

Review

# Polysaccharide Films/Membranes for Food and Industrial Applications

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**Abstract:** Membrane processes are extensively employed in a range of industrial and food applications. Due to growing environmental concerns and the introduction of regulatory measures, it is imperative to develop innovative membrane materials that can effectively replace petrochemical-based polymers, in line with the principles of a circular economy. The focus of this review is the use of polysaccharides for obtaining films/membranes for food and industrial applications using selected case studies. Besides the polysaccharides extracted from biomass, the valorization of agrifood residues and the use of plants adapted to arid lands (i.e., cactus) to produce polysaccharide films for food packaging is addressed. Moreover, microbial polysaccharides produced using renewable resources present a significant alternative to commercial hydrophilic membranes for gases and ethanol dehydration. To meet industry requirements, the mechanical and barrier properties of the films can be improved by the inclusion of inert impermeable fillers and/or the chemical modification of the polysaccharides. The adsorption of proteins, dyes, and pharmaceutical compounds using a cellulose-based polymer is discussed. Despite their unique characteristics, polysaccharide production costs are still higher than most synthetic polymers. This is a challenge that can be overcome by scaling up the production and by valorizing agro-industrial wastes and by-products to make the application of polysaccharide membranes/films in the food and industry sectors more widespread.

**Keywords:** polysaccharides; membranes; packaging; dehydration; adsorption



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## 1. Introduction

A membrane consists of a polymer film which can be used as a selective barrier in several separation processes. Membranes are used in gas permeation, water treatment, and in health and food applications; however, in food packaging they are normally called films.

The petrochemical-based polymers are still dominant in membrane development, which is not compliant with a circular economy approach. Therefore, the relevance of using environmentally friendly materials, such as polysaccharides, for the preparation of membranes is mandatory.

Polysaccharides can be extracted from biomass or produced by microorganisms. The latter, known as microbial polysaccharides, presents an excellent alternative to the other category of polysaccharides (animal, plant, or algae-derived products), since they are produced using renewable carbon sources under controlled operating conditions, assuring the quantity and the quality of the final products [1,2].

Due to their good barrier properties to gases (oxygen and carbon dioxide), biodegradability, and biocompatibility, polysaccharides have been used as membranes/films in

various applications across multiple sectors, including the chemical industry, food production, and medicine. Since they have a hydrophilic nature, they show low barrier properties against water vapor. Therefore, the improvement of polysaccharide-based membrane properties, namely mechanical and barrier properties to water vapor and resistance to liquid water, is needed. The strategies may involve the incorporation of various additives, such as lipids, the formulation of blends utilizing different polymers, the design of multilayered membranes, the application of nanoparticles, and the chemical modification of polysaccharides [3].

In this review, the use of polysaccharides for obtaining films/membranes for food and industrial applications is addressed using case studies based on the last 10 years of my research and discussing them with a new insight and in an integrated way. They are also complemented by studies from other research groups on the topic.

Besides the polysaccharides usually extracted from plants, algae, or animals, corn and tomato processing residues can be used to obtain arabinoxylans and cutin, respectively. They show different characteristics: arabinoxylans have hydrophilic properties and are very good barriers to gases, while cutin is hydrophobic, impeding water transport through the film.

The use of plants particularly adapted to arid lands and severely degraded soils, such as cacti, can be explored to obtain pectins and betalains. There are few studies concerning the development of active films incorporating betalains extracted from the cactus pear which can be used as sensors for spoilage through application as active/intelligent food packaging.

The importance of microbial polysaccharide production, which can advantageously be used as an alternative to other natural polysaccharides, is also addressed. They only depend on microbial cultivation parameters, which may be easily controlled and may exhibit enhanced properties, competing with both natural polysaccharides and synthetic polymers. Nevertheless, their use has been limited mainly due to their costs of production which are still higher than the traditional petrochemical-based polymers. To decrease the polymer production costs, several agro-industrial wastes and by-products have been proposed as substrates for microbial cultivation. Microbial polysaccharide films for food packaging and membranes for solvents and gas dehydration are presented.

The chemical modification of polysaccharides allows for less favorable properties to be addressed or new desired characteristics to be developed. This is especially known for cellulose due to its extremely low solubility in water and most common solvents. Cellulose-based polymers (ethers, esters) can be obtained by the substitution of their hydroxyl groups with other functional groups. The removal of proteins and dyes, as well as pharmaceutical compounds, using a recently developed cellulose-based polymer (dicarboxymethyl cellulose—DCMC) combined with membrane filtration is presented.

## 2. Development of Films/Membranes for Food Applications

### 2.1. Films for Food Packaging

Packaging is an essential component of the food industry. It functions as a protective container for products, safeguarding them from shocks, vibrations, and compressions. Moreover, packaging plays a critical role in the preservation of food by serving as a barrier against chemical and biological contamination. Ultimately, it enhances the shelf life of products throughout the distribution and storage processes, ensuring the maintenance of quality and safety while significantly minimizing food waste [4].

Packaging uses synthetic plastics, which are non-biodegradable and non-renewable materials creating serious environmental problems. Therefore, the search for alternative

materials in food packaging is mandatory, reducing the carbon footprint and using fewer toxic reagents [5].

#### 2.1.1. Films from Polysaccharides Extracted from Biomass

Polysaccharides are usually obtained from plants, algae, or animals (e.g., cellulose, starch, alginate, chitosan, and pectin) and have been widely used for edible and/or biodegradable film development.

Cellulose, a highly available polymer, is composed of D-glucose units linked by  $\beta$ -1,4 glycosidic bonds. It is known for its chemically modifying capacity and is receiving growing attention in the field of food packaging, due to its biocompatibility, sustainability, and biodegradability [6]. NatureFlex™ commercialized by Futamura are eco-friendly cellulose films, which are compostable and made from GMO-free renewable resources, providing an alternative to traditional petroleum-based plastics.

Starch consists of amylose and amylopectin, whose properties and ratios vary depending on the source [7]. Starch films have been developed from corn starch or cassava starch using solvent casting or extrusion processes [8–10]. These are more adequate for industrial production; however, plasticizers and other polymers are often added to obtain starch films with adequate mechanical and barrier properties. Films using a starch-based antimicrobial agent (OCSI) and environmentally friendly polymers (PVA, PBAT, and PCL) have presented excellent UV blocking, antimicrobial and mechanical properties, and low water vapor permeability, as well as good food preservation [11]. Commercial films MATER-BI, produced using starch, cellulose, and vegetable oils, are produced by Novamont company (Novara, Italy).

Alginate is obtained from brown algae and is composed of unbranched, linear binary copolymers of  $\beta$ -D-mannuronic acid (M) and  $\alpha$ -L-guluronic acid (G) residues linked by 1–4 glycosidic bonds. The source of the alginate affects the ratio of M and G residues, which impacts its physical and chemical properties. Alginate films have been used for packaging, extending the shelf life of food products [12–14].

Chitosan derives from the deacetylation of chitin. It is non-toxic and biocompatible, with antimicrobial and antifungal properties [15]. Chitin can be obtained from seafood industry waste, as well as by synthesis with different microorganisms. Properties such as the degree of acetylation (DA), nitrogen composition, N/C ratio, molecular size, and polydispersity are dependent on the source of chitin. In general, chitin has a DA above 90% and a nitrogen content of 7%. Chitosan has a DA of less than 40% and nitrogen content greater than 7% [16].

Pectin, mainly composed of galacturonic acid units, is a safe, edible substance commonly used in coatings due to its effective barrier properties against lipids, oxygen, and aromatic compounds [17]. Pectin can be extracted from the by-products of the fruit and vegetable industry, offering a valuable opportunity for resource optimization. The degree of esterification (DE) is an important parameter for pectin applications, and it is defined as the percentage of carboxyl groups esterified present in the structure of pectin [18].

Biopolymers frequently require enhancements to their mechanical and rheological properties, which can be achieved through molecular restructuring or the incorporation of food-grade additives. In addition to possessing the required mechanical properties, these films must ensure adequate permeability for water vapor and gases. The desired specific permeability characteristics can be attained by integrating inert impermeable fillers and/or reactive compounds into the polymer matrix. The inert fillers can reduce permeability by increasing the diffusion path, while the reactive compounds interact selectively with the diffusing species, increasing the time before significant permeability occurs [19].

The use of blends and multi-layers can produce new composite materials, with properties similar to synthetic polymers [20,21]. Blends and bilayer films of chitosan with pectin or alginate [22,23], gelatin [24,25], or whey and zein fiber [26,27] have shown higher water vapor permeability, better mechanical properties, and lower water solubility compared with chitosan films.

The use of different fillers (clays, nanocellulose, and metal oxides) with different polysaccharide films to reduce water vapor and gases (CO<sub>2</sub> and O<sub>2</sub>) permeabilities and improve mechanical properties have been studied by several research groups [28,29]. Composite films of polysaccharides (pectin and carrageenan); gellan gum, mica particles, and nanosheets [19,30]; chitosan films with montmorillonite (MMT) and essential oils [31]; and films with ZnO nanoparticles, obtained with an eco-friendly route using apple peel, tomato, and passion fruit extracts [32], have been produced. Nanoparticles (NPs) produced using plant extracts are less toxic since phytochemicals can form a protective layer that reduces the toxicity of nanoparticles (NPs) and enhances antibacterial activity through their synergistic effects in plant extracts [33,34].

### 2.1.2. Films from Polysaccharides Extracted from Cactus

Cactus plants are well adapted to arid lands and *Opuntia ficus indica* (OFI) can be found in Mexico and Mediterranean countries. The fruits, prickly pears, have been used for human consumption due to their bioactive compounds (e.g., ascorbic acid, betalains, flavonoids, and phenols) [35,36] with antioxidant, anti-cancer, and anti-inflammatory properties. Aiming at promoting cactus plantation in marginal lands of Mediterranean countries, with minimum pressure on the available water resources and to add value to the final product and waste, different extraction methods of mucilage and betalains from *Opuntia* spp. can be used [37]. Furthermore, there are no studies concerning the development of active films incorporating betalains extracted from the cactus pear which can be used as a food protector and/or as sensors for spoilage, having applications as active/intelligent food packaging [38].

Intelligent food packaging is an innovative technology designed to monitor the quality and safety of food throughout its shelf life. This technology utilizes indicators and sensors that detect physiological changes in food caused by microbial and chemical degradation. These indicators can signal the freshness of the packaged product through noticeable color changes, which can be easily recognized by both food distributors and consumers. However, most of the current indicators are made from synthetic materials. Therefore, it is crucial to enhance the sustainability of food packaging, and the selection of sensors should reflect this need [39–44].

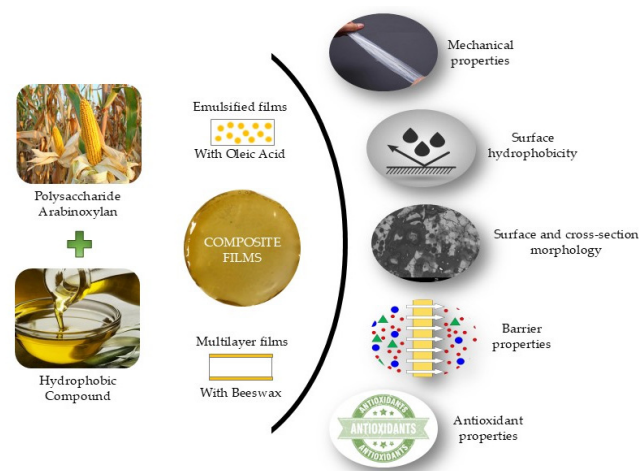
### 2.1.3. Films from Polysaccharides Obtained from Agrifood Residues

The valorization of residues from the agrifood industry to produce biopolymers is also particularly important. Corn fiber, a by-product from the wet milling of corn for food applications, is used in animal feed. However, it has valuable components that should be valorized, such as arabinoxylans and ferulic acid.

Arabinoxylans are obtained after the alkaline extraction of corn fiber followed by ultrafiltration for concentration and the partial removal of small compounds. The further purification of the arabinoxylans fraction was assessed by ultrafiltration operated in a diafiltration mode, at different temperatures and at different operation regimes (under controlled transmembrane pressure conditions and under controlled permeate flux conditions). The obtained arabinoxylans, with the optimized parameters of the purification step, were used to produce films, with different plasticizer contents (glycerol, 15, 30, and 50% dry basis) and citric acid as a crosslinking agent. In addition, films with the incorporation of ferulic

acid were developed to obtain active barriers with antioxidant activity. Due to their intense brownish color, they were decolorized using hydrogen peroxide and were characterized in terms of mechanical properties (tensile tests), water adsorption capacity, and permeability to water vapor, oxygen, and carbon dioxide. The films presented antioxidant activity, water vapor permeability, and mechanical properties similar to that of non-decolorized films [45].

Arabinoxylan, which was previously extracted from corn fiber through alkaline hydrolysis, was utilized to create composite and multilayer films. The composite films were produced by casting an oil-in-water emulsion containing oleic acid, while the multilayer films (composed of beeswax–arabinoxylan–beeswax) were made by submerging the arabinoxylan films in a beeswax solution. These films were characterized in terms of their antioxidant activity, optical and mechanical properties, surface hydrophobicity, and barrier properties against water vapor, gases, and ultraviolet–visible (UV–vis) radiation (Figure 1). The multilayer films exhibited enhanced barrier properties, improved UV–vis radiation resistance, and higher water contact angle values compared with the emulsion-based films. The prepared films presented good potential to be used as packaging materials for food products with a low water content [46].



**Figure 1.** Arabinoxylan films—composition and properties. Reproduced from [46].

Composite films of cellulose with arabinoxylans-rich extract from brewer’s spent grain showed higher hydrophobic behavior and antioxidant activity showing the potential to be used for active packaging. Wheat bran arabinoxylans were incorporated into chitosan films, providing health benefits due to their prebiotic properties [47].

Tomato pomace is a valuable and economical renewable resource that has been extensively studied for the extraction of polyester cutin, which is predominantly formed from long-chain hydroxy fatty acids. These fatty acids serve as excellent precursors for the synthesis of innovative hydrophobic biopolymers [48]. Furthermore, films composed of pectin and cutin, which emulate the natural structure of tomato peel, have been successfully developed through a casting methodology [49].

Monomers of cutin were extracted from tomato pomace and utilized to produce cutin/chitosan films from cutin extracts and commercial chitosan to be used in food packaging. This research shows the application of tomato pomace for biopolymer development and provides an application for the valorization of tomato processing waste [50].

#### 2.1.4. Films from Microbial Polysaccharides

Intensive research is being conducted on the production of microbial polysaccharides (gellan, pullulan, hyaluronan, bacterial cellulose, and bacterial alginate) as they represent an advantageous alternative to others recovered from animals or plants. They only depend

on the microbial cultivation parameters, which may be easily controlled and may present new or improved properties, being competitive with natural polysaccharides, as well as with synthetic polymers.

Nevertheless, their use has been limited due to their production costs which are higher than most of the traditional petrochemical-based polymers. To decrease the polymer production costs, several agro-industrial wastes and by-products have been proposed as substrates for microbial cultivation.

The use of bacterial cellulose (BC) as a biodegradable food packaging material has been reviewed recently. Although most studies are focused on biomedical applications, there is an enormous potential to apply BC in packaging, either as films or as reinforcing agents [51].

Gellan gum is a water-soluble polysaccharide produced by the bacterium *Sphingomonas* sp. It is composed of a repeating tetrasaccharide unit containing 1,3- $\beta$ -D-glucose, 1,4- $\beta$ -D-glucuronic acid, 1,4- $\beta$ -D-glucose, and 1,4- $\alpha$ -L-rhamnose [52]. GG-based materials have been used for food packaging applications to extend the shelf life of different food products, such as fish [53] and fruits and vegetables [54,55].

Other polysaccharides have emerged, namely the microbial polysaccharide (FucoPol) which was produced using glycerol, a surplus of the biodiesel industry [56]. The film-forming capacity of this new polysaccharide was evaluated, and the films obtained by casting were characterized in terms of their hygroscopic, mechanical, and barrier properties for food applications. Bilayer films of FucoPol and chitosan showed an improved performance in terms of water vapor and gas barrier properties (Table 1). The bilayer films have been shown to be less permeable to O<sub>2</sub> than FucoPol films, being good candidates to be used for the packaging of low moisture content products [57]. Similar findings were obtained for bilayer films of zein/chitosan and pectin/chitosan. This could be explained by the polar interaction of the interfacial polymer during the layers assembly which led to the formation of a dense structure of the two-layer film matrix [58,59].

**Table 1.** Barrier properties of FucoPol, chitosan, and bilayer films.

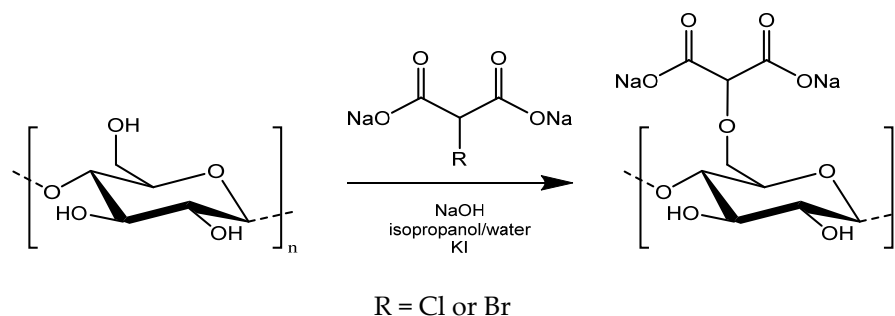
Film	$P_{O_2} \times 10^{16}$ (mol/m·s·Pa)	$P_{CO_2} \times 10^{16}$ (mol/m·s·Pa)	$P_w \times 10^{11}$ (mol/m·s·Pa)
FucoPol	1.9	6.5	0.8
Chitosan	2.4	15.0	4.1
Bilayer	0.5	5.8	1.7

$P_{O_2}$ —oxygen permeability,  $P_{CO_2}$ —carbon dioxide permeability,  $P_w$ —water vapor permeability.

## 2.2. Porous Adsorptive Membranes for Wine Clarification

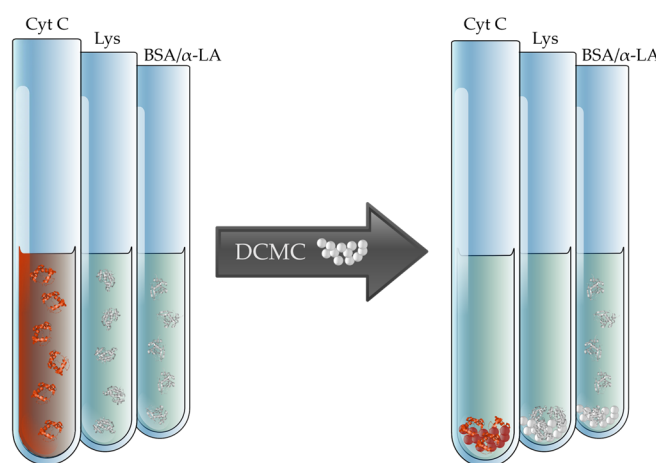
The presence of residual grape proteins in bottled white wines can turn them hazy during transport or storage. This is extremely negative for consumers and may have a severe economic impact. Thus, it is necessary to remove these proteins and the most common way is using bentonite, which can adsorb positively charged proteins from wines. However, this procedure produces large quantities of lees and between 1 and 10% of wine is wasted [60].

Cellulose constitutes the most abundant renewable polymer resource available worldwide and chemical modification allows for the less favorable properties to be addressed or new desired characteristics to be developed [61]. Recently, a dicarboxymethyl cellulose polymer (DCMC) (Figure 2) was developed and patented [62] which can perform ion exchange at low pH values of 2.5–3.5, which is the pH of white wine.



**Figure 2.** Schematic representation of the synthesis of DCMC [62].

Its capacity for the adsorption of cationic species was evaluated for proteins and model wine proteins [63,64]. DCMC successfully adsorbed positively charged proteins, cytochrome C (Cyt C), and lysozyme (Lys) and did not promote the adsorption of uncharged proteins,  $\alpha$ -lactalbumin ( $\alpha$ -LA) and bovine serum albumin (BSA), at pH 7.4 (Figure 3).



**Figure 3.** Adsorption of proteins with DCMC. Reproduced from [64].

A hybrid adsorption–membrane filtration (AMF) process can combine adsorption with filtration. This method provides an alternative to common fixed-bed columns, without sacrificing efficiency. It was evaluated for cytochrome C with over 95% removal.

However, the development of DCMC porous membranes would allow the coupling of adsorption and filtration, providing the reutilization of the membranes. For the porous membranes, a novel method based on Pickering emulsions recently developed with two different biopolymers (PVA and gelatine) [65,66] can be applied with DCMC. Emulsion templates attract great attention owing to the facile tailoring of pore size and distribution in polymeric materials [67]. Other methods have also been reported for the fabrication of porous membranes (phase inversion and electrospinning) with green solvents to substitute the toxic compounds normally used [68,69]. The production of porous DCMC membranes using these solvents would make the process even more sustainable.

### 3. Development of Polysaccharide Membranes for Industrial Applications

Polysaccharide-based membranes are increasingly recognized for their effectiveness in solvent and gas dehydration, as well as in wastewater treatment processes. They demonstrate remarkable capability in the removal of aromatic compounds, dyes, and heavy metal ions, highlighting their potential to address significant environmental issues.

These membranes may represent a critical advancement in the development of efficient purification technologies.

### 3.1. Membranes for Solvent and Gas Dehydration Processes

The dehydration of organic solvents is essential for the synthesis of pharmaceuticals, solvent extraction, and drying of final products. The solvents are usually azeotropic mixtures and conventional distillation cannot surpass the azeotropic concentration and has a high energy impact. Thus, alternative processes are required to accomplish an extremely low water content and pervaporation can be used due to its high selectivity. However, suitable membranes with high permeability and selectivity are needed since it has a high impact on the cost of the installation and the performance of the process.

Gas dehydration is also a critical issue in industry due to the corrosion problems that water in the presence of acid gases (H<sub>2</sub>S and CO<sub>2</sub>) may cause. Water vapor removal can be applied in diverse areas, namely in gas dehydration (flue gas and biogas), and air treatment for packaging and processing industries.

For dehydration, hydrophilic membranes of polyvinyl alcohol (PVA), polysulfone (PS), and polyamide (PA) are the most used [70]. However, the development of new materials that are stable and have a long lifetime is crucial [71]. Polysaccharides, such as chitosan and sodium alginate, have already been evaluated in pervaporation for solvent dehydration (i.e., ethanol, isopropanol, tetrahydrofuran, and acetone) with high selectivity and water flux [72,73]. The use of microbial polymers as membranes for pervaporation is reduced and only bacterial cellulose membranes have been referred for ethanol dehydration [74,75].

Membranes using the extracellular polysaccharide FucoPol, the glycerol by-product of the biodiesel industry, produced at a low cost, and with an abundant carbon source, were developed. These membranes showed a selectivity for water–ethanol of 100 (Table 2) for the dehydration of ethanol with 10 wt. % water, with the selectivity of water–ethanol (w–et) being the ratio of the permeabilities of water and ethanol through the membrane.

**Table 2.** Water–ethanol selectivity of the polysaccharide membranes (FucoPol and hybrid FucoPol) and a commercial membrane (PERVAP<sup>®</sup> 4101).

Membrane	[Water] <sub>feed</sub> (wt. %)	Selectivity (w–et)
FucoPol	10.2	100
Hybrid FucoPol	10.0	570
PERVAP <sup>®</sup> 4101	9.9	554

w–water, et–ethanol.

The permeability can be calculated using Equation (1):

$$J_i = P_i / \delta (p_{i,feed} - p_{i,perm}) \Leftrightarrow J_i = P_i / \delta (\gamma_i * x_{i,feed} * p_i^{sat} - p_{i,perm}) \quad (1)$$

where *i* is the target compound (water or ethanol), *J<sub>i</sub>* is the molar flux (mol/m<sup>2</sup>·s), *P<sub>i</sub>* is the permeability (mol/m·s·Pa), *p<sub>i,feed</sub>* and *p<sub>i,perm</sub>* are the partial pressures in the feed and the permeate (Pa), respectively, *δ* is the thickness of the membrane (m), *γ<sub>i</sub>* is the activity coefficient, *x<sub>i,feed</sub>* is the molar fraction in the feed stream, and *p<sub>i</sub><sup>sat</sup>* is the saturation vapor pressure (Pa).

To reinforce the mechanical and thermal properties, a hybrid membrane was developed incorporating a SiO<sub>2</sub> network by using a solgel method with (3-Glycidioxypropyl) trimethoxysilane (GPTMS) as a crosslinker silica precursor. These membranes were applied for ethanol dehydration by pervaporation with a high selectivity water/ethanol (570) (such as PERVAP<sup>®</sup> 4101 commercial membrane). Thus, they could emerge as a viable alternative

to commercial hydrophilic pervaporation membranes, presenting significant opportunities for advancements in the field.

Since, for industrial applications, it is necessary to evaluate the membrane lifetime, long-term operation data is indispensable. The results obtained for the hybrid polysaccharide membrane on 3 consecutive days are shown in Table 3.

**Table 3.** Hybrid polysaccharide membrane (Hybrid FucoPol) stability in consecutive pervaporation experiments.

Experiment Days	$P_w \times 10^{12}$ (mol/m·s·Pa)	$P_{et} \times 10^{13}$ (mol/m·s·Pa)	Selectivity (w-et)
1	7.7	0.14	570
2	9.7	0.53	182
3	14.7	6.2	24

$P_w$ —water permeability,  $P_{et}$ —ethanol permeability.

It is possible to notice that the hybrid polysaccharide membrane is not stable for long-term operation. There is a decrease in membrane selectivity from 570 to 182 after 24 h of operation and to a value of 24 after 36 h. This is due to the swelling of the membrane, allowing water and ethanol to permeate. This excessive swelling leads to the selectivity decrease due to the change in the membrane structure [76].

Gas dehydration, namely flue gas and biogas dehydration, was also studied using these membranes. Water vapor, pure gases ( $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2$ ), and gas mixtures at different conditions of relative humidity were used and the FucoPol hybrid membranes showed an excellent performance with permeability values below 3.0 barrer (1 barrer =  $3.35 \times 10^{-16}$  mol/m·s·Pa) and a high selectivity for water vapor with values of 4000 for  $\text{H}_2\text{O}/\text{CH}_4$  and 300 for  $\text{H}_2\text{O}/\text{N}_2$ . To assess the stability of the process, it was evaluated using the same membrane during 20 consecutive experiments (7 h each). The membrane maintained its gas barrier properties after long-term exposure to water vapor because the swelling was not so severe in this case [77].

Due to their hydrophilic characteristics and resistance to acidic solutions, these membranes could have potential for application in the dehydration of acids and solvent processing by nanofiltration. Chitosan membranes blended with deep eutectic solvents have been used for the pervaporation of polar/non-polar organic mixtures [78].

### 3.2. Membranes for Wastewater Treatment

The use of biopolymers as adsorption materials for the treatment of contaminated sites is a promising technology. A higher removal of certain pollutants, such as aromatic compounds, dyes, and heavy metal ions, has been achieved with polysaccharide-based materials compared with other commercial sorbents currently used in wastewater treatment processes [79]. Chitosan combined with polyacrylamide gel was used to remove dyes from industrial wastewater effluents, thus substituting the excessive cost of commercial adsorbents [80].

Chitosan hollow fiber adsorptive membranes containing Fe (III) were prepared for the removal of As (V) from aqueous solutions since both Fe (III) ions and chitosan exhibited a high affinity for arsenic [81].

The adsorption of herbicides was achieved using membranes composed of chitosan and alginate. These biopolymers can produce multilayer membranes, using the electrostatic forces between the anions of the carboxylate groups of alginates and the cations of the protonated amines of chitosan [82]. They were also used for the removal of the herbicides, Paraquat, Diquat, Difenzoquat, and Clomazone, which is strongly related to the electrostatic charge, partition coefficients, and dissociation constants of the herbicides. The

chitosan/alginate membranes may be interesting for the adsorption of different herbicides in each layer of the membrane, e.g., a positively charged herbicide can be adsorbed onto the alginate layer and a negatively charged herbicide can simultaneously be adsorbed onto the chitosan layer [83].

Silver nanoparticles (AgNPs) were incorporated into a cellulose membrane for the adsorption of pesticides (Cypermethrin, Paraquat, and Cartap). The AgNPs were synthesized using an eco-friendly method by reacting *Mentha piperita* (mint) extract with AgNO<sub>3</sub> aqueous solution. The high adsorption capacity obtained with the cellulose–AgNPs membrane makes it an excellent alternative for the remediation of wastewater polluted with pesticides [84]. The removal of dyes was accomplished using composites of chitosan, alginate, and cellulose with clays and zeolites [85–87].

Using the dicarboxymethyl cellulose (DCMC) polymer with commercial porous membranes, the adsorption of methylene blue (MB) was studied by coupling adsorption and filtration. An extremely high removal rate (95% MB) was achieved and DCMC was successfully reused in consecutive experiments, thus contributing to a more sustainable process [88].

This could also be applied in wastewater treatment for the removal of micropollutants. These are called emerging contaminants and consist of an extensive group of synthetic and natural compounds, including pharmaceuticals, personal care products, steroid hormones, and agrochemicals that negatively affect human and animal health. Many of them have basic atoms and are, therefore, easily protonated in wastewater.

The prevalence of pharmaceuticals in urban freshwater systems is a critical issue that demands urgent intervention. Conventional wastewater treatment is not able to eliminate these compounds, so they are commonly discharged with treated effluent into rivers, lakes, and estuaries [89]. While rivers have been studied more extensively, the streams that flow through cities tend to have higher concentrations of pharmaceuticals, posing greater risks to animals, plants, and human health. Thus, the treatment of these waters at the source will be the most suitable way of eliminating these contaminants, avoiding their dilution [90,91].

DCMC was successfully used for the removal of metformin (a medicine to treat type 2 diabetes), propranolol (a beta blocker used for heart diseases), and benzalkonium chloride (a surfactant used in laundry detergents and softeners for textiles) with over 60% efficiency.

A hybrid adsorption–membrane filtration process (AMF), coupling DCMC with a commercially available polyethersulfone (PES) porous membrane with 0.45 µm pore diameter, was evaluated for propranolol removal. The removal was highly effective (70%) due to the high adsorption capacity of DCMC. Three consecutive cycles of adsorption/desorption were completed and DCMC maintained the same adsorption capacity [92].

#### 4. Conclusions and Future Perspectives

In this review, the use of polysaccharides for obtaining films/membranes for food and industrial applications was presented. Besides the polysaccharides extracted from biomass, corn and tomato processing residues were used to obtain arabinoxylans and cutin, respectively. Another source of polysaccharides are plants particularly adapted to arid soils, such as cactus, and they were used to obtain pectins and betalains. Microbial polysaccharides were advantageously used to produce membranes for dehydration.

However, the improvement of these polysaccharide-based membranes/films, regarding their mechanical properties, resistance to liquid water, and permeability to water vapor and gases, is crucial. Strategies include the use of additives, blends with different polymers, design of multilayered membranes, use of nanoparticles, as well as the chemical modification of polysaccharides. The removal of proteins and dyes, as well as, pharmaceu-

tical compounds, using a cellulose-based polymer (dicarboxymethyl cellulose—DCMC) combined with membrane filtration was presented.

Although polysaccharides are an excellent alternative, their use has been limited mainly due to their production costs, which are higher than those of most petrochemical-based polymers. Waste valorization and scaling up production will be crucial to overcome this challenge. The widespread application of polysaccharides will have profound impacts on numerous sectors, fostering the development of new membranes/films, increasing the effectiveness of existing products, and minimizing environmental impacts.

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