REVIEW ARTICLE



# Pervaporation-based membrane processes for the production of non-alcoholic beverages

Roberto Castro-Muñoz<sup>1</sup>

Revised: 18 March 2019 / Accepted: 26 March 2019 / Published online: 10 April 2019 © Association of Food Scientists & Technologists (India) 2019

Abstract Nowadays, the interest in manufacturing nonalcoholic or low alcoholic content beverages from alcoholic beverages is a current challenge for food technologists; this is due to the fact that huge consumption of alcoholic beverages may produce health problems in the costumers. In principle, the post-fermentation ethanol removal from alcoholic beverages is carried out by means of evaporation or distillation. Such current dealcoholization methodologies are efficiently removing the ethanol, however, some organoleptic compounds can also be lost during the process. This makes the dealcoholization process highly sensitive in order to preserve the quality properties of the beverages. Thereby, membrane-based technologies, which use perm-selective barriers for the separation, have been highly promoted for such purpose. Pervaporation (PV) technology is indeed one of these technologies aimed for ethanol removal. Herein, the goal of this review is to provide a compelling overview of the most relevant findings for the production of non-alcoholic beverages (such as beer and wine) by means of PV. Particular attention is paid to experimental results which provide compelling feedback about the accurate ethanol removal and minimal changes on physicochemical properties of the beverages. Moreover, some theoretical basis of such technology, as well as key criteria for a more efficient dealcoholization, are also given.

Keywords Non-alcoholic beverages - Dealcoholization - Membranes - Pervaporation - Ethanol removal

#### Introduction

As it is well-known, the alcoholic beverages consumption has extremely increased according to the World Health Organization (WHO) (WHO [2011](#page-10-0)). Indeed, the current global consumption of alcoholic beverages is around 54.2 billion liters per year (Statista [2018\)](#page-10-0) that includes mainly beer and wine products, this consumption is expected to increase in coming years (Solov [2018](#page-10-0)). Typically, several types of drinks can be found in the category of alcoholic beverages, such as beer, spirits, wines (e.g. fortified wines, rice wine or other fermented beverages based on sorghum, millet, maize) and some other traditional high-alcohol content beverages (Cleophas [1999](#page-9-0)); among all these products, the highest consumption regards to the beer and wine. However, in the last decades, the market for nonalcoholic beverages, especially for beer and wine, has increased its demand based on the regulations to prevent health issues. It is well documented that high and excessive consumption of alcoholic beverages may result in different types of diseases, such as pancreas cancer, pancreatitis, hepatitis, fatty degradation of liver, cirrhosis, peptic ulcers, allergenic induction, increase of uric acid in plasma, promoting obesity and some other derivative harmful effects; especially, pancreatitis and cirrhosis in their acute form are frequently caused by high huge consumption of alcoholic beverages (Costanzo et al. [2010](#page-9-0); Partanen et al. [1997](#page-10-0); Sohrabvandi et al. [2012](#page-10-0)). In fact, these diseases are directly attributed to the alcohol content of such beverages. On the other hand, such beverages can also provide some positive benefits into human health related to their nutritional benefits, including hypolipidemic effect, anti-mutagenic and anti-carcinogenic effects, reduction of cardiovascular disease (cardioprotective effect), immune system

<sup>&</sup>amp; Roberto Castro-Mun˜oz food.biotechnology88@gmail.com; castromr@vscht.cz

University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6, Czech Republic

stimulation, anti-osteoporosis effect, and reducing risk of dementia (Sohrabvandi et al. [2012](#page-10-0)).

The production of non-alcoholic beverages can be carried out by altering the fermentation process (Purwasasmita et al. [2015](#page-10-0)), or by using special and immobilized yeasts (Strejc et al. [2013](#page-10-0)); unfortunately, to break the fermentation process restricts the production of some desirable compounds. In fact, at the brewery industry, the fermentation step is fundamental for the flavor formation and quality control of beers (Olaniran et al. [2017](#page-10-0)). Thereby, this practice is not so recommendable alternative for the production of non-alcoholic beverages, At this point, it is needed to carry out the ethanol removal after the complete beverage fermentation step; this may imply a dealcoholization through a downstream process.

The dealcoholization of beverages is a promising alternative for the production of non-alcoholic drinks which can preserve most of the organoleptic and nutritional values of the original beverages. Basically, the dealcoholization process mainly involves physical separation processes based on different driving force, such as (1) temperature (e.g. evaporation and distillation) (Andre´s-Iglesias et al. [2016;](#page-9-0) Belisario-Sánchez et al. [2012\)](#page-9-0), (2) pressure in which different membrane-based processes can be found (e.g. pervaporation (PV), nanofiltration, reverse osmosis), (3) pressure and temperature simultaneously (e.g. PV, membrane distillation), and (4) concentration (e.g. dialysis, membrane contactors, osmotic distillation, and diafiltration). All these membrane processes have been widely sought by food research community according to their advantages over conventional dealcoholization procedures (Lipnizki [2014](#page-10-0); Mangindaan et al. [2018\)](#page-10-0). Importantly, membrane-based processes are becoming emerging alternatives for the treatment and processing of food systems (Cassano et al. [2018;](#page-9-0) Castro-Muñoz et al. [2017,](#page-9-0) [2018a](#page-9-0); Castro-Muñoz and Fíla [2018](#page-9-0); Galanakis [2013](#page-9-0); Galanakis et al. [2015\)](#page-9-0), as well as the recovery of high-added value compounds from natural food products (Castro-Muñoz et al. [2016](#page-9-0), [2018b](#page-9-0); Galanakis [2013](#page-9-0), [2015;](#page-9-0) Galanakis et al. [2015\)](#page-9-0). In particular, PV is a potential candidate for efficient and selective ethanol removal from alcoholic beverages. Thereby, the aim of this review paper is to provide a perspective about the literature findings for the dealcoholization of alcoholic beverages (e.g. beer and wines) by means of PV, aiming the production of non-alcoholic beverages.

Particular attention is paid to experimental results which provide compelling ethanol removal and its effect on the physicochemical properties of the beverages. Moreover, some theoretical basis of such technology, as well as key criteria for a more efficient and selective dealcoholization, are also given.

## Ethanol removal using pervaporation: theoretical background and key features

Pervaporation (PV) is considered as a suitable and effective membrane-based technology to perform the separation of similar boiling points components contained in an ''azeotropic mixture'', where phase change from liquid to vapor takes place. Today, PV is considered an alternative to traditional processes, such as simple distillation, vacuum distillation, fractional distillation and steam distillation (Afonso et al. [2015\)](#page-9-0). Table [1](#page-2-0) displays the most relevant advantages and disadvantages of PV technology, highlighting that its low energy consumption and non-use of additional solvents encourage its use (Figoli et al. [2015\)](#page-9-0).

In principle, a binary or a multi-component liquid mixture is separated by partial vaporization using a dense non-porous membrane. The liquid feed mixture is in direct contact with the ''selective'' side of the membrane, while the permeate (collected at the other side of the membrane) is in a vapor phase, enriched by the species with a higher affinity with the membrane (hydrophilic or hydrophobic type). In theory, the transport of the permeating species takes place due to the driving force applied, which could be (1) vacuum or (2) sweeping gas (like nitrogen) and (3) temperature, and then condensed and recovered. Indeed, PV is combining two different separation processes, such as ''permeation'' and ''evaporation'' (Crespo and Brazinha [2015](#page-9-0)). The transport mechanism through dense polymeric membranes is described by the so-called solution-diffusion model (Crespo and Brazinha [2015;](#page-9-0) Wijmans and Baker [1995](#page-10-0)), which comprises three main steps: (1) adsorption of the target component from the mixture to the membrane "selective" layer on the basis of its chemical affinity, (2) diffusion of the target component through the membrane as a result of the concentration gradient, (3) desorption of the component at the permeate side of the membrane (Wee et al. [2008](#page-10-0)). The mass transport is governed by the chemical potential  $(\mu_i)$  gradient, the physical properties of the permeating component (1) and its concentration in feed and permeate side.

To date, PV has been efficient in separating different types of water-organic, organic–organic and organic–water mixtures (Castro-Muñoz et al. [2018b,](#page-9-0) [c](#page-9-0)). Importantly, the membrane material plays an important role in the separation by PV, as Fig. [1](#page-2-0) depicts.

The alcohol removal crucially comprises the use of nonporous hydrophobic membranes, which are able to remove organics (non-polar or less polar compounds), such as alcohols (e.g. ethanol, propanol, isobutanol, and isoamyl alcohol), aldehydes (e.g. acetaldehyde) and esters (e.g. ethyl acetate and isoamyl acetate). Indeed, PV finds its main application in food and cosmetic industries for the

<span id="page-2-0"></span>





Fig. 1 General drawing of hydrophilic and hydrophobic membranes for PV applications

extraction of aroma compounds (Lipnizki et al. [2002](#page-10-0); Raisi et al. [2009;](#page-10-0) Saffarionpour and Ottens [2018](#page-10-0)). When dealing with the dealcoholization of beverages, this implies an organic–water separation, it means, removing the ethanol from a complex aqueous solution. Actually, PV uses highly selective membranes removing the minor component in the mixture only, i.e. the ethanol is commonly the minor compound in alcoholic beverages.

To date, several hydrophobic nature polymers for the preparation of membranes have been proposed for the ethanol removal in aqueous solution models (Castro-Muñoz et al.  $2018c$ . Table [2](#page-3-0) displays some of these hydrophobic membranes and their performance.

As can be seen, polydimethylsiloxane (PDMS) based membranes have been the most sought for such separation. In fact, hydrophobic membranes are also well recognized as organophilic, for their ability to remove organics. Generally, the membranes cannot display high productivities in terms of permeate flux; however, they were highly selective displaying selectivity values between 2–15 towards ethanol, whereas the total fluxes were considerably low  $(0.060 - 3.3 \text{ kg/m}^2/\text{h})$ . At this point, polyether block amide (PEBA) and poly [1-(trimethylsilyl)-1-propyne] (PTMSP) polymers provide higher total fluxes. On the other hand, it is likely that membranes based on polybenzoxazines (PBZ) and butyl acrylate-styrene copolymer tend to provide better selective properties, having selectivities of about 10,000 and 16, respectively. Of course, as in all membrane-based technologies, several factors are playing an important role on the separation performance of membranes, such as operating conditions (e.g. temperature, vacuum pressure, feed flow rate, feed concentration) and membrane properties (e.g. nature, thickness) (Baker et al. [2010\)](#page-9-0). For instance, the permeation flux of ethanol tends to increase usually with the increase of feed concentration, similar behavior has been seen for the selectivity as well (Gu et al. [2009](#page-9-0)). This is because the flux of organic solvents (e.g. ethanol, butanol, acetone) is a result of increasing of the activity and partial pressure, hence enhancing the driving force for permeation, and thus allows higher fluxes (Zhou et al. [2011\)](#page-11-0). Figure [2](#page-4-0) shows clearly the effect of feed ethanol concentration on the performance of a hydrophobic membrane during the removal of ethanol.

On the other hand, the increase in temperature also enhances the permeation rates of polymeric membranes; however, the selectivity usually decreases. Temperature indeed causes a polymer chains motion in membranes; this promotes the permeation of other molecules across the membranes, and thus reducing the selective membrane properties (Baker et al. [2010\)](#page-9-0).

Feed concentration	Polymeric membrane Manufacture		Operating conditions	Total flux values $(kg/m^2/h)$	Selectivity (separation factor)	References
5 wt $%$ ethanol	<b>PEBA</b>	Homemade	40 °C Vacuum pressure $1.5$ mbar.	0.70	3.0	Gu et al. (2009)
$10 \text{ wt}\%$ ethanol	<b>PDMS</b>	Homemade	40 °C Vacuum pressure	0.50	5.0	Huang et al. (2009)
$10 \text{ wt\%}$ ethanol	<b>PTMSP</b>	Homemade	13 mbar. 50 °C Vacuum pressure	3.5	12	Claes et al. (2010)
10 wt% ethanol	Pervatech PDMS	Pervatech BV (The Netherlands)	$0.04$ mbar. 50 °C Vacuum pressure	3.3	6	
10 wt% ethanol	PERVAP <sub>4060</sub>	Sulzer ChemTech (Switzerland)	$0.04$ mbar. 50 °C Vacuum pressure	1.9	7	
5 wt $%$ ethanol	Pebax	Homemade	$0.04$ mbar. $25~^{\circ}\mathrm{C}$ Vacuum pressure	0.080	2.5	Le et al. $(2011)$
1 wt $%$ butanol $0.5 \text{ wt\%}$ acetone $0.15$ wt%	<b>PDMS</b>	Homemade	$0bar$ . $50 °C$ Vacuum pressure $2.8$ mbar.	0.275	10	Zhou et al. (2011)
ethanol 5 wt $%$ ethanol	Cross-linked PDMS	Homemade	40 $\degree$ C	0.080	8.5	Zhan et al. (2012)
5 wt $%$ ethanol	<b>PDMS</b>	Homemade	$60^\circ$ Vacuum pressure 0 bar	0.060	15	Sun et al. (2013)
4 wt $%$ ethanol	<b>PDMS</b>	Homemade	25 °C Vacuum pressure 20 mbar	0.100	8	Yadav et al. (2013)
5 wt $%$ ethanol	<b>PDMS</b>	Homemade	50 °C Vacuum pressure 330 mbar	0.800	5	Li et al. (2014)
5 wt $%$ ethanol	<b>PDMS</b>	Homemade	50 °C Vacuum pressure 2 mbar	0.090	5	Liu et al. $(2015)$
5 wt $%$ ethanol	<b>PDMS</b>	Homemade	$70~\mathrm{^{\circ}C}$ Vacuum pressure $10.1$ mbar	1.5	7.5	Zhang et al. (2015)
5 wt $%$ ethanol	Butyl acrylate- styrene copolymer	Homemade	30 °C Vacuum pressure 1.3 mbar	0.300	16	Samanta and Ray (2015)
$10 \text{ wt}\%$ ethanol	PBZ	Homemade	70 °C Vacuum pressure 13 mbar	0.035	10,000	Chuntanalerg et al. (2016)
6 wt% ethanol	<b>PDMS</b>	Homemade	40 °C Vacuum $pressure < 1$ mbar	0.900	8.7	Naik et al. (2016a, b)
$6\%$ wt $\%$ ethanol	<b>PDMS</b>	Homemade	40 °C Vacuum $pressure < 1$ mbar	0.35	8	Naik et al. (2016a, b)

<span id="page-3-0"></span>Table 2 Hydrophobic membranes tested for the ethanol recovery by PV. Adapted from Castro-Muñoz et al. ([2018c\)](#page-9-0)

<span id="page-4-0"></span>





Fig. 2 Effect of feed ethanol concentration on the pervaporation performance of a hydrophobic membrane. Taken from Baker et al. ([2010\)](#page-9-0)

#### Non-alcoholic beverage production through PV

#### Nutritional value of alcoholic beverages

This review addresses the most consumed beverages worldwide, such as beer and wine that have been dealcoholized by PV. In principle, the dealcoholization process seems to be an easy task; however, the organic compounds (related to nutrients) contained in such products make difficult the ethanol removal. For instance, the beer contains several nutritional compounds, including proteins, phenolics (e.g. antioxidants), certain minerals, anthocyanins, dietary fibers, some prebiotic compounds and B vitamins (Sohrabvandi et al. [2010a](#page-10-0), [b,](#page-10-0) [2012](#page-10-0)). Table 3 shows the most common nutrients that can be found in beer.

Moreover, a beer contains multiple desirable organoleptic compounds (e.g. alcohols, esters, carbonyl compounds, vicinal ketones) (Olaniran et al. [2017](#page-10-0)); finally, beer contains more protein and B vitamins than wine (Sohrabvandi et al. [2012\)](#page-10-0); however, the wine, as the main winemaking product, contains much more bioactive compounds (e.g. polyphenols, anthocyanins) than beer, e.g. red

Table 3 Nutrients contained in a normal beer. Adapted from Sohrabvandi et al. ([2012\)](#page-10-0)

<b>Vitamins</b>	Minerals		
Biotin $(2-7 \mu g/L)$	Calcium $(25-120 \text{ mg/L})$		
Vitamin $B_{12}$ , $B_6$ (5–10 mg/L)	Phosphorus $(11 \text{ mg/L})$		
Vitamin A, C, D, E, K $(< 0.06 \mu g/L)$	Magnesium $(50-90 \text{ mg/L})$		
Niacin $(16 \text{ mg/L})$	Potassium $(200-450 \text{ mg/L})$		
Folate $(20 \mu g/L)$	Sodium $(20-350 \text{ mg/L})$		
Thiamine $(0.2-7 \text{ mg/L})$	Iron $(1-5 \text{ mg/L})$		
Riboflavin $(1-61 \text{ mg/L})$	Zinc $(0.07-12 \text{ mg/L})$		
	Selenium $(0.7-13 \mu g/L)$		

wines contain about 1720 mg polyphenols  $L^{-1}$  (Lugasi and Hóvári [2003](#page-10-0)), which definitely provide biological benefits to the costumers, this is because such compounds are wellrecognized for their associated antioxidant capacity (Car-tron et al. [2003](#page-9-0); Paixão et al. [2007](#page-10-0)). Certainly, minimal content of phenolic compounds have been quantified in beers compared to wine (Bartolomé et al. [2000](#page-9-0)); however, more than 35 phenolic compounds (e.g. tyrosol, ferulic acid, HMF, trytophol) can be found in the beer (about 80–90% from malt and 10–20% from hops) (Sohrabvandi et al. [2010a,](#page-10-0) [b](#page-10-0)); whereas a huge range of phenolic compounds are contained in wines, such as catechin, epicatechin, p-coumaric acid, caffeic acid, gallic acid, fertaric acid, to mention just a few (Paixão et al. [2007](#page-10-0)). Table [4](#page-5-0) displays the main phenolic compounds that have been quantified in the wine:

The variety of phenolic compounds indeed depends on the kind of wine (e.g. red, white, sparkling, sherry, port, fruit, and brandy). Particularly, red wine is a complex mixture of flavonoids (e.g. anthocyanins and flavan-3-ols) and nonflavonoids (e.g. resveratrol, cinnamates, and gallic acid), while flavan-3-ols are the most abundant together with polymeric procyanidins composing up to 50% of the total phenolic constituents (Guilford and Pezzuto [2011](#page-9-0)). It has been reported that phenolic compounds provided by beer and wine are able to be absorbed and extensively metabolized by the human body (Nardini et al. [2006](#page-10-0), [2009\)](#page-10-0). Thanks to this, beneficial effects have been attributed to the consumption of beer and wine, such as anti-mutagenic and anti-carcinogenic effects,

Phenolic compound	Concentration (mg/L)			
Catechin	89			
Epicatechin	94			
Dimer B1	69			
Dimer B <sub>2</sub>	54			
Dimer B3	79			
Dimer B4	96			
Trimer C1	41			
Trimer <sub>C2</sub>	74			
Gallic acid	43			
Protocatechuic acid	12			
Caftaric acid	55			
Gentisic acid	54			
Caffeic acid	8			

<span id="page-5-0"></span>Table 4 Phenolic compounds contained in red wine. Adapted from Cartron et al. ([2003\)](#page-9-0)

cardioprotective effect, immunomodulation, and anti-osteoporosis effect (Sohrabvandi et al. [2012](#page-10-0)). Finally, the balance between alcohol and phenolic compounds in wine and beer may be critical in determining their antioxidant potential due to the fact that the alcohol displays pro-oxidant effects (van Golde et al. [1999\)](#page-10-0). Clearly, the ethanol removal from such beverages needs a highly selective technique, like pervaporation, to separate the alcohol without altering strongly the nutritional properties. The next section provides the state of the art of the literature findings in the field.

### Non-alcoholic beverages production through pervaporation

Through traditional techniques, the dealcoholization pretends the ethanol removal from alcoholic beverages (Lipnizki [2014\)](#page-10-0), without altering the sensory and nutritional properties of the original products. However, unfortunately, some of the valuable compounds can be considerably lost. For instance, Table [5](#page-6-0) shows a comparison of the polyphenols profile and associated substances in alcoholfree and standard beers.

Importantly, such non-alcoholic beers are nowadays produced by means of different approaches in order to suppress the alcohol production in the beer (Lehnert et al. [2009;](#page-10-0) Schmidtke et al. [2012](#page-10-0); Sohrabvandi et al. [2010a](#page-10-0), [b](#page-10-0); Strejc et al. [2013](#page-10-0)), such as:

- the use of special strains of fermenting yeasts,
- reducing fermentable fractions to nonfermentable fractions,
- reducing glucose content in the wort,
- heating of fermenting wort,
- high-temperature mashing,
- pressurization during fermentation,
- cold contact procedure, and
- periodic aeration of fermenting wort.

While the vacuum evaporation and vacuum distillation are used as post-fermentation technologies for removing out the ethanol (Andrés-Iglesias et al. [2015](#page-9-0); Blanco et al. [2016](#page-9-0); Lipnizki [2014\)](#page-10-0). All these techniques and procedures are definitely producing changes in nutritional value as well as the sensorial characteristics of the beverages (Brányik et al. [2012\)](#page-9-0). This is, therefore, the main reason that makes to the industry of looking for highly selective techniques to carry out the dealcoholization. According to Sohrabvandi et al. [\(2010a,](#page-10-0) [b\)](#page-10-0), several techniques are currently being proposed for the dealcoholization of beverages (e.g., beers and wines), such as vacuum distillation, water vapor-/gas-stripping under vacuum, adsorptive alcohol removal, dialysis, reverse osmosis, and osmotic distillation; while some of membranes processes, like dialysis, and reverse osmosis, are already industrial methods for beverage dealcoholization (especially beer).

In the case of pervaporation (PV) has not been widely studied for such purpose, but it has been proved to be a potential candidate for the partial dealcoholization of beverages (Blanco et al. [2016](#page-9-0)). For instance, Catarino et al. [\(2009](#page-9-0)) developed a process to extract aromas from an original beer through a polyoctylmethylsiloxane membrane. Several compounds were identified in beer, such as alcohols (e.g. ethanol, propanol, isobutanol, and isoamyl alcohol), esters (e.g. ethyl acetate and isoamyl acetate) and aldehydes (e.g. acetaldehyde). Similarly, Olmo et al. [\(2014](#page-10-0)) also demonstrated that PV meets the requirements for the recovery of aroma compounds from beer, e.g. isobutyl alcohol increased up to16% in the in alcohol-free beer, while ethyl acetate in low-alcohol beer increased up to 35.72%. In this sense, the extraction of beer aromas could be useful to adjust the aroma profile of the dealcoholized beers, which can, unfortunately, be lost during the processing.

Regarding the beverage dealcoholization by PV, Catarino and Mendes [\(2011b](#page-9-0)) investigated the production of low-alcohol content beer by means of an industrial plant. Basically, the plant involved a hybrid process, which included the integration of PV and distillation units, as Fig. [3](#page-6-0) shows.

The aroma compounds (e.g. amyl alcohol, ethyl acetate, isoamyl acetate, and acetaldehyde) were recovered using a PV polyoctylmethylsiloxane membrane (organophilic) (Catarino and Mendes [2011b](#page-9-0)). Such aroma compounds were then incorporated into the dealcoholized beer <span id="page-6-0"></span>Table 5 Comparison of the polyphenols content and its derivatives in alcohol-free and standard commercial beers. Adapted from Bartolomé et al.

([2000\)](#page-9-0)



(produced by conventional distillation). In such a way, this industrial methodology allowed to obtain a non-alcoholic beer containing less than 0.5 vol.% ethanol, and also a good flavor profile. It is important to mention that PV technology does not require in principle temperature to carried out the separation; nonetheless, the use of temperature may help to improve the permeate fluxes with a possible effect on components selectivity. At this point, the PV process at room temperature could help to avoid any compound degradation, as well as contribute to obtain higher selective performance.

As introduced previously, the wine has been also dealcoholized by membrane-based technologies, to produce non-alcoholic wine. Since wine contains more nutritional compounds than beer, the post-fermentation removal of ethanol has to be carefully performed, minimizing any possible degradation of bioactive compounds (e.g. polyphenols, antioxidants, etc.) (Gómez-Plaza et al. [1999](#page-9-0)); actually, wine also contains higher ethanol content compared to the beer. To date, different types of wines have been dealcoholized by means of PV; for instance, Tan et al. [\(2003](#page-10-0)) used a PV-PDMS membrane for wine dealcoholization. The dealcoholization process was conducted at 40 °C. The process was able to produce wine with  $3-7\%$  of ethanol while the average permeate flux was  $1.5 \text{ kg/m}^2/\text{h}$ . This flux is indeed in agreement with reported values for ethanol removal using PDMS membranes in diluted ethanol solutions (see Table [2](#page-3-0)).





Takács et al. ([2007\)](#page-10-0) reported the dealcoholization of semi-sweet Tokaji Harslevelu type wines (alcohol content 13.11% ABV) using pervaporation PERVAP membranes. Such commercial membranes are well recognized by its organophilic nature. The authors reported that temperature influences in PV performance in terms of flux and separation ability, e.g. higher permeate fluxes were commonly obtained at higher temperatures; whereas the ethanol selectivity of the membranes tended to decrease. No details about the physico-chemical properties of the dealcoholized wines were provided. On the other hand, Catarino and Mendes ([2011a\)](#page-9-0) used another PV organophilic (polyoctylmethylsiloxane supported in polyetherimide) membrane to recover the aroma compounds from wine. Afterward, the aroma compounds were added back to the dealcoholized wines, which was partially dealcoholized by means of nanofiltration (NF). Herein, the authors tested four different NF membranes (same molecular weight cutoff of 200 Da), which were manufactured by different membrane materials. In general, the integrated membrane processes were able to provide a high-quality and low alcohol content wine (ca. 7–8 vol.%) from a standard alcoholic wine (ca. 12 vol.% of ethanol).

In a different approach but still using integrated membrane-based systems, Salgado et al. ([2017\)](#page-10-0) proposed PV and NF steps for the preparation of flavored white wines (see Fig. [4\)](#page-8-0), which contained low alcohol contents.

The production process basically involved two NF processes for the sugar reduction of must, while PV was used for the aroma recovery. Such a procedure allowed to obtain a wine containing a final alcoholic degree between 10.2–10.5 vol.%. The conventional production commonly produces white wine containing ca. 12 vol.% ethanol concentration, it means, a small reduction in the alcohol content was observed; but definitely, a concentration of the aroma compounds has been noticed (Salgado et al. [2017](#page-10-0)). Particularly, this commercial organophilic (PERVATECH PV-SR1 spiral wound) membrane displayed low total flux  $(0.073 \text{ kg/h/m}^2)$ ; but it was able to recover some specific compounds, such as isoamyl alcohol, hexanal, 2-phenylethanol and benzaldehyde. This membrane was indeed highly selective to the alcohols compared to the aldehydes.

## Economic framework in beverages dealcoholization by means of PV technology

To provide an economic assessment about the involvement of PV itself in dealcoholization process is a difficult task due to the fact that most of the ongoing research has been done at lab scale. Moreover, the use of PV technology at large scale is usually combined with the distillation process. In this regard, Hoof et al. ([2004\)](#page-9-0) provided a

theoretical economic evaluation of a hybrid distillation/ pervaporation unit for used for specific separation (isopropanol-water mixtures), which resulted in the following costs:

- Operational costs mainly related to energy:  $\sim 17 \text{ e/ton}$ product
- Investment costs:  $\sim$  40 €/ton product
- Maintenance, including membrane replacement:  $\sim 13$ €/ton product

Furthermore, they stated that the costs, for draining solvents that form an azeotrope, using such a hybrid distillation-pervaporation process, can be around half of those associated with conventional distillation. In addition, considering the limited implementation, no investment costs can be mentioned for a full-scale installation. However, there is an increasing importance of ethanol removal from alcoholic beverages. Moreover, the high worldwide consumption of these beverages may minimize the final total investment cost.

On the other hand, PV process can also be useful for the recovery of aromas, in which beer and wine have been also considered as a source. For instance, the worldwide market for flavors and nutraceutical ingredients has been estimated about  $\epsilon$  13 billion in 2006, while more recently the US market has been projected around  $\epsilon$  5.5 billion in 2014 (food 36%, cosmetics and toiletries 27%, beverages 15%), which is expected to rise 3% per year (Brazinha and Crespo  $2014$ ; Castro-Muñoz et al.  $2018a$ ). In this regard, the food processing companies could extend the use of PV technology for other purposes contributing to decrease the final investment cost.

### Future directions and challenges in beverages dealcoholization by PV

To date, it is likely that beer and wine are the main alcoholic beverages that have been subjected to dealcoholization. This is probably due to the fact that they are the most consumed products worldwide. Definitely, their expected consumption and high nutritional content may play a crucial role in the implementation of pervaporation in dealcoholization processes (Mangindaan et al. [2018\)](#page-10-0). Moreover, the ability of PV in the recovery of aroma compounds could be an alternative to develop more efficient processes. Indeed, if the loss of aroma and some other organoleptic compounds are continued being an issue in beverage dealcoholization, the use of PV can be strongly needed for meeting the quality parameters of the non-alcoholic beverages.

Actually, it is challenging that most of the organic compounds are ethanol soluble meaning that there is no process which can guarantee the complete ethanol removal

<span id="page-8-0"></span>

**White must** 



**White wine** 

without altering the composition of the other organics (Müller et al.  $2017$ ). At this point, it is needed to start the use of other highly ethanol selective membrane materials, such as PBZ and butyl acrylate-styrene copolymer. In this way, low temperatures may provide higher selectivity as well as minimal degradation for highly sensitive compounds.

When dealing with the integration of PV into other technologies, such technology has proven to be able to be coupled with the conventional distillation, but also to other membrane-based technologies proposed for alcohol removal, such as osmotic distillation, nanofiltration, reverse osmosis and membrane contactors (Liguori et al. [2016,](#page-10-0) [2018](#page-10-0); Mangindaan et al. [2018](#page-10-0)). Interestingly, the coupling of such processes could produce high-quality and low-alcohol content beverages (Blanco et al. [2016;](#page-9-0) Brányik et al. [2012](#page-9-0)). Finally, it is clear that further research is required in order to adopt pervaporation in dealcoholization of beverages.

### Concluding remarks

This review has shown the role of the pervaporation technology during the dealcoholization of beverages. Apparently, dealcoholization procedure seems to be a simple process; however, the alcoholic beverages (e.g. beer, wine), as multicomponent colloidal solutions, make complicated the removal of alcohol. Also, the high affinity of the organics in ethanol produces strong changes on the organoleptic features of the final products. In this way, pervaporation could be used not only in dealcoholization, but also in the field of aroma recovery. Actually, it is quite possible that PV is going to be implemented in any dealcoholization process based on its ability to recover aromas, and thus meet the quality parameters of the non-alcoholic beverages.

Moreover, if the removal of ethanol is attempted to be highly efficient, the use of hydrophobic membranes is crucially needed in PV technology, these membranes can partially remove the alcohol from the drinks, but they can also perform the separation of the ethanol from other polar compounds. Finally, the food technicians should take into account other highly selective membrane materials toward ethanol, e.g. the ones provided in Table [1.](#page-2-0) Moreover, it is recommendable to operate the PV at the low temperatures which generally provides higher selectivity.

Acknowledgements R. Castro-Muñoz acknowledges the European Commission—Education, Audiovisual and Culture Executive Agency (EACEA) for his PhD scholarship under the program: Erasmus Mundus Doctorate in Membrane Engineering—EUDIME (FPA No 2011-0014, Edition V, http:/eudime.unical.it).

#### <span id="page-9-0"></span>Compliance with ethical standards

Conflict of interest The author declares no conflict of interest.

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