in the water and wastewater treatment industry to eliminate pathogens and carcinogenic free radicals.

4. Plasmonic Membranes

Researchers have recently investigated various applications of plasma-modified and plasmonic membranes. Most of these studies suggest using plasmonic membranes in thermal, adsorptive, and photocatalytic water purification and wastewater treatment processes. The applications of plasmonic and plasma-modified membranes are discussed in the following sections.

4.1. Application of Plasmonic Membranes in Thermal Water Treatment Methods

Water can be purified using thermal methods. Some commercially available thermal systems for water purification are multi-stage flash technology (MSF) and multi-effect distillation (MED), but these systems require a lot of thermal and electrical energy for their operation [98–100]. Membrane filtration processes such as reverse osmosis (RO) have been developed to solve the problems of conventional thermal evaporation methods for water purification. The key to the success of RO technology is that 3 to 6 kW h m^{-3} of power is required to overcome the osmotic pressure of saltwater, compared with the 15 kW h m^{-3} of power required to evaporate the same saltwater sample. However, despite the success of the RO technology, it has some weaknesses which prevent maximum water recovery of more than 40–50%. This limitation is because the rejected stream from the RO system has high salinity. Higher water recovery factors can be achieved in RO systems by applying higher hydraulic pressures, but there is a risk of forming deposits in the membrane module and consequently exceeding the membrane burst pressure, usually 60–70 bar [101–108]. Therefore, in recent years, membrane distillation technology (MD) has emerged, combining thermally based evaporation and membrane filtration methods. In the MD method, the water recovery factor is not limited by the osmotic pressure and can be increased to a value close to 90%. The high water recovery factor is achieved at a moderate operating temperature between 60 and 80 °C [109,110]. In the MD method, a net flux of water vapor is generated by contact between a warm and highly saline stream and the side of a membrane. The partial pressure gradient across the hydrophobic and microporous membrane is the reason for mass transfer in MD systems [107,109]. Figure 3 shows the operating principle of an MD system.

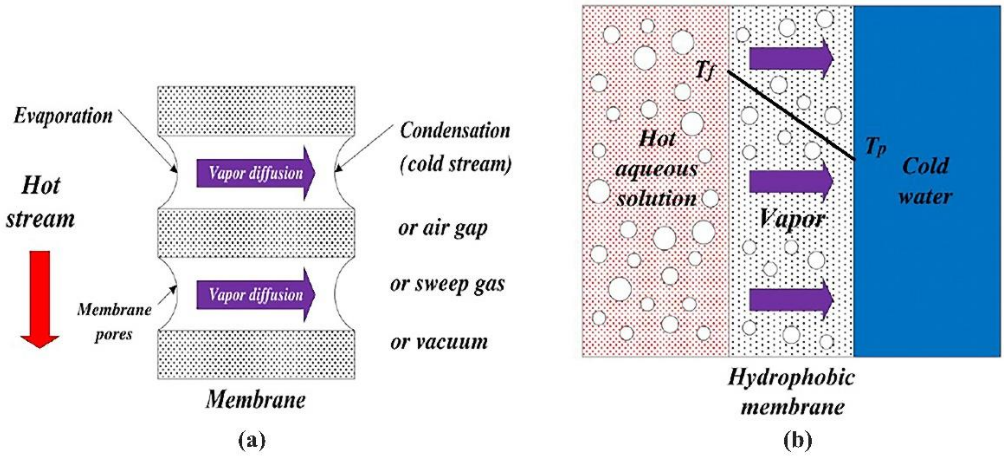

Figure 3. The working principles of an MD unit: (**a**) diffusion of vapors and prevention of passage of liquids through the membrane due to its hydrophobic properties; and (**b**) transfer of water vapor from the hot stream to the cold stream due to a partial pressure difference created by the temperature difference between different sides of the membrane. Reproduced with permission from [111]. Copyright © 2016, Elsevier.

Many publications have mentioned the applications of the MD method for the desalination of sea and brackish water. However, the MD technique is not limited to the desalination of water; MD can also be used in industrial wastewater treatment for purification, extraction, concentration, and final formulation of organic and inorganic compounds [112,113]. In the MD water treatment method, only the volatile components evaporate. In this way, suspended solids and non-volatile salts and dyes can be separated from the water; thus, the quality of the output water from MD systems is high. In addition, MD systems require little space compared with thermal processes, and the water vapor flow in such systems is not affected by the solute concentration in the feed water [114–117]. However, conventional MD systems have low thermal efficiency. This low thermal efficiency is due to the phenomenon of temperature polarization (TP), which occurs because heat is dissipated in the form of latent heat during the evaporation of water. The performance and efficiency of the MD system are also affected by the conductive heat flux through the membranes. However, conductive heat flux through the membranes has a more negligible effect on the system's efficiency than the removal of latent heat [118,119]. Due to the occurrence of TP in MD systems, the overall efficiency of such systems drops to 50%. However, the low operating temperatures of MD systems make the use of waste heat from industrial processes and heat from renewable sources an exciting option in such systems [107,120–123]. Using membranes modified with photothermal NPs is a solution for TP in MD systems. Photothermal NPs can convert localized light into heat. Therefore, they can be used as nano heaters on the surface of the membrane.

Recently, many efforts have been made to develop efficient photothermal NPs. Photothermal NPs were developed by changing the chemical composition of various materials and surface and structural changes. Recent studies have shown that using thermoplasmonic materials in MD units is beneficial for water evaporation through a photothermal interface. Thermoplasmonic materials can be used to generate heat with a beam of light. This effect is achieved due to the optically resonant plasmonic excitations in metallic NPs [107,124–129]. The use of thermoplasmonic-modified membranes reduces heat loss and recovers heat at the water–vapor interface. Thus, using such membranes improves the interfacial evaporation of water [107,130–132]. Figure 4 shows the mechanism of heat generation by irradiated light beams in plasmonic membranes.

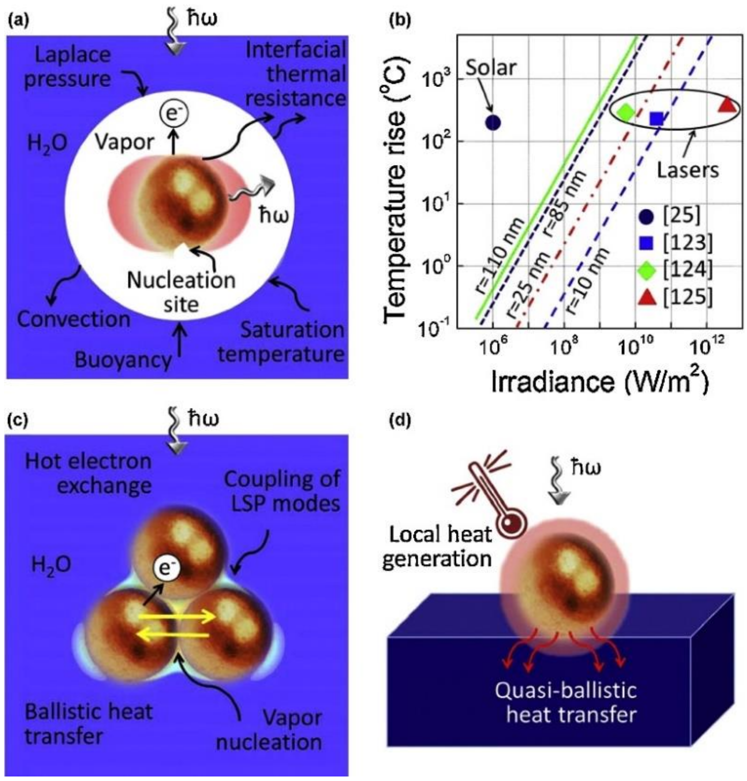

Figure 4. The mechanism of conversion of light to heat using plasmonic NPs: (**a**) Evaporation of water around a metallic NP due to the generation of a localized surface plasmon by photons; (**b**) Relationship between the degree of temperature increase for an Au NP and the irradiance of different wavelengths of light. Labels represent the size of the used gold NPs; (**c**) The thermal and optical interactions between different particles in a plasmonic MD unit; (**d**) Local heat generation in NPs smaller than the phonon mean free path in a substrate material. Reprinted with permission from [133]. Copyright © 2013, Elsevier.

Over the years, researchers have attempted to use solar energy for water treatment or desalination using various membrane technologies [23,74,98,134–140]. Some published articles have reported the successful use of plasmonic membranes in MD units. For example, Farid et al. [98] experimentally demonstrated using plasmonic titanium nitride NPs as a coating for hydrophilic porous membranes which evaporate water using solar energy. The system designed by Farid et al. included a photothermal TiN membrane and a 2D pathway for transporting the vaporized water. The designed system achieved a solar thermal conversion efficiency of 84.5%, and the portable water production rate in the system was 1.34 kg/m^{2}h. The high efficiency and water production rate were achieved using only cheap and readily available materials. Figure 5 shows a schematic diagram of the systems studied by Farid et al.

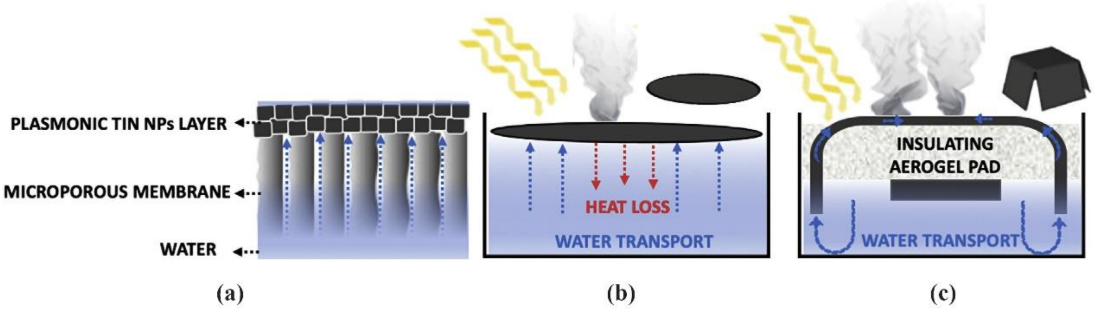

Figure 5. The experimentally studied systems by Farid et al.: (**a**) transport of water through the microporous channels of the membrane to the surface where plasmonic NPs are placed; (**b**) evaporation of water on the surface of the membrane due to the heat generated by plasmonic NPs in a non-insulated system; and (**c**) insulating the system using an aerogel pad to reduce the heat loss for enhancing vapor generation. Reprinted with permission from [98]. Copyright © 2020, Elsevier.

4.2. Application of Plasmonic Membranes in Adsorption Water Treatment Methods

The adsorption method is one of the most promising methods for removing dyes and toxic substances from water, such as heavy metal ions [141–145]. There are biodegradable and environmentally friendly adsorbents that adsorb the waste substances on their surfaces by physicochemical processes [141–144]. One of the most commonly used adsorbents in the industry is activated carbon, which exhibits exceptional performance, but its regeneration has a high cost [146]. Therefore, the use of cheap alternative adsorbents has been suggested by many researchers. Some of these materials are peanut shells, tea waste, and peels of fruits and vegetables such as garlic, lemons, and oranges [147–150]. One of the materials that has attracted the attention of many researchers is eggshell membranes (ESMs), which have a fibrillar structure. ESMs contain amino acid units and functional groups such as carboxyl (-COOH), amine (-NH_2), thiol (-SH), hydroxyl (-OH), and amide (-$CONH_2$). These functional groups act as sites for positive charges in an acidic solution and can adsorb anions of acids [151–154]. Alkaline wastes are adsorbed by forming an electrical double layer on the surface of the ESM. Thus, anions and cations, mainly found in industrial wastewater, can be removed by the double-layer structure of the ESM [22,155]. Although using ESMs as an adsorbent is promising, there are concerns about detecting the low concentrations of impurities in water. Modifying ESM with plasmonic NPs enables use of the surface-enhanced Raman scattering (SERS) technique, which is used to detect low concentrations of biological and chemical substances. SERS uses the scattering of light by plasmonic nanostructures to determine the concentration of various compounds [156,157]. Candido et al. [152] investigated the use of plasmonic chemically modified ESMs as adsorbents and evaluated the performance of the modified membrane for the detection of low concentrations of contaminants. It was found that the pH, mass of adsorbent, reaction time, and concentration of waste materials were important factors affecting the performance of ESM systems in waste removal. It was also found that the use of plasmonically modified membranes enables the detection of concentrations as low as 10–9 M using the SERS technique. The use of a rough metal substrate over a large area evenly distributes the hotspots and enables detection. In addition, silver-based flower nanoparticles (SFNPs) can be used, which have hierarchical structures. Figure 6 shows the scanning electron micrograph (SEM) of an SFNP and the absorption of UV and visible light spectra by the Ag nanoparticles.

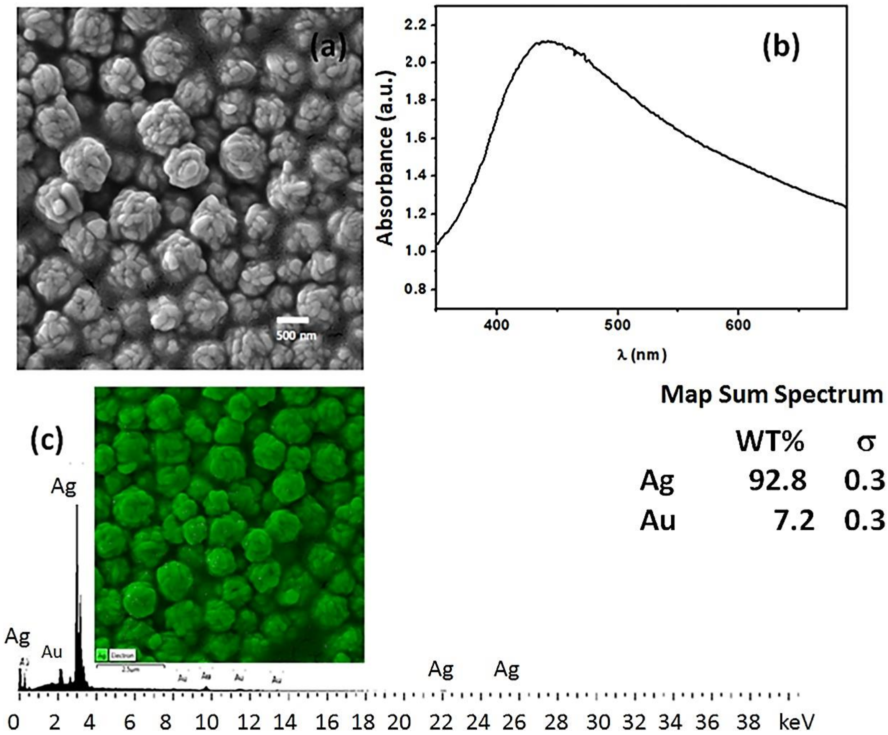

Figure 6. (**a**) SEM image of an SFNP; (**b**) absorbance of visible and UV spectra of light by SFNPs; and (**c**) energy-dispersive X-ray spectroscopy (EDS) of the same components, which are represented in the SEM image. The green color shows the silver particles. Reprinted with permission from [152]. Copyright © 2019, Elsevier.

4.3. Application of Plasmonic Membranes in Photocatalytic Water Treatment Methods

Nowadays, a wide variety of contaminants are released into the environment through wastewater. The release of contaminants such as antibiotics, analgesics, anti-inflammatory drugs, and organic dyes used in the cosmetic, leather, paper, textile, and food industries has raised new concerns due to genotoxicity to various organisms and other unpredictable effects [158–163]. Studies by Voogt et al. [164] showed that 44 different types of pharmaceuticals are present in the water cycle, and most of them are difficult to remove from water resources using conventional water and wastewater treatment methods [165]. Other studies also showed that up to 20% of consumed dyes enter water resources through wastewater [166,167]. As mentioned earlier, adsorbents such as activated carbon can be used to effectively remove pollutants such as dyes. However, the reproduction cost of effective adsorbents is high. As mentioned above, one method to treat wastewater containing dyes and other hazardous substances is using bioadsorbents such as ESMs. Another promising method to remove many pollutants from water is using photocatalysts in advanced oxidation processes (AOPs) for oxidative mineralization [168–171]. Bimetallic NPs, especially NPs containing metals such as gold and silver which have plasmonic properties, are excellent catalysts and stable materials [172–175]. When plasmonic materials are exposed to light, oscillation occurs in the electron cloud. Therefore, the extent of photocatalytic activity of plasmonic materials can be determined by the number of hot electrons generated by the localized surface plasmon resonance (LSPR) phenomenon. A higher number of hot electrons increases the photocatalytic activity of plasmonic materials [176–178]. Using semiconductor photocatalysts such as titanium dioxide, zinc oxide, and NPs of noble metals can also harness the full potential of solar photons [179,180]. Plasmonic materials cannot be used alone for wastewater treatment because the photocatalytic activity is insufficient to remove pollutants. Therefore, the use of plasmonic materials together with membranes is proposed. The photocatalytic properties of plasmonic materials can be used in membrane reactors (MRs) to effectively remove pollutants from water.

The most commonly used material for the fabrication of MRs is polyamide (PA). PA is very stable at various pH values, can operate in a low-pressure environment, and has an excellent salt rejection rate compared with cellulose-based membranes. However, PA is susceptible to fouling. PA membranes also need to be replaced frequently as they lose their resistance to chlorine [75,181,182]. As mentioned above, the antifouling properties of plasmonic materials have been demonstrated by various researchers. Therefore, modifying PA membranes with plasmonic materials can solve the fouling problem and reduce the number of replacements required. Amoli-Diva et al. [75] studied the applications of plasmonic materials in membrane filtration reactors for industrial wastewater treatment. Two antifouling membranes were prepared from commercially available PA modified by the in situ polymerization of polyacrylic acid (PAA) and the grafting process. Two plasmonic Au–Ag and Ag–Au photocatalysts were synthesized for the grafting process. Figure 7 shows the transmission electron microscopy (TEM) image of the plasmonic NPs used.

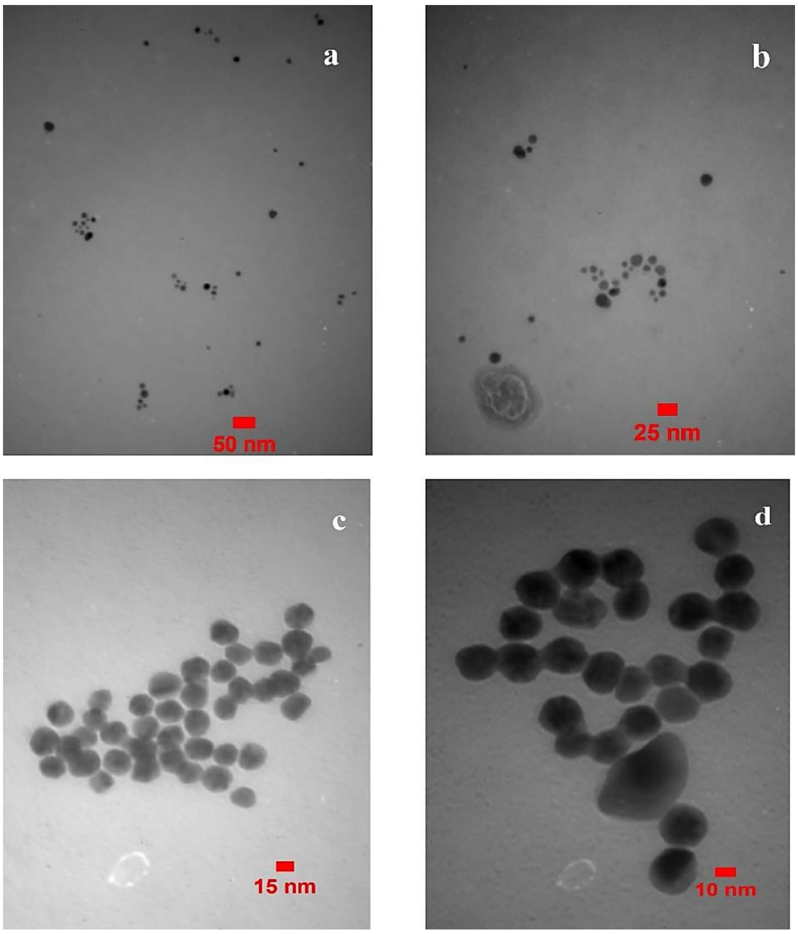

Figure 7. TEM image of the used plasmonic NPs: (**a**) Au NPs; (**b**) Ag NPs; (**c**) Au–Ag NPs; and (**d**) Ag–Au NPs. Reprinted with permission from [75]. Copyright © 2020, Elsevier.

In the experiment performed by Amoli-Diva et al., a xenon lamp was chosen as the optimal light source. The half-life, kinetic properties, and rate constant were evaluated for each synthesized photocatalyst. Reactions involving the photodegradation of ofloxacin (OFX), methylene blue (MB), and dyes present in industrial wastewater were studied using a dead-end unit MR. The antifouling and flux of the membranes produced were analyzed using actual pharmaceutical and textile wastewater samples. In addition, the anti-biofouling properties and the ability to deactivate *Escherichia coli* (*E. coli*) bacteria and their colonization zones were investigated. It was found that the output water of the modified plasmonic membrane in the MR unit was cleaner than the MR unit, which used a standard and unmodified membrane. Water flow was found to be more stable in systems using modified membranes than in systems using non-modified membranes. This more stable flow is due to the plasmonic-modified membranes' excellent antifouling and anti-biofouling properties. Figure 8 shows the experimental setup used by Amoli-Diva et al. [45].

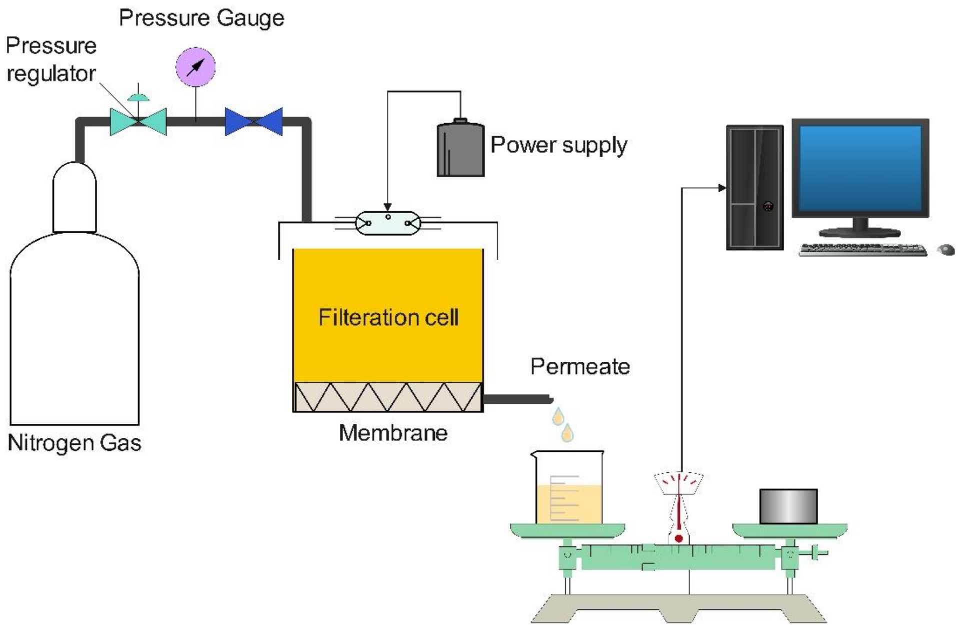

Figure 8. The experimental setup used in the study performed by Amoli-Diva et al. Reproduced with permission from [45]. Copyright © 2020, Elsevier.

5. Comparing Plasmonic Membranes with Other Water Treatment Methods

Unfortunately, there are scant data available to compare the overall performance of plasmonically modified membranes with other conventional water and wastewater treatment methods. Researchers have separately compared the performance of experimentally prepared modified membranes with unmodified membranes; however, the available data are too scattered to compare the performance of plasmonically modified membranes with unmodified membranes. As for plasmonic membranes in MD plants, studies by various researchers have demonstrated the superior performance of plasmonic membranes in wastewater and water treatment. For example, Farid et al. [98] studied the performance of TiN plasmonic membranes and compared their performance against polyvinylidene fluoride (PVDF) membranes in terms of steam generation in MD solar plants. Figure 9 presents a comparison of the steam generation performance of TiN membranes with the performance of PVDF membranes in MD solar systems.

As shown in Figure 9, the evaporation rate is higher in the system where TiN-modified membranes were used, which is due to the additional heat generated by the plasmonic activity of TiN NP under light exposure, based on the studies by Farid et al. With such an evaporation rate, 11 L of saltwater can be desalinated daily. This estimate was made considering a TiN membrane with a surface area of 1 m^2 exposed to sunlight for 8 h. In a similar study by Wilson et al. [138], a water evaporation efficiency of 80% was achieved when exposed to sunlight. They used a cheap setup consisting of a cotton-based membrane modified with plasmonic candle soot NPs. Figure 10 compares the performance of unmodified cotton membranes with modified membranes.

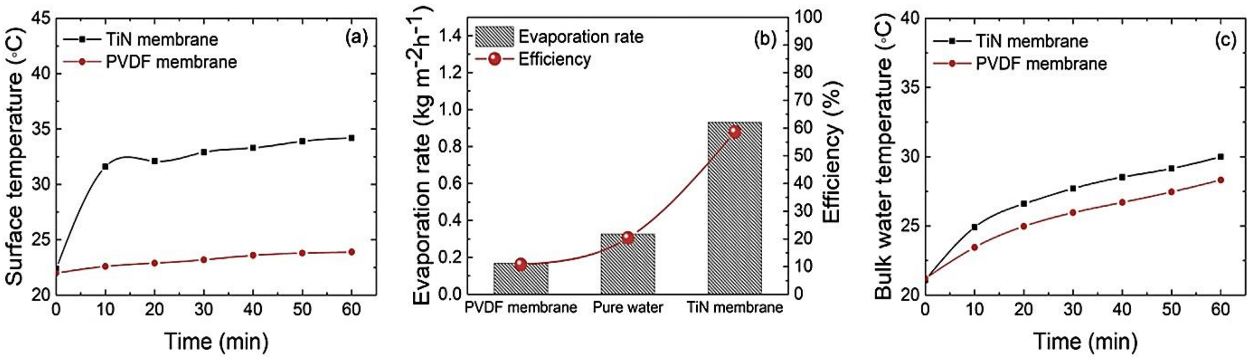

Figure 9. Comparing the solar vapor generation performance of TiN and PVDF membranes with the amount of vapor generated by leaving a sample of freshwater under sunlight: (**a**) dependence of the membrane's surface temperature to the duration of exposure to sunlight; (**b**) evaporation rates and efficiencies for various setups; and (**c**) dependence of the temperature of the bulk water to the duration of exposure to sunlight. Reprinted with permission from [98]. Copyright © 2020, Elsevier.

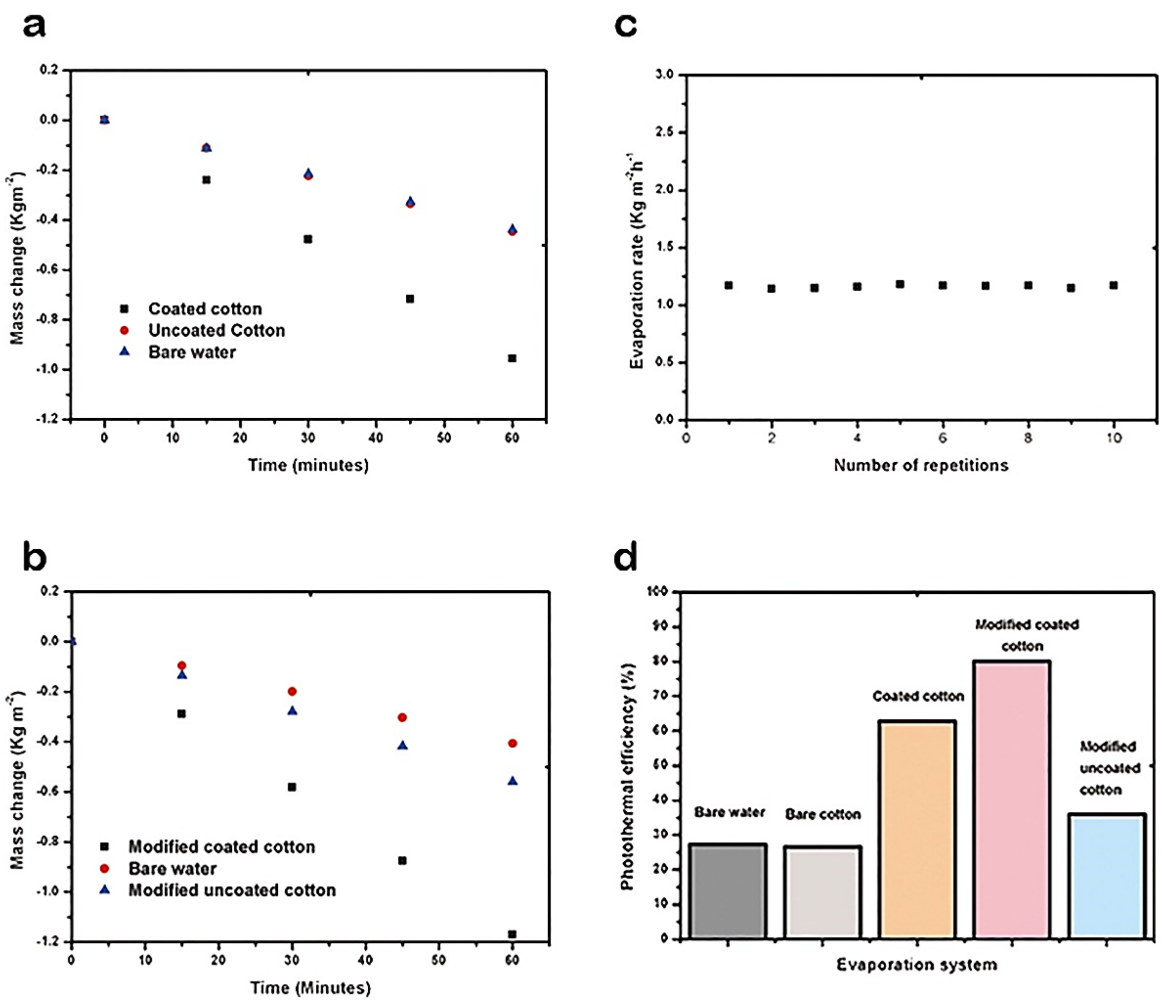

Figure 10. Comparing the performance of plasmonic-modified and non-modified cotton membranes based on the studies performed by Wilson et al.: (**a**) evaporation performance of different systems on the changes in the mass of liquid water; (**b**) performance of the plasmonic-modified system based on the rate of evaporation; (**c**) stability of the plasmonic-modified system in case of the rate of vapor generation; and (**d**) efficiency of different photothermal vapor generation systems. Reprinted with permission from [138]. Copyright © 2019, Elsevier.

Figure 10, similarly to Figure 9, demonstrates the superiority of plasmonic membranes in photothermal water purification methods. Wilson et al. also proposed and tested the usage of the designed membrane in wastewater treatment applications. Figure 11 shows the difference between untreated sewage water and treated water.

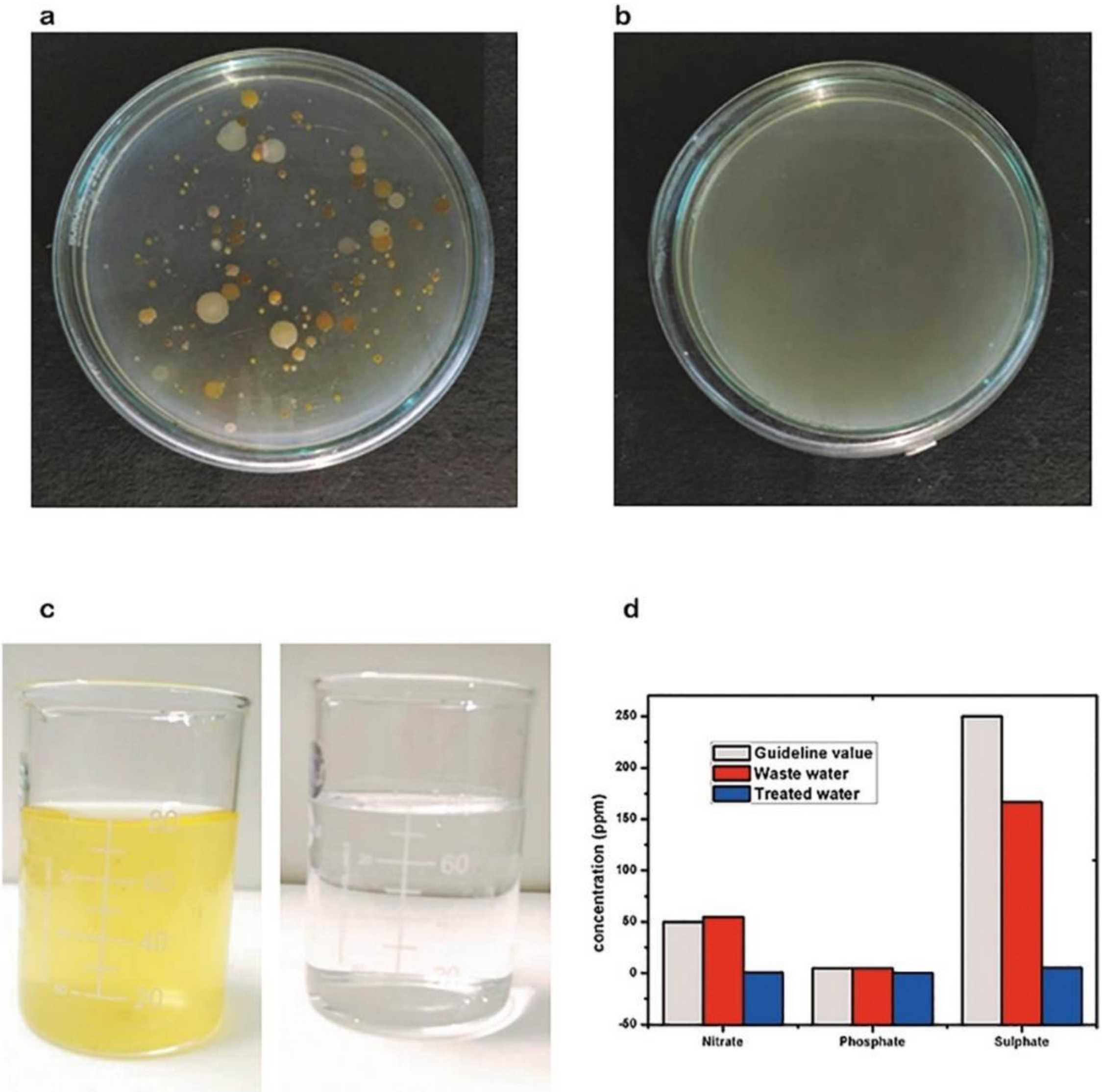

Figure 11. Application of the plasmonic-modified cotton-based membrane in wastewater treatment: (**a**) bacterial colonies in the wastewater; (**b**) nonexistence of bacterial colonies in the water treated with the designed membrane; (**c**) visual appearance of the wastewater and the treated water; and (**d**) performance of the designed membrane in the removal of nitrate, phosphate, and sulfate ions. Reprinted with permission from [138]. Copyright © 2019, Elsevier.

As a comparison between the performance of plasmonic and unmodified ESM membranes in adsorption processes for water and wastewater treatment, Candido et al. [152], as mentioned above, showed that using plasmonic membranes can simplify the detection of pollutants with concentrations as low as 10^{-9} M. Amoli-Diva et al. [75] studied the photocatalytic activity and kinetics of plasmonic membranes in a batch system for the degradation of OFX, MB, and MO. Figure 12 shows the performance of the developed plasmonic membranes compared with unmodified membranes in removing contaminants.

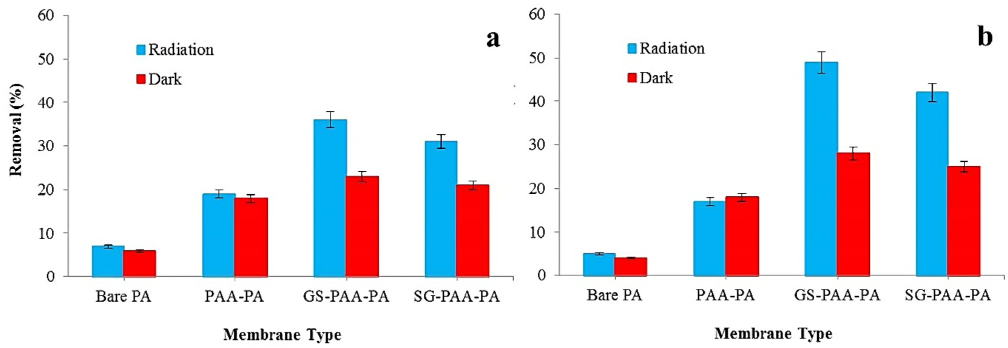

Figure 12. Comparing the removal performance of standard non-modified membranes with plasmonic-modified membranes. (**a**) performance of different membranes in case of removal of OFX; and (**b**) performance of different membranes in case of removal of MB. Reprinted with permission from [45]. Copyright © 2020, Elsevier.

The results obtained by Amoli-Diva et al. [75] indicate the superiority of Au-Ag (GS) and Ag–Au (SG) modified plasmonic membranes in the treatment of industrial wastewater.

6. Advantages and Disadvantages

As mentioned earlier, high efficiencies can be achieved by using plasmonic membranes in various water and wastewater treatment systems. Thus, plasmonic membranes can be used in MD plants, photocatalytic MRs, and membrane filtration units. In MD plants, forming plasmons by irradiating plasmonic NPs with light increases the efficiency of the evaporation process by generating heat. Metallic plasmonic NPs prepared mainly from Au and Ag NPs are exceptional catalysts and photocatalysts. Therefore, the use of membranes modified with such compounds in MR plants increases the efficiency of wastewater treatment. The use of plasmonic membranes in absorption wastewater treatment processes facilitates the monitoring of pollutant concentrations, because the unique properties of plasmonic materials enable the use of techniques such as SERS. Water flux is higher with plasmonic- and plasma-modified membranes, and frequent membrane replacement is not required due to the antifouling properties of such membranes. Plasmonic materials effectively control pathogens and prevent the growth of bacterial colonies. Therefore, plasmonic membranes are immune to biofouling and can be used to remove pathogens from water. Many researchers have investigated and developed cheap and environmentally friendly bioactive materials to replace more expensive commercially available membranes [83,98,138,139,183]. These membranes can be chemically modified and coated with a layer of plasmonic NPs to produce bioactive membranes with various desirable properties. However, the use of plasmonic membranes is not always advantageous. The main disadvantage of plasmonic membranes is that expensive metallic elements such as gold and silver are used for the fabrication of such membranes. Less expensive materials such as TiO_2 show good plasmonic behavior, but are not as effective as plasmonic NPs made with noble metals.

The second problem that can occur in systems using plasmonic membranes is the possible erosion of the membrane surface, which increases the risks associated with the slow release of the metallic NPs used in the membrane into the water and the environment. The cheaper Ag NPs used in plasmonic membranes are not as corrosion-resistant as Au NPs and can dissolve in water when exposed to specific compounds or ions. As a result, there is a risk of toxic Ag^+ ions entering the water and the environment. The application of plasmonic bioactive membranes, produced using fungi or other organisms, in industrial wastewater treatment plants is limited due to the presence of materials such as heavy metal ions in wastewater which are toxic to various organisms [184,185]. Table 1 summarizes the advantages and disadvantages of using plasmonic membranes in water and wastewater treatment.

Table 1. Some of the important advantages and disadvantages of plasmonic membranes.

Advantages of Plasmonic Membranes	Disadvantages of Plasmonic Membranes
High efficiency in various processes	The high price of metallic plasmonic NPs
Antifouling and anti-biofouling properties	Possibility of the release of metallic NPs or Ag^+ ions in water and environment
Excellent catalytic and photocatalytic activity	bioactive membranes prepared from organisms have limited application in industrial wastewater treatment
Antibacterial properties	Erosion of the plasmonic coating of membranes

7. Challenges

The main challenges ahead of using plasmonic membranes in the water treatment industry are the lack of data on the performance of such membranes compared with other water treatment technologies. There are only a limited number of published articles on plasmonic membranes, and data are lacking. Therefore, at this time, it is impossible to provide a comprehensive overview of the performance of plasmonic membranes. In addition, there is no standard method for designing and fabricating plasmonic membranes. The use of plasmonic NPs to modify membranes is a relatively new topic and is currently at the research and experimentation level, which means that the industrialization of plasmonic membrane technology requires further study. The price of metallic plasmonic NPs, mainly made using noble metals, is high; therefore, further research on cheaper alternative plasmonic materials is needed.

8. Perspective

Recently published papers on the applications of plasmonic membranes in water and wastewater treatment industries have reported promising results. Due to the severe problems caused by population growth, shortages of freshwater resources, and the uncontrolled release of wastewater into the environment, seeking new water treatment methods is necessary. Membrane technology is a promising solution to the problems currently faced by the water and wastewater treatment industries. Membrane technology can be combined with the currently available technologies for harnessing energy from renewable resources. Using a combination of renewable energy sources and membranes is one way to treat water cheaply. Modification technologies can further enhance the commercially available membranes. In the case of plasmonic-modified membranes, although promising results have been reported, there is still a fundamental gap in studies concerning the performance of plasmonic membranes compared with other water treatment methods and the design standards. Due to a lack of research on using plasmonic materials for membrane enhancements, scant data are available about the performance of plasmonic membranes in real-life operating conditions in industries. Due to the high price of noble metals used as plasmonic NPs, industries have not shown interest in developing plasmonic membranes. Developing cheap materials with excellent plasmonic properties that can be used as membranous coatings has attracted the attention of researchers and industries. Developing bioactive modified membranes which exhibit good performance can be a solution to environmental problems and complications caused by a shortage of freshwater. Subsequently, some research topics for future researchers interested in applications of plasmonic membranes in water and wastewater treatment industries and various aspects of them are suggested:

- Further experimental validation of currently available results obtained from various studies about plasmonic membranes.
- Writing codes and developing software packages or using currently available software packages to simulate the operation of plasmonic membranes and evaluate the performance of such modified membranes.
- Seeking cheap plasmonic materials that can be used for membrane modification.
- Comparing the performance of plasmonic membranes with other conventional water and wastewater treatment methods.

9. Conclusions

The population growth and shortage of freshwater resources are critical problems facing modern society. Additionally, the release of wastewater into the environment is a significant concern due to the enormous adverse effects of toxic materials in the water. The release of heavy metals, dyes, and pharmaceutical chemicals can damage the ecosystem and put the lives of humans and other organisms in danger. Thus, currently, wastewater treatment is an important consideration. Membrane technology is a promising solution to address the problems related to wastewater management and the shortage of freshwater resources. However, another problem is the high energy prices required to operate systems that use membranes for water purification. Additionally, membranes require frequent replacement due to fouling and biofouling, adversely affecting the water flux in membranes. To address these problems, researchers proposed modifying the membranes as a solution. Through modifications, many desired properties can be achieved in membranes. It has been known since 1877 that the transport properties of membranes can be changed by applying modifications.

Currently, there are many methods available for modifying membranes. One of these methods is plasma modification, which helps create membranes with antifouling properties. Advances in solid-state physics and chemistry have led to the discovery of an exciting property of metallic elements that can be exploited to make plasmons, which are quantums of plasma oscillations. Thus, a new field of study, named plasmonics, has emerged. Since then, various applications of plasmonic materials have been explored in many published studies. One of the exciting applications of plasmonic NPs is that they can be used to modify the surface of membranes to give them various properties. Plasmonic membranes have antifouling and anti-biofouling properties. They can generate heat under exposure to light and demonstrate brilliant catalytic and photocatalytic activity. Thus, they can be used in various applications that involve membranes, ranging from MD to catalytic and photocatalytic MR units. They can easily be integrated with technologies that produce energy from renewable sources. Thus, the efficiency of water purification systems that use plasmonic membranes is high. Plasmonic membranes can make monitoring the concentration of materials inside wastewater easier. Although many advantages can be detailed for plasmonic membranes, various drawbacks must be addressed in future research efforts. One of the main problems is the high price of plasmonic NPs, mostly fabricated using noble metals. Some researchers have proposed using cheap materials such as TiO_2, but due to a lack of data and research in the field of plasmonic membranes, a general comparison of the performance of various forms of plasmonic materials cannot be performed. The other problem is the release of the plasmonic coating of the membrane in the water resources and environment over time due to corrosion and erosion. Bioactive plasmonic membranes prepared from organisms are cheap and are not harmful to humans and the environment. However, the toxicity of materials in industrial wastewater can damage the organisms, or even kill them. As a result, such membranes have limited applications. Thus, future researchers in the fields of membrane technologies and plasmonics must consider the development of efficient bioactive plasmonic membranes to end the global water shortage and environmental issues caused by the uncontrolled release of wastewater. This article reviews the recent advances in plasmonic chemically modified bioactive membrane technology and provides and mentions the challenges of utilizing this technology in wastewater removal applications. This article also provides some perspectives to pave the way for future researchers interested in investigating various applications and aspects of plasmonic-modified membranes.

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