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Coupling nanofiltration and osmotic evaporation for the recovery of a natural flavouring concentrate from shrimp cooking juice

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ABSTRACT

A new combination of membrane-based processes was studied in order to produce a natural flavouring concentrate from shrimp cooking juice. The process associates a pre-concentration step by nanofiltration (NF) until a Volume Reduction Ratio (VRR) of 10 and a final concentration by osmotic evaporation (OE). Thanks to NF, the juice was partially concentrated and desalinated, allowing the further OE step to be run in optimal conditions: reduction of the quantity of water to be evaporated and of the production of brine, limitation of the loss of volatile compounds and improvement of sensory acceptability. During OE step run at pilot-scale, a 52% dry matter concentrate was obtained with aroma loss lower than 35%. Products were characterised by sensory and chemical analyses all along the process. With sensorial characteristics matching the expectancies, the concentrate could be incorporated at 2.5% in food preparations.

1. Introduction

To fulfil consumer's expectations for natural products, food industry strives to valorise natural resources and by-product streams allowing mention of “natural”, “clean label” on their range of products. Wastewaters like cooking juice are loaded in high added-value compounds for flavour industry (odorous volatile compounds, peptides, amino acids, nucleotides, organic acids and mineral compounds). To propose a complete shrimp flavour from shrimp cooking juices, both odorous volatile molecules and taste compounds are required.

Raw and fresh shrimps have a weak and mild fishy aroma. Various reactions like enzymatic hydrolysis, lipid oxidation, thermal reactions (Maillard reaction) and even environmental and microbial pollutions are involved to generate the familiar and characteristic shrimp aroma (Baek & Cadwallader, 1997). More than one hundred compounds have been found in shrimp aroma: pyrazines, unsaturated methylketones, sulphur-containing compounds and aldehydes seem to have a great impact on overall aroma (Kubota & Kobayashi, 1988; Kubota, Shuimaya, & Kobayashi, 1986; Kubota, Uchida, Kurosawa, Komuro, & Kobayashi, 1989). Baek and Cadwallader (1997) have determined 23 character-impact aroma compounds by an olfactometric method. Actually, the characteristic aroma of shrimps cannot be attributed to one single compound. It is a blend of molecules and deletion or over-concentration could affect sensory profile.

Taste (savoury) compounds complete flavour by enhancing aromas and giving length in mouth. Salted, sweet, acid, bitter and umami tastes are due to these non-volatile, low molecular weight and water soluble compounds. The umami taste is mainly given by sodium glutamate and reinforced by 5′-ribonucleotides. In addition to nucleotides, free amino acids, peptides, inorganic salts and sugars contribute to the crustacean and shellfish flavours (Fuke & Ueda, 1996; Spurvey, Pan, & Shahidi, 2000).

Pressure-driven membrane technologies have well-known potential to concentrate in favourable energetic conditions while simultaneously preserving product quality. In particular, nanofiltration (NF) especially seems to have a good potential of development for the recovery of valuable components from the effluents of seafood processing (Afonso & Borquez, 2002; Ferjani, Ellouze, & Ben, 2005) including both taste and aromas (Walha et al., 2011). Seafood cooking juices have been nanofiltrated to produce water with decreased chemical oxygen demand (COD) and to concentrate high valuable substances for flavour industry (Lin & Chiang, 1993; Vandanjon, Cros, Jaouen, Quéméneur, & Bourseau, 2002). But although the COD decrease was effective, the marine odour of the permeate revealed that aroma compounds permeated through the membrane (Vandanjon et al., 2002). With tighter membranes, reverse osmosis (RO) is expected to be more

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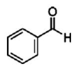
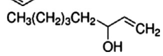
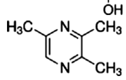
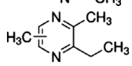
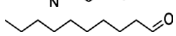
Table 1
Physical-chemical properties of the target flavour components.

	Structure	Chemical formula	MW ($g\ mol^{-1}$)	Molar volume ($Å^3$)	Boiling point (°C)	Log K_{ow}	Solubility in water 25 °C ($g\ L^{-1}$)	Vapour pressure 25 °C (Pa)
Benzaldehyde		C_7H_6O	106.12	101	178.62	1.48	6.95	169.0
1-Octen-3-ol		$C_8H_{16}O$	128.21	153.5	175.2	2.6	1.84	31.6
2,3,5 trimethylpyrazine		$C_7H_{10}N2$	122.17	124.7	171	0.95	15.2	80.9
3-Ethyl-2,5-dimethylpyrazine		$C_8H_{12}N_2$	136.20	–	180.5	2.07	1.47	99.0
Decanal		$C_{10}H_{10}O$	156.27	190.9	208.5	3.76	0.061	13.7

efficient than NF for the retention of low molecular weight solutes and aroma compounds. It has been studied for the concentration of various fruit juices in order to preserve nutritional and sensory qualities (Acosta, Vaillant, Perez, & Dornier, 2017). However, considering seafood cooking juices, salt is a limiting factor for concentration due to osmotic pressure (Vandanjon et al., 2002; Walha et al., 2011). Cros, Lignot, Razafintsalama, Jaouen, and Bourseau (2004); Cros, Lignot, Jaouen, and Bourseau (2006) proposed a preliminary desalination by electrodialysis that allowed organic matter to be concentrated up to 200 $g\ L^{-1}$. The process was recently evaluated economically by Bourseau, Massé, Cros, Vandanjon, and Jaouen (2014). However, such concentration level is still too low for flavour industry which demands concentrates that could be incorporated at 2% maximum in food preparations.

Osmotic evaporation (OE) seems promising to concentrate at dry matter (DM) content up to 60% while preserving most of the volatile compounds (Ali, Dornier, Duquenoy, & Reynes, 2003; Cissé et al., 2011). In OE, the solution to be concentrated is separated from a concentrated brine with low water activity by a porous hydrophobic membrane. The driving force for mass transfer is the difference in vapour pressure induced by the water activity gradient. The removal of water is commonly described by a three-step mechanism: (i) evaporation of water at the solution/membrane interface where vapour pressure is the highest, (ii) diffusion through the pores and (iii) condensation at the membrane/brine interface where vapour pressure is the lowest. The hydrophobic nature of the membrane combined with the low diameter of its pores prevent the penetration of liquid solution inside the membrane porosity. Thanks to the membrane contactor principle, the retention of non-volatile compounds is total and concentration levels higher than with pressure-driven membrane processes can be achieved. Few examples of industrial scale-up are known today, as OE is still limited by low evaporation flux especially at high DM content and by the production of diluted brine. Researches currently focus on these issues.

The purpose of this work is to combine the advantages of both technologies: in a first step, NF would help to concentrate the aromatic and savoury compounds without increasing salt concentration. A partially concentrated and desalinized concentrate would therefore be obtained. In the second step, OE would contribute to increase the DM content. Several advantages are expected from the preliminary NF: (i) partial concentration should reduce the quantity of water to be evaporated by OE and therefore limit the production of brine, (ii) second, partial desalination should increase sensory and nutritional acceptability and limit the evaporation of volatile compounds during OE. At each step, sensory quality of the retentate was considered in order to study the impact of each operation and to assess the end-product as a natural concentrated shrimp cooking juice. Whereas combining NF or RO with OE is reported for recovering aroma compounds or antioxidants in fruit juices (Cissé, 2010; Galaverna et al., 2008; Patil & Raghavao, 2007; Souza et al., 2013), there is no example of such combination regarding savoury juices, to the best of our knowledge.

2. Materials and methods

2.1. Raw material: shrimp cooking juice

A single batch of shrimp cooking juice from Capitaine Houat factory (Lorient, France) was constituted for all trials. It is a homogeneous blend of three cooking batches packed in plastic buckets and deep-frozen before storage at − 18 °C.

2.2. Chemical analyses

Dry matter (DM) was measured by desiccation in an oven at 105 °C up to constant mass (Memmert BE400 and BE600) and ashes by incineration at 550 °C during 7 h (Carbolite Advantec). Total nitrogen was analysed by Kjeldahl method. Chloride was measured by titration with $AgNO_3$ (DL53 Mettler). Analyses of amino-acids, sugars, organic acids and nucleic acids were done by HPLC in the Central Laboratory of Mane (Bar-sur-Loup, France). Analyses of cations (Ca, Mg, Na and K) by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) and of molecular weights distribution by gel filtration chromatography were assigned to an external laboratory. Other analyses included pH (Mettler Delta 340 pH meter), conductivity (340i WTW pH/conductivity meter) and turbidity (2100 AN Hach turbidimeter).

2.3. Flavour compounds analysis

Preliminary analysis by GC-MS allowed more than 70 volatile molecules to be identified among which 12 alcohols, 22 aldehydes, 10 ketones, 13 nitrogen compounds, 1 sulphur compound, 4 oxygenated compounds and 9 hydrocarbons.

Based on literature survey, it was decided to focus quantitative analysis on five molecules which play a major role in natural shrimp flavour and that could be easily quantified in the chromatograms (Table 1): benzaldehyde, 1-octen-3-ol, 2,3,5-trimethylpyrazine, 3-ethyl-2,5-dimethylpyrazine and decanal. 1-octen-3-ol is one of the most widely distributed volatile compounds in seafood products (Josephson, Lindsay, & Stuiber, 1984). Its mushroom odour is generally considered as desirable. Pyrazines have been reported as important flavour compounds in several seafood products; a strong impact of 2,3,5-trimethylpyrazine in the cooked shrimp aroma is noticed by Ishizaki, Tachihara, Tamura, Yanai, and Kitahara (2005).

To carry out SPME extraction, 10 mL of shrimp cooking juice were placed and sealed in 20 mL headspace vial (Supelco). Extraction temperature was 60 °C with 30 min of incubation time, followed by 30 min trapping and shaking, using a divinylbenzene/carboxen/polydimethylsiloxane fiber 30–50 μm (Supelco). Injections were automated

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