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Membrane technologies assisting plant-based and agro-food by-products processing:  
A comprehensive review

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1 **Membrane technologies assisting plant-based and agro-food by-products**  
2 **processing: a comprehensive review**

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21



## 22 **Abstract**

23 *Background:* Nowadays, membrane-based technologies (e.g. microfiltration,  
24 ultrafiltration, nanofiltration, membrane distillation, and pervaporation) have  
25 demonstrated to meet the requirements to be involved in different food and  
26 bioproduct processes.

27 *Scope and approach:* Several applications have been developed, including either  
28 separation, recovery or concentration of bioactive molecules from agro-food  
29 products and by-products, treatment of natural extracts, recovery of aromas from  
30 natural and processed products, production of non-alcoholic beverages, as the  
31 most popular ones. Therefore, the goal of this review is to give a comprehensive  
32 outlook of the latest developments focused on the separation, fractionation and  
33 concentration of several bioactive compounds contained in their original sources,  
34 as well as the food processes-assisted by membrane technologies.

35 *Key findings and conclusions:* Throughout this review, ongoing literature has been  
36 analyzed, discussing the relevant insights according to the type of membrane-  
37 based separation process, properties of molecules, membrane features and key  
38 factors influencing the separation performance of those technologies. Specific  
39 applications have been analysed and discussed, highlighting typical advantages  
40 and drawbacks over conventional technologies.

41

42 **Keywords:** *Agro-food products, high-added value compounds, microfiltration,*  
43 *ultrafiltration, nanofiltration, membrane distillation, pervaporation.*

44

## 45 **Nomenclature**

46 MF: Microfiltration

47 UF: Ultrafiltration

48 NF: Nanofiltration

49 MWCO: Molecular weight cut-off

50 MD: Membrane distillation

51 PV: Pervaporation

52

### 53 **1. Introduction**

54 The usage of membrane-based technologies has been nowadays considered for  
55 multiple approaches within the industrial processing of food products and by-  
56 products. Since couple of decades, different membrane-based techniques have  
57 been actively used for the separation, recovery and concentration of biologically  
58 active compounds (e.g. phenolic compounds, anthocyanins, carotenoids,  
59 antioxidants, polysaccharides) from agro-food products and their derivatives (e.g.  
60 wastewaters), clarification and concentration of natural extracts, recovery of  
61 aromas from natural and processed products, and production of non-alcoholic  
62 beverages (Castro-Muñoz et al., 2016; Figoli et al., 2006a). Recently, particular  
63 attention has been pointed out to recovery of high added-value compounds from  
64 agro-food by-products and the development of new products with a market value  
65 (Cassano et al., 2018; Santamaría et al., 2000). Thanks to their intrinsic properties,  
66 specific membrane-based techniques (e.g. MF, UF and NF) are currently  
67 considered as an emerging alternative to enhance the current valorization  
68 protocols, within sustainable strategies for biorefinery, providing remarkable  
69 advances in terms of environmental sustainability (Castro-Muñoz et al. 2018a;

70 Galanakis, 2013). On the other hand, emerging membrane processes, including  
71 MD and PV, are also involved in the strategies for the reclamation of bioactive  
72 molecules from food systems (Figoli et al., 2010; Galiano et al., 2019). Membrane  
73 technologies supply featured advantages over traditional separation processes  
74 (e.g. precipitation, coagulation, flocculation, evaporation, solvent extraction,  
75 adsorption, gravity sedimentation, centrifugation, among others)(Le & Nunes,  
76 2016), such as simple operating conditions in terms of pressure and temperature,  
77 thus preserving the biologically active properties of bio-molecules contained in  
78 natural and processed products, non-use of chemical (e.g. solvents) or biological  
79 agents, and, consequently, minimal risk of contamination. Moreover, membrane  
80 processes are recognized as highly selective techniques towards target solutes,  
81 with simple implementation, feasible scale-up, reduced number of operation steps  
82 and high energy savings (Van Der Bruggen et al., 2003).

83 In the light of the continuous and growing demand of both users and manufacturers  
84 for minimally-processed foods free of contaminants and health-promoting foods,  
85 the extraction of natural antioxidants using membrane-based technologies has  
86 been widely explored in the recent years (Cassano et al., 2019; Galanakis, 2015a).  
87 Therefore, the goal of this review is to provide a comprehensive outlook about the  
88 ongoing research works focused at enhancing the separation-extraction,  
89 fractionation and concentration of several bioactive molecules, as well as the food  
90 processing technologies-assisted by membrane technologies. By exploring the  
91 literature data acquired mainly at laboratory scale experiments, the performance of  
92 these processes is influenced by factors and parameters, which should be carefully  
93 considered, evaluated and optimized case-by-case for a real scenario and feasible

94 scale-up. Thereby, those aspects are fully addressed and discussed in detail  
95 according to the up-to-date literature insights.

96

## 97 **2. Microfiltration, Ultrafiltration and Nanofiltration**

98 Microfiltration (MF), ultrafiltration (UF) and nanofiltration (NF) are membrane  
99 processes based on the use of a perm-selective porous barrier, so-called  
100 “membrane”, through which fluids and solutes are selectively transported when a  
101 transmembrane pressure ( $\Delta p$ ) is applied. The membrane enables the partial  
102 fractionation of the feed bulk into two streams: a permeate stream, which contains  
103 the solvent (usually water) passing across the membrane accompanied by all  
104 those molecules presenting lower molecular weight than the membrane’s  
105 molecular weight cut-off (MWCO), and a retentate stream which contains all  
106 compounds partially or totally rejected by the membrane (**Figure 1**). The  
107 separation is based mainly on molecular size and to a lesser extent on shape and  
108 charge (Galanakis, 2015b).

109

110 **Figure 1.** General depiction of a pressure-driven membrane process.

111

112 MF membranes are typically characterized by nominal pore sizes of the order of  
113 0.1 -10  $\mu\text{m}$ . This process is commonly used to concentrate, purify or separate  
114 macromolecules, colloids and suspended solids from solutions (i.e. wine, juice and  
115 beer clarification in the food industry).

116 UF membranes have pore sizes in the range of 1-100 nm and are capable of

117 retaining species in the molecular weight range of 300-1,000,000 Da (i.e.  
118 biomolecules, polymers and colloidal particles as well as emulsions and micelles).  
119 NF is mainly used to separate ions and molecules in the molecular weight range of  
120 200-2,000 Da (pore sizes of NF membranes are in the range 0.5-2 nm) (Wei et al.,  
121 2018; Winter, Barbeau, & Bérubé, 2017). NF membranes have relatively high  
122 charge and are characterized by lower rejection of monovalent ions in comparison  
123 to that of multivalent ions.

124 Since MF, UF and NF membranes differ in the size of molecules they separate, the  
125 operating pressure involved is considerably different between the related  
126 processes. MF typically requires pressures between 110 and 300 kPa; operating  
127 pressures of UF are in the range of 150-500 kPa; a range from 500 to 1500 kPa is  
128 common for NF.

129 All these processes can be operated either in dead-end or in cross-flow  
130 configurations. In the dead-end filtration the feed is pumped perpendicularly onto  
131 the membrane surface; the retained particles tend to form a cake layer on the  
132 membrane surface whose thickness increases with the filtration time. Therefore,  
133 dead-end operation has to be run batch-wise to relieve the retained particles. In the  
134 cross-flow configuration the feed is pumped tangentially across the membrane  
135 surface so limiting the build-up of retained compounds on the membrane surface.  
136 As such the cross-flow operation allows for continuous process and it is a standard  
137 operation for most filtration processes in the food industry.

138 The performance of a membrane process is mainly evaluated by two parameters:  
139 the degree of separation (related to the retention during concentration) and  
140 productivity. The membrane productivity is characterized by the *permeate flux* (J, L



141  $\text{m}^{-2} \text{h}^{-1}$ ) which indicates the rate of mass transport per unit membrane area and  
 142 time:

$$143 \quad J = \frac{Q_p}{A} \quad (1)$$

144 where  $Q_p$  ( $\text{L h}^{-1}$ ) is the volumetric flow rate of permeate and  $A$  ( $\text{m}^2$ ) is the area of  
 145 the membrane.

146 The membrane selectivity is generally expressed in terms of rejection or retention  
 147 factor (R):

$$148 \quad R = \left(1 - \frac{c_p}{c_r}\right) \quad (2)$$

149 where  $c_p$  and  $c_r$  are the solute concentration in the permeate and retentate,  
 150 respectively. Rejection values range between 0 and 1 (or 0 and 100% if expressed  
 151 as percentage)(Castro-Muñoz, 2019b).

152 The volume concentration ratio (VCR) is defined as the ratio between the initial  
 153 feed volume ( $V_f$ ) and the volume of resulting retentate ( $V_r$ ) according to the  
 154 following equation (Sánchez, Carmona, Prodanov, & Alonso, 2008):

$$155 \quad VCR = \frac{V_f}{V_r} \quad (3)$$

156 The yield (Y) of a component, that is the fraction of a such component recovered in  
 157 the final retentate with respect the initial feed, is expressed as:

$$158 \quad Y = \frac{c_r V_r}{c_f V_f} \quad (4)$$

159 It is a function of VCR according to the following equation:

$$160 \quad Y = VCR^{(R-1)} \quad (5)$$

161 Some specifications and characteristics of MF, UF and NF operations are detailed  
 162 in **Table 1**.





163 The valorization of agro-food by-products is nowadays one of the primary  
164 challenges for scientists (Castro-Muñoz, 2018; Mirabella et al., 2014). In such a  
165 way, the interest in implementing the integral strategy “5-Stages Universal  
166 Recovery Process” has impressively raised in last years (Galanakis, 2015b). In  
167 particular, UF and NF are non-destructive techniques which can be applied in  
168 several steps of the above downstream processing; these processes are largely  
169 recognized for their ability to recover bioactive molecules from agro-food  
170 manufacturing wastes, also through their coupling in hybrid systems (Cassano et  
171 al., 2018; Dhillon et al., 2013).

172

173

174 **Table 1.** Main characteristics of MF, UF and NF processes. Adapted from (Castro-  
175 Muñoz et al., 2018).

176

177 **Table 2** reports the most recent literature data obtained at laboratory scale, in  
178 which different bioactive molecules have been successfully separated and thus  
179 recovered from agro-food wastes, including agricultural residues (fruit seeds,  
180 orange press liquor, grape marc, fermented grape pomace, etc.) and wastewaters  
181 (from corn, olive, artichoke, citrus and winemaking industry), and some other by-  
182 products (e.g. winery effluents, red wine lees) representing a rich source of  
183 phenolic compounds (Cassano et al., 2016a; Cassano et al., 2018). Interestingly,  
184 natural products have been also employed as potential sources of bioactive  
185 compounds (Castro-Muñoz & Fíla, 2018). Basically, these membrane operations  
186 provide high recovery efficiency being highly selective towards targeted derivative

187 polyphenols, including catechol, tyrosol, hydroxytyrosol, and phenolic acids (e.g.  
188 caffeic and p-cumaric).

189

190 **Table 2.** Bioactive molecules recovered from agro-food wastes using membrane  
191 technologies.

192

193 *2.1. Key parameters influencing the separation performance of pressure-*  
194 *driven membrane-based technologies*

195 Typically, the separation performance of pressure-driven membrane operations  
196 (like MF, UF and NF) in terms of permeation rate and solute rejection, depends on  
197 multiple factors, such as (Astudillo-Castro, 2015; Fane & Fell, 1987):

- 198 • *Physico-chemical properties of the feed stream:* this parameter strongly  
199 contributes to membrane fouling phenomena. In principle, the fouling is the  
200 main drawback of these processes since it produces a long term permeate  
201 flux decline caused by the accumulation of specific compounds on the  
202 membrane surface (Fane & Fell, 1987). It may occur due to the formation of  
203 a concentration polarization layer on the membrane surface, cake layer  
204 formation and/or partial or complete blockage of the membrane pores.  
205 Fouling results by particular types of interactions between the membrane  
206 and solutes contained in the bulk feed stream. Therefore, the  
207 physicochemical composition and the properties of individual feed  
208 molecules (i.e. nature, morphology, hydrophobic interactions, charge, zeta  
209 potential, etc.) have a meaningful effect on these interactions. For instance,  
210 phenolic-based molecules have shown adsorption properties on

211 polyethersulfone (PES) membranes due to weak polar interactions (Susanto  
212 et al., 2009; Cartalade & Vernhet, 2006). Also, polyphenols can interact with  
213 some other solutes (i.e. proteins, polysaccharides) to form up large particles  
214 which may have a negative effect during filtration.

215

216 • *Operating parameters:* Operating parameters including feed flowrate  
217 (cross-flow velocity), transmembrane pressure (TMP), temperature and feed  
218 concentration, have a key effect on membrane fouling, and thus affect both  
219 membrane selectivity and productivity. In general, an increase of the feed  
220 temperature produces a decrease in the fluid viscosity, as well as an  
221 increase of the diffusion coefficient of molecules: the effect of these two  
222 factors is to enhance mass transfer and to increase the permeation rate  
223 (Ramli & Bolong, 2016). For small pressures the permeation flux increases  
224 linearly with the applied pressures. As the pressure is increased flux shows  
225 a deviation from a linear flux-pressure behaviour and it becomes  
226 independent of pressure: at this limiting TMP or higher pressures, the  
227 permeate flux does not depend on the pressure anymore; the existence of a  
228 limiting flux can be attributed to concentration polarization and fouling  
229 phenomena (Astudillo-Castro, 2015). On the other hand, the retention of  
230 some molecules (e.g. phenolic-based solutes) tends to increase by raising  
231 the TMP (Díaz-Reinoso et al., 2009). This is due to the thin layer formation  
232 close to the membrane surface, which acts as an extra barrier and thus  
233 promote the retention of solutes (Bacchin et al., 2002).

234 The cross-flow velocity affects the shear stress at the membrane surface and,

235 consequently, the rate of removal of deposited particles responsible of flux decay:  
236 herein, an increase of cross-flow velocity has a large effect on flux. Finally,  
237 according to the film theory model the permeate flux decreases exponentially with  
238 increasing the feed concentration.

239 • *Membrane properties*: The intrinsic features of the membrane, such as  
240 surface topography, hydrophobicity/hydrophilicity, pore size and charge  
241 have an important influence on solute-membrane interactions, and  
242 consequently on membrane fouling. Hydrophobic polymeric membranes are  
243 the most used in this type of processes. In fact, many manufacturers (e.g.  
244 GE Osmonics, Nadir, Nitto-Denko, Lenntech, Toray) are using highly  
245 hydrophobic polymeric materials (i.e. polyamide, sulfonated polyether-  
246 sulfone, polypiperazineamide, polysulfone) for manufacturing of  
247 membranes.

248 Crucially, pore size is the primary feature that differentiates MF, UF and NF  
249 membranes (see **Table 1**). The membrane's ability to retain specific molecules is  
250 generally described by manufacturers in terms of molecular weight cut-off (MWCO)  
251 which is defined as the molecular weight of a solute 90% retained by a given  
252 membrane. However, molecules with the same molecular weight but different  
253 shapes and conformation (i.e. linear and spherical molecules) can be characterized  
254 by different permeabilities. In this sense, the MWCO is not a reliable tool to predict  
255 the separation capability of a membrane. Another important aspect is the  
256 asymmetric characteristic, which is that the membrane pores do not always  
257 possess a fair MWCO range across all membrane (Galanakis, 2015a). The  
258 asymmetric structure is generally related to the membrane preparation technique.



259 The asymmetric porous membranes can be obtained by means of wet phase  
260 inversion, one of the most common membrane preparation methods for NF and UF  
261 membranes (Blanco et al., 2006; Russo et al., 2019). Commercially, NF and UF  
262 membranes are normally prepared to possess an additional skin layer (i.e. dense  
263 selective barrier) to provide higher retention rates. This skin layer can be realised  
264 by manipulating preparation conditions (e.g. exposure time, humidity, polymer  
265 concentration) or ii) coating a top layer on the membrane' surfaces.

266 The surface roughness also influences the separation performance of NF and UF  
267 membranes. For instance, membrane fouling is promoted by rougher surfaces  
268 (Evans et al., 2008). The presence of protuberances on the surface of polyamide  
269 membranes may be responsible for fouling initiated by fouled matter capture.  
270 Interestingly, membranes based on cellulose acetate display smoother surfaces  
271 which are less susceptible to fouling.

272 Most of the membranes exhibit a net negative charge under common operating  
273 conditions; therefore, electrostatic forces take place between the membrane  
274 surface and some of the compounds present in the treated solution. The  
275 membrane surface charge is depending on type of membrane (i.e. functional  
276 groups present on its surface) as well as the pH and ionic strength of the bulk feed.  
277 This surface charge becomes relevant in case of charged molecules (e.g. proteins)  
278 present in the feed solution (Kanani, 2015). Finally, hydrophobic and Coulombic  
279 intermolecular interactions between the molecules and membrane surface (e.g.  
280 polyphenols- polyphenols, polyphenols-membrane) contribute to the molecule  
281 retention (Crespo & Brazinha, 2010).

282 NF membranes and tight UF membranes (in the range 1-3 kDa) have been



283 recognized for their capability to recover low molecular weight molecules (e.g.  
284 carotenoids, peptides, anthocyanins, low molecular weight phenols and sugars)  
285 from several types of agro-food products and by-products (see **Table 2**). For  
286 instance, Díaz-Reinoso et al. (2017) proposed a combination of UF and NF  
287 membranes with adsorption-desorption processes in order to recover and  
288 concentrate phenolic antioxidant compounds from white wine vinasses, as  
289 depicted in the flow diagram depicted in **Figure 2**. The final dried product  
290 contained 45% of phenolics (expressed as gallic acid equivalent, GAE) and  
291 presented a radical scavenging capacity equivalent to almost 2 g of Trolox (6-  
292 hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid). The proposed process  
293 allowed to reduce also the pollution load of the final effluent: indeed, COD and total  
294 solids were reduced of about 85% and 92%, respectively, with respect the treated  
295 effluent.

296

297 **Figure 2.** Layout scheme and overall mass balance of the developed process for  
298 the recovery and concentration of polyphenols from white wine vinasses (GAE,  
299 gallic acid equivalents; Trolox, 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic  
300 acid) (Díaz-Reinoso et al., 2017).

301

### 302 **3. Membrane distillation**

303 Membrane distillation (MD) is a thermally driven membrane process, in which  
304 hydrophobic microporous membranes are commonly used for separating non-  
305 volatile solutes. The temperature difference between separated solutions results in  
306 a vapour pressure difference, followed by the transport of vapour molecules from

307 the higher vapour pressure stream to the lower vapour pressure stream (**Figure 3**).  
308 For example, in seawater desalination, this technology operates at atmospheric  
309 pressures and temperatures below 100°C (Belessiotis, Kalogirou, & Delyannis,  
310 2016), which represents an attractive alternative to classic processes according to  
311 several advantages, such as lower operating temperatures and pressures when  
312 compared to conventional distillation and pressure-driven membrane processes  
313 (e.g. reverse osmosis), mild processing conditions for heat-sensitive food  
314 ingredients and reduction of energy consumption when using industrial waste  
315 energy or solar energy (Blanco Gálvez et al., 2009; Qtaishat & Banat, 2013).  
316 Moreover, a theoretical rejection of about 100% for non-volatile compounds makes  
317 this technique one of the most effective processes for desalination.

318 Typical membranes for MD applications are realized in flat-sheet or tubular  
319 configuration with hydrophobic polymers including polypropylene (PP),  
320 polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE). Typical pore sizes of  
321 MD membranes range between 0.2 and 1.0  $\mu\text{m}$ . The transmembrane flux through  
322 a MD membrane is related to the membrane pore size and other characteristic  
323 parameters by the following equation (Fawzy, Varela-Corredor, & Bandini, 2019):

$$324 \quad N \propto \frac{r^{\alpha} \varepsilon}{\delta_m \tau} \quad (6)$$

325 where  $N$  is the molar flux,  $r$  the mean pore size of the membrane pores,  $\alpha$  a factor  
326 whose value is 1 for Knudsen diffusion and 2 for viscous fluxes, respectively,  $\delta_m$   
327 the membrane thickness,  $\varepsilon$  the membrane porosity and  $\tau$  the membrane tortuosity  
328 (Lawson & Lloyd, 1997). According to equation (6) the thinner the membrane and

329 the greater the porosity of the membrane, the greater the flux rate. On the contrary,  
330 thicker membranes assure better heat efficiency limiting the heat loss by  
331 conduction through the membrane matrix.

332 When dealing with the use of MD in food and bioproducts processing, most  
333 ongoing developments are focused on the concentration of fruit juices (see **Table**  
334 **3**).

335

336 **Figure 3.** General drawing of a membrane distillation process.

337

338 At large scale, the concentration of fruit juices in the food industry is performed by  
339 multi-stage vacuum evaporation. This process results in a loss of fresh juice  
340 flavors, color degradation and the appearance of a “cooked” taste due to thermal  
341 effects. Since MD can be carried out at the atmospheric pressure and at a  
342 temperature much lower than the boiling point of the solution, it has received a  
343 great attention as technique for fruit juice concentration.

344 To date, many studies evaluating the performance of MD for juices concentration  
345 have been developed, including those performed on apple juice (Gunko et al.,  
346 2006), sugarcane (Nene et al., 2002) and orange juice (Deshmukh et al., 2011). In  
347 these studies MD has been carried out according to the direct contact membrane  
348 distillation (DCMD) configuration in which the permeate side of the membrane  
349 consists of a condensing fluid in direct contact with the membrane (cold distillate)  
350 separated by the hot feed. Gunko et al. (2006) observed an important temperature



351 dependence on the capacity of the DCMD process. Their results showed that at  
352 the same temperature difference between feed and permeate, higher fluxes are  
353 achieved while increasing the temperature of the feed.

354 Even though DCMD seems to be the most common configuration for the  
355 separation of water (as a volatile component), there are some reports evaluating  
356 the potential of vacuum membrane distillation (VMD) configuration as  
357 concentration process. In VMD, there is a vacuum pressure applied on the  
358 permeate side of the MD membrane while condensation takes place outside the  
359 membrane module. This configuration minimizes the conductive heat transfer  
360 across the membrane due to the low pressure on the permeate stream. According  
361 to the literature, VMD is a potential technique for gentle aroma compounds  
362 recovery from natural sources, e.g. black currant juice (Bagger-Jørgensen et al.,  
363 2004), as well as for sucrose concentration (Chen et al., 2018). According to the  
364 findings of Bagger-Jørgensen et al. (Bagger-Jørgensen et al., 2004), a linear  
365 relationship between permeate flux and the difference of the water vapour pressure  
366 can be found employing VMD. Additionally, the authors observed higher recovery  
367 rate of aromas compared to a conventional aroma recovery plant. For example,  
368 using VMD, it was possible to recover up to 83% of highly volatile compounds and  
369 38% of poorly volatile compounds.

370 Despite the satisfactory quality of the concentrated juices using this technique,  
371 there are still some drawbacks limiting the use of MD in the food industry, such as  
372 temperature polarization and membrane fouling. Especially, temperature  
373 polarization causes temperatures at the membrane surfaces to differ from the bulk

374 temperatures measured in the feed and in the distillate with significant loss in the  
375 driving force for transport regarding the imposed force. Both phenomena may  
376 provide a flux decrease as a result of membrane permeability reduction as a  
377 function of operating time. Moreover, another important issue in MD technology lies  
378 with the membrane and its long term anti-wetting performance to process liquids,  
379 which may influence the vapour transport through the pores (El-Bourawi et al.,  
380 2006). When dealing with the fouling phenomenon, enzymatic pretreatments, as  
381 well as the use of MF and UF as clarification steps for removing suspended solids  
382 and pectins from juices, allow to reduce the juice viscosity and to improve the  
383 evaporation flux during the MD concentration step.

384

385 **Table 3.** Overview of the latest uses of MD for the concentration of juices.

386

387 Quist-Jensen et al. (2016) evaluated the effect of an integrated two-step DCMD  
388 process on the quality of blood orange juice. Firstly, the clarification of the extract  
389 was performed by UF, in order to remove suspended solids and juice turbidity; the  
390 clarified juice, with an initial total soluble solids (TSS) content of about 9.5 °Brix,  
391 was pre-concentrated up to 24 °Brix and then concentrated up to 65 °Brix by using  
392 a MD laboratory bench plant equipped with two PP hollow fiber membrane  
393 modules (Enka Microdyn MD-020- 2N-CP) with a nominal pore size of 0.2  $\mu\text{m}$  and  
394 a membrane surface area of 0.1  $\text{m}^2$ . Such approach allowed to produce high  
395 quality concentrated juices, as in the final product the organoleptic, nutritional and

396 antioxidant properties of the fresh juice were efficiently preserved. The  
397 performance of the DCMD operation during the concentration of the juice in the  
398 range 24–65 °Brix is depicted in **Figure 4**. Thermal gradients of about 9 °C every 9  
399 h resulted in an evaporation flux of about 0.55 kg m<sup>-2</sup> h<sup>-1</sup> (Figure 4a). After this, the  
400 membrane cleaning at regular intervals (each 9 h) produced a good restoration of  
401 the initial flux. Flux decays were observed by increasing the TSS content (Figure  
402 4b) and juice viscosity (Figure 4c) confirming that at a higher TSS content the flux  
403 decrease primarily depends on juice viscosity and, consequently, on temperature  
404 and juice concentration. The formation of fouling layers offers an additional  
405 resistance to mass transfer and heat transfer contributing to a progressive flux  
406 decline.

407

408 **Figure 4.** Concentration profile of blood orange juice by DCMD. (a) evaporation  
409 flux (thermal gradient at time 0, 9, 18 and 27 h), (b) total soluble solids content and  
410 (c) viscosity as a function of operating time (Quist-Jensen et al., 2016).

411 Kozák et al. (2009) applied MD to produce a concentrated black-currant juice using  
412 a PP membrane module in hollow fiber configuration. It was reported that the  
413 increase in driving force (from 15 to 19 °C as thermal gradient) leads to an  
414 improvement of fluxes of about 80%. The microfiltered juice with 22°Brix TSS  
415 content was pre-concentrated by reverse osmosis (RO) and then concentrated by  
416 MD up to 58.2°Brix. All the analysed parameters, including density, total acidity and  
417 anthocyanins increased proportionally to the TSS content of the juice.

418

419 Other applications of MD in the food industry comprise the ethanol removal from  
420 fermentation broths, which is usually carried out by conventional distillation. Gryta  
421 and Barancewicz (2011) used PP capillary membranes to separate ethanol by MD  
422 during the fermentation of sucrose solutions with the participation of the yeast  
423 species *Saccharomyces cerevisiae*. Besides ethanol, propionic and acetic acids  
424 were removed from the broth to the distillate. The use of MD allowed to decrease  
425 the inhibitory effect of these compounds on microbial culture and reduce the cost of  
426 further concentration of alcohol. Interestingly, Purwasasmita et al. (2015) evaluated  
427 the potential of non-porous MD membranes (thin-film composite polyamide) in the  
428 beer dealcoholization process. In selected operating conditions (300 kPa feed  
429 pressure and 58 kPa vacuum pressure) the alcohol content was reduced from 5%-  
430 vol. to 2.45%-vol. in 6 h, with minimal loss of nutrients and flavoring components  
431 such as maltose and glycerol. The whole results clearly indicate that MD is suitable  
432 for the concentration of extracts (mainly fruit juices) by selective removal of water,  
433 recovery of aromas, and the removal of ethanol from specific processed  
434 feedstocks. However, in case of the selective recovery of aromas, and/or removal  
435 of ethanol aiming the manufacture of new processed products, pervaporation  
436 technology is likely the most sought technology. The next section addresses the  
437 latest findings in the field.

438

#### 439 4. Pervaporation

440 Pervaporation (PV), as a highly selective membrane separation technique, can  
441 selectively separate multicomponent azeotropic mixtures by partial vaporization  
442 using a physical barrier (so-called membrane). This perm-selective membrane can  
443 be either a non-porous polymeric or a non-porous inorganic (ceramic/zeolite)  
444 membrane. Unlike the previous membrane-based technologies, like MF, UF, NF  
445 and MD, in which porous membranes are used, PV is based on membranes with a  
446 non-porous structure. Indeed, PV uses a coupled mechanism of permeation and  
447 evaporation phenomenon (Kaippamangalath & Gopalakrishnapanicker, 2018;  
448 Wijmans & Baker, 1995). In pristine polymer-based membranes, the mass  
449 transport across the dense membrane has been well explained by the so-called  
450 solution-diffusion mechanism, in which specific properties (i.e. solubility, diffusivity)  
451 of the target molecules play a fundamental role (Wijmans & Baker, 1995).

452 To carry out the selective extraction of any component, the liquid azeotropic feed  
453 solution is in direct contact with the “selective” layer of the membrane, while  
454 vacuum is generally applied on other side of the membrane (i.e. permeate stream).  
455 Such permeate stream is in vapor phase and contains most of the permeating  
456 compounds with higher compatibility-affinity to the membrane (**Figure 5**). Since  
457 different species permeate through the membrane at different rates, substances at  
458 low concentration in the feed stream can be highly enriched in the permeate.

459

460 **Figure 5.** General schematic of a pervaporation process.

461

462 In terms of the real driving force for the PV process, the flux,  $J_i$ , of a specific  
463 compound  $i$  can be described by the following transport relation (Castro-Muñoz et

464 al., 2019):

$$465 \quad J_i = -L_i \frac{d\mu_i}{dz} \quad (7)$$

466 where  $d\mu_i/dz$  represents the chemical potential gradient of the target compound I  
467 across the membrane and  $L_i$  a phenomenological coefficient to be experimentally  
468 determined.

469 Taking into account the equilibrium conditions implied by the solution-diffusion  
470 model, the component  $J_i$  (expressed as  $\text{kg m}^{-2} \text{h}^{-1}$ ) can be derived as:  $J_i = \frac{P_i}{\delta} (p_{f,i} -$   
471  $p_{p,i})$  (8)

472 where  $P_i$  represents the permeability coefficient,  $\delta$  the membrane thickness and  
473  $(p_{f,i} - p_{p,i})$  the difference in partial vapour pressure of component  $i$  across the  
474 membrane.

475 The separation efficiency of PV membranes is usually expressed by means of the  
476 separation factor  $\alpha_{i,j}$ , defined as:

$$477 \quad \alpha_{i,j} = \frac{c_{p,i}/c_{p,j}}{c_{f,i}/c_{f,j}} \quad (9)$$

478 in which  $c$  represent the concentration (wt%) of a component  $i$  or  $j$  in the feed or  
479 permeate (Castro-Muñoz & González-Valdez, 2019). The corresponding  
480 enrichment factor is expressed as:

$$481 \quad \beta = \frac{c_{p,i}}{c_{f,i}} \quad (10)$$

482 The choice of the membrane material in PV is strongly correlated to the  
483 temperature and composition of the feed mixture as well as to the  
484 separation/purification target and the desired performance. Hydrophilic polymeric  
485 materials, including cellulose acetate (CA), polyvinyl alcohol (PVA), sodium



486 alginate, chitosan, poly lactic acid (PLA), facilitate the transport of highly polar  
487 compounds including water and alcohols. On the contrary, typical hydrophobic  
488 (also known as organophilic) membranes, including poly(octylmethylsiloxane)  
489 (POMS), polydimethylsiloxane (PDMS), polyether block amide (PEBA), or poly(1-  
490 (trimethylsilyl)- 1-propyne) (PTMSP), favor the preferential transport of non-polar  
491 compounds (or less polar molecules) (Castro-Muñoz et al., 2018c). Indeed, these  
492 kinds of membranes are thus preferred for the extraction of aroma molecules when  
493 they are contained in aqueous complex solutions (e.g. extract, juices, wines).  
494 Importantly, the chemistry and nature (e.g. hydrophobic or hydrophilic) of the  
495 targeted compounds will play a crucial role during the aroma extraction using PV  
496 (Baudot & Marin, 1997; Castro-Muñoz, 2019; Fouda et al., 1993). When organic  
497 molecules are concentrated and it is needed to selectively separate them from  
498 each other, it is suitable to use membranes with highly hydrophilic nature. These  
499 membranes can separate the molecules based on their polarity according to the  
500 polar functional groups, e.g. hydroxyl (-OH) groups. Nevertheless, some other  
501 aromas and complex organic compounds contained in agro-food products could  
502 also influence the extraction (Isici et al., 2006). Herein, the nature and chemistry of  
503 the molecules will have a significant effect on the yield of the process. Recent  
504 literature reviews have reported some of the primary aromas extracted and  
505 recovered from agro-food products, including extracts, wastes, by-products, fruit  
506 juices, and food processed products (e.g. wine, beer, cider, dairy products) by  
507 using different PV membranes (Castro-Muñoz, 2019).

508 Aroma compounds, including alcohols, esters and organic compounds, such as  
509 trans-2-hexenal, were preferentially permeated from apple juice through a

510 hydrophobic membrane with an active layer of PDMS by Bengtsson et al. (1989).  
511 Enrichment factor ( $\beta$ ) values were in the range of 44-125. Similarly, Cassano et al.  
512 (2006) investigated the performance of a commercial PDMS-based membrane in  
513 the recovery of aroma compounds from kiwifruit juice within an integrated  
514 membrane process where the depectinised juice was previously clarified by UF  
515 and then concentrated by osmotic distillation (OD). The enrichment factor for most  
516 of the aroma compounds detected in the permeate of the fresh juice resulted  
517 higher than that measured for the clarified and concentrated juice with the  
518 exception of 3-hexen-1-ol and (E)-2-hexen-1-ol. This result suggested the use of  
519 PV for the recovery of aroma compounds directly from the fresh juice before the  
520 clarification and concentration step. In all PV experiments, the enrichment factor of  
521 the alcohols resulted lower (10-40) than that measured for esters, such as methyl  
522 and ethyl butanoate (about 100). For all samples, the permeate flux increased  
523 linearly in the investigated range of operating temperatures (20-40 °C). For the  
524 concentrated juice the total flux resulted slightly higher than that measured for  
525 fresh and clarified juice (**Figure 6**). A similar behavior was also observed by Figoli  
526 et al. (2010) in the processing of fresh kiwifruit juice by PV with a composite  
527 membrane having an active layer made of styrene-butadiene-co-styrene (SBS) of  
528 about 40  $\mu\text{m}$  coated on a commercial UF support of PVDF. For this membrane the  
529 highest recovery factor was reached at an operating temperature of 30 °C.

530

531

532



533 **Figure 6.** Effect of temperature on the total flux in the processing of fresh, clarified  
534 and concentrated kiwifruit juice by PV (Cassano et al., 2006).

535

536

537

538 Aroujalian and Raisi (2007) investigated the effect of key parameters such as feed  
539 temperature, permeate pressure and feed flow rate on the pervaporative recovery  
540 process of volatile aroma compounds from orange juice by using a commercial  
541 PDMS membrane. Results indicated that increasing of Reynolds number from 500  
542 to 2500 had a very slight increasing in flux and enrichment factor of aroma  
543 compounds in the permeate. On the other hand, total and partial fluxes increased  
544 significantly when feed temperature was increased from 25 to 50°C: this  
545 phenomenon was attributed to an increase of the free volume in the PV membrane  
546 which in turn increases the diffusion rate of individual permeating molecules and  
547 high permeation fluxes. The selectivity of all aroma compounds increased also with  
548 temperature and this change was attributed to the activation energy of each  
549 component. As expected, an increasing of permeate pressure decreased the  
550 driving force of the permeation through the membranes leading to a reduction in  
551 the permeation flux. For some aroma compounds, such as hexanal, ethyl acetate  
552 and ethyl butyrate, the enrichment factor increased when vacuum pressure was  
553 raised.

554 Similar results were also obtained by Raisi et al. (2008) in the recovery of aroma  
555 compounds from pomegranate juice by using POMS and PDMS membranes. The



556 POMS membranes produced a higher aroma enrichment factor but lower  
557 permeation flux compared to the PDMS membranes.

558 Additionally, the influence of the feed temperature on the PV recovery was  
559 analyzed using the Arrhenius model. It was found out that the activation energy  
560 ( $E_a$ ) of the molecules was positive, which reveals that any feed temperature  
561 increase must cause higher permeation flux values. Interestingly, the apparent  $E_a$   
562 of the recovered aromas was higher than water molecules, indicating that the  
563 transport of these specific aromas across the membrane is higher temperature  
564 dependent comparing to water molecules. In general, when activation energy  
565 parameter is high, the permeation flux will be more sensitive to temperature  
566 variations; therefore, aroma molecules are likely more sensitive to this parameter  
567 (Raisi et al., 2008; Raisi et al., 2009). Importantly, the feed operating temperature  
568 plays a key role for the performance of a PV membrane since primarily influences  
569 the solubility and diffusion coefficients of the components across membrane  
570 (Wijmans & Baker, 1995). In addition to this, the separation of thermolabile  
571 molecules is more recommendable at lower operating temperatures, in order to  
572 prevent their thermal degradation.

573 Coffee is well recognized for its characteristic perfume notes related to several  
574 molecules, e.g. 2-methylbutanoic acid, 2-methylpropanal, hexanal, (E)-2-nonenal,  
575 to mention just a few. Thereby, it has been also used for the separation of key  
576 flavor and aroma molecules. Organic molecules, like 2-5-dimethyl pyrazine, and  
577 2,3-butanedione, that give sensorial features (e.g. creamy, sweet, nutty-like,  
578 buttery, and milky) were recovered using a commercial Pervatech BV membrane  
579 (Weschenfelder et al., 2015). This commercially available PDMS membrane had a



580 high selectivity towards 2,3-butanedione ( $\beta=45$ ) and 2-5-dimethyl pyrazine ( $\beta=42$ ).  
581 Additionally, such membranes offered relatively moderate organic permeation flux  
582 (of about  $0.432 \text{ kg m}^{-2} \text{ h}^{-1}$ ).

583 Within the food beverage production, valuable aromas are normally recovered and  
584 then concentrated using traditional distillation, and finally blended to clarified juice  
585 (De Vasconcelos Facundo et al., 2009). Some agro-food effluents are potentially  
586 considered as new feedstock of aroma molecules. For example, Souchon et al.  
587 (2002) applied the PV process to the deodorization of a cauliflower blanching  
588 effluent in order to recover valuable food flavouring compounds such as dimethyl  
589 disulfide, dimethyl trisulfide and S-methyl thio-butyrate. Hydrophobic PDMS and  
590 PEBA membranes showed high selective affinity for S-methyl thio-butyrate, with  
591 enrichment factors of 307 and 1200, respectively, when a model solution with three  
592 sulfur compounds was tested. In fact, this membrane was selected for PV  
593 experiments on the industrial effluent. In this case the selectivity resulted five times  
594 lower than the one obtained on model solution probably due to the formation of an  
595 important boundary layer. However, the odour quality of the permeate was  
596 completely modified and the retentate was significantly deodorized regards to the  
597 feed.

598 Recently, Dawiec-Liśniewska et al. (2018a) evaluated the extraction of aromas  
599 from fruit juice hydrolates by using PV on both laboratory and semi-technical scale.  
600 The hydrolate derivatives were obtained from several horticultural products (e.g.  
601 blackcurrant, plum, cherry and apple fruits), which usually contain a wide category  
602 of aromatic-based compounds. 37 different aroma compounds were identified and  
603 quantified in the blackcurrant hydrolate, while 14 and 20 organic compounds were



604 identified in cherry and apple derivatives, respectively. Commercial hydrophobic  
605 PDMS membranes (Pervap 4060, Sulzer, Germany) used in both laboratory and  
606 semi-technical scale, exhibited extremely high separation affinity for organic  
607 molecules, like heptan-1-ol ( $\beta \sim 1131$ ), hexanal ( $\beta \sim 3678$ ), pentan-1-ol ( $\beta \sim 5800$ ), and  
608 butyl acetate ( $\beta \sim 8602$ ). These membranes displayed a total permeate flux of at  
609 least  $0.180 \text{ kg m}^{-2} \text{ h}^{-1}$  that can be further increased up to  $0.450 \text{ kg m}^{-2} \text{ h}^{-1}$ ,  
610 depending on operating temperature. The results of the economic analysis  
611 demonstrated that PV is a profitable and feasible option for aroma recovery from  
612 fruit hydrolates.

613 Some processed products from the food beverage industry, including beer, cider  
614 and wine, are also currently explored for the recovery of aroma compounds  
615 (Catarino et al., 2009; Catarino & Mendes, 2011a; Paz et al., 2017). Catarino et al.  
616 (2009) evaluated a commercial brand beer as a candidate for extracting a wide  
617 range of esters (e.g. isomyl acetate, ethyl acetate), alcohols (e.g. isoamyl alcohol,  
618 propanol, isobutanol,) and aldehydes (e.g. acetaldehyde). The extraction of  
619 aromas was aimed to achieve the sensorial properties of low-alcoholic content  
620 beer, which is prone to lose some of these aromas caused by ethanol removal  
621 (Castro-Muñoz, 2019). PV experiments were performed by using a  
622 polyoctylmethylsiloxane/polyetherimide (POMS/PEI) composite asymmetric  
623 membrane and the effect of operating conditions on the process performance was  
624 analysed according to the response surface methodology (RSM) approach. In  
625 optimized conditions of feed temperature, feed velocity and permeate pressure  
626 ( $12.4 \text{ }^\circ\text{C}$ ,  $0.45 \text{ m s}^{-1}$  and  $1.0 \text{ mbar}$ , respectively) the permeate flux was predicted to  
627 be  $7.26 \text{ kg m}^{-2} \text{ s}^{-1}$ , while alcohols and esters selectivity was in the range 1.31-3.39



628 and 14.46-17.10, respectively. Experimental results, obtained in optimized  
629 operating conditions, resulted in a good agreement with the predicted values of the  
630 regression model. Experimental results for the runs performed at the optimal  
631 operating conditions mostly agreed with the predicted values.

632 In another approach Catarino & Mendes (2011b) investigated the manufacture of  
633 non-alcoholic beer with a corrected natural flavour profile by using an industrial set-  
634 up. Firstly, the aroma compounds were extracted from the original beer by using a  
635 POMS/PEI membrane, and later mixed with the previously dealcoholized beer.  
636 Such integrated methodology permitted to achieve a flavored non-alcoholic beer  
637 with minimal content of alcohol (<0.5 vol.% ethanol). A similar approach was used  
638 to improve the aroma profile of dealcoholized wine samples (Catarino & Mendes,  
639 2011a).

640 Two different commercial beers differing in ethanol concentration (a special beer  
641 ~5.5% ABV and a reserve beer ~6.5% ABV) were processed by using a  
642 hydrophobic PDMS Pervatech membrane to extract aroma molecules which were  
643 then mixed to a low-alcohol beer (less than 1% ABV) and an alcohol-free beer  
644 (less than 0.1% ABV) to enhance their sensorial and organoleptic quality (Olmo et  
645 al. 2014). Three individual flavor components were analysed in detail (ethyl  
646 acetate, isoamyl acetate, isobutyl alcohol) and selectivities were predicted  
647 considering solubility parameters of polymer and compounds. The theoretical  
648 calculation of relative selectivities from solubility parameters can provide useful  
649 information about the design of the process and the selection of the membrane in  
650 order to reach high productivities and selectivities. Similarly, Salgado et al. (2017)  
651 used a PV spiral-wound membrane module with a PDMS based membrane (PV-



652 SR1, Pervatech) for the extraction of aroma precursors (i.e. hexanal,  
653 isoamylalcohol, 1-hexanol, benzaldehyde, benzyl alcohol and 2-phenylethanol)  
654 from grape must. Such aromas were recovered and then blended into low alcohol  
655 white wines obtained by reducing the sugar content in grape must through NF  
656 membranes, in order to produce a full flavored white wine with reduced alcohol  
657 content. The PV membrane did not offer high permeation rates ( $\sim 0.073 \text{ kg m}^{-2} \text{ h}^{-1}$ ),  
658 however, the final product showed an aroma content similar to the original grape  
659 must with the exception of benzaldehyde and 1-hexanol.

660 All these studies confirm the great potential of PV-based membrane processes for  
661 recovering/extracting aromatic-based compounds from different natural feedstocks  
662 by means of non-porous membranes. Furthermore, PV can also assist other types  
663 of food processing processes, e.g. when dealing with the removal of ethanol to  
664 produce non-alcoholic drinks and beverages based on wine and beer.

665

## 666 **5. Conclusions and future trends**

667 Membrane-based processes, including MF, UF, NF, MD, and PV, have  
668 demonstrated to meet the recovery and extraction requirements of biologically  
669 active compounds, such as phenolic-based molecules and their specific  
670 derivatives, as well as aroma compounds from natural products and agro-food by-  
671 products.

672 In general, MF technology finds its main application as a pre-treatment technique  
673 to separate macromolecules, colloids and suspended solids from solutions, which  
674 subsequently are processed by UF and NF technologies for a better reclamation of  
675 smaller high-added-value molecules. Particularly, PV proved its ability to recover



676 different types of aromatic-based compounds, in which the membrane nature, as  
677 well as the polarity of the target solutes, play a key role on both productivity and  
678 selectivity of the process. In addition, MD and PV processes have been also  
679 addressed as potential candidates for the production of non-alcoholic beverages.  
680 Comparing with conventional techniques, membrane-based separation operations  
681 are economically profitable not only in terms of extraction but also because these  
682 emerging processes do not demand the usage of further agents or/and destructive  
683 compounds. Thus, the extraction and recovery of valuable molecules from different  
684 sources are both industrially sustainable and environmentally friendly, making  
685 membrane-based processes meaningful for the integral management of products  
686 and by-products derived from the food industry. It is likely that the prominent  
687 raising worldwide demand towards these valuable solutes will promote a wider  
688 application of such membrane-based operations in this field.  
689 Concluding remarks related to the different processes analysed in this context are  
690 reported in the following.

- 691 • *Microfiltration, ultrafiltration and nanofiltration*: Scientists in the field should  
692 put an effort into treating real feed solutions in order to provide a more real  
693 proximity performance of membranes. In such a way, future developments  
694 will provide relevant insights into the possibility of considering these  
695 processes for large scale applications. Importantly, fouling phenomena  
696 represent the main drawback when real complex solutions (e.g. agro-food  
697 by-products and wastewaters) are processed. At this purpose,  
698 "membranologists" are looking for novel membrane materials in membrane  
699 preparation, as well as facing the weakness of the existing ones, to develop



700 and manufacture new smart membranes that could mitigate membrane  
701 fouling and therefore less prone to be fouled. Another important aspect  
702 regards to the purity of the recovered solutes, which will primarily need  
703 further purification for a specific application.

704 • *Membrane distillation*: This process can overcome the limits of thermal  
705 processes (e.g. evaporation, distillation). Thanks to the effective operation at  
706 low temperatures, MD can be integrated with alternative energy sources for  
707 example waste energy or solar energy. This feature may make it more  
708 promising for industrial implementation. On the other hand, most of MD  
709 studies regard the concentration of aqueous model solutions at lab scale;  
710 thereby, investigations on complex feed solutions are encouraged. There  
711 are still some issues that must be overcome, such as scaling, fouling and  
712 pore wetting in a long-term MD processing. At this point, the main research  
713 challenge is to create new membrane materials with improved porosity and  
714 higher hydrophobicity using low-thermal conductive polymers to minimize  
715 heat loss.

716 • *Pervaporation*: This process finds the low permeation rate as its main  
717 drawback compared to other membrane-based technologies (e.g. MF, UF  
718 and NF), while its strength comprises the high selectivity. Importantly, two  
719 main issues should be addressed by future studies in the field. The first one,  
720 real feed solutions (e.g. agro-food products and by-products) should be  
721 tested in order to provide a more realistic approximation of membranes'  
722 performance over long-term operation. On the other hand, as a second  
723 issue, scientists should preferentially report the PV performance data based





724 on permeance and selectivity. Such parameters are a preferred way of  
725 reporting pervaporation results, in which membrane' performance does not  
726 depend on the operating parameters (including driving force). In this way, a  
727 fair comparison of different studies can be given.

728

### 729 **Conflict of Interest**

730 The authors declare no conflict of interest.

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732

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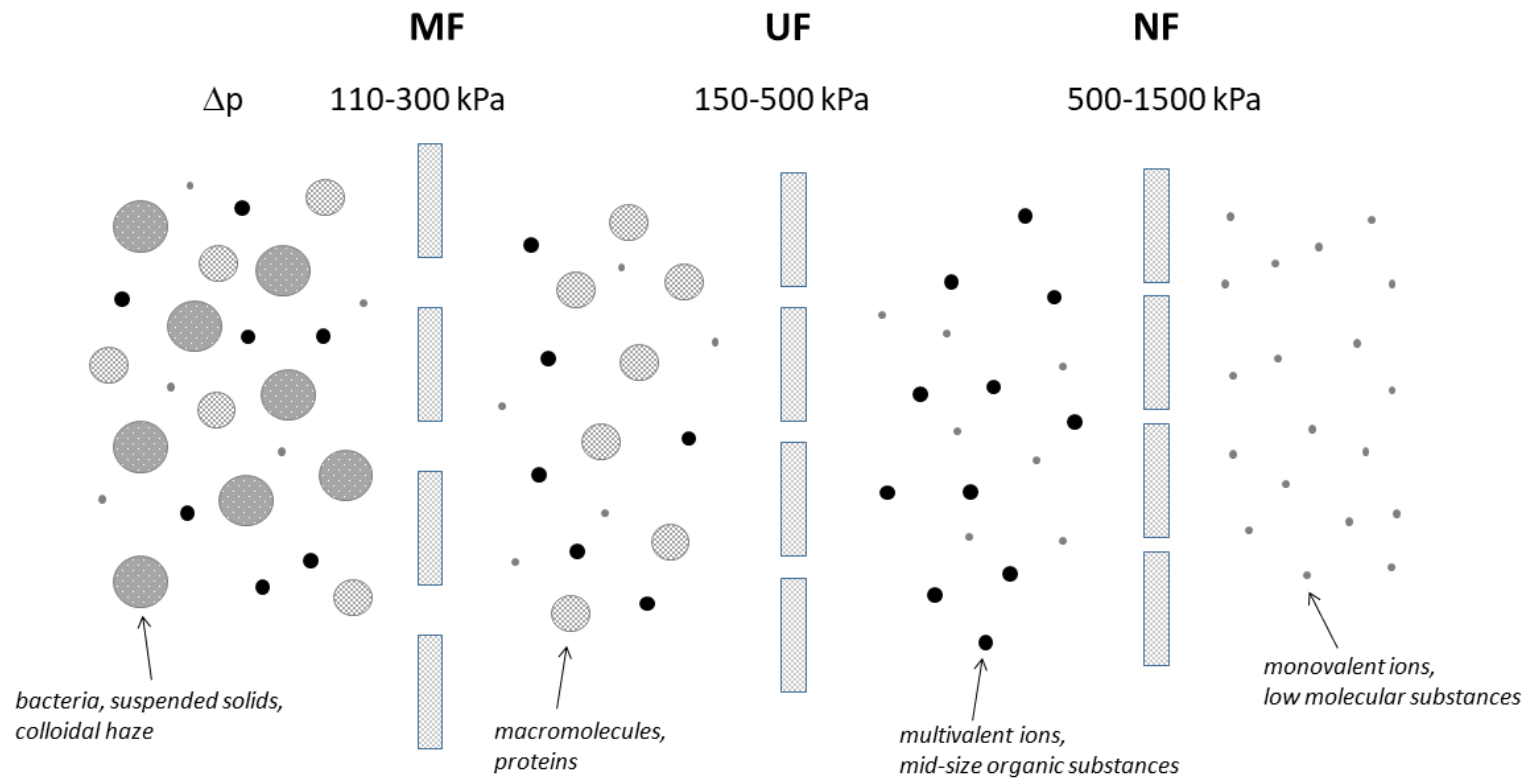
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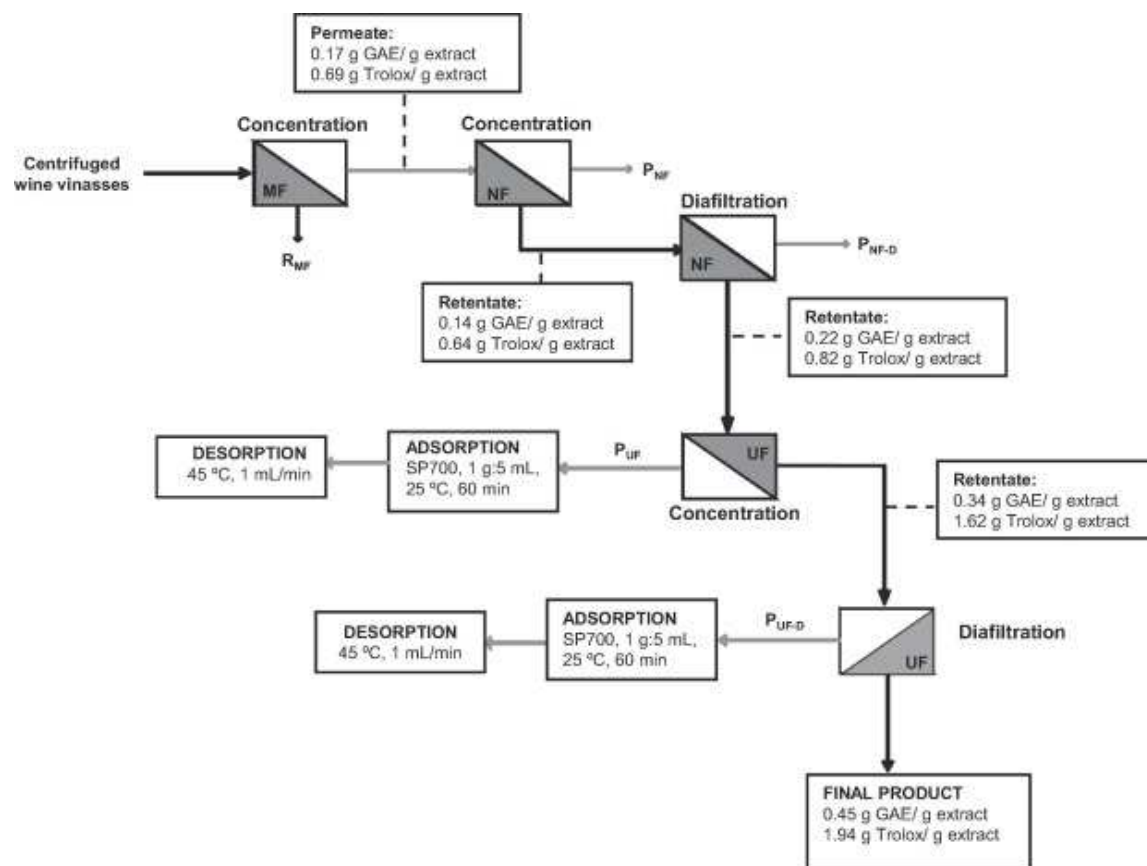
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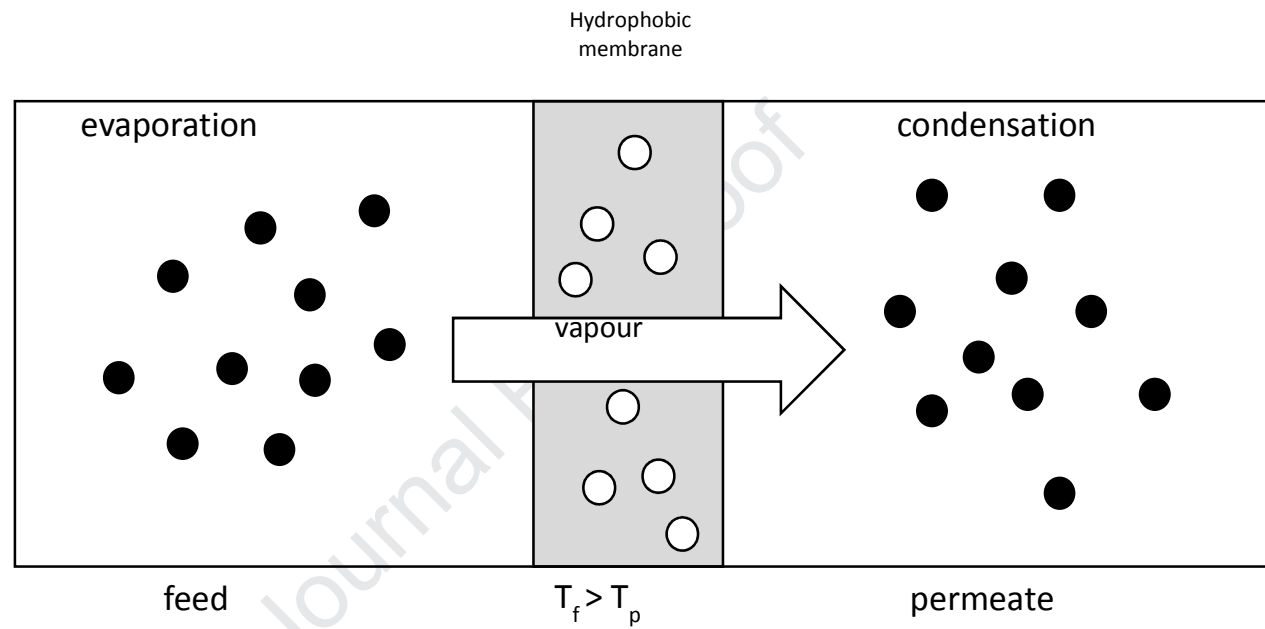
**Figure 1.** General description of pressure-driven membrane process.

**Figure 2.** Layout scheme and overall mass balance of the developed process for the recover and concentration of

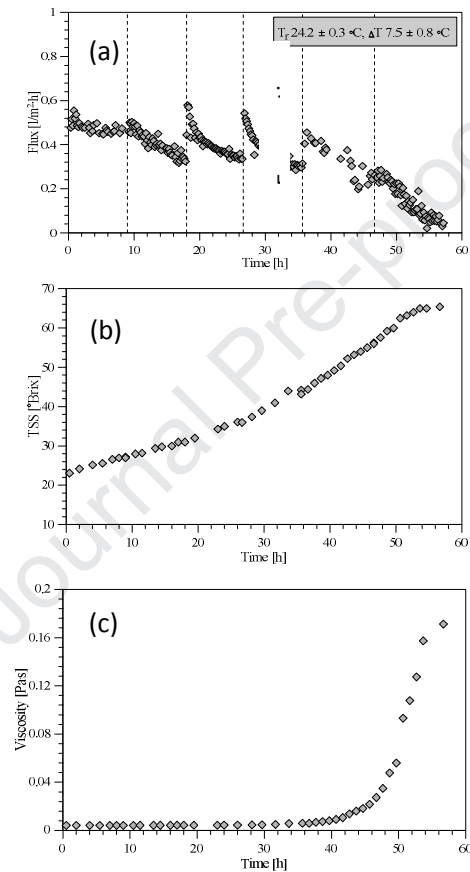


polyphenols from white wine vinasses (Díaz-Reinoso et al., 2017).

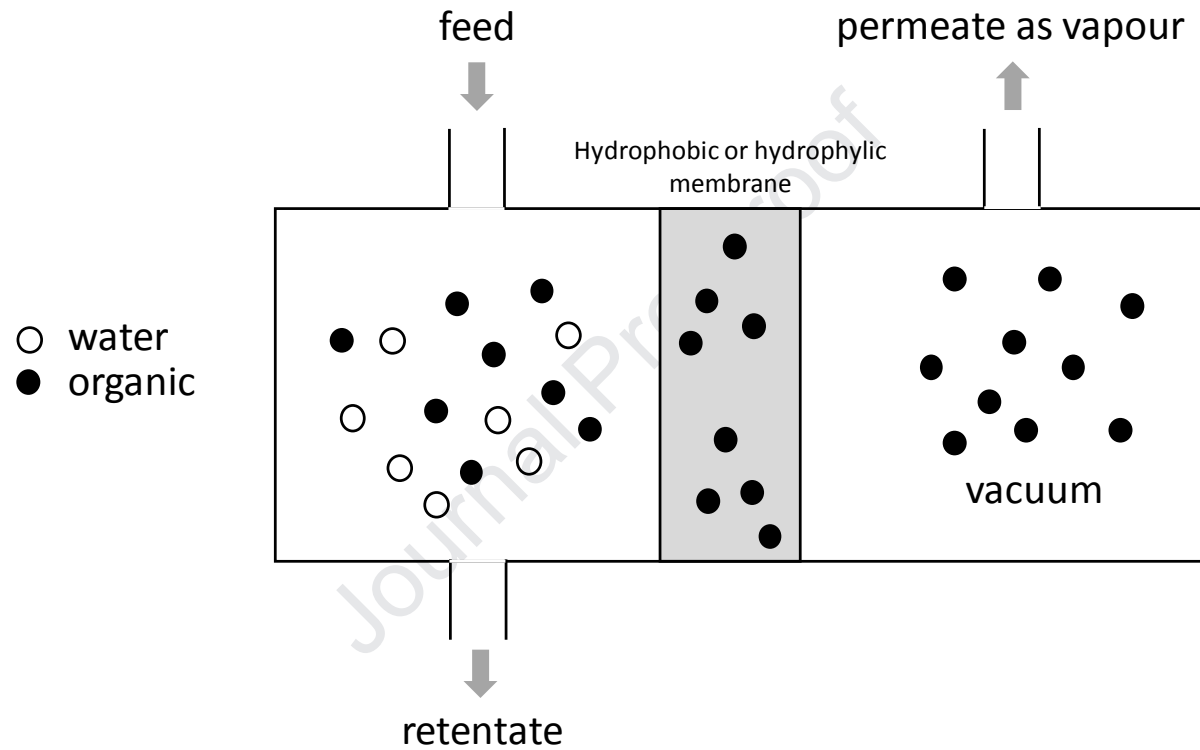
**Figure 3.** General drawing of a membrane distillation process.



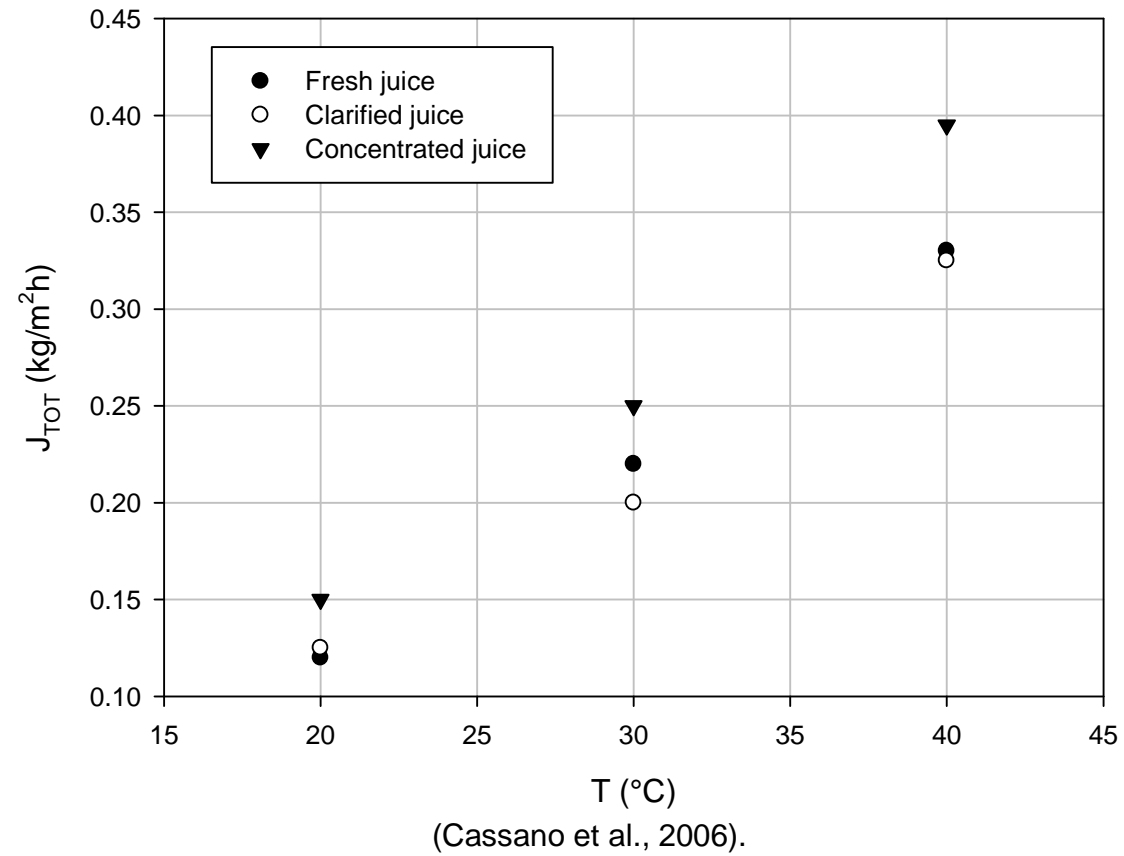
**Figure 4.** Concentration profile of blood orange juice by DCMD. (a) evaporation flux (thermal gradient at time 0, 9, 18 and 27 h), (b) total soluble solids content and (c) viscosity as a function of operating time (Quist-Jensen et al., 2016).



**Figure 5.** General schematic of a pervaporation process.



**Figure 6.** Effect of temperature on the total flux in the processing of fresh, clarified and concentrated kiwifruit juice by PV



**Table 1.** Main characteristics of MF, UF, NF, MD and PV processes.

Membrane process	Driving force	Mass transfer mechanism	Membrane	Water permeation flux (L/m <sup>2</sup> h)	Applications
Microfiltration	Pressure difference, 110-300 kPa	Convection	Porous; pore size 0.1-10 μm	500-10,000	Clarification, pre-treatment, sterilization
Ultrafiltration	Pressure difference, 150-500 kPa	Convection	Porous; pore size 1-100 nm	100-2,000	Concentration, fractionation of macromolecular solutions
Nanofiltration	Pressure difference, 500-1500 kPa	Diffusion/convection	Porous; pore size <2 nm	20-200	Concentration, purification of low molecular weight organic compounds, removal of multivalent ions
Membrane distillation	Partial pressure gradient	Evaporation diffusion/condensation	Hydrophobic micropores	0.1-30	Desalination, concentration
Pervaporation	Chemical potential or concentration difference	Adsorption/diffusion/desorption	Non-porous thin film	0.1-5	Separation of mixtures of volatile liquids



**Table 2.** Bioactive molecules recovered from agro-food by-products and wastewaters using membrane technologies.

<i>Recovered molecule</i>	<i>Recovery rate</i>	<i>Agro-food waste</i>	<i>Membrane process</i>	<i>MWCO/Material/ Configuration</i>	<i>Membrane nature:</i>	<i>Reference</i>
Phenolic compounds	45.7 %	Nixtamalization wastewaters	Integrated membrane process:			Castro-Muñoz et al., 2016; Castro-Muñoz & Yáñez-Fernández, 2015)
			MF	0.2 µm / Polysulfone / Hollow fiber	Hydrophobic	
			UF	100 kDa / Polysulfone / Hollow fiber	Hydrophobic	
Phenolic compounds	> 70 % > 80 % > 30 % > 60 %	Fermented grape pomace	UF	1000 Da / Thin-film / Spiral wound	Hydrophilic	Díaz-Reinoso et al. (2009); Díaz-Reinoso et al. (2010)
			UF	1000 Da / Ceramic (titania) / Tubular	Hydrophilic	
			NF	250 Da / Polyamide-polysulfone / Spiral wound	Hydrophilic	
			NF	350 Da / Polyamide-polysulfone / Spiral wound	Hydrophilic	
Hydroxytyrosol, protocatechuic acid, caffeic acid, tyrosol and p-cumaric acid	48.3 % 8.7 % 33.5 %	Olive mill wastewaters	UF	4 kDa / polyethersulphone / Flat sheet	Hydrophobic	Cassano et al. (2011)
			UF	10 kDa / Regenerated cellulose / Flat sheet	Hydrophilic	
			UF	10 kDa / Polyethersulphone / Flat sheet	Hydrophobic	
Hydroxycinnamic acids, o-diphenols	81 % 77 % 56 %	Winery sludge from red grapes	UF	100 kDa / Polysulfone / Flat sheet	Hydrophobic	Galanakis et al. (2013)
			UF	20 kDa / Polysulfone / Flat sheet	Hydrophobic	
			UF	1 kDa / Composite fluoropolymer / Flat sheet	Hydrophobic	
3,4-DHPEA, p-HPEA, 3,4-DHPEA-EDA, verbascoside, and total phenols	--	Olive mill wastewater	Integrated membrane process:			Servili et al. (2011)
			MF	0.3 µm / Polypropylene / Tubular	Hydrophilic	
			UF	7 kDa / Polyamide-polysulfone / Spiral wound	Hydrophilic	





Chlorogenic acid, Cynarin, Apigenin-7-O-glucoside	100 %	Artichoke wastewaters	Integrated membrane process:			Conidi et al. (2014)
			UF	50 kDa / Polysulfone / Hollow fiber	Hydrophobic	
			NF	400 Da / Polyethersulfone / Spiral wound	Hydrophobic	
			NF	150-300 Da / Polyamide / Spiral wound	Hydrophilic	
Gallic acid, chlorogenic acid and epigallocatechin gallate	> 85 %	Artichoke wastewaters	NF	400 Da / Polyethersulphone / Spiral wound	Hydrophobic	Cassano et al. (2015)
Hydroxytyrosol, procatechuic acid, catechol, tyrosol, caffeic acid, and p-cumaric acid	100 %	Residues from mate tree	NF	150-300 Da / Thin-film/ Spiral wound	Hydrophobic	Prudêncio et al. (2012)
		Olive mill wastewaters	Integrated membrane process:			Cassano et al. (2013)
			UF	0.02 $\mu\text{m}$ / PVDF / Hollow fiber	Hydrophobic	
			UF	1 kDa / Composite fluoropolymer / Flat sheet	Hydrophobic	
			NF	Salt rejection >97% / Thin-film / Spiral wound	Hydrophobic	
Hydroxytyrosol, procatechin acid, tyrosol, caffeic acid, p-cumaric acid, oleuropein and some other low MW polyphenols.	78 %	Olive mill wastewaters	Integrated membrane process:			Garcia-Castello et al. (2010)
			UF	200 nm / Al <sub>2</sub> O <sub>3</sub> / Tubular	Hydrophobic	
			NF	578 Da / Polyethersulphone / Spiral wound	Hydrophobic	
Hydroxycinnamic acids and flavonols.	40 %	Olive mill wastewaters	UF	25 kDa / Polysulfone / Spiral wound	Hydrophobic	Galanakis et al. (2010)
	71 %		UF	10 kDa / Polyethersulfone / Spiral wound	Hydrophobic	
	81 %		UF	2 kDa / Polyethersulfone / Spiral wound	Hydrophobic	
	99 %		NF	120 Da / Polypiperazine/ Spiral wound	Hydrophilic	
Anthocyanins, flavonoids	> 90 %	Orange press liquor	NF	180 Da / Polyamide-polysulfone / Spiral wound	Hydrophilic	Conidi et al. (2012)
	> 80 %		NF	300 Da / Polypiperazine amide thin-film composite / Spiral wound	Hydrophilic	
	> 80 %		NF	400 Da / Polyethersulfone / Spiral wound	Hydrophobic	
	> 70 %		NF	1000 Da / Polyethersulfone / Spiral wound	Hydrophobic	
Anthocyanins (cyanidin-3-glucoside chloride, myrtillin chloride and peonidin-3-	> 65 %	Orange press liquor	NF	Na <sub>2</sub> SO <sub>4</sub> rejection > 25-50 % / Polyethersulfone / Spiral wound	Hydrophobic	Cassano et al. (2014)



glucoside chloride), flavanones						
Chlorogenic acid, Apigenin-7-O-glucoside	100 %	Artichoke wastewaters	NF	200-300 Da / Polyamide / Spiral wound	Hydrophilic	Conidi et al. (2015)
Caffeoylquinic acid, flavonoids, chlorogenic acid, cynarin	> 40 %	Artichoke brines	NF	1000 Da / Polyethersulfone / Spiral wound	Hydrophobic	Cassano et al. (2016b)
	> 62 %		NF	400 Da / Polyethersulfone / Spiral wound	Hydrophobic	
	> 99 %		NF	300 Da / Piperazineamide / Spiral wound	Hydrophobic	
	> 95 %		NF	150-300 Da / Cross-linked polyamide/ Spiral wound	Hydrophilic	
	> 93 %		NF	150-300 Da / Cross-linked polyamide/ Spiral wound	Hydrophilic	
	> 80 %		White vinasses	NF	200 Da / Polyethersulfone / Spiral wound	
Phenolic compounds	21 %	Winery effluents	MF	0.4 $\mu\text{m}$ / Polyimide / Hollow fiber	Hydrophilic	Giacobbo et al. (2015)
	5 %	Winery effluents	MF	0.2 $\mu\text{m}$ / PVDF / Hollow fiber	Hydrophobic	Giacobbo et al. (2017a)
Phenolic compounds	>90 %	Racking wine lees	Integrated membrane process:			Giacobbo et al. (2017b)
			UF	10 kDa/Fluoropolymer/Tubular	Hydrophilic	
			UF	1000 Da/ Fluoropolymer /Tubular	Hydrophilic	
			NF	200-300 Da/Polypiperazine/Tubular	Hydrophilic	
	57 %	Olive mill wastewaters	UF	3 kDa/Regenerated cellulose/Flat sheet	Hydrophilic	Ochando-Pulido & Martínez-Férez (2017)
Phenolic compounds	97-98 %	Apple pomace extract	NF	150-300 Da/Polyamide thin film composite/Spiral wound	Hydrophilic	Uytbroek et al. (2018)
Quinic acid	92 %	Apple pomace extract	NF	150-300 Da/Polyamide thin film composite/Spiral wound	Hydrophilic	
Catechin	78 %	Apple pomace extract	NF	150-300 Da/Polyamide thin film composite/Spiral wound	Hydrophilic	
Epicatechin	87 %	Apple pomace extract	NF	150-300 Da/Polyamide thin film composite/Spiral wound	Hydrophilic	



**Table 3.** Overview of the latest uses of MD for the concentration of juices.

<i>Type of application:</i>	<i>Membrane configuration:</i>	<i>Pretreatment step:</i>	<i>Operating conditions:</i>	<i>Highlighted inputs:</i>	<i>MD configuration:</i>	<i>Membrane material:</i>	<i>Reference:</i>
Apple juice concentration	Flat sheet	Enzyme treatment, UF as prefiltration	Feed: 50-70°C Permeate:10-30°C	Maximum concentration for solids content of 50%.	DCMD	PVDF	(Gunko et al., 2006)
Sugarcane concentration	Flat sheet	-	Feed: 75°C Permeate:25°C	Continuous removal of water (10 kg/m <sup>2</sup> h).	DCMD	PP	(Nene et al., 2002)
Orange juice concentration	Flat sheet	Prefiltration	Feed: 40-70°C Permeate:20-30 °C	Increase in feed flow rate reduces concentration polarization and fouling phenomenon.	DCMD	PTFE	(Deshmukh et al., 2011)
Aroma recovery	Flat sheet	UF as pre-filtration	Feed: 10-45°C Pressure:0.7-3 kPa	Recovery of highly volatile aroma compounds (ranged from 68 to 83%) from black-currant juice.	VMD	PTFE	(Bagger-Jørgensen et al., 2004)
Sucrose concentration	Hollow fiber	-	Feed:70°C Permeate:100 kPa	Maximum sucrose concentration of 50°Brix (from a feed starting solution of 10°Brix solution).	VMD	alumina	(Chen et al., 2018)
Black-currant juice concentration	Hollow fiber	Enzyme treatment MF as pre-filtration RO as pre-	Feed:26-30°C Permeate:11°C	Concentration from 22 up to 58.2 °Brix.	DCMD	PP	(Kozák et al., 2009)

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		concentration					
Orange juice concentration	Hollow fiber	UF as pre-filtration	Concentration from 24°Brix up to 65 °Brix.	DCMD	PP	(Quist-Jensen et al., 2016)	

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Vacuum membrane distillation (VMD), direct contact membrane distillation (DCMD).

Journal Pre-proof

## Highlights

Membrane-based technologies as emerging tool to recovering functional molecules.

Tight UF and NF membranes as the high efficiency extraction of phenolic molecules.

Membrane distillation assists the concentration of valuable molecules.

Pervaporation meets the requirements for the selective extraction of aromas.