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Chapter

Cellulose Acetate Membrane Preparation for Wastewater Treatment

Ibtissem Ounifi, Fatma Khaled, Malak Kahloul, Hanen Ajari and Amor Hafiane

Abstract

For a long time, humans have used cellulose, as a natural, renewable, and transformative polymer, for scientific development to create new technologies. Cellulose is the most abundant biopolymer on Earth, accounting for more than 50% of terrestrial biomass. For this reason, the treated cellulose (cellulose acetate (CA)) was used in the membrane preparation for water desalination. However, membrane preparation has recently attracted big attention of several research groups. In this case, cellulose acetate (CA), as an inexpensive hydrophilic biopolymer, was chosen as a polymer for preparing the membranes via the inversion phase, since it offers an efficient purification benefit with low energy consumption and less cost. The purpose of this chapter is to describe the various types of membrane preparation based on cellulose acetate, with pathogens, bacteria, and heavy metal (cadmium), and the applications of these membranes in the treatment of contaminated water, to ensure a clean water supply for both human and industrial uses.

Keywords: cellulose acetate, membrane preparation, water desalination, hydrophilic

1. Introduction

In the last decades, the population, industrial, agriculture, and urbanization growth has continually increased and by adding climate change and policymaking problems, the fresh and accessible water has been reduced in the world. As a result of global problems, the demand for pure water increased more and more [1, 2]. As well as the problem of heavy metal contamination of water supplies, which is producing major environmental and health implications, there is a growing need for water desalination technology [3–5].

Recently, membrane separation technology has been attracting considerable attention due to its potential applications over conventional approaches such as thermal desalination. Membrane process becomes widely used in water desalination, wastewater treatment, pharmaceutical filtration, and even in energy generation due to their different properties such as being environmentally friendliness, simple operation, high efficiency, low operating cost, low energy use, and scalability [5–7].

Generally, the basic function of the membrane is to separate different mixtures by concentrating small molecules and allowing others to pass. This specific function as well as the other advantages makes membrane technology the principally used application in many separation membrane processes such as water, wastewater treatment, and adsorption, which gives it an important part in the water challenge (**Figure 1**) [8, 9].

Membrane separation is based on an efficient and scalable methodology by using some or without no harmful chemicals by-product formation. Membrane fabrication from synthetic polymers becomes very much used in various separation membrane applications. Generally, several polymers are used such as polysulfone (PSF), polypropylene (PP), polyethylene (PE), polyethersulfone (PES), polyvinylidene fluoride (PVDF), polyvinyl alcohol (PVA), chitosan (CS), and cellulose acetate (CA) [10–13]. Nowadays, CA polymers attract a big interest in membrane fabrication due to their biodegradable polymer propriety, hydrophilicity nature, and large disposability, since cellulose is an organic compound widely existing on the earth [14, 15]. Generally, cellulose can be found in plants, wood, algae, tunicates, and even in bacteria, and its annual extracted production can attain around 7×10^{10} tons per year, which highly encourages the use of cellulose [16]. As result, many researchers focused on membrane fabrication with the derivatives of cellulose such as cellulose acetate CA [14, 15].

The CA membrane's performance was improved by its high chemical and mechanical stability, high hydrophilicity with desalination pilot, high transport properties, low protein adsorption, excellent water affinity, and excellent film-forming property [17]. Due to their desirable properties and environmental aspects, CA-based membranes have been used in several separation membrane applications, such as gas separation [18], reverse osmosis (RO) [19], nanofiltration (NF) [20], ultrafiltration (UF) [21], microfiltration [22] (MF), pervaporation [23], and ion exchange [24], and it has been widely used to electrospun fibers for it simple processing [25].

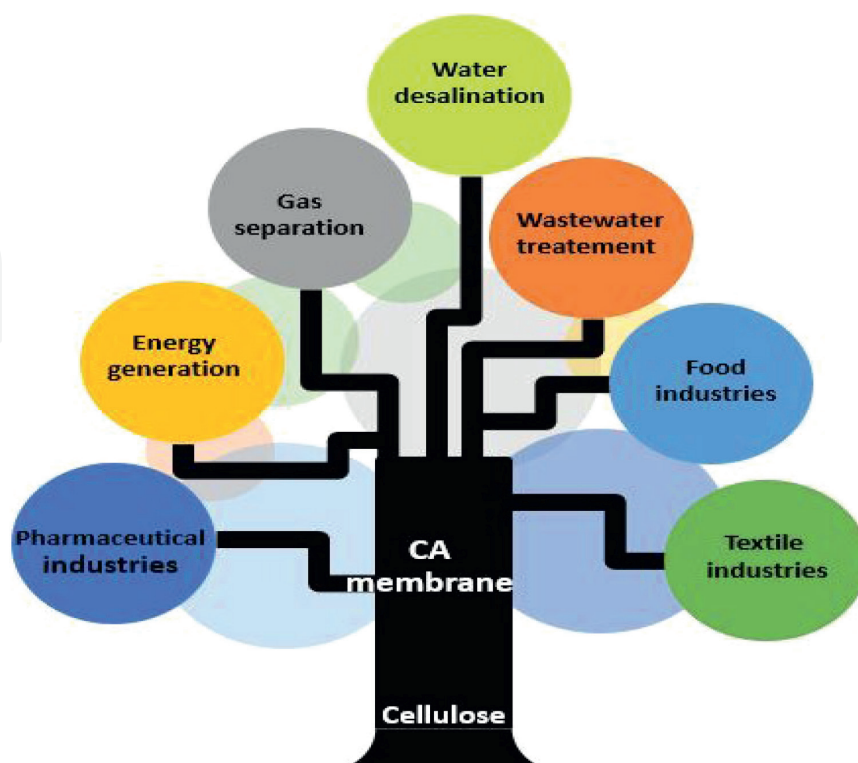


Figure 1.
CA-based membrane applications.

More than separation membrane technology, CA has the potential to be used securely in industrial and biomedical applications such as wound dressing [26] because of its low toxicity and biodegradability [27].

The wide use of CA membranes can be attributed to the fact of the importance of CA in the manufacturing of biodegradable membranes, besides the possibility of improving membrane performance through the addition of additives [28], particularly nanomaterials. Therefore, to improve the CA membrane's performance, the membrane solution can be mixed with the right organic or inorganic additives in the phase inversion technique [29]. To modify CA membranes, some researchers changed the solvent in the membrane casting solution. Various solvents were tentatively used to prepare CA casting solution, including N-methyl pyrrolidone (NMP), N, N-dimethyl acetamide (DMAc), and a composed solvent of N, N-dimethylformamide (DMF), acetone, or 2-propanol [30–32].

This chapter aims to present the cellulose properties and its derivatives and to study the use of CA polymer in the preparation of polymeric separation membranes applied in different applications. First, a presentation of CA properties will be carried out. Then, different CA membrane configurations will be described. Thus, the performance of the fabricated CA membrane in those applications will be studied and discussed, by taking the CA as the used polymer on the one hand and as an additive on other hand.

2. Cellulose acetate properties

Cellulose acetate is an important cellulose derivative due to its properties, which are used in different applications. It is extracted from natural and renewable resources. Further, CA is a natural polymer that is characterized by flexibility, biodegradability, non-poisonous, and eco-friendly [33]. Due to its important and unique benefits of biodegradability and nontoxic behavior [27], CA has been used frequently in membrane manufacturing, especially in the treatment of water and wastewater technologies. Several polymers have been used in the membrane synthesis, but the CA membranes are the most preferred according to their durability, low cost, and especially required hydrophilic and biodegradability properties [34, 35].

Also, by dint of their chemical and thermal stability (**Figure 2**), good water retention capacity, and great resistance to methanol permeation, the CA membranes are used in many other applications such as gas separation for CO₂ capture or nitrogen generation [36] and fuel separation for fossil fuel and natural gas drainage [37].

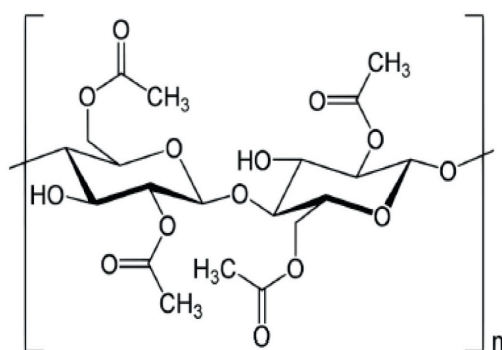


Figure 2.
Chemical structure of cellulose acetate.

Several molecular weights of CA were used in membranes synthesis, which are usually attained at 30,000 g mol⁻¹ [38], 37,000 g mol⁻¹ [39], 50,000 g mol⁻¹ [40], and 60,000 g mol⁻¹ [41], between 39 and 40% of this weight is acetyl.

One of the main properties of CA is the possibility of dissolution in green solvents with the ability to use in the phase inversion technique for membrane fabrication [42, 43]. Totally, bio-based NF membranes were fabricated using CA as a base membrane and several glycerol derivatives (monoacetin, diacetin, triacetin, and glycerol-formal) as green solvents 2-methyl tetrahydrofuran as a co-solvent to reach rejection in the NF range [44]. The greatest membranes resulted from diacetin solvent by permeability in the range of 5.5–12.8 L m⁻² h⁻¹ bar⁻¹ and >90% Rose Bengal rejection.

3. Configuration of CA membranes

3.1 Flat sheet

Flat sheet membranes in general take the shape of flat sheets. The membrane's top surface is an active layer for fluids or particulate/fluid separation. The membrane surface is the top upper layer that performs the function of separation. However, flat membranes are often prepared by pouring the polymeric solution onto a substrate. The most important characteristics of this type of membrane are high specific flow rates, high production rates, and easy placement of the membrane in the module. There are several approaches for preparing flat sheet membranes, but generally phase inversion or thin film composite and interfacial polymerization are the essential methods used to manufacture these membranes.

3.1.1 Phase inversion

Generally, asymmetric porous polymer membranes are developed by the phase inversion process. This occurs when an initially homogeneous polymer solution changes its conditions thermodynamically.

During phase inversion, a single-phase casting solution causes the formation of two different phases: a polymer-rich phase or a solid membrane matrix and a poor phase in terms of polymer or liquid polymer [45]. This technique enables the change from a liquid to a solid state. The transition from one liquid to two liquids starts the solidification process. Slowly, the rich polymer phase forms the solid matrix during this liquid-liquid transition. The poor phase is then removed by successive washing steps and leaves room for the pores of the membrane. The morphology of a membrane has a significant impact on its characteristics and performance.

Therefore, membrane synthesis's major objective by phase inversion process is to obtain membranes with specific properties such as microfiltration, nanofiltration, or ultrafiltration that ensure good thermal, chemical, and mechanical resistance properties and to develop adequate membranes properties with a surface layer for reverse osmosis and gas separation [46].

The CA membranes prepared by using this technique have been used as microfiltration [46], ultrafiltration [47], nanofiltration [42], forward osmosis [48], reverse osmosis [49, 50], and gas separation membranes [51].

In order to create an asymmetric microfiltration membrane structure, a selective layer and a prefilter layer were combined in one membrane to examine the effect of the CA-based solvent system on membrane morphology during membrane

manufacturing. A pseudo-ternary mixture of CA, methyl formate (MF), 2-propanol, and water was used to create asymmetric microfiltration of CA membranes by using a single-layer dry casting process. The created membranes were composed of uniform pore size isotropic regions that are covered by protective skin.

The selective layer of these newly created CA microfiltration membranes fronted the solid-liquid interface in contrast to other asymmetric membranes, or the upper area facing the liquid-gas interface showing larger pore sizes. The denser bottom layer acted as a selective layer while the upper layer functioned as a prefilter. The membranes' structural analyses were carried out using scanning electron microscopy. A theory regarding the actions occurring in the casting solution during membrane development is addressed in depth along with the membrane structures that have been observed [52].

Using the phase inversion method, Ounifi et al. [53] successfully synthesized ultrafiltration membranes from a casting solution containing CA at concentrations ranging from 15 to 21 wt.%. With the rise in CA, the membrane's performance, such as permeability, contact angle, and water content, decreased, but the PEG rejection rose at higher polymer concentrations during membrane formation (**Figure 3**).

CA-NF membranes [54] were prepared via the NIPS inversion process. The asymmetric structure of CA-NF membranes, which are composed of a dense top layer supported by a porous sublayer, was clearly elucidated by the SEM images (**Figure 4**). By increasing the CA concentration in the polymeric doped solution used for membrane synthesis, membrane characterization demonstrated the membrane's hydrophilic nature and decreased porosity. The membrane's permeability to pass water through it was decreased due to reduced porosity, while the salt retention was increased. The size exclusion mechanism dominates the transport mechanism through the membrane.

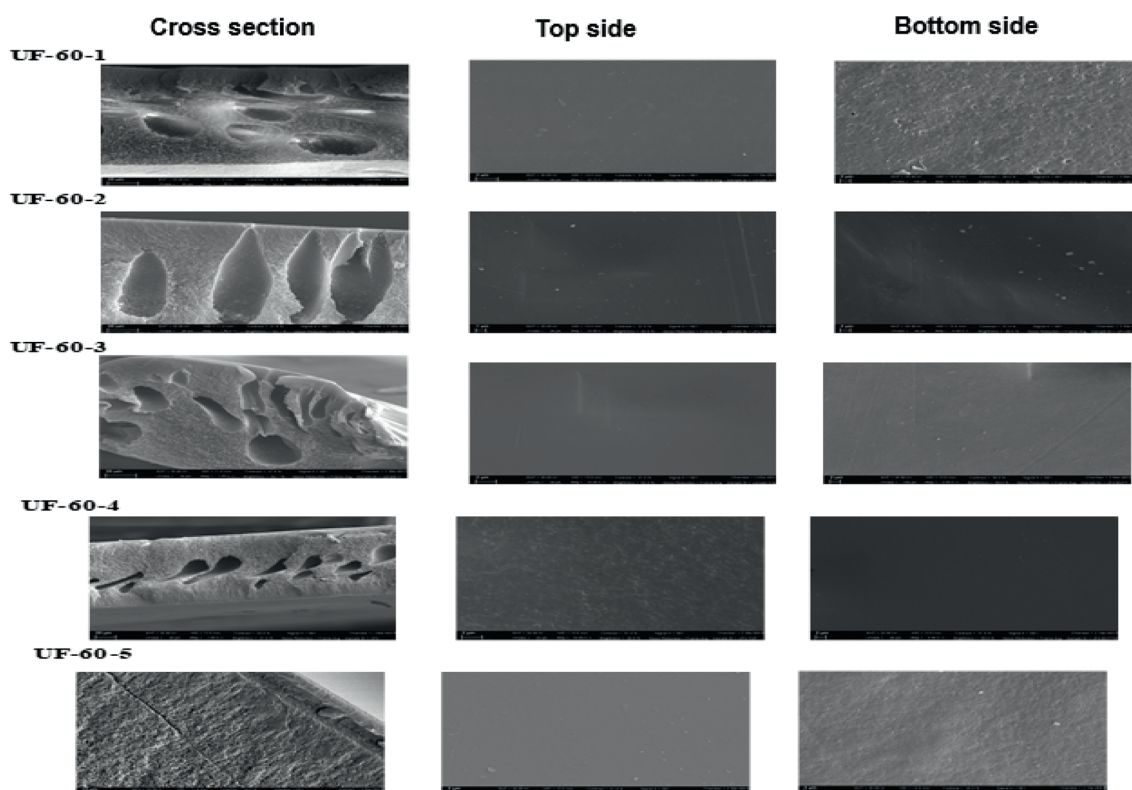


Figure 3. Scanning electron microscopy (SEM) image of ultrafiltration membranes with different polymer concentrations [53].

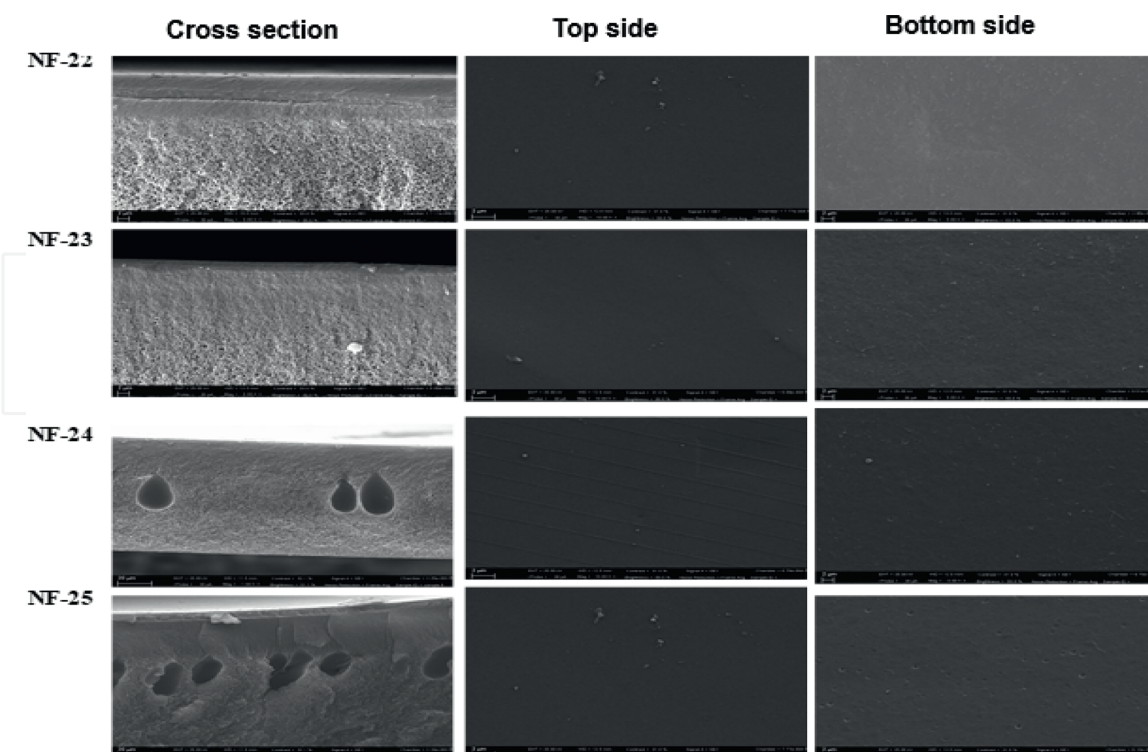


Figure 4. SEM image of nanofiltration membranes with different polymer concentrations [54].

The order of rejection is determined by the hydration energy and hydrated radii of the ions and is as follows: $R(\text{Na}_2\text{SO}_4) > R(\text{CaCl}_2) > R(\text{NaCl})$.

In numerous studies, the phase inversion method used in the CA membranes preparation is carried out in conjunction with the inclusion of certain additives such as composites, nano-materials, surfactants, and polymer additives to change the prepared membranes with this approach.

TiO_2 nanoparticles were dispersed in the CA casting solutions to prepare hybrid CA/ TiO_2 membranes by phase inversion. Investigations were done into how TiO_2 affected the morphology and thermal stability of CA/ TiO_2 membranes. According to SEM findings, the addition of TiO_2 nanoparticles makes the hybrid membrane more porous due to increase in the average pore size. The TGA results show that the interaction between TiO_2 nanoparticles and CA exists. Thus, the hybrid membrane's thermal stability was enhanced by the addition of TiO_2 nanoparticles. In addition, PWF membranes show that the addition of TiO_2 nanoparticles leads to an increase in water permeation [49].

CA-silica (SiO_2) hybrid membranes (CA- SiO_2) were synthesized by promoting in situ condensations between silanols of the SiO_2 precursor and COH or acetate groups of the CA polymer. The results showed molecular water strongly linked to hydrogen bonds with SiOH groups, which leads to a drastic drop in the membrane hydraulic permeability, from 57 to 10 $\text{kg h}^{-1} \text{m}^{-2} \text{bar}^{-1}$. In comparison with the CA membrane, the inclusion of 5 and 10 mol% silica enhanced the hydraulic permeability from 32 to 82 $\text{kg h}^{-1} \text{m}^{-2} \text{bar}^{-1}$ [55].

In order to remove Cd^{2+} , Ounifi et al. developed fouling-resistant UF membranes. Phase inversion was used to prepare the UF membranes by combining PAA and CA. The strong hydrogen interactions between the two polymers were observed with FT-IR. The membranes showed highly porous and asymmetric structures with

affected morphologies as a result of PAA concentration. In fact, by increasing the PAA concentration from 0 wt.% to 15 wt.%, the porosity increased from 44.5% to 75.6%. Contact angle and pure water flow measurements showed that adding PAA to the CA structure improved the membrane hydrophilicity. The result ultimately demonstrated that the prepared membrane with a higher PAA content (M15) showed superior permeability and HA rejection characteristics [56].

3.1.2 Interfacial polymerization

Interfacial polymerization (IP) is a very practical and useful simple technique to create a thin layer for nanofiltration and reverse osmosis membranes. Interfacial polymerization always has been of a particular interest in industrial manufacturing.

The thin layer on the membrane surface is formed by the reaction and co-polymerization of two reactive monomers. In recent years, the development of thin-film composite by interfacial polymerization has received attention due to significantly improved membrane properties such as selectivity and fouling resistance. This method involves several steps, and it is easy to apply. To create the hydrophilic NF membranes, the substrate, typically used in MF or UF (cellulose acetate, polysulfone, etc.), must first be dissolved in a solution containing a highly reactive, water-soluble, and bifunctional or trifunctional monomer. In the second step, the membrane is submerged in a solution comprising an additional mono- or polyfunctional monomer that is soluble in an organic water-miscible solvent. The two monomers thus combine to form a dense polymer layer at the water/organic solvent interface. To finish the polymerization reaction and cross-link the resulting structure, this layer is then frequently heated. The layer that forms on the surface of the membrane limits the progression of the reagents, resulting in a dense thin layer of around 50 nm. This is an advantage of interfacial polymerization.

The porous layer is usually made from PSF polymer. However, interfacial polymerization can also be performed on CA-based support, or CA can be used as a selective layer on a support.

According to Ounifi et al., the interfacial polymerization of *m*-phenylenediamine (MPD) and trimesoyl chloride (TMC) on CA-UF successfully produced innovative flat-sheet thin film composite membranes for nanofiltration (TFC-NF) (**Figure 5**). The thin film composite TFC had a smooth surface, as evidenced by the SEM and atomic force microscopy (AFM) pictures. The polyamide thin top layer preparation conditions, such as reaction time and TMC concentration, have a significant impact on the membrane performance, specifically the water permeability and salt rejection of the TFC-NF. When TMC concentration and IP reaction time were increased, the water permeability fell from 23.35 to 10.44 L m⁻² h⁻¹ bar⁻¹. Additionally, the salt rejections of these TFC membranes were adjusted from 69.23% to 92.43% and from 19% to 55.54% by using Na₂SO₄ and NaCl, respectively. A cadmium solution retention test has been done. The test result showed that this type of membrane could be used for the removal of heavy metals. The rejection of cadmium depends on the nature of the associated anion: $(\text{Cd}(\text{NO}_3)_2) < R(\text{CdCl}_2) < R(\text{CdSO}_4)$, and it can reach a maximum of 97.76% for CdSO₄ [57].

According to Ounifi et al., the interfacial polymerization technique was used to successfully prepare a number of polyamide TFC membranes. The CA-UF was used as a substrate. The PA layer over the CA-UF substrate was created by using the monomers of 1,3-cyclohexanebis(methylamine) (CHMA) and TMC in the aqueous phase and organic phase, respectively (**Figure 6**). Many concentrations of CHMA, namely 0, 0.2, 0.5, 1, and 2 wt.%, were used on membranes preparation. (The CHMA concentration

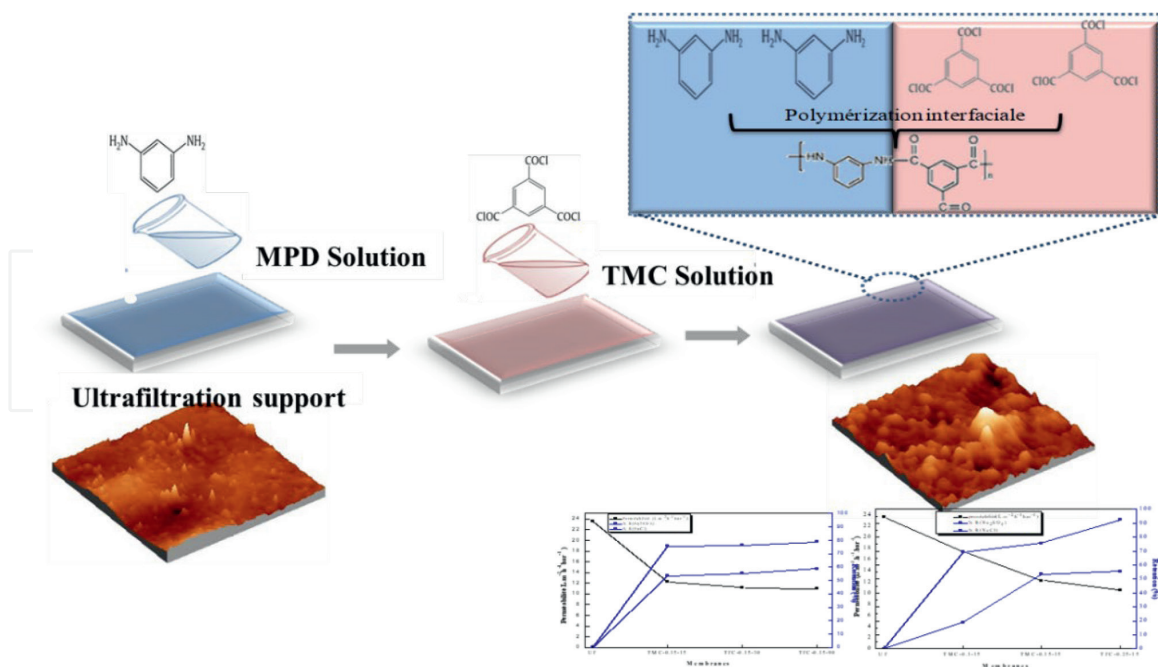


Figure 5. Mechanism of polymerization interfacial and AFM image of membrane CA and TFC.

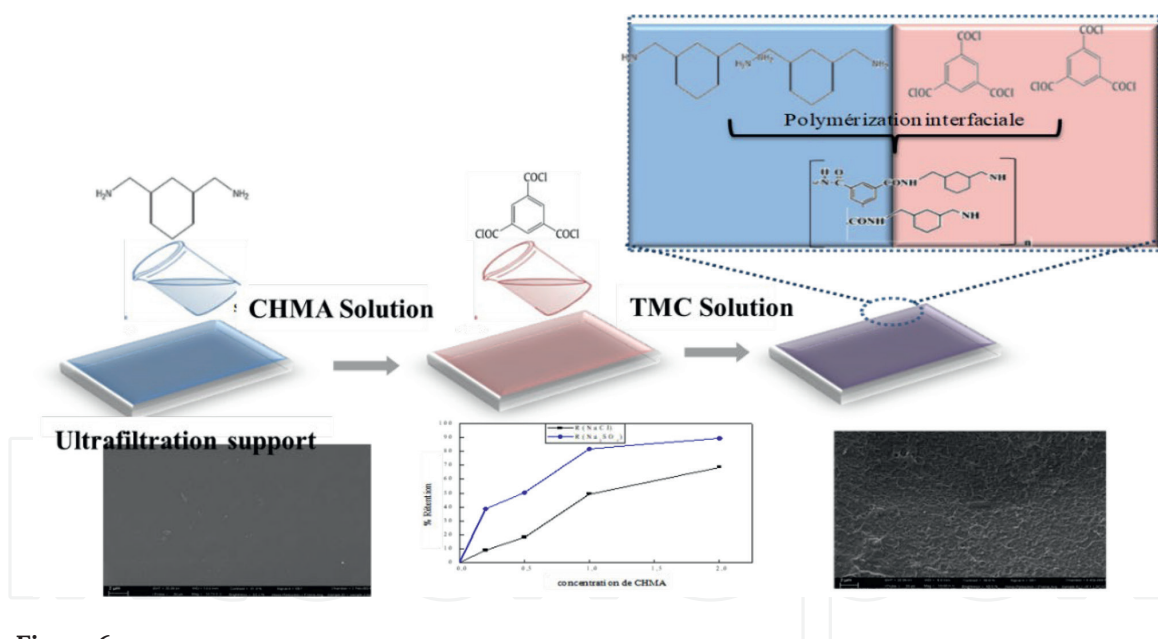


Figure 6. Mechanism of polymerization interfacial and SEM image of membrane CA and TFC.

effect on the membranes' physicochemical characteristics was investigated by using SEM, FT-IR, water absorption, water permeability, porosity, contact angle, specific salt rejection rate, and dye molecules.) SEM analyses showed that increase of the CHMA concentration increased the membrane's rugosity and thickness. The TFC-2 membrane exhibited the typical polyamide membrane morphology identified by the "ridge and valley" structure. The FT-IR findings revealed novel absorption bands specific to polyamide, which ensure the successful use of interfacial polymerization. The hydrophilicity of the membrane was enhanced by the increase in CHMA concentration, but the porosity and permeability of the membrane decreased. For example, the membrane water permeability varied from 36.02 for the CA-UF membrane to 17.09 L h⁻¹ m⁻² bar⁻¹

for TFC-2. All TFC membranes had a higher rate of salt and dye rejection than the CA-UF membrane. The TFC-2 membrane demonstrated a desalination rate of up to 89%, whereas the malachite green and Congo red were rejected at rates of 89 and 85%, respectively. The concentration of the molecule and the feed solution's pH affect the separation performance. Overall, the CHMA could be a potential candidate to develop TFC membranes for desalination and water treatment [58].

Ounifi et al. studied the influence of support pores size and monomer type on the thin-film composite membranes performance. By using CA membrane as support and the cyclohexane-1,3,5-tricarboxylchloride (HTC) monomer as an organic phase, a series of polyamide-cellulose acetate thin-film composite nanofiltration (TFC-NF) membranes were created for this purpose by the interfacial polymerization technique. The effect of the cellulose acetate concentration of 15, 16.5, 18, 19.5, and 21.5% on the pore size of the membrane support was studied. As monomers, m-phenylenediamine (MPD), piperazine (PIP), and 1,3-cyclohexanebis (methylamine) (CHMA) were investigated. In comparison with other produced membranes, the results showed that the PIP/HTC membrane is more hydrophilic and has a more intense granular and convex structure with a rougher surface. The concentration of cellulose acetate affected both the porosity and the water flow. The reduction in porosity was consistent with the results of the SEM analysis (**Figure 7**), contact angle, and permeability. The retention of NaCl, CaCl₂, and Na₂SO₄ was evaluated for the membrane performances. Comparing the Na₂SO₄ rejection with CaCl₂ and NaCl, the former is higher than the latter [55].

3.2 Hollow fiber

Another membrane configuration, in which CA can be used as the main polymer, is a hollow fiber membrane. The hollow fiber membrane (HF) has three main advantages, which have led to the hollow fiber design becoming a popular choice:

1. Compared with flat and spiral-wound modules, hollow fiber modules offer a substantially higher ratio of membrane area to unit volume (several thousand M²/M³), resulting in higher production per unit volume of membrane module;
2. They may be back-washed to recover the flux due to their self-supporting;
3. They also have good operational flexibility [59].

The two main approaches for preparing HF are thermally induced phase separation (TIPS) and non-solvent-induced phase separation (NIPS). For NIPS, the solvent is extracted from the solution using a non-solvent, which is a substance that is miscible with the solvent but cannot solubilize the polymer. When the solvent is removed from the solution, the polymer concentration rises to its solubility limit, causing precipitation. The membrane structure can be tuned by adjusting preparation parameters such as polymer concentration, non-solvent system strength, and the use of additives [60].

For TIPS, phase separation is achieved by lowering the temperature of the dope solution. To obtain a homogeneous solution, the polymer is dissolved in one or more solvents, commonly referred to as diluents, at a high temperature. The dope solution is then extruded at high temperature through a spinneret and cooled in a low-temperature-quenching bath. At low temperatures, the diluents are unable to solubilize the polymer, and the solution undergoes phase separation [61]. After the hollow fiber membrane is formed, the remaining solvent is removed with cleaning baths [62].

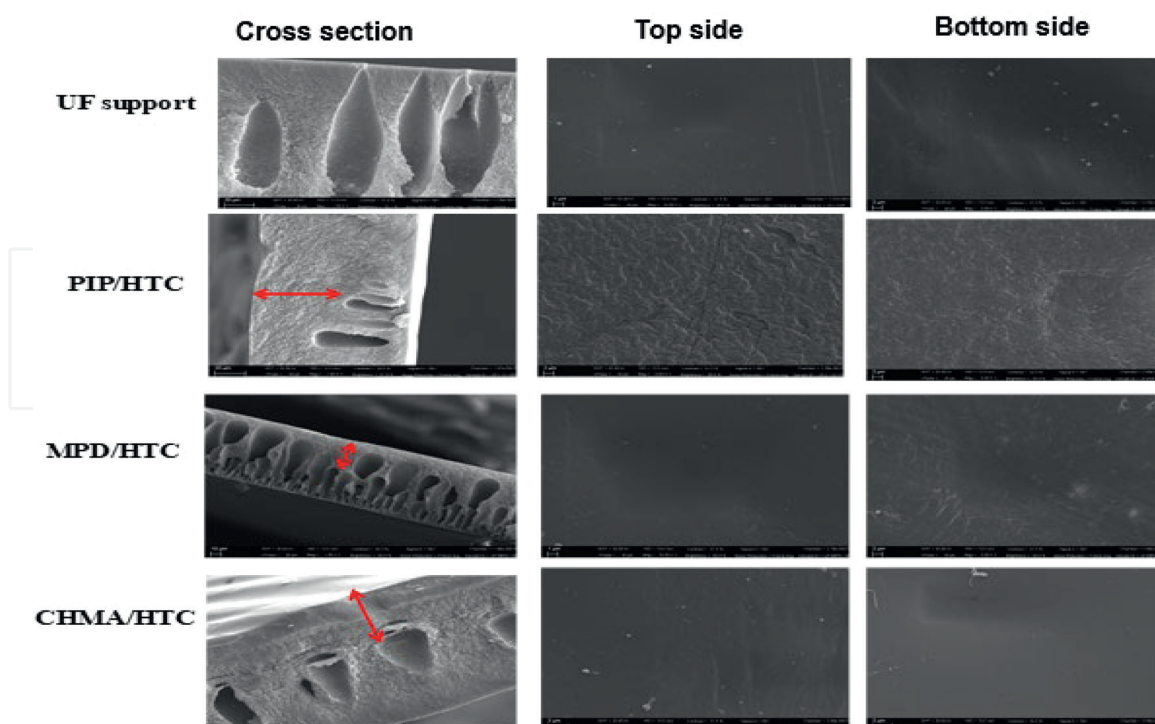


Figure 7. SEM image of TFC nanofiltration membranes with different monomers [55].

The diluent properties and solution viscosity both play a significant role in determining the final membrane morphology.

A homogeneous braid-reinforced CA hollow fiber membrane was formed through a non-solvent-induced phase separation process, and the membrane was prepared using an HMR method, in which CA solution was coated on the outer surface of the CA braid [63].

CA hollow fiber membranes were fabricated via various spinning processes such as dry-jet wet spinning [64], well-known dry-jet wet spinning process, dry-wet phase inversion method [65], coating a separation layer on the high-strength hollow tubular braid, melt spinning, and stretching process [66].

Most CA hollow fiber membranes were prepared using the dry-wet phase inversion method (**Figure 8**), but their poor mechanical properties limited their use in engineering practice. As a result, a hollow fiber membrane with high mechanical properties was required. Many studies have been conducted in order to improve the mechanical properties of hollow fiber membranes. For example, coating a separation layer on the high-strength hollow tubular braid was a relatively simple and effective method. There were two interfaces between the braid and the separation layer: the homogeneous-reinforced (HMR) interface and the heterogeneous-reinforced (HTR) interface. According to Fan et al. [63], the homogeneous-reinforced (HMR) method and the heterogeneous-reinforced (HTR) technique were combined to reduce the weaknesses of each and achieve the best results. Fan et al. produced a BR CA hollow fiber membrane by coating the “hybrid” braid with CA solution. The two-dimensional braided technique was used to prepare the “hybrid: braid, which consisted of CA filaments as the homogeneous fiber and PAN filaments as the heterogeneous fiber as shown in **Figure 9** [63]. SEM image results illustrated the cross section of BR CA membranes made by CA filaments and PAN filaments process in various braid compositions as shown in **Figure 10**.

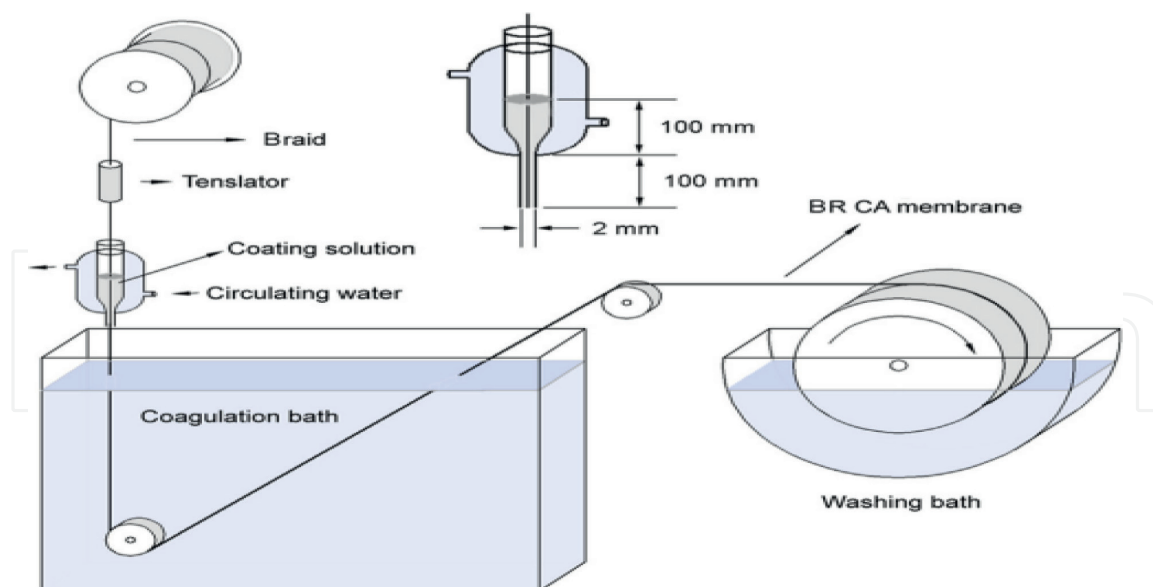


Figure 8.
 Spinning apparatus set up [63].

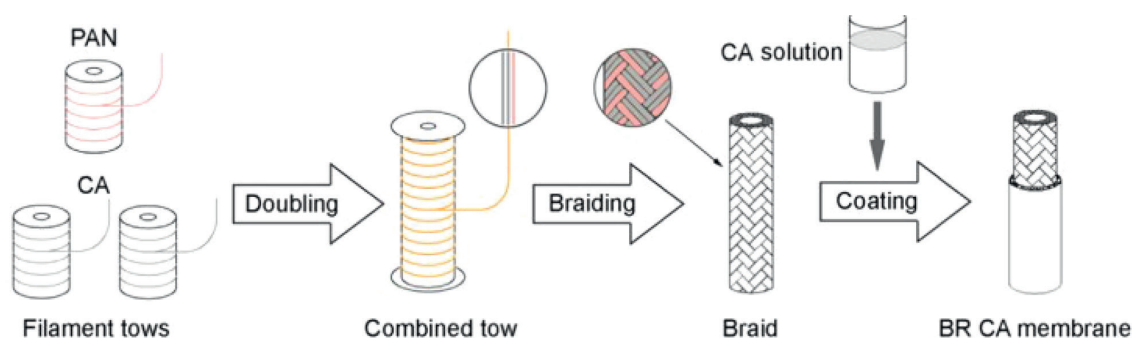


Figure 9.
 BR CA membrane process with CA filaments and PAN filaments.

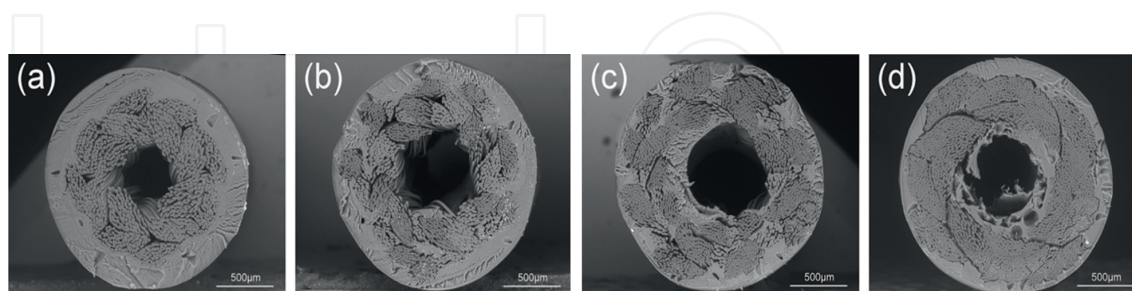


Figure 10.
 SEM images of cross section of BR CA membranes with various braid compositions.

The mechanical properties and permeation performance of BR CA hollow fiber membranes with different fiber ratios in the braid and different CA concentrations in the coating solution were investigated, giving a new approach to obtaining BR CA membranes with desirable performance.

The potential and application of commercial hollow fiber membrane products are majorly affected by three factors:

1. The thickness of the functional separation or selective layer and its substructure morphology;
2. The pore size and pore-size distribution of the functional separation or selective layer;
3. The chemistry, mechanical, and physicochemical characteristics of the membrane material.

CA-based NF hollow fiber membranes have been studied and explored for FO applications. Heat treatment at 60°C for 60 min slightly affects the membrane pore radius and the thickness of the outer skin layer. Further, heat treatment at 95°C for 20 min results in the significant shrinkage of pores on the membrane surface with a denser outer skin layer. The resultant fiber has a high rejection to NaCl and MgCl₂ and low pure water permeability in the NF tests. The CA-NF hollow fiber heat-treated at 95°C shows promising FO performance. However, this type of CA-NF hollow fiber membrane shows a decrease in performance ratio with increasing saline concentration in the feed due to the more severe internal concentrative concentration polarization. These preliminary results manifest the potential of the CA NF hollow fiber membranes for concentrating various feed solutions in the FO process. Future works should aim at the reduction of the dense layer thickness to enhance the flux while maintaining a low salt leakage rate [67].

3.3 Electrospinning

Electrospinning has drawn much attention and has already been as one of the most important techniques for manufacturing nonwoven polymer nanofiber membranes (synthetic or bio-based) with diameters ranging from nanometers to microns. Many geometries of the membrane are available for a diverse range of applications, along with agri-food, biomedical, cosmetics energy, and even textiles. For producing fibers from a polymer solution and synthesizing sophisticated three-dimensional structures, the electrospinning process employs thousands to tens of thousands of volts of high-voltage electrostatic repulsion [68]. A large number of synthetic polymers, including polycaprolactone (PCL), poly-lactic acid, and poly(lactic-co-glycolic acid), as well as more than 200 different types of natural polymers and composites, such as gelatin, silk fibroin, chitosan, and collagen, have been used in electrospinning technology systems. With this technology, the synthesized nanofibers have a high surface area-to-volume ratio, excellent plasticity, and flexibility, which can improve cell adhesion, proliferation, and differentiation activities. Lyu and his coworkers [69] studied the electrospun cellulose acetate (CA) membranes with optimized porous structures. They demonstrated that the synergy between pore size and porosity is extremely crucial for moisture-induced electricity generation from porous AC membranes. They will also prove that the pore size and porosity of CA membranes can be easily adjusted using a compression process. Several investigations are made to apply the membranes manufactured by electrospinning. For example, Cheng et al. [70] developed a new type of deacetylated cellulose acetate (DA)-polydopamine (PDA) composite nanofiber membrane by electrospinning. The membrane was employed as a highly efficient adsorbent for removing methylene blue (MB) from an aqueous solution. The results demonstrated the successful development of a uniform PDA coating layer on the surface of DA nanofibers. After 30 h of adsorption, the

DA-PDA nanofiber membrane had an adsorption capacity of 88.2 mg/g at a temperature of 25°C and a pH of 6.5, which is approximately 8.6 times greater than that of DA nanofibers. The experimental results revealed that the DA-PDA composite nanofiber's adsorption behavior followed Weber's intraparticle diffusion model, the pseudo-second-order model, and the Langmuir isothermal model. According to a thermodynamic analysis, endothermic, spontaneous, and physisorption processes were observed. Furthermore, Acid black 172 was removed from simulated wastewater using electrospun CA/P(DMDAAC-AM) composite nanofibrous membranes [71, 72]. The equilibrium adsorption capacities were 116, 159, and 192 mg/g, respectively, when the proportion of P(DMDAAC-AM) to CA was 20, 30, and 40wt.%. When the proportion of P(DMDAAC-AM) was 40%, the average fiber diameter was 185 nm, and the maximum adsorption capacity could reach 231 mg/g. CA/P(DMDAAC-AM) nanofibrous membrane adsorption capacities had a well-linear relationship with P(DMDAAC-AM) content and fiber diameter.

4. Additives

CA has been widely used in the manufacture of membranes due to its hydrophilicity, adsorption properties for dyes, heavy metals, and low cost. But in order to improve permeability, separation efficiency, and antifouling properties, in many studies, the CA membranes were prepared by adding certain additives such as composites, nanomaterials, surfactants, and additive polymers to modify the membranes [73, 74].

Ounifi et al. [56] developed new UF membranes incorporating polyacrylic acid (PAA) as an additive. The strong hydrogen interactions between the two polymers observed with FT-IR and the presence of a single peak in the DSC results confirmed the homogeneity of the mixture of CA and PAA in the prepared membranes. The membranes presented asymmetric and highly porous structures with morphologies affected by the PAA concentration. Indeed, the porosity and permeability increase proportionally with the concentration of PAA and inversely proportionally with the contact angle. The benefits of blending CA with PAA have also been observed in terms of antifouling properties, allowing efficient flux recovery from the membrane fouled by the treatment.

CA/TiO₂ hybrid membranes were prepared by phase inversion method, by dispersing titanium dioxide (TiO₂) nanoparticles in the CA casting solutions [49]. The influence of TiO₂ on the morphology and thermal stability of CA/TiO₂ membranes has been studied. The SEM results show that the addition of TiO₂ nanoparticles causes the hybrid membrane to become more porous due to the increase in average pore size. The thermogravimetric analysis results show that the interaction exists between TiO₂ nanoparticles and AC. Moreover, the thermal stability of the hybrid membrane was improved by the addition of TiO₂ nanoparticles. PWF membranes show that the addition of TiO₂ nanoparticles leads to an increase in water permeation.

Ultrafiltration membranes based on CA modified with zinc oxide (ZnO) were prepared [75]. Membrane performance was improved. For example, mechanical stability, morphology, contact angle, and porosity were evaluated on modified CA and ZnO samples. The increase in ZnO concentration in the membrane leads to a decrease in permeability and an increase in hydrophobicity compared with the pure CA membrane.

To improve antifouling properties and shedding, PVP has been added to CA to produce a membrane [76]. PVP is a hydrophilic additive polymer and blowing agent.

From the results, it was significant that the membrane mixed with the hydrophilic additive PVP was improved and showed better properties in terms of surface roughness, hydrophilicity, and thermal and mechanical stability.

CA membranes modified with various surfactant additives, including Span-20 (sorbitan monolaurate), Tween-40 (polyoxyethylene sorbitan monopalmitate), and Tween-60 (polyoxyethylene sorbitan monostearate), were prepared [77]. The effect of these additives has been studied. The membranes' behavior depends on different concentrations of surfactant. It was found that even adding small amounts of the studied surfactant additives could affect the separation behavior and cause the formation of macro voids and finger-like structures. In addition, the effect of surfactant hydrophobicity on the contact angle and water permeation flux of the prepared

Polymer	Additives	Preparation technique	Membrane	Application	Ref
CA	Ag-Nps	NIPS	UF	Water flux 1651 L m ⁻² h ⁻¹	[78]
CA	SiO ₂	NIPS	UF	Permeability increased from 32 to 82 kg h ⁻¹ m ² bar ⁻¹	[79]
CA	SiO ₂	TIPS	Gaz separation	CO ₂ /CH ₄ selectivity 80	[80]
CA	Fe ₃ O ₄	Vacuum filtration of nanoparticles on commercially CA membrane	NF	Cu ²⁺ Rej.: 63.2% Cd ²⁺ Rej.: 64.1% Cr ⁶⁺ Rej.: 70.2%	[81]
CA	TiO ₂	NIPS	UF	Removal of Chrome (VI)	[82]
CA	ZnO	Casting	UF	Antibacterial activity against <i>S. aureus</i>	[83]
CA	ZrO ₂	NIPS	Hollow fiber	Arsenic removal of 87.24% and permeability of 89.94 L m ⁻² h ⁻¹ bar ⁻¹	[84]
CA	CNT	NIPS	RO	5 g L ⁻¹ NaCl Rej.: 96% Flux: 100 L m ⁻² h ⁻¹ (35 bar, 0.01 wt% CNT)	[85]
CA	GO	NIPS	RO	The synthetic seawater salt rejection of the CA membrane modified with 1 wt.% GO at 25 bar was 1.8 times greater than that of the pure CA membrane	[86]
CA	PVA	NIPS	Hemodialysis	42.484 L/m ² h is the maximum flow of water CA-PVA rejected up to 95% of bovine serum albumin	[87]
CA	PEG	NIPS	UF	The pure water flux elevated from 49.5 Lm ⁻² h ⁻¹ to 62.2 Lm ⁻² h ⁻¹ Rejection result for chromium metal of 31.89%	[88]

Table 1.
Some of the additives used with CA in membrane preparation.

membranes was investigated. It was found that the membrane derived from tween-60 has the lowest flux and the highest contact angle (77.2°) (**Table 1**).

5. Application of CA membranes

5.1 Microfiltration

In a combination with solvents, CA and polyethyleneimine (PEI), which were cross-linked by polyisocyanate, were combined to prepare a modified MF membrane via phase inversion approach. PEI may provide linkage sites for affinity-separated ligands in the membranes or serve as ligands for metal chelation, endotoxin elimination, or ion exchange [46]. The impacts of phase inversion time, induced by water vapor mixed with PEI and cross-linking agent concentrations, were examined in the function of membrane performance. The prepared blend membranes with 0.15 wt.% PEI/CA ratio and 0.5 wt.% cross-linking agent/PEI were used to adsorb Cu^{2+} and bovine serum albumin (BSA) separately. The produced membrane has a maximum Cu^{2+} ion adsorption capacity of 7.42 mg/g dry membrane. The maximum BSA adsorption capacities are of 86.6 and 43.8 mg/g dry membranes for the membranes with and without the chelating Cu^{2+} ion, respectively.

Commercial CA-MF membranes were modified to exhibit antibacterial capabilities by being impregnated with thymol mixed in very critical solvents. Batch impregnation was used, and the thymol loading was watched as a result of treatment pressure and time. *Staphylococcus aureus* and *Pseudomonas aeruginosa* were used to test the obtained materials' antibiofilm capacities, while *S. aureus* and *Escherichia coli* were used to test the blockage of membranes in contact with bacteria. The finding's results showed a quick impregnation process that could achieve high thymol loads in just 0.5 h at 15 MPa and 20 MPa. A concentration of 20%Thymol exhibited high antibiofilm effects against the tested strains without changing the membrane performance. The research revealed that the specified polymer could benefit from these potent antibacterial characteristics. These great antibacterial characteristics could be sustainably added to the commercial polymer structure of membranes. The results revealed also that the modification of the commercial cellulose acetate microfiltration membranes with polymeric structure might be successfully accomplished in a quick (30 min) and safe environmental procedure. Therefore, modified membranes with great antibacterial properties may find use in the biomedical sector and venting.

5.2 Ultrafiltration

Ultrafiltration (UF) membrane was prepared by Ounifi et al. [53] by using the Manjikian process by phase inversion with progressively rising polymer concentrations (cellulose acetate at 15, 16.5, 18, and 21 wt.%). At increased polymer concentrations, the membranes showed improved ability to retain pathogenic and epidemiological Uro Pathogenic *Escherichia coli* (UPEC). The membranes with the highest polymer concentration were able to hold back an entire discharge of 8 U-log bacteria.

By using a blending method, new fouling-resistant UF membranes were prepared for cadmium (Cd) remediation by fusing the adaptability of CA with the intricate characteristics of poly(acrylic acid) (PAA). Since CA/PAA membranes performed better than pure CA membranes in UF experiments, it was clear that the mixture had

many advantages. Overall, PAA is a useful additive for creating CA membranes with enhanced antifouling exploitable qualities for the UF treatment of heavy metal-polluted water bodies.

The synthesis, evaluation, and use of polyurethane (PU) with cellulose acetate blend membranes are all part of this research [89]. Numerous characterization methods, including contact angle measurement, ATR-FTIR, water content, and SEM, have been used to investigate the characteristics of PU-CA blend membranes. It was determined that common functional groups were used as raw materials (polyurethane and cellulose acetate) in the PU-CA mixed membrane. It was demonstrated by contact angle measurements and water content values that cellulose acetate's addition decreased the hydrophilicity of the modified membranes, due to the fact that pure polyurethane membrane has higher hydrophilicity than pure cellulose acetate. Aqueous potassium dichromate solution used for ultrafiltration as one of the promising separation techniques was used to test the suitability of the modified membranes. The PU-CA1 membrane was experienced varied chromium ion concentrations for many pH and pressure levels. At pH 3 and 0.4 MPa, process economy was attained for all chromium ion concentrations. The PU-CA1 membrane offers a novel option for the treatment of effluents in the textile sector due to its specific properties.

5.3 Nanofiltration

CA-NF membranes were prepared by using the phase inversion method [54]. The concentration of cellulose acetate was varied from 22 to 25 wt.% to investigate its effect on the characteristics of the membrane. The membrane's characterization revealed that the increase of CA concentration makes it more hydrophilic and decreases the membrane's porosity, consequently reduction in water permeability across the membrane, while it increases salt retention. Knowing that the size exclusion mechanism predominates the transport mechanism through the membrane, the ions rejection was sequenced in the following order: $R(\text{Na}_2\text{SO}_4) > R(\text{CaCl}_2) > R(\text{NaCl})$ as determined by the hydration energy and hydrated radii of the ions. CA NF membranes have been successfully used for cadmium elimination. The rejection rate of removed cadmium reached up to 98%. In terms of steric hindrance and hydration energy, the results revealed that the Cd^{2+} coupled with divalent anions is more rejected than that coupled with monovalent anions. Additionally, the retention reduces from 10^{-4} to 10^{-2} mol L^{-1} when the salt concentration rises. Thus, the obtained results showed the feasibility of using developed CA-NF membranes in removing cadmium from contaminated water.

In order to increase the rejection of Pb^{2+} ions and the permeation flux, vinyl triethoxysilane-graphene oxide/gum arabic was added as filler to modify the CA-based membranes [39]. By using NF membranes, the rejection of Pb^{2+} ions was studied by investigating the effect of contact time, pH, permeation flux, and pressure. It was demonstrated that at pH 9, the rejection reached its optimal with 97.6%, whereas a low pH of 1–6 was not required for salt rejection. The permeation flux was $8.6 \text{ l m}^{-2} \text{ h}^{-1}$, which is caused by concentration and pressure variations. The special characteristics of the nanomaterial-based membrane and their connection to the current treatment methods offer great prospects to revolutionize water and wastewater treatment as well as to further our knowledge of membrane performance. NF-CA may present a significant factor in the commercial manufacturing of membranes.

5.4 Reverse osmosis

Diainabo et al. aimed to evaluate the pertinence of CA, PSF, and silica (SiO₂) for the treatment of wastewater. For this purpose, PSF-blended CA hybrid filtration membranes (CA/PSF) also PSF-blended and SiO₂-embedded CA hybrid adsorption membranes (CA/PSF-SiO₂) were prepared and characterized for Cu²⁺, Fe²⁺, and Zn²⁺ ions elimination from contaminated aqueous solutions. Phase inversion method was used to prepare the membranes by using CA, PSF, and SiO₂ granules that were dissolved in N, N dimethyl formamide (DMF). Different pore sizes were detected at the rough surfaces and cross sections of the membranes, which were studied by using SEM. The obtained membranes' porosity and pore sizes ranged from 26.8 ± 0.3 to 81.1 ± 0.3 μm and from 1.26 to 1.38%, respectively, based on the differentiation. The membranes' contact angles varied from 49 to 76 degrees on their glass side and from 56 to 77 degrees on their air side [90].

More than 90% of the initial metal ions present in the polluted solution, which was concentrated at 40 mg L⁻¹, were taken up by the hybrid filtration polymer membranes. CA/PSF-SiO₂ membranes were used in adsorption tests. These substances were found to have a higher capacity for adsorption than many other well-known adsorbents in the literature, which reach 70 mg g⁻¹ for CA/PSF 85/15-SiO₂ for Cu²⁺.

Membrane type	Membrane properties	Application fields	Retention	Ref
Electrospun CA	Hydroxyapatite (CA/HAp) nanocomposite	Fe ³⁺ and Pb ²⁺ ions rejection	99.7–95.46%	[92]
Electrospun nanofibrous membranes of CA	Grapheme oxide/magnetite/hydroxyapatite/cellulose acetate nanofibrous membranes	Cr ⁶⁺ , Se ⁴⁺ and methylene blue removal	93.4–95.1%	[93]
CA	Deacetylated cellulose acetate (DA)@polydopamine (PDA)	Methylene blue removal	88.15 mg/g	[94]
CA	CA/MOFDPC (metal-organic frame work derived porous carbon) membranes	Methylene blue removal	87.9%	[95]
CA polymeric membrane	Polymeric membrane impregnated with ZnO nanostructures	Methylene blue removal	75%	[96]
CA	Cellulose acetate/grapheme oxide/sodium dodecyl sulphate (CA/GO/SDS)	phosphate removal	87.2%	[97]
CA	Cellulose acetate/block copolymer membranes	Polyphenols	67.4314 mg/g	[98]
CA based mixed matrix membrane (MMM)	Fe-Al-Mn@chitosan/cellulose acetate	Fluoride de contamination	Excellent defluoridation performance	[99]
CA phthalate (CAP)	Poly(methacrylic acid) (PMAA)/CA	Heavy metals Cu ²⁺ , Hg ²⁺ and Cd ²⁺	4.8 mg/g	[100]

Table 2.
 Some of the application of CA membrane.

Finally, it was discovered that the removal of Cu^{2+} , Fe^{2+} , and Zn^{2+} could be improved by coupling adsorption with ultrafiltration in the tangential mode. This allowed for a removal efficiency of 95% for Cu^{2+} at a low metal concentration of 60 mg L^{-1} , and at a high metal concentration of 900 mg L^{-1} , it showed a great removal efficiency of around 98% [90].

In order to remove metal ions from AMD, the performance of a combined filtration and adsorption process was studied. The prepared membranes showed the ability to extract metal ions from polluted solutions at low concentrations of 40 and 60 mg L^{-1} , whereas at a high concentration of 900 mg L^{-1} , the filtration was ineffective. The selected adsorbents demonstrated a better adsorption capacity than those from other adsorbents used in the literature. The maximum adsorption efficiency with the CA/PSF 85/15- SiO_2 membrane reached between 35 and 70 mg g^{-1} of Cu^{2+} , Fe^{2+} , and Zn^{2+} ions, while the adsorbent had the lowest capacity adsorption with CA/PSA- SiO_2 membrane reached from 20 to 61 mg g^{-1} toward the same metal ions. By combining the effects of the filtration and adsorption membranes, a great metal uptake was obtained at both low and high concentrations. Consequently, this strategy presents a reliable method to remove the metal from aqueous solutions, and it has potential use in industrial wastewater treatment (Table 2) [91].

6. Conclusion

Cellulose acetate (CA) as one of the most important derivatives of cellulose was studied. Cellulose acetate has attracted big attention as a polymer due to its desirable properties, which are biodegradability, hydrophilicity, environment friendliness, sustainability, and low cost. In this chapter, the use of cellulose acetate CA in the fabrication of polymeric membranes was studied. CA-based membranes are applied in different fields such as gas separation, pharmaceutical industries, adsorption, water treatment, and wastewater treatment by using separation membrane processes such as nanofiltration, ultrafiltration, microfiltration, and reverse osmosis; in addition, the different configurations of fabricated CA membranes such as flat sheet, hollow fiber, and electrospinning were discussed. Also, the different preparation methods such as phase inverse and interfacial polymerization approaches were outlined. The CA-based membrane has the property to be blending to organic and inorganic additives. To improve the performance of the CA-based membranes and their structural properties, different cost-effective additives were added to the polymeric solution, especially nanomaterials. The obtained CA-based membranes with additives were characterized by their high biodegradability, high hydrophilicity, and high retention of dye molecules and heavy metals.

Abbreviations

CA	cellulose acetate
PS	polysulfone
PP	polypropylene
PE	polyethylene
PES	polyethersulfone
PVDF	polyvinylidene fluoride
PVA	polyvinyl alcohol

CS	chitosan
PCL	polycaprolactone
PDA	polydopamine
MF	microfiltration
IP	interfacial polymerization
NIPS	and non-solvent-induced phase separation
TIPS	thermally induced phase separation
UF	ultrafiltration
NF	nanofiltration
OR	reverse osmosis
HF	hollow fiber membrane
TFC	Thin film composite
NMP	N-methyl pyrrolidone
DMAc	N, N-dimethyl acetamide
DMF	N, N-dimethylformamide
PIP	Piperazine
CHMA	1,3- cyclohexanebis (methylamine)
TFC	M-phenylenediamine
MPD	trimesoyl chloride
HTC	cyclohexane-1,3,5tricarboxylchloride
PEI	polyethyleneimine
PAA	poly(acrylic acid
MB	methylene blue
PU	polyurethane
SEM	scanning electron microscopy
AFM	atomic force microscopy
FTIR	Fourier-transform infrared spectroscopy
TAG	thermogravimetric analysis
HMR	homogeneous-reinforced

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