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Version last revised: 18.10.2019

Roberto Castro-Muñoz, Grzegorz Boczkaj

Chapter 9

Pervaporation in food processing

9.1 Introduction

Over the last years, there is a challenging need in finding and developing new suitable technologies for food processing. This need relies on implementing more efficient manufacture protocols, high quality food products together with less elaboration/processing times and costs [1, 2]. Therefore, several technologies are being developed to create new alternative processes for the processing and formulation of diverse food systems. When dealing with the manufacture of foods, the proposed methods should minimally interact with the specific food ingredients and molecules contained in the products [3, 4], such as bioactive, nutraceutical, and functional compounds. Among all these biomolecules, specific targeted molecules present high added value, such as aromas, essentials, and alcohols, among others, due to their importance on the applicability and functionality within the biological processes as well as the physicochemical qualities of the products [5], the latest playing an important role in customer's attention. For example, aromas, essentials, and fragrances influence customers' positive emotions in terms of attributes, perception, and thus driving food acceptance [6]. Such food ingredients are normally recognized as volatile compounds which are naturally present in a wide variety of fruits and vegetables, being part of the entire plant organs [7]. Aromas, essentials, and fragrances are categorized and formed by esters, aldehydes, ketones, alcohols, lactones, terpenoids, and carotenoid-based derivatives [8, 9]. Such solutes display excellent stability in the original food system (e.g., fruits and vegetables) since they are linked to sugars in the way of glycosides [10, 11], that is, bound volatile molecules or glycosylated aroma molecules, and hence they do not release any aroma. Therefore, their extraction from the natural source requires physical extraction protocols (e.g., temperature), chemical agents (e.g., acidification), and biochemical methods (e.g., enzymes), which are identified as the most effective [5]. Once aromas are released by glycoside hydrolysis, aromas are present in their volatile form, resulting in a challenging recovery due to their low thermal stability, reactivity, and volatility. Herein, pervaporation (PV) has become a promising way to recover and selectively separate

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from the original source since this technology needs the membrane as the unique barrier of separation together with a driving force (i.e., vacuum pressure) [12, 13]. PV has seen its wide implementation in other fields of chemical engineering, such as purification of solvents [14], improving the conversion of chemical reactants in chemical and biochemical reactions [15, 16], salt removal for seawater desalination [17], and process intensification [18]. In theory, PV partially vaporizes the components in the complex mixture, which are preferentially separated based on their infinity [19]. **Apart from the membrane-solute affinity, diffusivity, and solubility properties of the target molecule across the membrane are also fundamental for the mass transport** [16].

AU: Please check the sentence “Apart from the membrane-solute affinity, diffusivity, and solubility . . . membrane are also fundamental for the mass transport” for clarity.

Very recently, the research community has documented that PV meets the requirements for the recovery of aromas [8]. Besides this, the author has highlighted two relevant insights: (i) PV can efficiently extract more than 70 different types of aroma solutes, and (ii) specific organophilic/hydrophobic membranes, such as polydimethylsiloxane (PDMS), polyether block amide (PEBA), and poly(octylmethylsiloxane) (POMS), offer the best yield in recovering such volatile molecules from agro-food systems. Therefore, the purpose of this chapter is to introduce the reported cases of study at extracting aromas from food systems and their derivatives, paying attention to the type of source and the membrane concept used for the successful separation. Nonetheless, PV usage is not only limited to the separation of aromatic-base solutes, it has been reported that this technique is also able to separate organic compounds at diluted concentration in aqueous systems [20]. This permits to extend the application of the technique into defined food manufacturing processes, for example, the production of nonalcoholic drinks. Thus, this chapter also reveals the current evidence in removing ethanol from traditional alcoholic beverages, aimed at producing alcoholic-free drinks.

9.2 Pervaporation in aroma separation

As it is well known, aromas, flavors, essentials, and fragrances are particularly important within the beverage and food industries since it is the first contact for the consumer’s acceptance, together with the physical aspect of the food. Such acceptance is inherently a consequence of multiple biochemical reactions in the human being, as illustrated in **Figure 9.1**. However, the interest of these high value-added solutes not only deals with food processing but also in other commercialized products, such as cosmetic and pharmaceutical, which definitely need an enrichment of aroma compounds.

Most of the aromas are low-molecular-weight molecules, which may present either cyclic or linear structure with reactive functional groups [22]. **Table 9.1** enlists some of the typical aromas contained in food systems. Since these molecules are generally organic compounds, they can be recovered by PV membranes, especially hydrophobic or organophilic, which facilitate the transport of organics and less polar compounds [23],



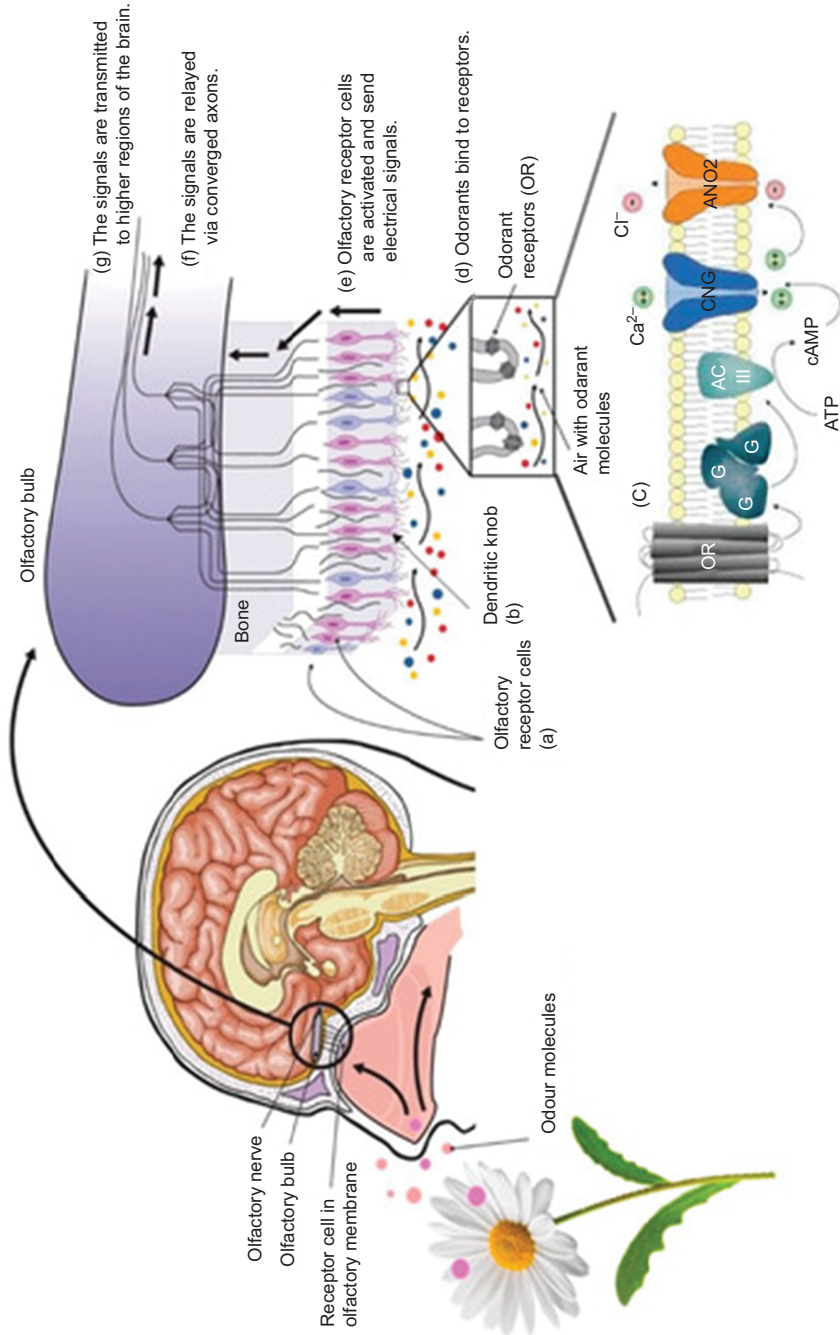


Figure 9.1: Schematic illustration of olfactory transduction mechanism [21].

Table 9.1: Typical aroma solutes present in agro-food systems.

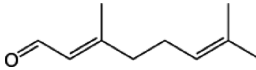
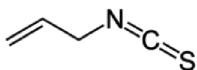
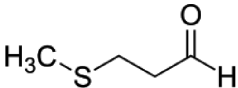
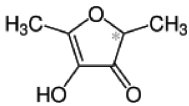
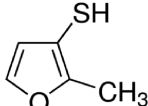
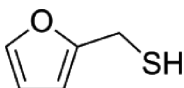
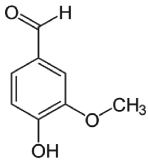
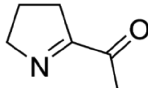
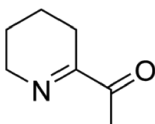
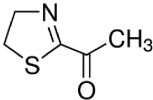
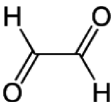
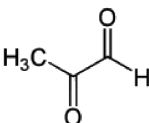
Aroma molecule:	Molecular weight: (g mol ⁻¹)	Chemical structure:
3,7-Dimethyl-2,6-octadienal	152.24	
Allyl isothiocyanate	99.15	
Methional	104.17	
Furaneol	128.13	
2-Methyl-3-furanthiol	146.16	
2-Furfurylthiol	114.16	
Vanillin	152.15	
2-Acetyl-1-pyrroline	111.14	
6-Acetyl-1,2,3,4-tetrahydropyridine	125.17	



Table 9.1 (continued)

Aroma molecule:	Molecular weight: (g mol ⁻¹)	Chemical structure:
2-Acetyl-2-thiazoline	129.17	
Glyoxal	58.04	
Methylglyoxal	72.06	

as represented in Figure 9.2. Importantly, PV is widely used for the removal/recovery of the minor components contained in a complex mixture, which is directly related to the most permeating species across the membrane.

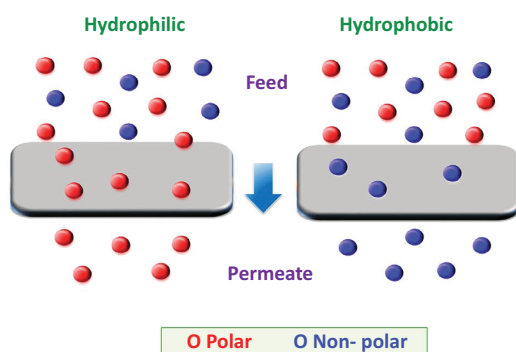


Figure 9.2: Types of pervaporation membranes and their affinity toward molecules depending on their polarity.

The overall performance of the PV process in recovering aromas depends directly on the operating conditions (including feed composition, vacuum pressure, and temperature) and the intrinsic properties of the membrane (e.g., structure and morphology). However, both the chemistry and nature (hydrophilic or hydrophobic) of



the target organic aroma also influence the performance of the PV membrane and thus their recovery efficiency. For example, lactones, characterized by fruity notes, are identified as the most hydrophobic molecules according to their high carbon presence together with oxygenated heterocycle [24]. While esters, also characterized by fruity top-notes, possess a highly hydrophobic nature since they contain less than eight carbon atoms, and oxygen atoms. Finally, alcohols and aldehydes are characterized by hydrophilic nature; this is thanks to the wide number of polar groups (e.g., hydroxyl) [25, 26].

To date, various agro-food systems, including fruit juices, extracts, and commercial products (e.g., beer, wine, and dairy products), have been primarily explored as potential sources of aromas. In 1989, Bengtsson and coworkers initiated the assay of PV as a way of recovering organics from apple juice [27]. After the PV operation, a permeate sample enriched in alcohols, *trans*-2-hexenal, and esters was obtained. Thanks to the use of hydrophobic PDMS membrane, the implemented process was able to offer aroma recovery efficiencies ranging from 49% to 100% (see Table 9.2). It is important to note that PV is likely to offer very low permeation rates with the aroma separation (lower than $0.5 \text{ kg m}^{-2} \text{ h}^{-1}$); this is attributed to the nonporous structure in PV membranes.

Methylthiobutanoate, which releases a characteristic cheesy odor and a musty flavor, together with diacetyl were separated from a dairy solution [28]. At this time, the authors applied an organophilic PV process utilizing PDMS and PEBA membranes. Herein, it was identified that the membranes exhibited a low enrichment (β) factor due to the hydrophilic nature of both flavor molecules. In a subsequent research development, the same group reported the ability of PDMS membranes in separating ethyl acetate with a β factor ranging from 230 to 280 [29]. Besides, these hydrophobic membranes concurrently recovered other valuable molecules [30, 31], including isoamyl acetate, ethyl-2-methylbutanoate, and hexyl acetate. The membranes eventually showed a recovery rate of approximately 80%. In the case of isoamyl acetate, it is generally sought for its typical notes to banana fruit and jackfruit [32]. Interestingly, other types of polymer membranes, such as hydroxy-terminated polybutadiene (HTPB)-based polyurethaneurea (PU), have revealed a higher affinity in extracting ethyl acetate [33, 34]. The obtained β factor (of about 655) toward the ester allows the expectation of a higher recovery yield compared to that of the PDMS membrane. According to the authors' remarks, HTPB-PU copolymer promotes the transport of the ester due to the fact that it possesses apolar hydrophobic segments for the specific absorption of organics (like aromas). Especially, hydrophobic and flexible soft segments (polyol) stand out for the facilitated organic diffusion [33]. This agrees based on their permeation fluxes, for example, HTPB-PU had a permeation of $0.256 \text{ kg m}^{-2} \text{ h}^{-1}$, which represents a higher value in comparison with PDMS membrane (maximum permeation of $0.012 \text{ kg m}^{-2} \text{ h}^{-1}$) [29]. Specific polymer, like poly(vinylidene fluoride-co-hexafluoropropene), owns the suitable intrinsic properties to preferentially permeate ethyl acetate faster compared to other organophilic polymer membranes [35],



Table 9-2: Various natural agro-food systems processed for aroma separation via pervaporation.

Aroma solute:	Natural source:	Membrane material:	Productivity (permeate flux, β) ($\text{kg m}^{-2} \text{h}^{-1}$)	Enrichment factor: (β)	Reference
Ethanol, butanol, isopentanol, hexanol, hexanal, <i>trans</i> -2-Hexenal, butyl acetate, hexyl acetate, ethyl acetate, ethyl butanoate, ethyl-2-methyl butanoate, butyl butanoate	Apple juice model solution	PDMS	0.107	44–125	[27]
Methylthiobutanoate, diacetyl	Dairy model solution	PDMS	0.040	15	[28]
			–	19	
		PEBA	0.015	17	
			–	18	
Ethyl acetate	Model solution	PDMS	0.006	230	[29]
			0.012	280	
Vanillin	Bioconversion culture broth	PEBA	0.128	12.6	[37]
Ethyl acetate, isobutanol, isoamyl alcohol, methyl lactate, hexanol, furfuraldehyde, 2,3-buthaneidol, 5-methyl-furaldehyde	Muscat wine	POMS	0.001	10–160	[38]
Ethyl-2-methyl butanoate	Apple juice model solution	POMS	–	3400	[39, 40]
Ethyl butanoate				2000	
Isoamyl acetate				2900	
Hexyl acetate				3400	

(continued)

Table 9.2 (continued)

Aroma solute:	Natural source:	Membrane material:	Productivity (permeate flux, factor: β) ($\text{kg m}^{-2} \text{h}^{-1}$)	Enrichment factor: (β)	Reference
2-Hexenal	Apple juice model solution	PDMS	0.045	380	[41]
		POMS	0.035	700	
		PEBA	0.002	150	
Linalool	Wine must	POMS	–	150	[42]
1-Hexanol				200	
Methyl anthranilate)	Grape juice model solution	PDMS	0.055	15	[43]
Dimethyldisulfide	Model solution	PEBA	0.0005	–	[44]
S-Methyl butanoate			0.0007		
Dimethyltrisulfide			0.002		
Isobutyl acetate	Bioconversion culture broth	POMS	0.003	400	[45]
S-Methyl thio-butyrate	Cauliflower blanching water	PDMS	0.0006	307	[46]
		PEBA	0.0002	1200	

Linalool	Blueberry model solution	PDMS	-	231	[47]
<i>d</i> -Limonene				135	
1-Heptanol				405	
1-Hexanol				203	
<i>trans</i> -2-Hexenal				267	
Ethyl acetate				290	
<i>cis</i> -3-Hexenol	Tea extract	POMS	0.003	120	[48]
Ethyl acetate	Model solution	PDMS	0.008	-	[49]
Ethyl acetate	Tropical fruit juice	Commercial Pervap 1070	0.077	124	[50]
Ethyl butanoate			0.077	410	
Ethyl hexanoate			0.055	213	
Menthone	Model solution	PDMS	0.001	1200	[51]
β -Ionone			-	6	
Citronellal			0.002	20	
Ethyl acetate	Diluted flavor systems	POMS	0.065	105	[52]
Ethyl butyrate				98	
2-Hexenal				90	
Benzaldehyde				70	
Hexanol				40	
2-Methyl-1-butanol				30	

(continued)

Table 9.2 (continued)

Aroma solute:	Natural source:	Membrane material:	Productivity (permeate flux, factor: β) ($\text{kg m}^{-2} \text{h}^{-1}$)	Enrichment factor: (β)	Reference
Methyl butyrate	Strawberry model solution	Commercial Pervap 1070	0.25	90	[53]
Ethyl butyrate			0.15	55	
Methyl butanoate	Clarified kiwifruit juice	Commercial Pervap 1060	0.13	120	[54]
1-Hexen-1-ol				20	
(E)-2-Hexen-1-ol				20	
1-Hexanol				50	
Ethyl butanoate				100	
Bergapten, linalool, linalyl acetate, limonene	Bergamot peel oils	Commercial GFT 1070	–	–	[55]
(E)-2-Hexenal	Kiwifruit juice	SBS composite	0.001	70	[56]
(E)-2-Hexen-1-ol				55	
1-Octen-2-ol				32	
1-Hexanol				80	
Ethyl acetate	Model solution	HTPB-PU	0.250	550	[34]
Limonene, ethyl butanoate, methyl hexanoate, ethyl benzoate, hexanal, heptanal, benzaldehyde, 1-octen-3-ol, methyl acetate	Cashew apple fruit	PDMS	0.17	–	[57]

Ethyl acetate	Orange juice	PDMS	0.0001	11	[58]
Ethyl butyrate			0.0001	6	
Hexanal			0.0001	6	
Limonene			0.0035	12	
Linalool			0.0001	8	
α -Terpineol			0.0001	5	
<i>E</i> -2-Hexen-1-ol	Bilberry juice	PDMS	-	120.6	[59]
<i>n</i> -Hexanol				236.9	
<i>E</i> -2-Hexen-1-al				46.3	
Linalool				49.3	
Phenyl acetaldehyde				5.5	
Benzyl alcohol				4.2	
<i>Z</i> -3-Hexen-1-ol				27.4	
<i>trans</i> -Hex-2-en-1-ol	Bilberry juice	PDMS	-	194	[60]
Linalool	Bergamot essential oil	Commercial	0.20	28	[61]
Linalyl acetate		Pervap 1070	0.25	55	
3-Methyl butanal	Pomegranate juice	PDMS	0.140	23	[62]
Isopentyl acetate			0.200	21	
<i>n</i> -Hexanol			0.050	19	
α -Ionone			0.040	9	

(continued)

Table 9.2 (continued)

Aroma solute:	Natural source:	Membrane material:	Productivity (permeate flux, factor: β) ($\text{kg m}^{-2} \text{h}^{-1}$)	Enrichment factor: (β)	Reference
3-Methyl butanal	Pomegranate juice	PDMS	0.0005	15	[63]
Isopentyl acetate			0.0005	16	
<i>n</i> -Hexanol			0.0004	15	
α -Ionone			0.0002	6	
1-Methyl-1-pyrrole, methylpyrazine, furfural, 5-methyl-2-furancarboxyaldehyde, 2-formyl-1-methylpyrrole.	Coffee extract	PDMS	0.001	–	[64]
Propanol	Beer	POMS	0.28	1.4	[65]
Isobutanol				2.9	
Isoamyl alcohol				3.6	
Ethyl acetate				24.7	
Isoamyl acetate				35.7	
Acetaldehyde				5.3	

Propanol	Beer	POMS	0.13	1.4	[66]
Isobutanol				2.5	
Isoamyl alcohol				3.0	
Ethyl acetate				8.2	
Isoamyl acetate				20.5	
Acetaldehyde				0.3	
α -Pinene	Lemon Juice	POMS	0.0004	22	[67]
β -Pinene			0.0003	18	
Limonene			0.0009	16	
Vanillin	Reaction system	PEBA	0.39	4.2	[68]
1-(3H)-Isobenzofuranone	Brown crab boiling juice	Commercial Pervap 4060	-	39.9	[69]
2,5-Dibutylfuran				29.9	
<i>cis</i> -Geranyl acetone				37.0	
4-Methyl-2-pentanone				52.8	
2,7-Dimethylnaphthalene				20.1	
4-Methylthiazole				32.7	
<i>i</i> -Amyl alcohol	Pineapple juice	Commercial PDMS	0.0002	75	[70]
Methyl 2-methylbutanoate		Pervaptech BV	0.002	80	
Methyl hexanoate			0.0002	25	
Ethyl acetate			0.0003	100	

(continued)

Table 9.2 (continued)

Aroma solute:	Natural source:	Membrane material:	Productivity (permeate flux, factor: β) ($\text{kg m}^{-2} \text{h}^{-1}$)	Enrichment factor: (β)	Reference
Ethyl acetate	Model solution	Commercial Pervap 1060	0.0004	7	[71]
Ethyl acetate	Beer	Commercial	0.0003	42	[72]
Isobutyl alcohol		PDMS		32	
Isoamyl acetate		Pervaptech BV		9	
2,3-Butanedione	Soluble coffee solution	Commercial	0.432	45	[73]
2,3-Pentanedione		PDMS		7	
3-Methylbutanal		Pervaptech BV		8	
Benzaldehyde				4	
Acetaldehyde				5	
Furfural			2		
2-5-Dimethyl pyrazine				42	
5-Methyl furfural				1	
Benzaldehyde, 1-hexanol, isoamylalcohol, hexanal, benzylalcohol, 2-phenylethanol	Grape must	Commercial PDMS Pervaptech BV	0.100	1–2	[74]

Ethyl alcohol	Sugarcane molasses fermentation broth	Commercial Pervap 4060	0.100	1	[75]
Ethyl acetate			<0.100	3	
Isoamyl alcohol			0.150	1.5	
Isoamyl acetate			0.100	1	
Pentan-1-ol	Plum, apple, blackcurrant and cherry hydrolysates	Commercial Pervap ECO 001BP	0.008	5800	[76]
Hexanal				3678	
Butyl acetate				8602	
Heptan-1-ol				1131	

for example, this membrane had an ester permeation rate in the range of $2 \text{ kg m}^{-2} \text{ h}^{-1}$. This permeation rate is tremendously high for PV membranes focused on the separation of organic aromas [36].

Fruits are likely the natural agro-food products that contain a major variety of aromas and fragrances; this is the case of kiwifruit, in which more than 50 aromas have been determined [77]. Specific compounds, such as 3-penten-2-ol, 3-hydroxy-2-butanone, 3-methyl-2-butenal, 2-hexanol, nonanal, 3-methyl-1-butanol, 2-methyl-1-butanol, 3-methyl-2-butanone, 3-methyl 3-buten-2-one, and octane [78], are among the key organics present in this natural product. Based on its potential for different aroma notes, the kiwifruit was proposed by Cassano's group, who extracted methyl butanoate, ethyl butanoate, 1-hexen-1-ol, (*E*)-2-hexen-1-ol, and 1-hexanol from kiwifruit juice, which was previously clarified by ultrafiltration [54]. Cassano and coworkers at this time used a Pervap 1060 membrane which exhibited β factors of 120, 100, 20, 20, and 50, respectively. Once such solutes were extracted in a permeate, they were blended into a concentrated kiwifruit juice to enrich such a minimally processed product.

A few years later, a new membrane material was assayed by Figoli et al. [56] for the separation of aroma molecules of kiwifruit juice. In this study, a thermoplastic elastomer, like styrene-butadiene-co-styrene (SBS), was employed as a selective separation layer exhibiting low permeation flux. The permeating molecules displayed a temperature dependency, which means that the permeation increases by temperature increment. Unlike Cassano's approach, the selectivity in SBS membrane for (*E*)-2-hexenal, (*E*)-2-hexen-1-ol, 1-octen-2-ol, and 1-hexanol was better than the PDMS membrane.

Orange is another important source of essences, which include acetaldehyde, ethyl acetate, acetal, and ethyl butyrate, as the main aromatic components [79]. In particular, ethyl acetate, ethyl butyrate, hexanal, limonene, linalool, and α -terpineol were obtained from orange juice by Aroujalian en Raisi [58]. A PDMS membrane was used in this study; this membrane apparently revealed a β increase toward ethyl butyrate as a function of vacuum pressure increase.

Chemicals representing the major components (identified as 3-methyl butanal, isopentyl acetate, *n*-hexanol, and α -ionone) in aroma essence for pomegranate juice were acquired using three POMS and two PDMS membranes [62]. At this point, POMS were potentially able to offer higher β values in extracting the solutes than PDMS. Similar to other studies, the results revealed that the permeate flux was directly dependent on the operating temperature according to the Arrhenius model; in other words, an increase in temperature resulted in an enhanced permeation of organics. The process analysis also proved that the apparent activation energy of the aroma solutes was higher than that of water, concluding that the aroma transport is more temperature sensitive than water [62, 80]. Apart from such analysis, temperature greatly influences the solubility and diffusion coefficient of species [81]; this means that both parameters tend to increase by temperature increment.



Raisi and Aroujalian [63] analyzed the PV extraction process of aroma ingredients from pomegranate juice. The authors found out that the affinity between a PDMS membrane and aromas were given as: isopentyl acetate>3-methylebutanal>n-hexanol>water. This membrane-aroma affinity resulted in a higher sorption ability of the molecule in the membrane, and hence facilitated permeation. These findings were in agreement with the Hansen solubility parameters [63].

Lemon is widely defined by its multiple aromas, fragrances, and essential oils. In fact, lemon is recognized as one of the most enriched sources of essential oils together with other citric products including bergamot, lime, sweet orange, tangerine, and mandarin [82]. Among the wide range of aromas, lemon contains mainly terpenes, such as limonene, γ -terpinene, *p*-cymene, and α -citral [83], to mention just a few of them. Knowing its potentiality as a source of terpenes, lemon juice was subjected to pervaporative separation of α -pinene, β -pinene, and limonene. Rafia and coworkers utilized a POMS polymer membrane which showed an enhanced β and permeation values when driving force increased [67]. Differently from PDMS membranes, temperature increment in POM membranes preferentially promoted water transport compared to aromas, compromising the β factor. Therefore, the recovery of terpenes was recommended to be done at low temperatures.

A large list of chemical solutes (e.g., carboxylic acids, pyrroles, pyridines, and chlorogenic acids) is responsible for the characteristic taste and aroma of coffee [84]. This product is recognized, commercialized, and consumed worldwide. Of course, the research has recently exerted interest in this typical product for the extraction of chemicals. In this case, 2,3-butanedione and 2-5-dimethyl pyrazine were intentionally separated via organophilic PV (PDMS Pervaptech BV membrane). Weschenfelder et al. [73] notified that the commercial membrane demonstrated high selectivity for 2,3-butanedione and 2-5-dimethyl pyrazine according to the β values of 45 and 42, respectively. Together with the acceptable selectivity, the membrane also exhibited a suitable permeate flux of about $0.432 \text{ kg m}^{-2} \text{ h}^{-1}$.

In addition to the exploration of fruits and natural extracts for the extraction of aromas and fragrances, different wastes, residues, and by-products from agro-foods are pointed out as feasible feedstock of these high added value organics based on the recent trends in food waste valorization for the manufacture of chemicals and materials [15, 85]. These wastes are inherently the result of the various food treatments (such as washing) and derived processing processes (such as peeling, and pressing) [86, 87]. In 2002, Souchon and coworkers pioneered the use of food wastes for the separation of organic molecules via PV technology [46]. They acquired characteristic solutes, including *S*-methyl thio-butyrates, dimethyl trisulfide, and dimethyl disulfide, from cauliflower blanching residues. These sulfur-based components were extracted using PEBA and PDMS membranes, in which they were highly selective for methyl thio-butyrates showing β factors of 1,200 and 307, respectively. The main lack of the membranes was related to their low productivities in terms of permeation.

AU: Can the phrase "Differently from PDMS membranes, temperature increment . . ." be changed to "Contrary to PDMS membranes, temperature increment"?

AU: The term "derived processing processes" in the sentence "These wastes are inherently the result of . . ." seems redundant. Please rephrase.



Essences were successfully separated from the oil extract of bergamot peels [55]. To cope with the complex extraction from this waste, Figoli and coworkers applied an enzyme treatment to preliminary extract bergapten, linalool, linalyl acetate, and limonene. At this point, the properties of PDMS membranes were found adequate to recover the aromas [61]; the study once again confirmed that temperature tends to improve the transport of organic components and hence obtain higher β factors.

Martínez et al. [69] introduced the pervaporative separation of various categories of chemicals from brown crab boiling juices. Several alcohols, aldehydes, esters, ketones, furans, hydrocarbons, naphthalene derivatives, sulfur compounds, and terpenes were identified and quantified by the authors. The results denoted that Pervap 4060 membrane efficiently separated specific solutes, including 1-(3H)-isobenzofuranone, cis-geranyl acetone, 2,5-dibutylfuran, 4-methyl-2-pentanone, 2,7-dimethylnaphthalene, and 4-methylthiazole (see Table 9.2).

Very recently, fruit juice hydrolates were exploited by Dawiec-Liśniewska et al. [88] as a source of aromatic compounds. The aromas are generally present in such by-products around 1wt.% of the total extract. Based on this, the diluted organics are frequently desired to carry out PV process. Dawiec-Liśniewska et al. [88] concentrated diverse aromas from plum, apple, blackcurrant, and cherry fruit derivatives employing a laboratory and semi-technical scale PV setup [88, 89]. To sum up, the authors identified and quantified more than 30 different molecules in the blackcurrant hydrolates, and around 20 and 14 types of molecules were analyzed in apple and cherry hydrolates, respectively. Using a hydrophobic Pervap membrane, impressive β factors for several compounds were estimated of about 5800, 3678, 8602, and 1131 for pentan-1-ol, hexanal, butyl acetate, and heptan-1-ol, respectively. As expected, the highly selective properties of the membranes commonly bring low permeation properties, in this case, the membrane had a flux of $0.180 \text{ kg m}^{-2} \text{ h}^{-1}$, and it could be raised (ca. $0.450 \text{ kg m}^{-2} \text{ h}^{-1}$) when temperature increased.

When the target deals with the enrichment of commercial products, a good alternative can be the extraction of aromas from particular processed food systems. Beer, wine, and cider have been some of the explored at separating specific aromas [65, 90, 91], however, the PV application has been also extended at producing novel market products attending specific needs of the costumers, for example, the production of nonalcoholic products. The following section compiles some case studies addressing such scope.

9.3 Pervaporation in the production of nonalcoholic drinks

According to recent reports of the World Health Organization (WHO) [92], consumption of typical alcoholic beverages has tremendously raised in society. A current



report notifies that the global consumption of alcoholic beverages is calculated to about 54.2 billion liters per year [93], in which beer and wine are found as the most consumed products [94]. Since it has been argued that high consumption of alcoholic beverages contributes to specific diseases, including pancreatitis, hepatitis, fatty degradation of liver, cirrhosis, peptic ulcers, allergenic induction, among others [95–97], there is a current trend in attending consumers' necessity at producing products with similar physicochemical properties but alcohol-free. To date, it has been demonstrated that the best option for manufacturing such nonalcoholic beverages with the post-fermentation removal of ethanol from the commercial products. In this way, PV has been devoted at selectively removing the ethanol; for instance, Catarino and Mendes [66] carried out the manufacture of low-alcohol content beer utilizing a hybrid industrial plant. Figure 9.3 illustrates the developed process implementing PV; this system comprised assisting distillation units with PV.

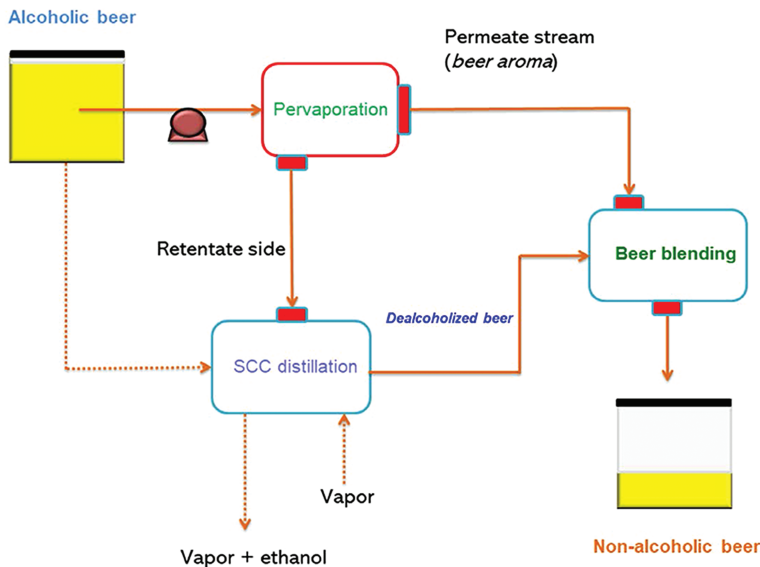


Figure 9.3: PV assisting the production of nonalcoholic beer. Adapted from [98].

In this work, the authors extracted the aromas from conventional beer using a PV-POMS membrane and subsequently blended into the dealcoholized beer. The industrial protocol led to manufacturing an alcohol-free beer presenting less than 0.5 vol% ethanol, meeting an acceptable flavor profile [66]. Similarly, PV was eventually utilized by Catarino en Mendes [90] for the separation of the aromatic components from wine. Here, the aroma extraction has been referred to meet the organoleptic attributes of the dealcoholized wine.



More recently, PV acted as a fundamental unit operation in processing full-flavored low alcohol white wines [74]. PV was able to separate targeted molecules, for example, benzaldehyde, 1-hexanol, isoamylalcohol, hexanal, benzylalcohol, 2-phenylethanol, from the grape must, and later embed them intentionally in the fermentation stage. The blending step helped to produce nonalcoholic wines with featured sensorial properties.

Commercial beers, such as special beer (presenting 5.5% ABV) and reserve beer (presenting 6.5% ABV) were subjected to pervaporative processing to acquire aromas and flavor ingredients (isobutyl alcohol, ethyl acetate, and isoamyl acetate). Here, such organic were once again blended in low-alcohol beer (presenting <1% ABV) and an alcohol-free beer (presenting < 0.1% ABV) to enhance their sensory attributes [72]. By performing a sensory evaluation, the products proved to have good acceptance.

AU: The sentence "Here, such organic were once again blended in low-alcohol beer . . .to enhance their sensory attributes" is incomplete. Please check.

9.4 Concluding remarks

In this chapter, new readers in the field have acquired an outlook of the most recent developments at using pervaporation technology in food processing. In general, PV has devoted to assisting extraction processes of aromas and fragrances from natural food products, as well as their main derivatives (wastes, residues, and by-products) produced with the manufacture and elaboration processes. When using organophilic membranes (e.g., POMS, PEBA, and PDMS), PV has proven to meet the basic requirements as a promising technique in selectively separating more than 70 food aroma ingredients from diverse complex agro-food systems. Apart from aroma extraction, PV can be defined as an alternative method for the elimination of ethanol from alcoholic drinks aiding to produce ethanol-free products. This is due to the fact that PV can partially remove out the alcohol from the beverages. To finalize, the resulting enriched nonalcoholic products (like beer and wine) with aromas extracted via PV have demonstrated to reach the quality attributes for potential commercialization.

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