

The Application of Membrane Technology in the Concentration and Purification of Plant Extracts: A Review

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Abstract

The obtained plants and by-products during food and agricultural manufacturing processes are sources for many bioactive components that attract industrial and academic interest. The essential method of obtaining these bioactive components is the extraction process by using solvents. The efficiency of the extraction processes mainly depends on the choice and selectivity of these solvents. However, the most challenging step is recovering the components from the solvent to obtain the active part and pure products. In this recovery process, many methods were applied, such as evaporation and adding assistant chemicals, which had many downsides as energy consumption and unwanted product. Consequently, membrane technology such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), membrane distillation (MD), and osmosis distillation (OD) has been applied as a new approach in concentrating plants extract. Since this new approach has proved its efficiency in this field, the main objective of this paper is to provide a review of academic studies that have addressed using different membrane techniques to concentrate the plant extracts.

Keywords

microfiltration, ultrafiltration, nanofiltration, membrane distillation, osmotic distillation, reverse osmosis, plant extracts, concentration

1 Introduction

Plants and their active ingredients have attracted people for many years. People have used plants in treating many diseases and relieving pain, and using plants for these purposes is as old as humanity. Moreover, the connection between people and their search for drugs in nature dates from the far past [1]. For centuries, plants have been of great interest to humans as flavors, fragrances, dyes, preservatives, and pharmaceuticals [2]. Today, medicinal plants are of great importance due to their significant properties as a great source of therapeutic phytochemicals that may lead to new drugs' development. Much research indicates that most phytochemicals from plant sources such as phenols and flavonoids have a positive effect on health and cancer prevention [3], treatment of diabetes [4], cardiovascular diseases [5], in addition to their role against bacteria and pathogens [6].

To begin with, the study of medical plants begins with pre-extraction and extraction procedures, which are the main steps in the processing of bioactive ingredients from plants. There are many methods used in these extraction and separation processes such as, maceration, infusion, percolation and decoction, Soxhlet extraction or

hot-continuous extraction, microwave-assisted extraction (MAE), ultrasound-assisted extraction (UAE) or sonication extraction, accelerated solvent extraction (ASE), supercritical fluid extraction (SFE) [7].

The second steps in obtaining these active substances are purification and concentration; for instance, the crude extracts from solvent extraction are unusable immediately, and intensive treatment such as purification or refining is required. Achieving the usability of a plant-based material involves concentrating on the desired products and removing unwanted materials alongside separating products from an organic solvent. Therefore, making an extracted-plant material usable is, generally, the most challenging aspect of producing natural compounds. The conventional purification approaches include distillation, evaporation to remove solvents, or the usage of additives such as caustic for oil refining processes. Distillation requires a significant amount of energy. Adding chemicals such as caustics to crude extracts can also lead to undesirable results, including molecular cross-linking and rearrangements resulting in a decrease in nutritive value and

the formation of toxic compounds. Furthermore, from an environmental point of view, conventional processes of obtaining active substances from plants consume large amounts of water and chemicals and create heavily contaminated effluents [8].

In recent years, researchers have paid a lot of attention to membrane technology, and they have considered it an environmentally benign technology for purifying natural extracts. For two decades, researchers have used various membrane-based technologies to separate, restore and concentrate bioactive compounds (such as phenolic compounds, anthocyanins, carotenoids, antioxidants, and polysaccharides) from Agri-Food products and their derivatives (such as wastewater), clarification and concentration of natural extracts, recovery of odors from natural and processed products, production of non-alcoholic beverages [9]. In other words, membrane technologies represented a viable alternative to conventional techniques due to the low operating and maintenance costs, moderate operating conditions of temperature and pressure, ease of control and expansion, and highly selective separation. In particular, pressure-driven membrane processes, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [10].

In addition to plant extracts, the waste resulting from food and agricultural-industrial processes is a significant source of active substances, such as antioxidant components, carbohydrates, sugars, pectins, proteins, and phenolic compounds. These active substances exist widely in vegetables (artichoke, olive, maize, etc.), fruits (grapes, apple, pear, cherries, berries, etc.) [11]. Recently, pressure-driven membrane processes, such as MF, UF, and NF, have been applied to the treatment of agro-food wastewaters. These processes have proven their effectiveness and ability to extract and recover these active substances for reuse in the medical, pharmaceutical, and food fields [12].

2 Microfiltration and ultrafiltration

Microfiltration (MF) membranes are used to retain colloidal particles as large as several micrometers. MF overlaps conventional filtration for the separation of small particles. Regarding MF and UF pores' size, microfiltration membranes (MF) have the largest pores, and ultrafiltration (UF) membranes are the next largest. Furthermore, UF and MF look similar, and in fact, they are more alike than they are different. Nonetheless, their different historical background kept them very distinct to practitioners and membrane manufacturers [13].

MF is one of the oldest pressure-driven membrane applications practiced commercially, where it comes second after dialysis [13]. MF can remove micrometer-sized matter, such as suspended particles, major pathogens, large bacteria, proteins, and yeast cells based on the principle of physical separation [14]. Microfiltration grew out of the discovery of nitrocellulose in 1846. Later on, cellulose nitrate membranes were reported by Frick in 1855. Early cellulose nitrate membranes were prepared by dipping a test tube in a collodion solution. Surprisingly, some old materials are still used today [13]. MF membranes are of average pore size between 0.1 and 10 μm where pores are distributed uniformly throughout the membrane. Moreover, MF is done under a pressure gradient of 1–3 bar following the sieving mechanism [15]. The wide range of pore size in these films has allowed them to be applied in many fields such as desalination [16], wastewater treatment [17, 18], and in the food field, especially in the milk and juice concentration and clarification fields [19–21], in the purification of pharmaceuticals [22], and as downstream processing in biotechnology [23].

Historically speaking, the first real ultrafiltration membrane was born in the early 1960s [24]. UF is a process in which a high molecular weight component is rejected by using a fine porous membrane. This process aims at separating water and fine solution from macromolecules and colloids [25]. Ultrafiltration is also one of the membrane separation techniques that separates, purifies, and concentrates solutions between microfiltration and nanofiltration. Furthermore, ultrafiltration membranes reject the molecular weight 500~500000 Da. The approximate diameter of the pore is about 0.001~0.1 μm , the operating pressure difference is generally 0.1~0.8 MPa, and the diameter of the separated component is about 0.005~10 μm [26]. Numerous polymer membranes have been developed for ultrafiltration applications. Polyethersulfone (PES) has been developed as a commonly employed material in ultrafiltration processes for protein separation. Due to its mechanical strength and physicochemical stability, polysulfone (PSF) is an excellent UF membrane material because of its film and membrane forming properties and high mechanical and chemical stability [25].

Although the first aim of developing ultrafiltration membranes was purifying water, it has been applied in many fields since its inception. Among the first applications of ultrafiltration are the recovery of protein and its concentration from cheese whey [27]. Another application in the food industry is gelatin filtration [28], egg

processing [29, 30], and also, commercial ultrafiltration which was applied to clarify the fruit juice [31]. Additionally, the ultrafiltration applications within biotechnology downstream processing are also increasing.

In comparison to the conventional processes, microfiltration and ultrafiltration can bring the following benefits: separation can be done without changing the temperature and pH of the solution and without chemical additives, thus reducing production costs and solving the problem of waste treatment, improving the product quality, and reducing labor costs [31]. However, filtration processes face the problem of membrane contamination, depending on the ratio of particle size to that of the membrane pore. Thus, particles may completely block, partially close, or internally constrict pores.

3 Nanofiltration (NF)

The term NF first appeared commercially by FilmTec (now Dow Chemical Company) in the mid-1980s to describe a new line of membrane products with properties between UF and RO membrane. Since the term NF was not known in the 1970s, such membrane was initially categorized as either loose/open RO, intermediate RO/UF, or tight UF membrane [32, 33].

Regarding its features, the molecular weight cut off (MWCO) of the NF membrane is about 200–1000 Da, which corresponds to pore sizes between 0.5 and 2 nm. Furthermore, Unlike UF and RO membranes which generally carry no charge on their surface, NF membrane often carries positive or negative electrical charges [34]. In most cases, NF membranes are negatively charged in neutral or alkaline conditions and positively charged in highly acidic conditions. Given this, the separation of NF membrane is governed by three distinct mechanisms, namely the steric hindrance (or size sieving), electrostatic (Donnan) exclusion, and dielectric exclusion.

The first generation of NF membranes was manufactured in the early 1970s from cellulose acetate (CA) or its derivatives. These membranes were manufactured based on the well-known dry and wet phase inversion technique of Sidney Loeb [35, 36]. But the poor biological and chemical stability of cellulose-based membranes has limited the range of industrial applications since these membranes have always suffered from constant changes in water flow and solute rejection during operation. Because of these reasons, a second-generation NF membrane was developed based on non-cellulosic materials. This film is a thin film composite (TFC) that consists of three different

layers; a selective layer of ultra-thin polyamide (PA), a small porous inner layer on the upper surface, and the third one is a polyester non-woven underlayment [37]. Generally, the overall structure should have good resistance to acids, bases, oxidation and reduction, high pressures, and sometimes resistance to high temperatures [38]. Since the the first appearance of these films, significant efforts have been devoted to improving their properties and composition. This, in turn, has led to the production of NF films with different separation properties, allowing applications for various industrial processes, and today there are many manufacturers of NF membrane.

As NF membranes differ in many aspects such as materials, morphology, transfer/separation mechanism, and applications, the characterizations of the membrane pore structure; pore radius, pore density, pore shape, pore length, and tortuosity are essential in light of understanding the process. Therefore, characterization methods are major to support the interpretation of dissolved transport, membrane fouling, etc. Several characterization methods that are based on direct automated observation and experimental methods have been applied too. Moreover, various methodologies have been used to investigate this characterization like the gas adsorption-desorption technique, also known as Brunauer-Emmett-Teller (BET) procedure, which allows direct measurement of the pore size distribution. Reverse surface impregnation combined with transmission electron microscopy (TEM) allows direct measurement of pore size and distribution. Atomic force microscopy (AFM) allows direct measurement of pore size, distribution, surface roughness, topography, and force interactions between the membrane and colloids. In addition, many methods and methodologies studied the chemical composition and physical properties of these membranes [39, 40].

Regarding the usage of NF, it has been mainly applied in the procedures of the drinking water purification process, such as treatment and softening water [41, 42] and removing micro-pollutants [43], removing sulfate and electrolytes from seawater [44, 45], and separating heavy metals from contaminated water [46]. Furthermore, using NF membranes in a non-aqueous has also held strong potential in several industrial applications since the 1990s. Due to the lower energy costs involved in the organic solvent membrane processes, a growing interest in applications including solvent recovery in the petrochemical and oleochemical industries [47]. Recovery of polyphenols and valuable components from agro-food by-products [48], as well as separation and purification

of valuable products in the pharmaceutical industry can be observed [49]. Generally, the trend in nanofiltration research has increased since 2007. Besides, nanofiltration membranes continue to see an increasing interest in their use as a separation tool [50].

4 Membrane distillation (MD) and osmotic distillation (OD)

MD and OD are non-pressure-driven membrane processes. They are capable of concentrating liquid foods and non-food aqueous solutions under ambient temperature and pressure, enabling them to preserve the original organoleptic properties of the product. Therefore, they represent attractive processes for concentrating solutions that contain heat-sensitive compounds, such as fruit juices and pharmaceuticals. Unlike pressure-driven membrane processes, the driving force of separation is the difference in vapor pressure across the membrane resulting from either a temperature gradient (in MD) or water activity, i.e., osmotic pressure (in OD) [51].

Historically speaking, on 3 June 1963, the MD process was identified by Bodell, who filed a US patent to describe a device that produces potable water from an importable aqueous mixture [52]. Later on, on the fifth of May 1986, a workshop was held in Rome to find a unique name for a process, previously known by different names such as membrane distillation, thermal evaporation (PV), and membrane evaporation. In that workshop, the term ‘membrane distillation’ was chosen for the distillation process in which two sides of the membrane are separated (liquid and gaseous phases) by a porous membrane [53]. Membrane distillation can be classified into four types according to the specific method used to activate the vapor pressure gradient across the membrane, which represents the driving force for this process.

4.1 Direct contact membrane distillation (DCMD)

In DCMD, the condensation liquid, often using pure water, is colder than the feed stream where the condensation and the feed liquid are in with the hydrophobic membrane. This point makes this type of formation is known as direct contact membrane distillation, and the vapor pressure difference is maintained on both sides of the membrane by applying a temperature difference [54–56].

4.2 Vacuum membrane distillation (VMD)

It is also a thermally driven process in which the feed solution is in contact with one side of the membrane.

The vacuum is applied on the other side using vacuum pumps, wherein the applied vacuum pressure is less than the equilibrium vapor pressure. Therefore, condensation occurs outside the membrane unit [54, 57].

4.3 Air gap membrane distillation (AGMD)

In this type of distillation, a stagnant air layer is placed between the side that breaks through the membrane and the condensation wall to reduce heat loss by conduction. The presence of the air gap acts as thermal insulation between the membrane and the condenser wall greatly reducing the heat loss and improving the separation efficiency [54, 58].

4.4 Sweeping gas membrane distillation (SGMD)

In this process, on the one hand, the evaporated water molecules are collected through a cold inert gas in the condensation chamber. On the other hand, the hot feed solution is circulated on one side of a micropore membrane and a cold scrub gas on the other side of the membrane. Although not much work has been done regarding SGMD, this configuration has the advantages of relatively low conductor heat loss with low mass transfer resistance [54, 59].

OD is a non-thermal technique used to remove water from aqueous solutions (i.e., concentrate wastewater and recover valuable components). In OD, a small porous aqueous membrane separates two aqueous solutions with different dissolved concentrations. Moreover, the OD process can be operated under atmospheric pressure and ambient temperature. The driving force is the gradient of vapor pressure across the membrane obtained with a hypertonic salt solution on the permeable side. The hydrophobic nature of the membrane prevents pore penetration by aqueous solutions, resulting in the formation of vapor/liquid interfaces at the entrance to the pores. Under these conditions, a clear water flow occurs from the high vapor pressure side to the low side resulting in concentrated feed and dilution of the hypertonic salt solution [60, 61]. OD technology is also called isothermal membrane distillation, osmotic membrane distillation, osmotic evaporation, and gas membrane extraction [62]. Generally, many components were used to prepare the osmotic solutions, like sodium, potassium, magnesium, and calcium salts, and some organic liquids such as glycerol or polyglycols. The basic requirements for osmotic solutions are non-volatile and have high osmotic activity to maintain a low vapor pressure and maximize the driving force. These solutions must be thermally stable and preferably non-toxic, non-corrosive, and low-cost [63–65]. Advantages of OD

compared to other separation methods. The processes can be summarized as follows:

1. high selectivity;
2. ability to operate at ambient pressure and temperature;
3. no use of chemical additives;
4. simple mechanical design;
5. no or less degradation of heat-sensitive components;
6. possibility to achieve the higher concentrated feed.

In addition to both MD and OD, there is the so-called osmotic membrane distillation (OMD), which is a mixture of DCMD and OD. It is an isothermal process in which the membrane is brought into contact with the hot aqueous feed solution to be treated and a cold osmotic solution. Moreover, the driving force for vapor transport in OMD is the partial pressure difference between the feed and the brine. This difference is caused by the decrease in the activity of the water in the brine. Therefore, OMD can be performed at room temperature and atmospheric pressure without degradation of heat-sensitive components and loss of some volatile components from liquid foodstuffs. This process is proposed to remove water from liquid foods such as fruit and vegetable juices, milk, instant coffee, tea, and various non-food non-heat-resistant aqueous solutions. Inorganic salts (NaCl , CaCl_2 , MgCl_2 , and MgSO_4) or organic solvents (glycerin and polyglycerin) can be used as removal solutions [66, 67].

Furthermore, most of the studies that dealt with the membrane distillation process focused on studying and evaluating process variables. These studies examined the effect of temperature, pressure, flow, membrane properties, and pollution rather than evaluating the quality of plant extracts and their content of active ingredients after the membrane distillation process. For example, Zhao et al. [68] studied the effects of operating temperature, vacuum pressure, and feed concentration was investigated using a plate membrane module to concentrate Ginseng extracts aqueous solution using a VMD. In addition, the technology of scanning electron microscopy analysis was used to observe membrane fouling. It was found that both gas-liquid two-phase flow at the feed side and gas back-washing within the membrane module are effective ways to control membrane fouling. Also, DCMD was applied to concentrate traditional Chinese medicine extract (TCM), in which the transmembrane flux, under various operation conditions, was measured in real-time during the concentration process [69]. However, most of the studies

examined the concentration and clarification of the juices, not on the concentration of medicinal plants extracts.

In addition to all the previous applications, the membrane technologies (MF, UF, NF, MD, OD) have found an application in the concentration and recovery of active substances from plant extracts. Table 1 [70–78] and Table 2 [79–90] summarize some of the studies which examine the application of the mentioned technologies in the concentration of plant extracts.

5 Integrated membrane processes

In a lot of research, membrane unit operations have been combined into integrated systems, and this has led to many benefits such as reducing energy consumption and improving product quality, processability and selectivity at the same time. Among these studies we mention:

1. Three-step filtration process was applied to concentrate oleuropein from olive leaf extract. micro-filtration system through 0.2 nm pores size membrane (Microporous membrane TOPER Model, diameter 300 mm, CN). The microfiltration permeate was submitted to a cross-flow ultrafiltration system of MWCO 5 kDa (GE Power & Water, ZeeWeed 1500 Minnetonka, MN). Finally, the ultrafiltration permeate feed a cross-flow nanofiltration System of MWCO 300 Da (GE Osmonics, HL2540TF, Minnetonka, MN). Results revealed that a large portion of phenolic compounds was recovered in the permeate fraction of the UF process. The nanofiltration retentate showed high polyphenol and flavonoid contents. Based on the content of solute in feed and retentate fractions of NF membrane, oleuropein was concentrated approximately 10 times to reach 1685 mg/100 g extract [91].
2. The pectin extract obtained from red currant marc by-products was concentrated by separating the membrane with two flat-sheet RO membranes (ACM2-TRISEP and SG composite (CHEZAR spol. s r., Bratislava, Slovak)), one flat-sheet NF membrane (DL) (CHEZAR spol. s r., Bratislava, Slovak) and one spiral-wound NF membrane. The results showed that the concentration by RO resulted in an increase of the total soluble solids (TSS) content to 4.28 Brix, while for NF, the corresponding level was 8.88 Brix. The membrane resistance and the fouling resistance were the determinants relative to the gel resistance [92].

Table 1 Application of microfiltration and ultrafiltration membranes in the concentration of plant extracts

Membrane type	Material	Pore size	Applied pressure	Temp	Raw material	Targeted product	Findings/results	References
Microfiltration	Ceramic	0.2–1.4 μm	50–200 kPa	50–60 °C	Watermelon juice	Lycopene	41 times more concentrated, 34 times pure than the initial juice	[70]
Microfiltration	Hollow Fiber	150 kDa	35–172 kPa	–	Tea leaves	Total polyphenol	80% of the total polyphenol content, 75% of the EGCG purity in permeability	[71]
Ultrafiltration	Millipore type GS 0.22 Millipore Type HA 0.45	22 nm, 45 nm	5 bar	23 °C	Grape seed	Polyphenol	Maximum amounts of polyphenols (11.4% of total seed weight) were obtained. The application of ultrasound reduced in 44% the total resistance during ultrafiltration of the membrane of 20 kDa.	[72]
Microfiltration and ultrafiltration (ultrasound-assisted)	Flat membrane (cellulose acetate)/ MF polyethersulfone/ UF	0.22, 0.3, 0.8 μm (MF) 5, 10, 20, 30 kDa (UF)	0.7 bar (MF) 6 bar (UF)	25 °C	Green tea extract	Phenolic compounds	Steady turbidity values (<5 NTU) after 30 days. No tea cream formation of storage under refrigeration.	[73]
Microfiltration and ultrafiltration	Flat sheet regenerated cellulose (Millipore), PSF and polyacrylonitrile (PAN) membranes	45 nm	3 bar	–	<i>Sabia officinalis</i> L. (Labiaceae family) <i>Viscum album</i> L. (Loranthaceae)	Antioxidant activity	Concentrated extracts had a very high radical scavenging activity (TEAC = 351.87–479.04 μmol Trolox/mL and 345.14–426.18 μmol Trolox/mL extract) for sage hydro-alcoholic and mistletoe extract respectively. PAN membrane has the greatest retention degree for bioactive compounds.	[74]
Ultrafiltration	FSM0, 15PP (fluoro polymer) UV050 (polyvinylidene UPI50 (PES)	0.15 μm 50 kDa 150 kDa	12–30 bar	–	Grape seed extract	Polyphenols	Very high retentions of phenols (87–91%)	[75]
Ultrafiltration	polysulfone (PS) PES	100 kDa 50 kDa	1–2 bar/step 1 0.8, 1.5, 3 bar/step 2	50 °C	pulp of papaya	Lycopene	Lycopene retention was higher than 90%. The best UF performance was obtained with the PS 100 membrane, pressure 1 bar.	[76]
Ultrafiltration	Cellulose membrane	0.2 μm	–	–	Citrus peel	pectin	4.2% of galacturonic acid increased 0.6% of pectin decreased.	[77]
Microfiltration and Ultrafiltration	Hydrophilic amphoteric nylon 6,6	200, 450 nm 25, 50, 100 and 500 kDa	MF: 0.1 Mpa UF: 0.5 MPa	26±2 °C	Black tea extracts	Polyphenols	UF-500 increased retention of original colour and recovery of polyphenols.	[78]

Table 2 Application of nanofiltration and reverse osmosis membranes in the concentration of plant extracts

Membrane type	Material	Pore size	Applied pressure	Temp	Raw material	Targeted product	Findings/results	References
Nanofiltration	Polyamide	400, 700, 1000 Da	–	–	Grape seed	Phenolic	Procyanidin rejection was 96.36±0.87%. The antioxidant activity was increased around 2.24 times.	[79]
Nanofiltration	PS with SBA-15-NH ₂ SeIRO MPF-36, Koch membrane	600–1500 Da	10 bar	–	<i>Geranium robertianum</i> and <i>Salvia officinalis</i>	Phenolics, flavonoids, and antioxidants	>70% rejection rate of polyphenols and flavonoids; >88% antioxidants, 85.9% rejection rate of polyphenols and flavonoids, >90% antioxidants	[80]
Nanofiltration	NF245 and NF270	200–400 Da	10–17 bar	5–50 °C	Blueberries	Polyphenols	Complete rejection of phenolic compounds with good permeability	[81]
Nanofiltration	DuramemTM 200	200, 300, 500 Da	20, 40 bar	50 °C	Rosemary extracts	Phenolics and antioxidants	Complete rejection of rosmarinic acid and other antioxidant components	[82]
Nanofiltration	NP010, NP030, TFC-S, NF200, Desal DL, Desal DK	200–1000 Da	0–40 bar	20±2 °C	Artichoke brines	Flavonoids and caffeoylquinic acids	<92% rejections of total caffeoylquinic acids, flavonoids, and cynarin	[83]
Nanofiltration	PVDF membrane (NF) PAM membrane (MF)	150 and 300 Da (NF) 400 nm (MF)	300 kPa (NF) 300 kPa (MF)	20±2 °C	Strawberry juice	Anthocyanin, phenolics, and antioxidants	Retention up to 95% of anthocyanin; phenolics and antioxidants increased 20% and 50%	[84]
Nanofiltration	NF 90 Filmttec, Dow Chemical Company, São Paulo, SP, Brazil	200–300 Da	800 kPa	25 °C	Alcoholic extract of Pequi (<i>Caryocar brasiliense</i> Camb.)	Polyphenols and carotenoids	Rejections of around 10% for carotenoids and 15% for polyphenols, retention coefficient around 100% and 97% of carotenoids and polyphenols	[85]
Reverse Osmosis	Membrane R25A polyamide	500 Da	5 bar	25 °C	Camu-camu	Vitamin C	A concentrated camu-camu (CC) with high vitamin C and cyanidin-3-glucoside was obtain, in an increment respectively 7.0 and 4.5 times higher. The concentration of phenolic compounds was increased by 3.2 times, and anthocyanins in 6.5 times.	[86]
Osmotic Distillation	Hydrophobic, polypropylene, hollow-fiber membrane	0.2 µm	–	35–45 °C	Grape juice, Apple juice, and Roselle extract	Increased concentration of dissolved solids	The final total soluble solids (TSS) contents achieved were 660, 570, and 610 g/kg for grape juice, apple juice, and roselle extract, respectively.	[87]
Forward Osmosis	A flat membrane module (developed by Osmotek, Inc., Corvallis, OR)	–	–	27±2 °C	Rose petals	Anthocyanin	The forward osmotic concentration using the membrane resulted in minimum degradation of anthocyanin.	[88]
Osmotic evaporation (OE)	Hollow polypropylene membrane	0.04 µm	–	23±2 °C	Three different tea extracts (medicinal Rosil No. 6, Black, and Forest Fruit teas)	Phenolic	Tea concentration of 40% (w/w) was reached in 5 h, with a constant water flux and without losing the phenolic content or antioxidant potential.	[89]
Reverse Osmosis	RO 99 and X20	–	average 40 bar	30 °C	Beetroot peel extracts	Betalains, phenolics, and antioxidants	>90% betalains recovery; >99% phenolics and antioxidants recovery	[90]

3. Integrated membrane process that includes micro-filtration, reverse osmosis, and osmotic distillation for producing concentrated sage (*Salvia fruticosa* Miller) extract. A multi-tube ceramics membrane (Schumasiv, Pall Corporation, NY, USA) with a pore size of 0.45 μm and an effective area of 0.125 m^2 was used to examine this process. The RO membranes were flat sheet ACM2 Membranes (DDS, Silkeborg, Denmark) in a sandwich unit with 0.18 m^2 effective area and 93% NaCl salt retention. Finally, this study concluded that sage extract could be concentrated up to 32.4 w/w% successfully by using the suggested integrated membrane process. However, the loss of a remarkable and valuable compound (30–40%) can be observed during the reverse osmosis process in the applied conditions. In contrast, osmotic distillation retained more than 90% of the total polyphenol content, flavonoid content, antioxidant activity, and almost all the determined individual polyphenols, with more than 90% retention [93].
4. The phenolic compounds were purified and concentrated from aqueous Jamun (*Syzygium cumini* L.) seed extract by an integrated membrane process. Six commercial flat sheet polymeric membranes were used; on the one hand, three were ultrafiltration membranes with molecular weight cut-off (MWCO) of 25, 50, and 100 kDa. On the other hand, the other three were nanofiltration membranes characterized by MWCO of 1000, 400, and 250 Da. The experimental results showed that the operating conditions affected permeate flux, recovery of polyphenols, purity, and antioxidant activity of the phenolic extract. Ultrafiltration experiments at lower operating pressures (276 and 414 kPa) using 100 kDa membrane resulted in the recovery of 59–66.7% of total polyphenol content in the clarified extract with the purity of 49–58.3% starting from an extract purity of 39.2%. The clarified extract could be concentrated successfully about three times higher using a 250 Da nanofiltration membrane at a volume concentration ratio of three. Moreover, the present study revealed that the UF/NF integrated membranes process succeeded in clarifying and concentrating the obtained phenolic extract from Jamun seed with enhanced purity and antioxidant activity [94].
5. The concentration of anthocyanin using different membrane processes, individually and in combination with each other, was studied by using a polyamide membrane (with 99% NaCl retention character) in case of RO process and FP-100, polyvinylidene-difluoride (PVDF) membrane of nominal molecular weight cut off 10,000 in UF process, and hydrophobic polypropylene (PP) membrane of pore size 0.2 μm for the OMD process. The aim of observing the integrated membrane process involving clarification by UF, preconcentration by RO, and the final concentration using OMD was having it as an attractive alternative compared to the membrane processes operated alone. The hybrid process achieved the anthocyanin concentration from 40 mg/100 mL to 980 mg/100 mL (increase in 25-fold concentration, from 1 to 26 B). The hue angle and chroma of the color were increased in the case of UF. Besides, it had also increased by using the UF process combined with OMD or with RO-OMD [95].
6. The aqueous extract of yerba mate was clarified by sequential treatment with three microfiltration membranes. These microfiltration membranes were polyethersulfone, ultrafiltration 1 (vinylidene polychloride) (30–80 kDa), and ultrafiltration 2 (Ceramic (zirconia oxide) (40 kDa). Then, the stability of the yerba mate extract was tested, and It was filtered and concentrated using a reverse osmosis membrane (polyamide-polyethersulfone). Moreover, the results showed that The turbidity of the clarified membrane extracts was less than 36 NTU, and there was no significant difference in the phenolic compound content between the crude and clarified extracts. Ultrafiltration membrane 1 (vinylidene polychloride) performed the best. It also produced extracts with the lowest loss in phenolic compounds (18%) and turbidity (99.9%) while maintaining a stable permeate flux. The reverse osmosis membrane concentrated the polyphenols and the extract solids three times. Besides, the clarified yerba mate extract maintained its phenolic compound stability and decreased turbidity over 30 days of storage [96].
7. An integrated membrane process to recover phenolic compounds from Goji (*Lycium barbarum* L.) leaves aqueous extract was evaluated by ultrafiltration. Then, it was treated with three membranes of flat-plate PES with MWCO in the range of 0.3–4.0 kDa to remove sugar compounds from polyphenols and improve the antioxidant activity of the produced fractions. Among the selected membranes, a 1 kDa membrane had the best performance regarding the purification of polyphenols from the clarified aqueous extract. The

rejection by this membrane of TSS and total carbohydrates was in the range of 15.8–25.3%, and it was decreased by increasing the volume reduction factor (VRF). On the other hand, the retention values for total polyphenols and total antioxidant activity (TAA) were in the range of 73–80%, and they were increased by increasing the VRF [97].

8. A three-stage hybrid membrane process for the concentration of ethanol-water extracts of the Echinacea plant has been investigated. In the first stage of the hybrid process, ethanol removal from the neat extract was achieved by PV, which gave an ethanol-free aqueous product containing suspended alkyl amides, suitable for marketing in tincture form. In the second stage of the hybrid process, the precipitated alkyl amides were removed from the first stage product by MF. In the third stage, the microfiltration permeate was concentrated several-fold by osmotic distillation. It was followed by adding back of the microfiltration retentate containing the precipitated alkyl amides to the osmotic distillation retentate. As a result, this gave a highly concentrated product suitable for marketing in capsule form [98].
9. To recover and concentrate monomeric anthocyanins and total phenolics from grape marc, the integration of pressurized liquid extraction (PLE) with membrane separation technology was used. Two preliminary procedures aimed at selecting the best NF membranes and evaluating a sequential process to enhance NF performance. In these studies, four NF membranes were tested in terms of permeate mass flux and retention index of total monomeric anthocyanins and total phenolics. Then, the usage of MF and UF processes were evaluated as alternatives to improve the concentration performance and reduce membrane fouling in the NF step. The results obtained suggest that the sequential process based on the previous MF with the MV020 membrane of the grape extract obtained by PLE, followed by the NF step with the NP010 membrane, was the most effective in the concentration of bioactive compounds. Where this process provided excellent results in terms of permeate flux, concentration factor (2.4), retention coefficients of monomeric anthocyanins (78.2%) and total phenolics (71%), and high antioxidant capacity (52%) compared to other tested NF membranes. The conclusion was that the integration of PLE with the projected sequential process (MV020-NP010) is efficient

for the recovery and concentration of bioactive compounds from grape marc, and promising for obtaining functional products with high added value [99].

10. To concentrate and purify the fructooligosaccharides (FOS) from yacon root extract, ultrafiltration (UF) with nanofiltration (NF) were combined with and without the use of discontinuous diafiltration (DF). After UF, 63.75% of the saccharides from the initial feed were recovered in total permeate. DF did not largely influence FOS retention during NF (it increased from 68.78% without DF to 70.48% with DF), but decreased glucose and fructose retentions, from 40.63 to 31.61% and 25.64 to 18.69%, respectively, which was desirable, allowing greater purification of FOS in the retentate. The yield of total saccharides in the final retentate after combined UF and NF processes was 50.89% and of FOS was 51.85%, with 19.75% purity. The results indicate that combining UF and NF is a promising technique for concentrating yacon saccharides. However, improving FOS purity requires more diafiltration steps [100].
11. Microfiltration (a minnitech polysulfone hollow fiber module (pore size 0.05 μm and area 0.5572 m^2), and ultrafiltration (molecular weight cut-off of 5 KDa, area 0.25 m^2) were followed by adsorption were applied to concentrate and purify the di-acylate cyanidin from red cabbage, which allowed the pigment concentration to increase three times higher than the initial concentration (from 32.05 to 32221.45 $\text{mg ECyn-3-glu L}^{-1}$). This finding was much higher than the one reported in the literature. Furthermore, this concentrated fraction also exhibited higher antioxidant activity (up to 8.81 mmol ET mL^{-1}) comparing the raw extract [101].

6 Key factors influencing the performance of membrane processes

There are many factors that influence the performance of membrane processes. The first factor is physico-chemical composition of the feedstream: It plays a critical role in the fouling of the membranes, especially at the concentration of extracts rich in phenolic compounds. In such extracts, phenolic compounds have demonstrated their adsorption on Polyethersulfone MF membranes. Phenolic compounds may interact with each other or with other compounds (i.e., polysaccharides), which forms large particles. These particles have a negative impact during the filtration process. The second factor operating parameters: such as transmembrane pressure, cross-flow velocity, and temperature have a strong

influence on membrane fouling, and consequently, on membrane productivity and selectivity. The third factor is membrane features and properties such as hydrophilicity/hydrophobicity, surface topography, charge, and pore size [102].

7 Weakness of membrane-based technologies and research gaps and the current trends

Despite the great potential of membrane technology and its applicability in removing/recovering nutrients from wastewater and sludge and extracts concentration, it has multiple critical points. The first one is the purity restrictions: membrane processes rarely produce two pure streams, which means that one of the streams always contains a minor amount of an undesired component. The second point is fouling, which still form the biggest challenge to the membrane technology. The third point is thermal, mechanical, and chemical limitations, where membrane modules cannot operate at relatively high temperatures. This problem is an outcome of most membranes being based on polymers, and most of these polymers do not maintain their physical integrity at temperatures higher than 90–100 °C. The fourth point is energy consumption-cost relationship. Despite the low operating costs, the available membrane modules are high and require large capital [102].

Moreover, most investigations and applications are still in the experimental (lab and pilot) stage, and it has not yet reached an industrial level. Thus, we should make more efforts to find suitable, effective membranes. We should also aim at finding membranes with more selectivity for nutrient removal/recovery from wastewater and extract concentration. The academic community should develop new stable membranes and conduct studies on membrane cleaning strategies and refreshing [103].

8 Conclusion

Recently, multiple techniques have been developed for extracting and concentrating bioactive compounds from

plants, including the usage of ultrasound, supercritical fluids and microwaves, and membrane separation processes. Among these processes, membrane processes represent a viable alternative to conventional technologies. Adopting the new technologies is due to its lower costs of operation and maintenance, moderate operating conditions of temperature and pressure, ease of control and upgrading, high selectivity and minimal thermal damage of the processed solutions, and high quality of the obtained products. MF, UF, NF, and RO have been extensively investigated for the recovery, separation, and concentration of active compounds from different plant matrices in addition to offering new and interesting perspectives of combined different membrane operations such as combine UF with (MF or NF), or introduction of the NF step before RO, and combining membranes with conventional separation techniques such as a combination of enzymatic treatment or evaporation with RO.

This paper has reviewed and discussed different membrane processes and their types. It has also shown the applications of membrane technology in separating and concentrating active compounds from plant extracts. According to the literature, the results showed the efficiency of many membrane processes in this field as it was found that the combination of various membrane unit processes successfully meets the requirements of recovery, purification, and concentration of valuable compounds from different plant sources and the production of concentrated liquid fractions of importance for commercial use in the food, pharmaceutical, and cosmetic industries.

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