

# **Membrane Separation for Food, Medical/Pharmaceutical Applications**

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### **ABSTRACT**

This paper reviews recent work on food, medical/pharmaceutical applications of artificial nanofiber membranes development. The invention of nanofibers opened avenues, towards its application in food and medical/ pharmaceutical. Nanofiber can be made from a wide range of natural, synthetic and hybrid polymers with a variety of physical, chemical and mechanical properties. Nanofibers are one-dimensional materials in fiber shape with diameters in the nanometer range (10 m-9 m). These nanofibers have unique properties such as a high surface-to-volume ratio which offers large surface area, nanoporosity and mass transport properties. Specific attention is given to the application of the membranes in food technology such as, (dairy Industries, purifications of fruit juices etc.) drug delivery systems, cell membranes for cancer treatments, artificial organs, tissue engineering and including its possible uses to fight against corona. Emerging membrane-based organ-on-chip technology was also discussed. The membrane materials, fabrication methods and the current state of the art were also reported and future prospects were discussed. **Keywords:** Membranes; Food industry; Medical; Dairy; Wine; Polymers; Ceramic membranes

## **INTRODUCTION**

Membrane separation processes operate without heating and use less energy than conventional thermal separation processes such as distillation, sublimation or crystallization. The separation process is purely physical and both fractions (permeate and retentate) can be used. Separation is based on principles that rely on the chemical and physical properties of particles and molecules. Membrane filters are very thin microporous sheets of film attached to a thicker porous support structure [1]. At its most basic, a membrane serves as a sieve, separating solids from liquids forced through it. Not only can membranes separate solids from liquids, but they can also separate soluble molecules and ionic particles of different sizes from each other.

## **MATERIALS AND METHODS**

Membrane can be divided into three groups.

**•**Biological membranes

- **•**Artificial membranes
- **•**Theoretical membranes

Artificial or synthetic membranes can be divided as composite membranes and nanocomposite membranes. Further membranes can be divided as:

#### **Biological membranes**

- **•**Cell membrane and intercellular membranes
- **•**Mucous membranes
- **•**S-layer
- **•**Serous membranes and mesothelial surrounded organs

#### **Artificial membranes**

- **•**Reverse osmosis
- **•**Filtration (microfiltration, ultrafiltration)
- **•**Distillations
- **•**Pervaporation
- **•**Dialysis

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- **•**Emulsion liquid membranes
- **•**Membrane based solvent extraction
- **•**Membrane reactors
- **•**Gas permeation
- **•**Supported liquid membranes

All these membranes are well discussed in the literature and well known. Each of these categories is distinguished by the size of the species they retain. Retention is based on the pore size of the membrane [2].

Membrane Contactor (MC) technology is common in the medical field. In many applications, the contactor is not even called a contactor but is referred to by other names, depending on the specific application in which it is deployed. Examples include blood oxygenator (the earliest use of a membrane contactor), gas transfer membrane, membrane degasser, membrane deaerator, Membrane Distillation (MD) device, Osmotic Distillation (OD) device, membrane gas absorber, membrane extractor and membrane humidifier pervaporation technique by using non porous perm selective membrane is also used for liquid feed mixture. For the food industry, the following applications are being researched.

- **•**Alcohol removal from wine
- **•**Aroma recovery from fruit juices, beer, herbal and flowery extracts
- **•**Restoration of aroma components during fermentation

Pervaporation is, despite its successes and potentials, so far not established in the food industry. Pullulan is also a naturally occurring substance derived from the fungus Aureobasidium pullulans and its film is sometimes referred to as 'edible packaging'. Pullulan is a polysaccharide polymer consisting of maltotriose units, also known as  $\alpha$ -1,4-;  $\alpha$ -1,6-glucan'. Three glucose units in maltotriose are connected by an α-1,4 glycosidic bond, whereas consecutive maltotriose units are connected to each other by an  $\alpha$ -1,6 glycosidic bond (Chemical formula:  $(C_6H_{10}O_5)_n$  (Figure 1).



Pullulan is an important polysaccharide with unique characteristics. This natural biopolymer is water-soluble, nonmutagenic, non-immunogenic and non-toxic. Pullulan is a former film and binder popular in the production of breath strips [3]. However, it also appears as an ingredient in cosmetics and beauty products because of its solubility in water and adhesive properties.

#### **Applications of pullulan**

- **•**Edible films
- **•**Pharmaceutical applications
- **•**Production of antimicrobial films from
- **•**Medical applications
- **•**Biotechnological applications
- **•**Oral delivery of insulin by polymer-coated liposomes

In terms of biodegradable membranes/nanofibers, the material's biocompatibility is more important than its structure.

#### **Polymeric membrane preparation techniques**

The polymeric membrane preparation techniques are common. Table 1 shows organic membrane preparation techniques which are widely in use (Figure 2).



**Table 1:** Membrane preparation techniques.



Figure 2: Morphology (SEM image): A) Sintering, B) Stretching, C) Track-etching, D) Solution coatings (cross section of a composite membrane and E) Phase inversion.

**Sintering:** This technique is based on the compression of the particles of the selected material and heating to a temperature below the melting point to induce a sticking of the particles. Membranes are fabricated from powders of polymeric materials such as Polyethylene (PE) and Polypropylene (PP) or inorganic materials such as stainless steel, metal oxides and graphite or glass [4].

**Track-etching:** The principle of the technique is the irradiation of a thick film of polymer, usually polyester or polycarbonate. This technique presents uniform pores with cylindrical geometry.

**Stretching:** This technique is used to prepare porous membranes starting from extruded dense films of semicrystalline polymers such as PE, PP and PTFE.

**Solution coatings:** This technique allows the preparation of composite membranes, the use of which is desirable when the separation to be accomplished is controlled by the diffusion rather than by the size of the species to be separated. Membranes prepared with this technique are formed by two different materials, the first of which is a thin dense film made of a selective polymer deposited on top of the second one, which merely acts as porous support. Dip coating, spray coating, spin coating and plasma polymerization are some of the procedures for preparing composite membranes [5].

**Phase inversion:** This is the most versatile technique which allows the preparation of all kinds of membranes. Only polymeric membranes can be prepared via this technique.

#### **Ceramic membranes**

Ceramic membranes are a relatively new technology being developed in the filtration field. Ceramic membranes are very resistant to the action of aggressive media (acids, strong solvents). They are very stable chemically, thermally and mechanically and biologically inert. Even though ceramic membranes have a high weight and substantial production costs, they are ecologically friendly and have a long working life.

Ceramic membranes are generally made as monolithic shapes of tubular capillaries. Ceramic membranes are made from inorganic materials (such as alumina, titania, zirconia oxides, recrystallized silicon carbide or some glassy materials). By contrast with polymeric membranes, they can be used in separations where aggressive media (acids, strong solvents) are present [6]. They also have excellent thermal stability which makes them usable in high temperature membrane Ceramic membranes are used for membrane separation processes in the dairy industry, due to their excellent pH, temperature and cleaning tolerance, in addition to high flux performance; however, in recent years, polymeric membranes have become increasingly popular due to their costeffectiveness operations.

#### **Nanofibers/Scaffold**

The invention of nanofibers opened many avenues, such as its application in water treatment, medical/pharmaceutical, separation of gases, membrane technology, etc.

Nanofibers are one-dimensional materials in fiber shape with diameters in the nanometer range (10 m-9 m). These nanofibers have unique properties such as a high surface-to-volume ratio which offers large surface area, nanoporosity and mass transport properties. Nanofiber can be made from a wide range of natural, synthetic, and hybrid polymers with a variety of physical, chemical and mechanical properties. Electrospinning is an easy electro hydrodynamic fabrication method used for producing nanofibers by an electrical force.

Nanofibers can be prepared from various materials, but in the pharmaceutical and biomedical context, they typically comprise a polymeric carrier loaded with a drug. There are various techniques for fabricating pharmaceutical nanofibers.

- **•**Drawing
- **•**Phase separation
- **•**Melt-blown technology
- **•**Template synthesis
- **•**Force spinning
- **•**Freeze-drying
- **•**Electrospinning

Spinnerets play a major role in the performance of the ENFs (Electrospun Nanofibers). Though electrospun nanofibers are used in drug delivery, there are some limitations, such as the low production yield and the clogging of the needles used as spinnerets. Moreover, since the electrospun nanofibers are intended for clinical applications, it is of significant importance to ensure inter-batch homogeneity and implement high-quality control methods. To overcome these problems, a new approach, named Needleless Electrospinning (NLES), is proposed. NLES is the process of fabricating nanofibers by electrospinning a polymeric solution directly from an open liquid surface. The spinnerets utilized in NLES are divided into two groups *i.e.,* rotating and stationary spinnerets. NLES have advantages over conventional ES, such as a higher production yield and the absence of needle-associated problems. However, the process is not fully proven in controlling electrospun fiber quality and the process is multivariate. Controlling electrospun fiber quality and homogeneity is challenging and requires further investigation [7].

By using polymer reagent routes, ceramic nanofibers are fabricated by the sol-gel technique. Polymeric solutions containing precursors are electrospun and afterward, the nanofibers go through either chemical conversion or thermal treatment to synthesize ceramic nanofibers. The presence of oxygen in the air then proceeds to the conversion of ceramic particles to oxides. Pyrolyzing in an inert atmosphere  $(N_2 \text{ or } Ar)$ changes the polymer structure to a carbon and graphite structure (Table 2).

Rasouli et al. discussed the emerging applications of nanofibers in biomedical and healthcare fields. Recent research on nanofibers is going on drug delivery systems and controlled drug release, tissue - engineered scaffolds, dressings for wound healing, biosensors, biomedical devices, medical implants and skincare, as well as air, water and blood purification systems. Attention is also given to different fibers (e.g., mesoporous, hollow, core shell nanofibers) fabricated from various materials and their potential biomedical applications. In recent years, synthetic polymers have been developed into nanofibers by the electrospinning method and evaluated as a scaffold material for regenerative medicine. Recently many articles concerning nanofibrous scaffold and their applications in the medical field have appeared in different scientific journals (Figure 3).



**Table 2:** A comparison of the various modification methods for nanofibers.



Considering the unique characteristic properties of nanofibers, Leena et al. discussed its applications in beverage industries such as filtration of beverage products, beverage packaging materials, encapsulation matrices for protection of aromatic and volatile compounds in the beverages, nanosensors for quality assessment, monitoring storage conditions and pesticide detection. Yadav et al. discussed the recent advancements in the fabrication of carbon nanofibers and additionally pointing out their significance for applications in biomedical sector for their potential role in the future. Bhattarai et al. summarized several key aspects of electrospinning in the use of electrospun fibers in drug delivery with a special emphasis on electrospun nanofibers impregnated with nanoparticles [8].

da Silva discussed the developments on the preparation and application of electrospun nanofibers in food-related areas, including filtration and other separation strategies, enzyme immobilization, enzymatic membrane reactors, sensors and encapsulation of food bioactive compounds and cells. Nanofibers have desirable mechanical properties suitable for application in the food packaging industry.

#### **Cell membranes**

The cell membrane is one of the most fundamental constituents in biological systems, creating the physical boundaries of cells. Cell membranes are made from thin sheets of fatty molecules

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called lipids; they are at least ten times thinner than an iridescent soap bubble and yet allow cells to collectively form organisms as diverse as bacteria, trees and people. Cell membranes are very selective about what they let pass through, using tiny embedded proteins as gatekeepers. Membranes repair dings to their structure automatically and change thickness to pass signals from the outside environment to the cell's interior.

#### **Polymer electrolyte membrane**

Polymer membranes may be functionalized into ion exchange membranes by adding highly acidic or basic functional groups, e.g., sulfonic acid and quaternary ammonium, enabling the membrane to form water channels selectively transport cations or anions, respectively.

#### **Membranes in food technology**

Membrane technology has made a tremendous impact on the food industry over the last several years. The separation of materials for different applications has become an important industrial operation. Considerable progress continues to be made in membrane technology and newer applications for existing systems are being discovered as the trend is to create integrated systems which utilize several different membrane types within a process. Membrane separations can be used either as alternatives to conventional techniques or as a novel technology for processing new ingredients and foods. Membrane separations are considered green technologies. In many cases, membrane processes are more advantageous than traditional technologies. Guiga and Lameloise discussed the main membrane technologies available for food applications in the liquid phase, including membrane materials, modules and implementation [9].

The technology can be applied to several production methods, mainly in;

**•**Milk-solids separations in the dairy industry

**Table 3:** Comparison of important characteristics of different membrane modules.

- **•**Treatment of waste water from food industry treatment
- **•**Juice clarification and concentration
- **•**Concentration of whey protein
- **•**Sugar and water purification and waste management

Several filtration mediums exist as well as many types of membrane configurations. Knowledge of the various membrane technologies and how they are used in the food industry can enhance overall production and offer cost-cutting options for various separations. The module design or support structure of a membrane is critical to its performance. Some factors to consider include flux (the rate of permeation), the solids content of the process fluid, cost, cleanability and scalability. Food industry applications use four basic module designs: Spiral-wound, tubular, hollow-fiber and plate-and-frame styled systems.

Spiral-wound, mainly for dairy and other soluble protein processing, polysaccharide gum concentration and most RO and NF applications.

**Tubular systems:** Its accounts are about 10% to 15% of food industry applications. Tubular designs have a porous outer structure with a semi-permeable membrane coating on the inside of the tube. Tubular membranes are suitable for beverage clarification or the reverse osmosis of pulp-containing juices.

**Plate-and-frame and hollow-fiber systems:** These are among the miscellaneous configurations that make up the remaining percentage of designs used in food industry applications. In plate-and-frame styles, flat-sheet membranes are affixed to both sides of a porous plate and sandwiched in a holder. Hollow-fiber membranes are similar to tubular membranes except that the hollow fibers are much smaller. The fiber's inside diameter may range from 0.5 mm to 1.1 mm as opposed to 12.5 mm to 25 mm for the tubular design (Table 3).



Reverse osmosis has become a standard process in the food industry. It is used to purify water for plant operations, to concentrate cheese whey proteins or milk in the dairy industry, for sugar concentration in the cereal processing industry, for the concentration of juices and wastewater treatment in meat and fish processing industries. Reverse osmosis deserves a special look because of its suitability for a wide variety of applications.

## **RESULTS AND DISCUSSION**

#### **Dairy industries**

The dairy industry is a predominant part of the food industry. Membrane applications in the dairy industry date back to the late 1960s with the concentration of milk proteins to make Ricotta cheese, using the MMV process based on ultrafiltration, named after its inventors, Maubois, Mocquot and Vassal among the different membrane technologies encountered in this sector, Microfiltration (MF) is a very attractive separation process. It offers various fractionation possibilities of milk components and makes it possible to ensure dairy products' safety and create products with high added value and new functionalities [10].

Membrane separation technology is a technology with suitable properties for dairy products in which solution is passed through a membrane of microscopic pores and pressure applied to separate the components. Milk is a complete food and its products are a good source of essential nutrients for human health and raw materials for other industries. At present, the application of membrane separation technology in the dairy industry mainly includes sterilization of dairy products, the concentration of dairy products, desalination of whey, fractionation of milk protein, yogurt concentration, etc. Each milk protein has different nutritional functions and specific functional properties. Different molecular weight cut-off membranes are used to fractionate and purify milk proteins to obtain different proteins, such as whey protein, serum albumin, lactoglobulin/-lactalbumin, egg albumin, Simmond wood protein, etc.

### **Treatment of wastewater treatment industries**

The dairy industry is the major wastewater source in food processing that contains a large amount of organic matter and nutrients. Dairy or food industry wastewater usually does not contain commonly cited hazardous and toxic chemicals. Dairy proteins are valuable products and used as high-value food additives, nutraceuticals and therapeutics. Catenacci et al. presented a critical review focusing on applications of membrane-based technologies for the innovative management of wastes produced by the dairy sector.

Membrane separation usually plays an important role in secondary biological treatment as it is energy-saving and straightforward and has zero wastewater emissions. During this processing, protein and sugar are also recycled from the wastewater. Membrane with a different molecular weight cut off plays different roles in wastewater treatment. Table 4 shows the applications of different membrane separation technologies for wastewater treatment.

**Table 4:** Applications of different membrane separation technologies in dairy wastewater treatment.



Andrade et al. evaluated the application of Membrane Bioreactor (MBR) as secondary and Nanofiltration (NF) as tertiary treatment for the reuse of dairy wastewater, focusing on determining the best NF operating conditions. It was observed that the proposed treatment system's (MBR+NF) overall efficiencies for COD were 99.9% and 93.1% for total solids. The final treated wastewater could be reused for cooling, steam generation or washing of external areas and trucks. A few recent processes used to treat wastewater from food industries, including main operating conditions including major findings.

Kuwait Institute for Scientific Research (KISR) installed a complete system including biological treatment, Powdered Activated Carbon (PAC) and submerged membrane microfiltration system (CMF) using propylene membrane for treating the dairy effluent. The average removal efficiencies of the system for Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Dissolved Solids (TDS) were 98.8%, 92.5%, 96.7%, respectively (Figure 4).



Figure 4: Integrated (CMF-S) treatment systems.

This integrated system is shown in Figure 4, in which feed water passes through an aeration tank for biological treatment. The specifications of these membrane modules as the following.

- **•**Number of modules: 4
- **•**Membrane types: Polypropylene
- **•**Membrane area: 13 meach
- **•**Filtration direction: Outside in
- **•**Fiber outside diameter: 650 μm+30 μm
- **•**Fiber inside diameter: 390 μm+20 μm
- **•**Number of fiber: 14,500 nominal

The treated effluents will flow through PAC to suppress any lactose in the feed effluent and reduce any odor in the feed water. So far, not a single separation technique could be successfully used as a stand-alone system. NF will be successful only in combination with other suitable processes. In the future, the focus should be given to the design and fabrication of new generation surface engineered superior composite membranes to cater to the need of new applications by fine-tuning their morphology and physicochemical properties like hydrophilicity, porosity, charge density and thermo-mechanical stability and thereby introducing distinct functionalities (such as antimicrobial, photosensitive or adsorptive capabilities) [11].

### **Electrodialysis**

Electrodialysis and pervaporation techniques are also used in the dairy industry. Electrodialysis is a unit operation applied for the separation or concentration of ions in a solution, based on their selective electromigration through semipermeable membranes under the influence of a potential gradient. Electrodialysis is a comparatively new process in terms of its use in the food industry. Some applications of ED in the food industry are as follows.

- **•**Demineralization of milk and whey
- **•**De-acidification of fruit juice
- **•**De-ash sugar solutions

Electrodialysis operation has been widely used for demineralization in the dairy industry and has successfully been applied to desalinate skimmed milk. It has shown that the technique is useful in the demineralization of dairy products. Bazinet et al. reported that electrodialysis with filtration membranes could be applied to recover molecules with bioactive properties such as antioxidant, anticancer and antihypertensive peptides. It was also reported that electrodialysis, an electrochemical separation process with charged membranes stacked to separate ionic species from aqueous solutions and uncharged components when an electrical field is applied, is a new membrane separation process with numerous applications in the food, nutraceutical and beverage industries.

#### **Fruit juices**

Membrane-based operations have proven to be attractive alternatives to the conventional clarification and concentration methodologies of fruit juices from both economic and qualitative points of view. Pressure-driven membrane operations, membrane distillation, osmotic distillation and pervaporation have been widely investigated in the last few decades to replace conventional technologies used in the fruit juice processing industry (*i.e.,* clarification, stabilization, concentration and recovery of aroma compounds). Conidi et al. discussed membrane-based operations in the fruit juice processing industry.

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Advantages of membrane technology over traditional separation:

- **•**High selectivity based on unique separation mechanisms
- **•**No thermal stress of processed fluids due to moderate operating temperatures
- **•**No use of chemical additives
- **•**Easy scale-up
- **•**Compact and modular design
- **•**Low energy consumption

Advantages of membrane tech for the juice clarification pressuredriven membrane operations, including MF, UF, NF and RO are widely used. Recently other membrane operations such as Osmotic Distillation (OD), Membrane Distillation (MD) and Pervaporation (PV) have been used for juice concentration and recovery of aroma compounds.

#### **Flavors/Aroma recovery (liquid food processing)**

Among the available techniques for flavor recovery in the food industry, distillation or stripping, pervaporation, supercritical fluid extraction and adsorption showed potential for selective recovery of the flavor components from liquid food streams. Different chemical substances contribute to particular flavor perceptions, as is depicted in Figure 5.



#### **Miscellaneous**

The amnion is a membrane that closely covers the embryo when first formed. It is the innermost layer of the placenta and consists of a thick basement membrane and an avascular stromal matrix. Amniotic membrane transplantation has been used as a graft or as a dressing in different surgical subspecialties. Amniotic membrane transplantation is a good and viable option in ocular surface diseases and reconstruction. The amniotic membrane, used as a graft or as a substrate for the cultivation of limbal corneal cells, has shown encouraging results, Gheorghe et al. suggested that Trattler et al. reported that the use of amniotic membranes, including non-healing epithelial defect, Herpes Simplex Virus (HSV) and Herpes Zoster Virus (HZV) keratitis,

neurotrophic keratitis, Stevens-Johnson syndrome/toxic epidermal necrolysis and dry eye disease [12].

Scientists have developed new fluorescent probes that prove the existence of cell membrane structures called 'lipid rafts', allowing researchers to study how toxins and viruses invade cells.

Portland state university researchers used advanced electron microscopy to create a 3-D reconstruction of a membrane protein at an unprecedented level of resolution, setting the stage for the development of drugs that could target the protein more effectively to treat a variety of diseases scientists have developed new fluorescent probes that prove the existence of cell membrane structures called 'lipid rafts', allowing researchers to study how toxins and viruses invade cells.

Since the outbreak of COVID-19, there has been a worldwide shortage of face masks particularly the N95 ones worn by health care workers. Due to shortages, many healthcare workers have repeatedly worn the same N95 mask, even though they are intended for single use. Researchers from the King Abdullah university of science and technology have developed a membrane that can be attached to a regular n95 mask and replaced when needed. Researchers placed the silicon-based template over a polyimide film and used a process called reactive ion etching to make pores in the membrane, with sizes ranging from 5 nm-55 nm. The researchers then peeled off the membrane and attached it to an N95 mask. They claim that their new filter is more efficient in blocking virus particles than the standard N95 masks.

The ongoing COVID-19 epidemic worldwide necessitates the development of novel effective agents against SARS-CoV-2. ACE2 is the main receptor of SARS-CoV-2 S1 protein and mediates viral entry into host cells. Wang et al. claimed that the membrane nanoparticles prepared from ACE2-rich cells have the full capacity to block SARS-CoV-2 infection. Wang group demonstrated that it is an easy-to-achieve membrane nanoantagonist for curbing SARS-CoV-2, which enriches the existing antiviral arsenal and provides new possibilities to treat COVID-19. Multiple major health organizations recommend the use of Extracorporeal Membrane Oxygenation (ECMO) support for COVID-19-related acute hypoxaemic respiratory failure.

### **Conclusion**

Membrane operations provide the dairy industry with reliable, safe, clean and sober processes. Membrane separations can be used either as alternatives to conventional techniques or as a novel technology for processing new ingredients and foods. Membrane separations are considered green technologies.

In many cases, membrane processes are more advantageous than traditional technologies. The use of pullulan in the biomedical field has seen recent expansion due to its nonimmunogenic, non-toxic, biocompatible and inert nature. In comparison to dextran, the degeneration rate of pullulan in serum is quicker than that of dextran. It seems that in the future, the functionalized chitosan nanofibers will open a new avenue for a wide range of applications in tissue engineering and regenerative medicine. There have been significant interests in the uses of polymeric membranes in the medical/pharmaceutical area. Most applications of membranes have been reported in drug delivery systems, artificial organs and tissue engineering.

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