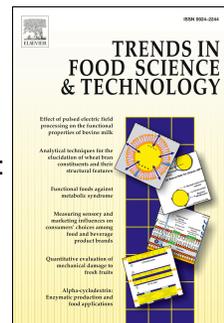


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Membrane technologies assisting plant-based and agro-food by-products processing:
A comprehensive review

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

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19

20

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 (Δp) 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 μm . 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 (L h^{-1}) is the volumetric flow rate of permeate and A (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 μm . 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, α a factor
326 whose value is 1 for Knudsen diffusion and 2 for viscous fluxes, respectively, δ_m
327 the membrane thickness, ε the membrane porosity and τ 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 μm and
394 a membrane surface area of 0.1 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⁻² h⁻¹ (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, δ 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 (Isci 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 (β) 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 μm 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 m s^{-1} and 1.0 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.

731

732

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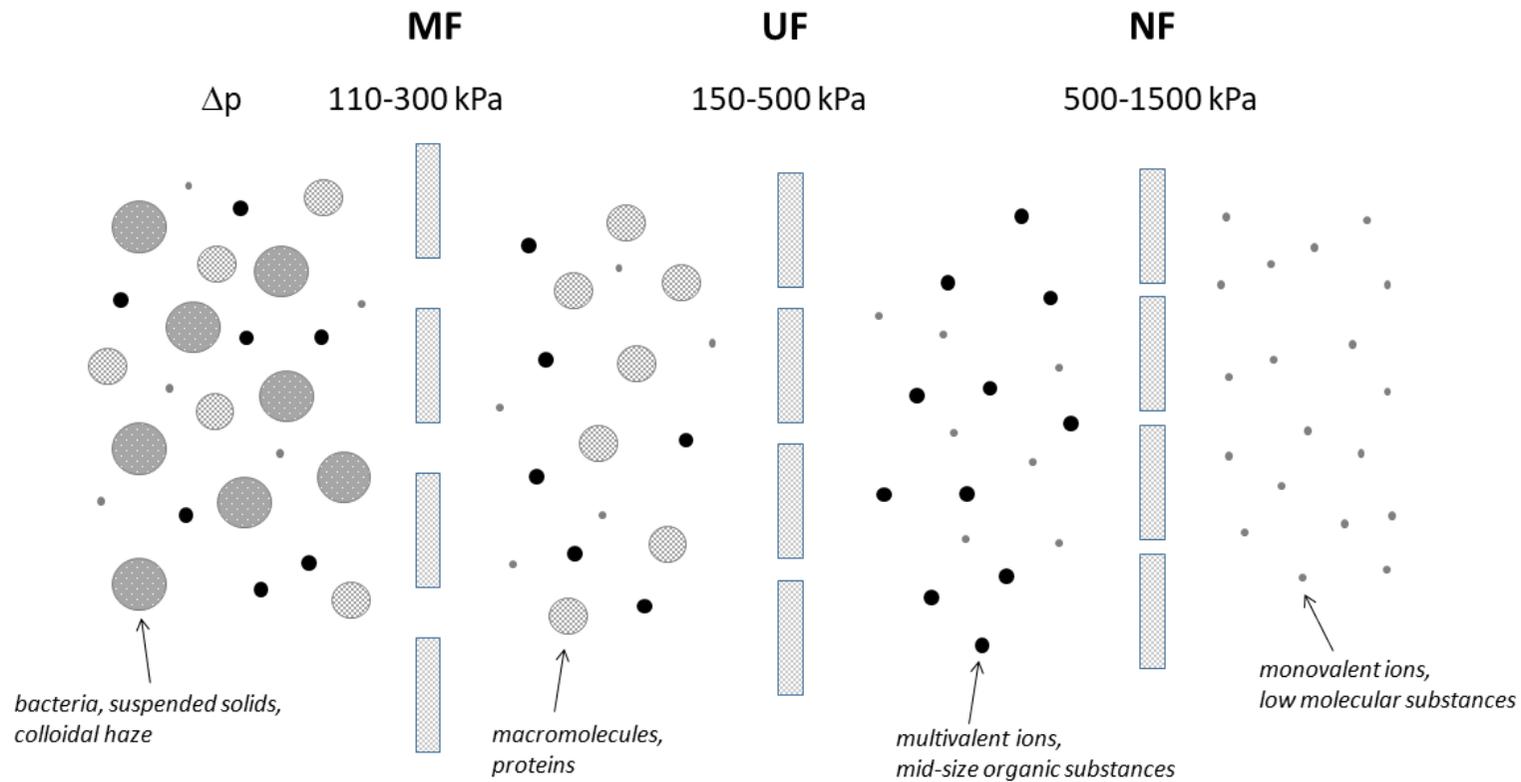
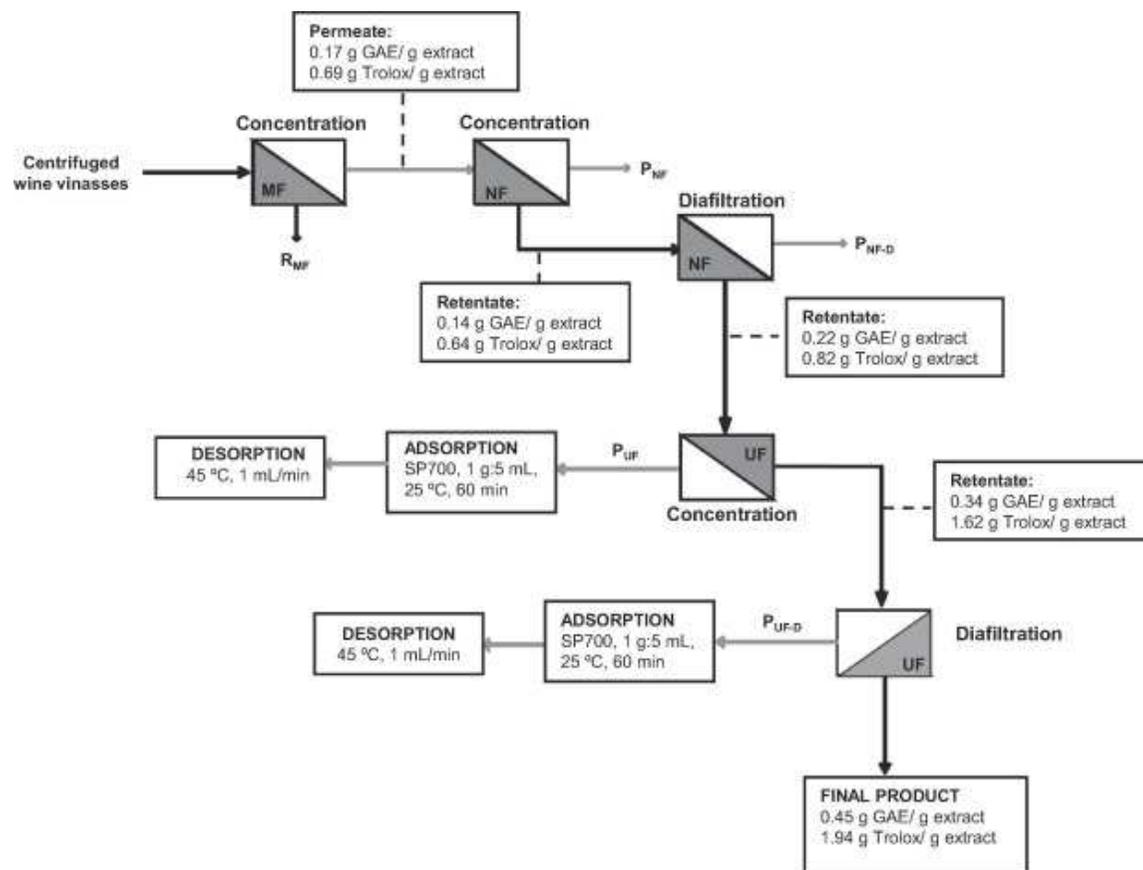
Figure 1. General description of pressure-driven membrane process.

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.

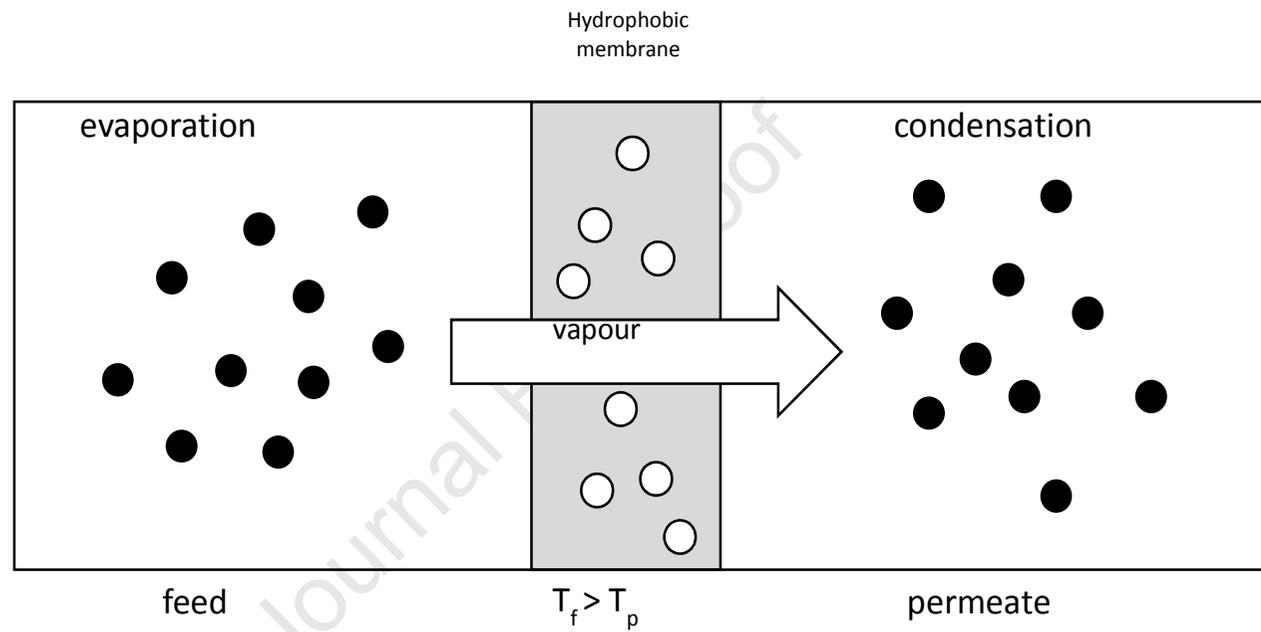


Figure 4. Concentration profile of blood orange juice by DCMD. (a) evaporation flux (thermal gradient at time 0, 9, 18 and 27 h), (b) total soluble solids content and (c) viscosity as a function of operating time (Quist-Jensen et al., 2016).

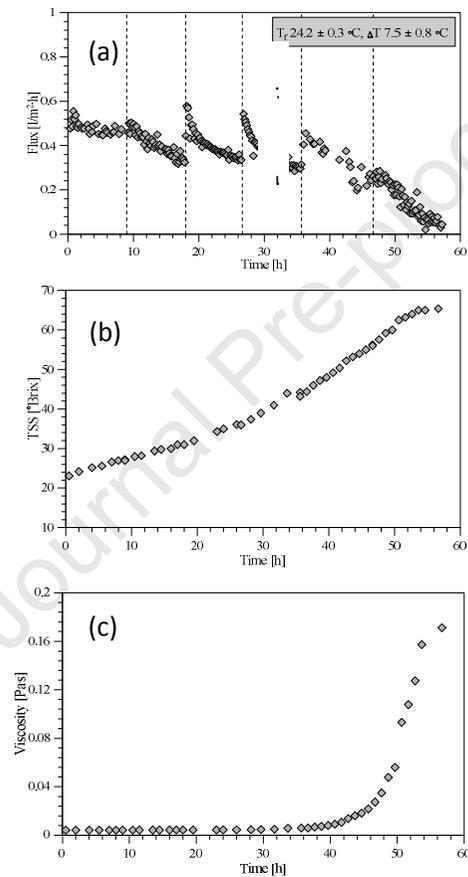


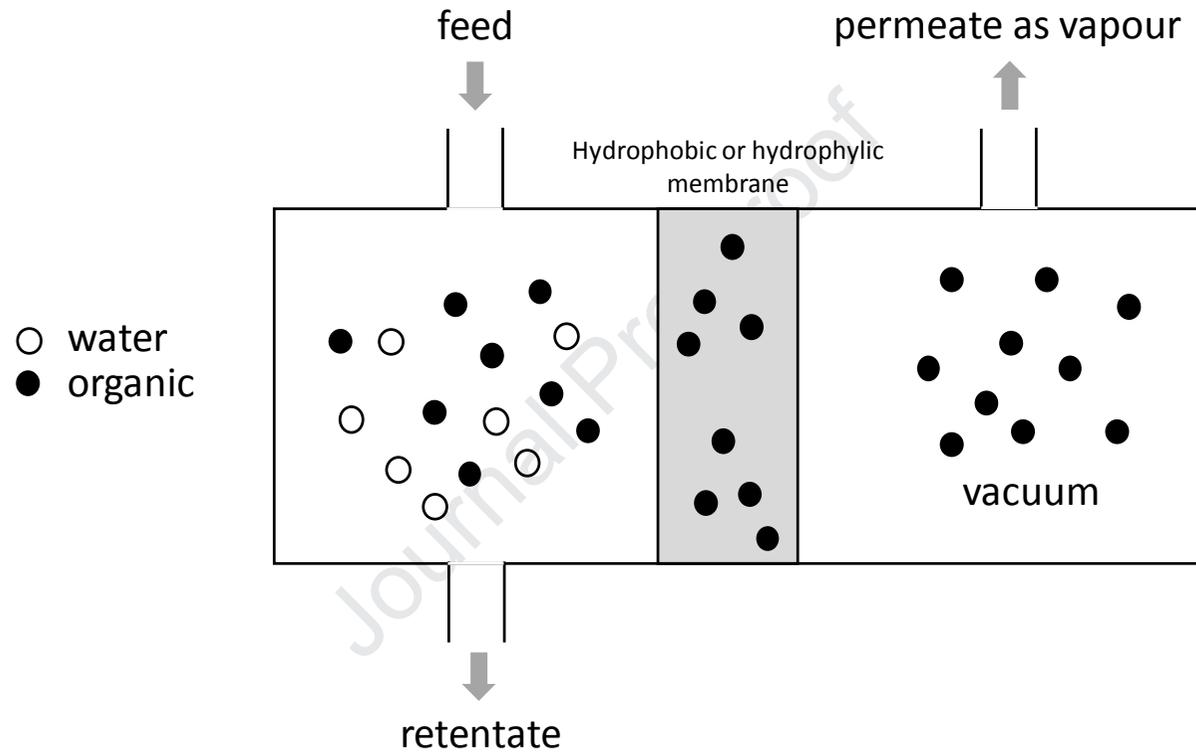
Figure 5. General schematic of a pervaporation process.

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

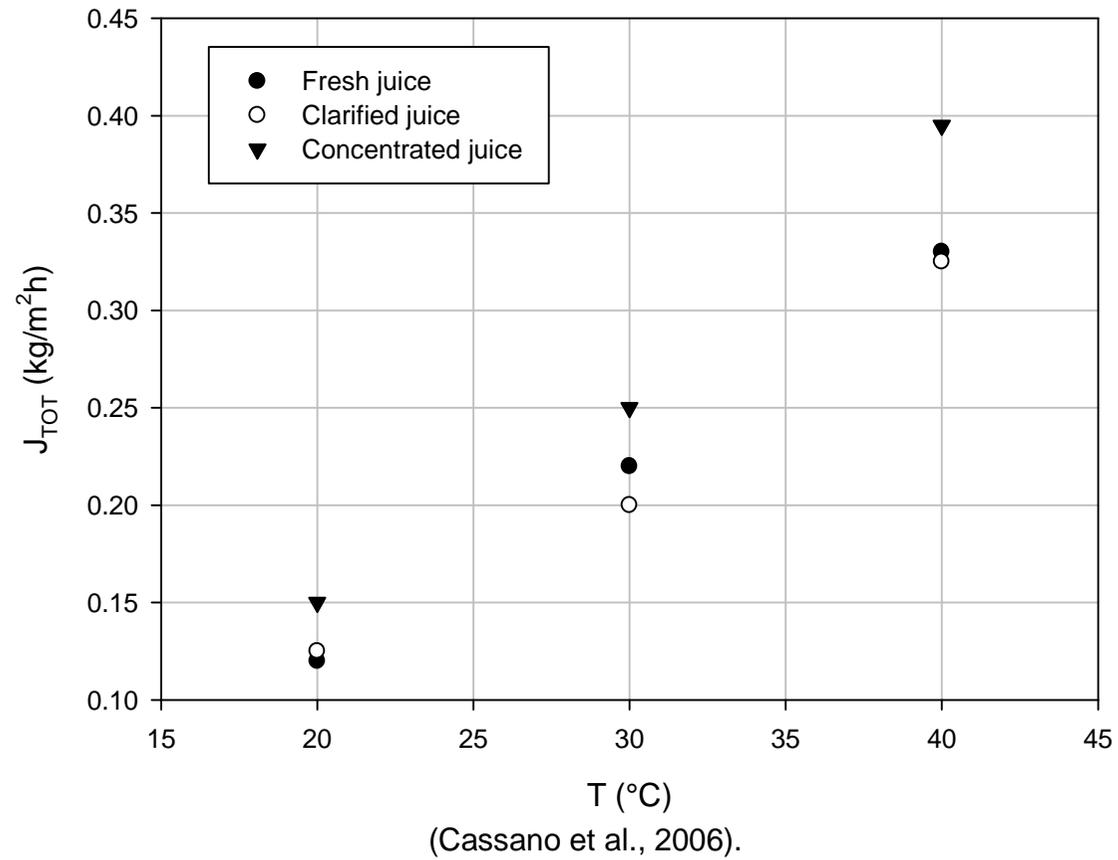


Table 1. Main characteristics of MF, UF, NF, MD and PV processes.

Membrane process	Driving force	Mass transfer mechanism	Membrane	Water permeation flux (L/m ² 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 %	Fermented grape pomace	UF	1000 Da / Thin-film / Spiral wound	Hydrophilic	Díaz-Reinoso et al. (2009); Díaz-Reinoso et al. (2010)
	> 80 %		UF	1000 Da / Ceramic (titania) / Tubular	Hydrophilic	
	> 30 %		NF	250 Da / Polyamide-polysulfone / Spiral wound	Hydrophilic	
	> 60 %		NF	350 Da / Polyamide-polysulfone / Spiral wound	Hydrophilic	
Hydroxytyrosol, protocatechuic acid, caffeic acid, tyrosol and p-cumaric acid	> 80 %	Olive mill wastewaters	NF	150-300 Da / Thin-film / Spiral wound	Hydrophilic	Cassano et al. (2011)
	48.3 %		UF	4 kDa / polyethersulphone / Flat sheet	Hydrophobic	
Hydroxycinnamic acids, o-diphenols	8.7 %	Olive mill wastewaters	UF	10 kDa / Regenerated cellulose / Flat sheet	Hydrophilic	Galanakis et al. (2013)
	33.5 %		UF	10 kDa / Polyethersulphone / Flat sheet	Hydrophobic	
	81 %		UF	100 kDa / Polysulfone / Flat sheet	Hydrophobic	
3,4-DHPEA, p-HPEA, 3,4-DHPEA-EDA, verbascoside, and total phenols	77 %	Winery sludge from red grapes	UF	20 kDa / Polysulfone / Flat sheet	Hydrophobic	Servili et al. (2011)
	56 %		UF	1 kDa / Composite fluoropolymer / Flat sheet	Hydrophobic	
	--		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 µm / 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 ₂ O ₃ / 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 ₂ SO ₄ 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 μm / Polyimide / Hollow fiber	Hydrophilic	Giacobbo et al. (2015)
	5 %	Winery effluents	MF	0.2 μm / 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 ² 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)



		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)

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.