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Abstract

Background

Food-processing gaseous effluents are rich in **flavoured volatile organic compounds** (FVOCs). The discharge of these FVOCs is unwanted as they can contribute to the environmental olfactive pollution.

Scope and Approach

Their recovery would then enable their **valuation** through the strengthening of the organoleptic properties of the finished products or their use in other products, as well as **reducing the pollution** linked to their discharge. However, there are only a few documents in the literature concerning **food aromas recovery** from gaseous effluents. This paper reviews the **used or potential technologies** for the recovery of aromas from gaseous effluents in the food-processing industry.

Key Findings and Conclusions

The technologies that are already applied in the food processing industry for aroma recovery from gaseous effluents are the **vapour permeation** and the **condensation**. The **adsorption** and the **absorption** are technologies used for scrubbing volatile organic compounds, which can be potentially used for gaseous aroma recovery.

Review on the potential technologies for aromas recovery from food industry flue gas

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Keywords: aroma recovery, gaseous effluent, food process, volatile organic compounds

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Introduction

Food aromas are typically mixtures of odourous volatile organic compounds (VOCs) with a molecular size lower than 400 Da (Cros, Lignot, Bourseau & Jaouen, 2005; Landy, Courthaudon, Dubois & Voilley, 1996; Lipnizki, Olsson & Trägårdh, 2002a). The involved molecules are mainly alcohols, aldehydes, carboxylic acids, furans, fatty acids, esters, ethers, hydrocarbons, ketones, lactones, pyrazines and terpenes (Karlsson & Trägårdh, 1997; Longo & Sanromán, 2006). Regarding the previous definition, the term flavoured volatile organic compounds (FVOCs) refers to aromas in the present text.

Food industry processes often involve the production or the transformation of FVOCs, which are generally important for the organoleptic properties of the product such as aroma and flavour. The generation of FVOCs in the food industry is mainly due to fermentation and cooking/thermal processes. In the fermentation processes, including the production of dairy and alcoholic drinks and bakery products (Ardö, 2006; Rowe, 2005), FVOCs are produced by microorganisms via enzymatic and chemical reactions mainly from amino acids (Smit, Smit & Engels, 2005). The generation of FVOCs in thermal processes is achieved by Maillard reactions, caramelisation of sugars, amino acids degradation of Strecker and lipids oxidation and degradation (Ardö, 2006; Rowe, 2005).

Some industrial downstream processes used for the conservation and/or the concentration of process products can lead to the loss of generated FVOCs by evaporation or solubilisation causing the modification of sensory properties of the concerned products. Furthermore, the produced odourous effluents are rejected outside the plant, contributing in certain cases to the olfactory and environmental pollution. In order to comply with the evolution of environmental regulations (Khan & Kr. Ghoshal, 2000; Souchon, Pierre, Samblat, Bes & Marin, 2002; Yeom, Lee, Song & Lee, 2002) and to guarantee the quality of the final product for more and more consumers (Brazinha, Alves, Viegas & Crespo, 2009; Pereira, Ribeiro Jr, Nobrega & Borges, 2006), food industries are focusing on the recovery of the FVOCs from effluents before their rejection. In addition, the recovered FVOCs can be considered as natural aromas with a market value 20 to 50 times above those of identical molecules produced by chemical synthesis (Lipnizki, Olsson & Trägårdh, 2002b; Song, Song & Lee, 2003).

In the scientific literature, the recovery of FVOCs in food processes has been widely developed in the production of fruit juice beverages (Aroujalian & Raisi, 2007; Pereira et al., 2006; Ribeiro Jr, Lage & Borges, 2004; Sampaio, Garruti, Franco, Janzanti & Da Silva,

2011; Song et al., 2003; Souchon, Pierre, Athes-Dutour, & Marin, 2002). The review paper of Karlsson & Trägårdh (1997) gives an overview of the seven available technologies for that purpose, as well as their advantages and limitations. Very little information is available on the recovery of FVOCs from food processes gaseous effluents, except in some industrial patents. The aim of this review is to present the technologies that can potentially be used in food industries for the recovery of FVOCs from gaseous effluents. First, common technologies are presented. Then, other potentially interesting technologies are discussed.

Common technologies

Membrane technologies

The membrane technology used to separate volatile organic compounds (VOCs) from gaseous effluents is called vapour permeation (VP). In this process, the compounds of interest are separated from the gaseous mixture thanks to their more favourable penetration through the membrane. They are then continuously condensed on the other side of the membrane, generally in liquid form (Brüschke, 2001).

In this technology (^{see Figure 1}), the separation is also ensured by the specific migration gradient of each compound between the two sides of the dense membrane. In the recovery stage, all the compounds that crossed the membrane are continuously desorbed under reduced pressure or taken towards the condenser progressively by a sluggish vector gas.

VP has been developed as a derivative of pervaporation (PV) in which the compounds to recover are in a feed liquid phase. The principles of these two processes are basically the same since both are membrane-based; different compounds of a fluid mixture are separated thanks to their different solubility and diffusivity in a specific dense membrane. The main technical difference between VP and PV processes lies in the state of the VOCs' mixture to recover, respectively, vapour and liquid. In addition, according to Ribeiro Jr et al. (2004), the VP is less influenced by the membrane polarisation phenomenon, which can be severe in PV.

PV has been known since the beginning of the 20th century (Kober, 1917) and was first developed in the industry to recover and concentrate VOCs from aqueous mixtures. The importance of this industrial process has grown in the past three decades, as shown by hundreds of European and American patents generated during that period (Jonquières et al., 2002). The existing scientific literature on PV confirms the importance of this method.

However, it is important to remember that the majority of these studies were achieved on model aqueous solutions, as shown by Pereira et al. (2006) in their review paper.

According to the review of Jonquières et al. (2002), the number of European and US patent applications on VP represents 50% and 20%, respectively, of the total patent applications on both PV and VP technologies. This shows the interest of industrials for VP. The VP process is also widely used in industry for nitrogen production, hydrogen recovery and natural gas or pollutant VOCs capture (Sun L-M. & Thonnelier J-Y., 2004).

In both processes, the relative diffusion coefficients and the relative solubility in the membrane material of each compound determine the separation efficiency of the membrane. The effects of these two parameters on the selectivity of the membrane for gaseous compounds are opposite. According to R. W. Baker, Wijmans, and Kaschemekat (1998), small molecular size compounds are characterised by diffusion coefficients that are higher than those in large molecular size compounds, because these latter interact with more segments of the polymer chains of the membrane. However, these interactions increase their solubility in the material. Consequently, the material of the membrane influences the balance between diffusion coefficient and solubility. Therefore, according to the previous authors, membranes with rigid polymers (polysulfone/polyimides) favour important diffusion coefficients for small molecular size VOCs while the ones with rubbery polymer enable significant relative solubility for large molecular size VOCs. Since most of the FVOCS encountered in the food industry are included in a wide range of molecular size and various chemical classes, the choice of suitable membrane for FVOCS recovery is of a considerable interest.

Numerous studies have investigated the development and the improvement of new membrane materials to separate a wide variety of mixtures, such as presented in the review paper of P. Bernardo, Drioli, and Golemme (2009) for VP. Before that, Karlsson & Tragardh (1993) reviewed studies on the modelling and applications of PV to aroma recovery in water mixtures. Other studies have also appeared since the beginning of 2000 on the efficiency of the various membranes used in PV, as shown in the paper of Brian Bolto, Hoang, and Xie (2011). More recently, Yampolskii (2012) and R. W. Baker and Low (2014) published detailed review papers on the evolution of membrane materials for separation of gaseous mixtures compounds. Membranes for VP are basically classified into three groups: rubbery polymer membranes, glassy polymer membranes and inorganic membranes (Faiz & Li, 2012). In their review paper, P. Bernardo et al. (2009) listed the most important glassy (cellulose

acetate; polyperfluorodioxoles; polycarbonates; polyimides; poly(phenylene oxide); polysulfone) and rubbery (poly(dimethylsiloxane); ethylene oxide/propylene oxide – amide copolymers) polymers used in industrial membranes for VP.

The industrial implementation of vapour permeation has rapidly grown from the early 1980s to today (Paola Bernardo & Clarizia, 2013). These processes are widely used for petrochemical and chemical industries in the production and purification of halogenated VOCs and monomers (R. Baker, 2006; P. Bernardo & Drioli, 2010; Bessarabov D.G., 1999; Jonquieres et al., 2002; Ohlrogge, Wind & Brinkmann, 2010). However, they can also be used in the food industry to enhance and/or refine the flavour of some products.

In the literature, PV is the most investigated of the two processes for the food industry, especially for the recovery of aromas of fruit juices (Aroujalian & Raisi, 2007; Börjesson, Karlsson & Trägårdh, 1996; Pereira et al., 2006; Sampaio et al., 2011), dealcoholisation of beverages (Karlsson & Tragardh, 1996) and deodourisation of wastewater (Souchon, Pierre, Athes-Dutour et al., 2002). PV is also used for the selective recovery of aromas in dairy products, as presented in the paper of Baudot and Marin (1996).

Concerning VP, although the major industrial applications concern chemistry and petrochemistry, a potential application for food processes can be considered since there is a wide variety of commercialised membranes and the possibility to set up hybrid systems. Some studies were already published on VP separation of gaseous FVOCs met in food processing. Will and Lichtenhaler (1992) used the VP to selectively achieve the separation of propanol-methanol-water gaseous mixtures. Bhaumik, Majumdar, and Sirkar (2000) and Gales, Mendes, and Costa (2002) also published papers on the recovery of methanol, ethanol, *n*-butanol and ethyl acetate from gaseous stream using VP. The recovery of ethyl acetate, propyl acetate and butyl acetate by VP was also investigated by Song et al. (2003).

Although the principles of PV and VP are nearly the same, their industrial capital cannot be compared. Indeed, these technologies are applicable at two different stages of the process involving different investments and maintenance. Sander and Janssen (1991) evaluated the total cost of PV to be higher than VP while Suematsu, Kimura, and Nitta (1998) had the opposite finding. The evaluation of these technologies needs to be updated in order to adjust their costs. However, up-to-date, reliable information about VP and PV current markets are difficult to find, especially for new membrane materials, which are still seldom developed at the industrial scale. A review on the new generation membranes for VP applications is

proposed by B. Bolto, Hoang, Gray & Xie (2015). The most recent advances in the VP framework are mainly focused on new membrane materials, in particular the supported ionic liquid membranes (Dahi et al., 2014; Krull, Fritzmann & Melin, 2008; Matsumoto, Inomoto & Kondo, 2005; Matsumoto, Ueba & Kondo, 2009; B. Wang, Lin, Wu & Peng, 2008). Nevertheless, they mostly investigate the separation of some organic molecules than FVOCs recovery.

Condensation

In the condensation process, which is devoted to recover VOCs from a gaseous mixture, the gas stream treated is cooled until its temperature decreases below the boiling temperature of the compound(s) to recover. Techniques used to cool the studied gas are based either through the contact with a cooling surface (heat exchanger) or by using a cooling pure gas such as nitrogen. The VOCs are then concentrated in the resulting liquid phase.

The gas condensation can also be achieved only by pressurisation or in combination with the cooling gas. As explained in the review paper of Karlsson & Trägårdh (1997), the compounds of the gaseous mixture can be selectively fractionated depending on the temperature and the pressure applied in the condenser chamber. This application was described several decades ago in the paper of Hasting (1978) for the recovery of aromas evaporated during the concentration of apple and oranges juices.

There are numerous studies concerning the modelling and the optimisation of the parameters influencing the heat transfer during condensation. These works mainly concern air conditioners, water chillers and heat pumps, but the principle of condensation is basically the same. The review of Cavallini et al. (2003) summarises results of published papers on the effects of the condenser tubes geometry on the condensation process. This process is strongly influenced by the composition of condenser tubes and by the flow rate of the fluid to treat. The reviews of Miyara (2008) and Thonon (2008) analyse papers presenting new refrigerants composed of hydrocarbons that could be used to improve the condensation properties.

When the condensation is achieved using liquid nitrogen, the process is usually called cryo-condensation, as it appears in the US patents published during the three last decades (Budwig, Temperini & Rushmore, 2002; Carns & Tuot, 1993; Mandralis, Yunker & Westfall, 2003; Mazurek, Temperini, Barfuss & Rushmore, 2000; Siedlecki & Meinhold, 1977). These patents deal with the recovery of natural aromas from gaseous effluents of cocoa, nuts and

coffee. The recovery process is achieved by spraying liquid nitrogen on the gaseous effluent to reach temperatures lower than -80 °C and thereby form aroma frost particles.

It is commonly considered that condensation is an expensive technique and the choice of this technique is driven by the characteristics of the gas to be treated. In their review, Khan and Kr. Ghoshal (2000) mentioned the fact that condensation is more financially efficient for the recovery of high concentrated VOCs (>5000 ppm) with a boiling temperature above 37 °C at atmospheric pressure. However, these authors considered the additional costs of the gas cooling or pressurisation when the compounds to recover have a low boiling temperature/pressure.

Patents are the only type of publications mentioning the independent use of this technology for the recovery of FVOCS from gaseous effluents. All articles present the condensation in combination with VP or PV.

Other potential technologies

As mentioned, the most common technologies for aroma recovery are membrane-based techniques and the (cryo-)condensation. However, other technologies exist in the gas separation field, as suggested by the review paper of Karlsson & Trägårdh (1997), in which alternative technologies for the aroma recovery such as distillation are presented. This last technology is not directly applied to the gaseous effluents but is used for the regeneration of the solvent for the capture of aromas in certain technologies, and will not be described in the present paper.

Furthermore, since aromas are basically FVOCS, other technologies might be considered by analogy with technologies commonly encountered for the VOCs capture in depollution processes. The review papers of Khan and Kr. Ghoshal (2000), of X. Wang, Daniels, and Baker (2001) and the book section of Revah and Morgan-Sagastume (2005) suggest that for non-destructive VOCs separation, the adsorption and absorption techniques can be applied, in addition to the condensation and membrane-based techniques. Incidentally, these latter works also compare different technologies regarding their applicability, considering the VOCs' concentrations and the gas flow rate to be treated.

Adsorption

The adsorption technology is based on the interaction of the component to attach itself (i.e. the adsorbate) to a solid substrate (i.e. the adsorbent). It is commonly admitted that the adsorbate is held on the adsorbent surface and/or adsorbent pores by Van der Waals forces, leading generally to a fast and reversible equilibrium.

From Khan and Kr. Ghoshal (2000), X. Wang et al. (2001) and Revah and Morgan-Sagastume (2005), it appears that the adsorption is the most appropriate technology for treating stream with a very low VOCs concentration. However, its main drawback is that a desorption step is required to regenerate the adsorbent. This regeneration step requires high temperatures to desorb the fixed components from the substrate, leading to a considerable energy cost. Moreover, certain polar components can be adsorbed irreversibly on the sorbent (e.g. alcohols on activated carbon), which can lead to the appearance of artefacts when the sorbent is reused (e.g. benzaldehyde and acetophenone from Tenax TA (Desmet, Schelfaut, Górecki & Sandra, 2009)).

The adsorption technology is used in some applications of the food-processing industry, but only for aroma recovery from liquid stream or for wastewater treatments. This technology is presented as used in batch mode. To the best of the authors' knowledge, no article or patent investigates gaseous aroma recovery.

A review of the main adsorption processes is presented in the paper of Soto, Moure, Domínguez, and Parajó (2011). It is focused especially on the framework of phenolic compound recovery. According to their paper, the substrates that may be used to reversibly capture VOCs are activated carbons, minerals (such as zeolites) and resins. In the literature, it is noticed that the activated carbon processes are the most used and studied, as illustrated by the works of Zuim et al. (2011), Carpiné, Dagostin, da Silva, Igarashi-Mafra, and Mafra (2013), Diban, Ruiz, Urtiaga, and Ortiz (2008) and Edris, Girgis, and Fadel (2003). Furthermore, a review of the activated carbon-based processes is proposed by Dabrowski, Podkoscielny, Hubicki, and Barczak (2005), while an analysis of less conventional processes is presented in the paper of Ahmaruzzaman (2008).

Absorption

In the absorption technique, a gas stream is put into contact with a liquid in order to transfer one or several gaseous components into the liquid phase. Such techniques are used

mainly as air depollution treatment to remove pollutant volatile components and they are commonly referred to as scrubbing techniques, while it seems that they are not used for aroma recovery in the food-processing framework. According to Trambouze and Euzen (2004), the most common absorbing devices are falling films, bubble columns, stirred tanks, static mixers, plate columns, packed columns and spray columns. For the VOCs absorption, the packed columns (e.g. see Freudenthal, Otterpohl, and Behrendt (2005)) and the spray columns (e.g. see Batcha, Yih, and Raghavan (2007)) are mainly encountered because they ensure the best transfer characteristics, which are strongly influenced by the interfacial area and contact time.

It is suggested that the absorption devices can be used as a single operation with a reactant dissolved in the liquid phase (to enhance the absorption and/or neutralise the dissolved VOCs, like humic substance for odour control in (Freudenthal et al., 2005)) or can be used with a non-reacting liquid. The absorption stage is generally coupled with a desorption device, in order to regenerate the absorbing liquid. It is worth mentioning that in several applications, the gas-liquid absorption device is used to pre-concentrate the stream in order to increase the removal efficiency of an ulterior device. Such a configuration was studied by X. Wang et al. (2001), using a packed column prior to a membrane separator.

However, the absorption processes are recognised for their high removal efficiency (up to 98%, according to Khan and Kr. Ghoshal (2000)), for their relatively simple equipment requirement as well as for their wide concentration and flow rate range. Their critical drawbacks have been described by Khan and Kr. Ghoshal (2000).

Their operating costs may be high due to the low VOCs concentration in the gas stream. In such a case, a large amount of absorbing liquid is required. Consequently, a very cheap liquid has to be used. Therefore, water is usually selected but it significantly limits the absorption process since mostly the hydrophilic VOCs are absorbed. As mentioned earlier, the alternative is to link the absorber with a desorption device (e.g. a distillation column), but that increases the investment and the operating costs. Furthermore, their removal efficiency is strongly dependent on the selected absorbing liquid (with any potential additive or reactant).

Not only does the absorbing liquid need a good absorption rate and capacity (depending on the sample diffusivity and the solubility, which are generally not well known), it also has to meet several other constraints such as the price (if not regenerated), the viscosity, the volatility, the non-flammability, the degradability (if regenerated), the toxicity, etc.

Furthermore, it must be generally recognised as safe and edible for use in food products. The selection of an optimum absorbing liquid is, therefore, very complicated, and thorough investigations must generally be carried out for each kind of gas stream to be treated.

As mentioned earlier in this paper, the composition of effluents in food-processing industries is complex and heterogeneous due to the variety of chemical function of their FVOCs. Although the technologies used for the recovery of these FVOCs from gaseous effluents are in limited number, the choice of the appropriate technology remains a critical step. This choice depends on various parameters (^{see Table 1}) such as the flow rate of the effluent to be treated, the concentration of VOCs in this effluent and of course the global cost of the implementation.

The recovery efficiency is defined here as the proportion of VOCs extracted compared to the initial quantity in the treated gas. According to this definition, it appears that in Table 1, all of the technologies detailed in this paper allow a 90% recovery efficiency. Absorption seems to be an adequate technique for the recovery of VOCs from gaseous effluents with high volumetric flow rate (up to 2800 m³/minute), while membrane technologies are recommended for the smallest flows (<50 m³/minute). The latter is also referenced for the treatment of gaseous effluents with low VOCs concentrations, while the other three are applicable on effluents with ranges of concentrations from 20 to 500 ppm and even larger.

Conclusion

Numerous technologies exist nowadays for the recovery of VOCs from industrial effluents. In food processing industries, this operation mainly concerns liquid effluents and is achieved through the application of technologies such as pervaporation, distillation, adsorption and solvent extraction. Gaseous effluent treatment is generally carried out in industrial environments for cleanup before release. The technologies that can be applied in the food processing industry for aroma recovery from gaseous effluents seem to be condensation, adsorption, absorption and vapour permeation. Vapour permeation is mainly used for chemical, petro-chemical and pharmaceutical industries, but its technical characteristics suggest a potential application for the food processing industry. The use of condensation is limited to effluents with high aroma component concentrations and is very expensive because of the cooling necessity. The adsorption technology requires a desorption step for the

recovery of the fixed aromas and the regeneration of the adsorbing phase. The absorption, although easy to set up, is also limited by numerous technical and financial constraints depending on the absorbent.

It is not suitable to compare these four technologies on the basis of their potential investment costs because of the variability of the required equipment (condenser unit, vacuum pump, refrigeration unit, membranes, etc.) for each of them. The comparison of operation costs per volume unit is more realistic. Therefore, the operation cost of adsorption seems to have the best cost/benefit ratio when taking into account the reached recovery efficiency and the charge in VOC of the treated effluent.

Thorough investigations are still required for the development of technologies that are applicable for the recovery of gaseous FVOCs in food processing, taking into account the range of concentration, the capacities, the solvent regeneration and the financial cost of the industrial implementation.

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Figure 1. Schematic representation of the flow diagram combining technologies that can be used for recovery of VOCs from gaseous effluents: (1) membranes technologies; (2) condensation; (3) absorption; (4) adsorption; (A) absorption tower; (B) solvent regenerator; (C) adsorption column.

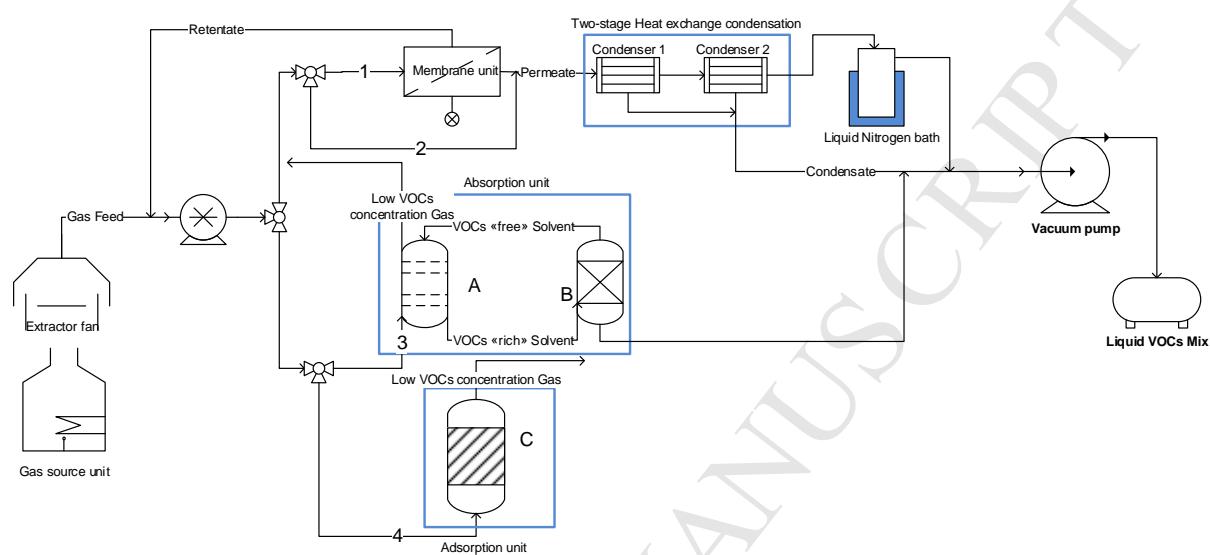


Figure 1.

Table 1. Comparison of the characteristics of the potential technologies for recovery of FVOCs from gaseous effluents in food-processing industries.

Technologies	Membranes Separation	Condensation	Adsorption	Absorption
Industrial applications	Environmental depollution; solvent recovery;	Environmental depollution; recovery of food aroma gases	Environmental depollution; capture of COVs from alcoholic fermentation	Environmental depollution; odour reduction
Type of VOCs	Alcohols; alkanes; aromatic hydrocarbons	Aromatic hydrocarbons; Ketones; aldehydes; alcohols; furan; Alkyl-pyrazines	Esters; Aldehydes; Alcohols; aromatic hydrocarbons	Alkanes; Aromatic hydrocarbons; chloro-hydrocarbons
Mediators	Polymer membranes	Liquid nitrogen	Activated carbon; porous resin	Water; high-boiling hydrocarbons
Volumetric flow rate range (m³/min)	6-42	3-560	3-170	60-2800
Recoveries efficiency	>90%	>95%	99%	95 to 98%
Ideal concentration range (ppm)	>30	2500-10000	800-10000	500 to 15000
Approximative investment cost (10⁴€)	17.5 to 25.6	60 to 70	14 to 26	/
Approximate annual operating cost (€/m³)	400-800	550-3500	280-1000	280-1000
Advantages	Cyclic operating; easy recycling of membranes; no additive required; no further treatment of recovered VOCs; operates under mild conditions	Ideal for high concentrated gas stream	Good recovery efficiency	Easy to set up; reuse of absorbent liquid; used in a wide range of concentration
Disadvantages	Costly and rarely available membranes; susceptibility of membranes to fouling and bacterial growth (inducing clogging and possibly VOCs alteration)	High energy consumption; important cooling fluid use; not suitable for compounds with boiling points above 37 °C	Less selectivity; poor regeneration of adsorbent; use of additive solvent for desorbing; susceptible to clog; not suitable for cyclic operation; require humidity control	Use of large amount of absorbing liquid; need of post-treatment for regeneration of absorbing liquid
References	(Ahlers & Rautenbach, 1994; Cha, Malik, Bhaumik, Li, & Sirkar, 1997; Degrève, Everaert, & Baeyens, 2001; Khan & Kr. Ghoshal, 2000; Majumdar, Bhaumik, & Sirkar, 2003; Suematsu et al., 1998)	(Barfuss, Mazurek, Rushmore, & Temperini, 2000; Budwig et al., 2002; Carns & Tuot, 1993; Degrève et al., 2001; Khan & Kr. Ghoshal, 2000)	(Bes; Degrève et al., 2001; Khan & Kr. Ghoshal, 2000)	(Freudenthal et al., 2005; Khan & Kr. Ghoshal, 2000)

Highlights

- Gaseous effluents treatment usually concerns cleanup before discharge.
- Little few information is available on recovery of food aromas from gaseous effluents.
- Vapour permeation, condensation, adsorption and absorption work for aroma recovery from gaseous effluents.
- The choice of an appropriate technology depends on its technical and financial constraints.