

# 1 Immersed membranes configuration for the microfiltration of fruit-based 2 suspensions

3  
4 **Keywords:** Fruit-based suspensions; Microfiltration; Immersed membranes; Productivity; Selectivity.

## 5 6 **Highlights:**

- 7 • Interest of using immersed membranes microfiltration for fruit-based suspensions
- 8 • Productivity in line with what is reported in other domains
- 9 • Selectivity goals towards compounds of interest reached
- 10 • Interesting alternative to conventional cross-flow filtration for small production units

## 11 12 13 **Abstract**

14 Microfiltration is widely used to ensure the athermal stabilization, clarification and concentration of various  
15 fruit-based suspensions (e.g. fruit juices, food by-products, wine). However, the performances of membrane  
16 filtration remain highly challenged by membrane fouling. To prevent membrane fouling, cross-flow  
17 filtration is generally performed. Nevertheless, this intensive working mode is considered as highly energy  
18 consuming due to the intensive pumping required to circulate the suspension at high velocities. In the light  
19 of this, immersed membranes configurations have been developed in many fields, as they allow working in  
20 energy-friendly operating conditions. Thus, this work investigated for the first time the performances of an  
21 immersed membranes configuration for fruit-based suspensions microfiltration, in terms of productivity  
22 (membrane fouling, permeate flux) and selectivity (clarification, concentration of bioactive compounds).  
23 This study focused on three fruit-based suspensions: a grapefruit juice and two winery by-products.  
24 Concerning the process selectivity, pilot-scale experiments showed that immersed membranes filtration  
25 allowed producing retentate and permeate of quality as least as good as the one related to conventional  
26 cross-flow filtration. Concerning the process productivity, cross-flow filtration allowed reaching higher  
27 fluxes compared to immersed membranes filtration, in accordance with the conventional order of magnitude  
28 specific to each configuration. Immersed membranes configuration could find interesting applications  
29 within small production units of fruit juices and/or industries dealing with the valorization of low added-  
30 value byproducts thanks to its various advantages (high compactness, easy handling and mobility, low  
31 investment and operational costs).

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33 **List of abbreviations and variables:**

<b>A</b>	Absorbance ( <i>I</i> )
<b>C</b>	Total monomeric anthocyanins content, malvidin-3-glucoside equivalent (mg.L <sup>-1</sup> )
<b>C<sub>i</sub></b>	Concentration of targeted compounds in the initial feed suspension
<b>C<sub>p</sub></b>	Concentration of targeted compounds in the permeate
<b>DF</b>	Dilution factor ( <i>I</i> )
<b>DM</b>	Dry Matter (g.L <sup>-1</sup> )
<b>J</b>	Permeate flux (m.s <sup>-1</sup> )
<b>l</b>	Path length (1.0 cm)
<b>MW</b>	Molecular weight of malvidin-3-glucoside (463.3 g.mol <sup>-1</sup> )
<b>NTU<sub>i</sub></b>	Turbidity of the initial feed suspension (NTU)
<b>NTU<sub>p</sub></b>	Turbidity of the permeate (NTU)
<b>PES</b>	Polyethersulfone
<b>C<sub>clarif</sub></b>	Clarification ratio (%)
<b>C<sub>ret</sub></b>	Retention ratio (%)
<b>R<sub>h</sub></b>	Total resistance to permeation (m <sup>-1</sup> )
<b>Red Extract</b>	Red grape pomace extract
<b>Seed Extract</b>	Grape seeds extract
<b>SIS</b>	Insoluble suspended solids
<b>TMP</b>	Transmembrane pressure (Pa)
<b>VRR</b>	Volume reduction ratio

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<b>ε</b>	Molar extinction coefficient of malvidin-3-glucoside (28 000 L.mol <sup>-1</sup> .cm <sup>-1</sup> )
<b>μ<sub>p</sub></b>	Permeate dynamic viscosity (Pa.s)
<b>μ<sub>s</sub></b>	Suspension dynamic viscosity (Pa.s)

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35 **1. Introduction**

36 Microfiltration is widely used to ensure the clarification and the concentration of various fruit-based  
37 suspensions such as fruit juices [1–4], agro-food by-products [5–7] or wine [8]. This solid-liquid separation  
38 technic allows producing high quality products thanks to its high selectivity and low operating temperatures.  
39 However, the performances of membrane filtration remain highly challenged by membrane fouling. Fruit-  
40 based suspensions, well known to be heterogeneous suspensions containing colloids and larger suspended  
41 insoluble solids (SIS) dispersed in a continuous aqueous phase, are considered as highly-fouling  
42 suspensions. During the microfiltration of such complex suspensions, particles deposition is considered as  
43 one of the main causes of membrane fouling [9,10]. This type of fouling is mainly governed by the balance  
44 between convective forces (permeate flow), leading particles to the membrane, and back-transport forces,  
45 removing particles away from the membrane surface [11].

46 So far, a wide range of filtration configurations have been studied for fruit juices microfiltration, such as  
47 cross-flow filtration using organic plane [3,12] or hollow-fiber [13,14] membranes, organic or inorganic  
48 tubular membranes [2,4,15–20]. In spite of the diversity of membrane shape and material and operating  
49 conditions, the use of high shear stress at the membrane surface is always a common feature to enhance the  
50 back-transport mechanisms and thus increase the permeate fluxes [21]. However, this intensive working  
51 mode is well known to be highly energy consuming due to the intensive pumping required to circulate the  
52 suspension at high velocities. Moreover, the use of important shear forces, leading to high turbulences, has  
53 been reported to induce particles size modifications [22,23] that could impact the suspension characteristics  
54 (fouling propensity, nutritional and sensorial properties).

55 In the light of this, immersed membranes configuration (out-to-in filtration) could be an interesting  
56 alternative for the microfiltration of fruit-based suspensions. In this configuration, the membrane (plane or  
57 hollow fiber) is immersed in the suspension and the filtration is generally ensured by permeate suction at  
58 constant flux. Thus, the filtration operation is conducted in conditions close to that of dead-end filtration,  
59 associated with limited back-transport forces and low permeation fluxes. Despite the low permeation fluxes  
60 commonly applied in such conditions, immersed membranes filtration have been widely and successfully  
61 used in many fields, notably for water treatment (e.g. drinking water production) and for wastewater  
62 treatment for the filtration of more heterogeneous suspensions (e.g. microalgae suspensions [24–26],  
63 activated sludge [27–30]). For these applications, the relatively low productivity is generally offset by the  
64 great packing density of the membrane , by the low cost of organic membranes and the low energy  
65 consumption of the process [31,32].

66 However, as far as the authors are aware, the performances of this filtration configuration remain little  
67 studied for fruit-based suspensions microfiltration. At present time, no studies have yet characterized the  
68 productivity and the selectivity of this filtration configuration for such applications. In the light of this, the  
69 aim of this work was to investigate the performances of an immersed membranes configuration for fruit-  
70 based suspensions microfiltration, in terms of membrane fouling and selectivity. This study focused on three  
71 different agro-food suspensions: a grapefruit juice, a grape pomace extract and a grape seeds extract. Firstly,  
72 a specific experimental strategy was conducted in order to define the optimal operating conditions of this  
73 system. Secondly, based on the previously identified operating conditions, filtration performances were  
74 analyzed in terms of membrane fouling and selectivity (clarification and/or concentration of targeted  
75 compounds). Finally, a comparison of immersed membranes configuration performances with conventional  
76 side-stream membranes configuration ones (cross-flow filtration) was proposed and discussed.

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## 78 **2. Material and methods**

### 79 **2.1. Fruit-based suspensions**

#### 80 **2.1.1. Selection**

81 Three agro-food suspensions were studied in this work: a grapefruit juice, a red grape pomace extract and a  
82 grape seeds extract.

83 Grapefruit juice was chosen as it is among the most popular citrus fruits worldwide [33,34]. Moreover, its  
84 microfiltration offers several applications in fruit-juices industries as it allows producing a high quality  
85 clarified and stabilized juice rich in phenolic compounds (mainly naringin and narirutin [35]) and a  
86 concentrated pulpy fraction rich in carotenoids (mainly lycopene and beta-carotene [36]). These products  
87 find useful applications in industries (pharmaceutical, cosmetic, food) thanks to their therapeutic, nutritional  
88 and sensorial properties.

89 Red grape pomace and grape seeds extracts were chosen for being among the main by-products (produced  
90 by considerable tonnage) by winery industries [37]. Their valorization is a major economic and ecological  
91 challenge, for which clarification is a key pre-treatment step. The microfiltration of red grape pomace and  
92 grape seed extracts allows producing a clarified permeate rich in phenolic compounds, notably flavonoids  
93 like tannins and anthocyanins in the case of red grape pomace extract [38,39]. This permeate can be easily  
94 valorized through additional extraction steps.

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#### 96 **2.1.2. Procurement, characterization and conservation**

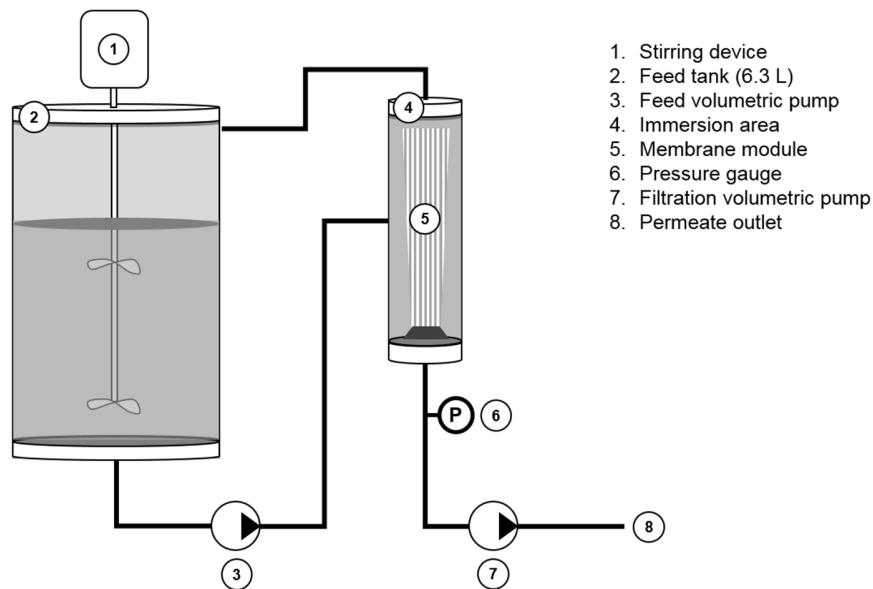
97 In this study, grapefruit juice was produced by squeezing Star Ruby grapefruit (*Citrus grandis* (L.) Osbeck)  
98 (Spain) in a semi-industrial extractor (Automatic orange juicer, model 32, SANTOS, Vaulx-en-Velin,  
99 France) and pre-filtered through a stainless steel sieve (1 mm mesh size). Red grape pomace extract (named  
100 Red Extract) and grape seeds extract (Seed Extract) were purchased from a local distillery located in the  
101 South of France. The extracts were obtained by industrial solid-liquid extraction in sulphited water, after  
102 grinding of the raw residues generated during winemaking (i.e. red grape pomaces and grape seeds). The  
103 three suspensions were stored at - 20 °C and thawed before use. Their main physicochemical and chemical  
104 characteristics (dry matter (DM), turbidity (NTU), pH, Brix degree (°Brix), dynamic viscosity ( $\mu_s$ ) and  
105 suspended insoluble solids (SIS)) were determined according to the protocols and methods described by  
106 [37,40].

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108 **2.2. Immersed membranes filtration experiments**

109 **2.2.1. Experimental set-up**

110 A schematic illustration of the experimental equipment is presented in figure 1. It consisted of a 6.3 L stirred  
111 stainless steel feed tank, linked to a 1.8 L external filtration unit containing immersed organic hollow-fiber  
112 membranes (main characteristics given in table 1). A low flow-rate pump (item 3 on figure 1, 520S IP31  
113 peristaltic pump, Watson-Marlow, Massachusetts, USA) allowed the juice flowing through the filtration  
114 unit (flow velocity of  $3.5 \times 10^{-2} \text{ m.s}^{-1}$ , corresponding to Reynolds number of around 500 in the external  
115 filtration unit with hollow-fiber membranes and to a  $3 \text{ s}^{-1}$  shear rate at the membrane surface). A cryostat  
116 connected to a water jacket on the recycling loop, maintained the system at a constant temperature of  $25 \pm$   
117  $2 \text{ }^\circ\text{C}$ .



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119 **Figure 1:** Schematic illustration of the immersed membranes microfiltration pilot unit.

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**Table 1: Main characteristics of the immersed membranes.**

<b>Configuration</b>	Hollow-fiber
<b>Material</b>	PES
<b>Average pore size (<math>\mu\text{m}</math>)</b>	0.1
<b>Intrinsic membrane resistance <math>R_m^*</math> (<math>\text{m}^{-1}</math>)</b>	$2.9 \times 10^{11}$
<b>Water permeability * (<math>\text{L} \cdot \text{h}^{-1} \cdot \text{m}^2 \cdot \text{bar}^{-1}</math>)</b>	1240
<b>Manufacturer</b>	Polymem (France)
<b>Filtration area (<math>\text{m}^2</math>)</b>	$1.8 \times 10^{-1}$

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\* Experimental measurement at 25 °C

130 The out-to-in permeation flow was ensured by pump suction (item 7 on figure 1, 621F/RE IP55 peristaltic  
131 pump, Watson-Marlow, Massachusetts, USA) at constant flux ( $J$ ). The evolution of membrane fouling  
132 during the filtration runs was estimated through the monitoring of the transmembrane pressure evolution  
133 (TMP) by an Almemo 2690-8 computer-controlled device (Ahlborn GmbH, Germany) connected to a  
134 pressure sensor. Indeed, according to Darcy's law (equation 1), in constant flux experiments, an increase of  
135 TMP is directly related to an increase of the resistance to permeation:

$$136 \quad J = \frac{TMP}{\mu_p \cdot R_h} \quad (1)$$

137 With  $J$  the permeate flux ( $\text{m} \cdot \text{s}^{-1}$ )

138  $TMP$  the transmembrane pressure (Pa)

139  $\mu_p$  the permeate dynamic viscosity (Pa.s)

140  $R_h$  the total resistance to permeation ( $\text{m}^{-1}$ ).

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### 142 **2.2.2. Definition of the optimal operating filtration conditions: flux-stepping experiments**

143 Pre-filtration tests were performed to define the optimal operating flux of the immersed membranes filtration  
144 system for each suspension. As stated before, membrane fouling is mainly governed by the equilibrium  
145 between convective and back-transport forces. Even if important permeate fluxes are generally needed to  
146 ensure the process sustainability, excessive fluxes can be counterproductive as they lead to important  
147 convective transports of foulant particles toward the membrane surface. Choosing an optimal permeate flux  
148 is therefore of crucial interest to control membrane fouling. Among all flux concepts that have been studied  
149 to guide permeate flux selection, critical and threshold fluxes concepts are particularly adopted when dealing  
150 with immersed membranes filtration [21].

151 Critical flux is defined as being the flux below which TMP remains strictly constant. According to many  
152 studies, critical flux is generally really low and its determination is time consuming [21,41,42], which limits  
153 its industrial application. In the light of this, the authors focused on the concept of threshold flux, defined  
154 as being the flux at which the rate of fouling increases significantly. This flux concept is more applicable  
155 for industrial applications, as less time is required for its determination and it generally matches with  
156 acceptable value of fluxes.

157 Threshold flux identification was based on progressive increasing flux-steps under total recycle mode  
158 (retentate and permeate were systematically returned to the feed tank) and on simultaneous TMP monitoring.  
159 In this work, the filtration was initially operated at a constant permeate flux of  $2 \text{ L.h}^{-1}.\text{m}^{-2}$  for 10 minutes.  
160 After 10 minutes, the flux was increased and the filtration was operated for another 10 minutes. Like so,  
161 flux was gradually increased at 10-minutes intervals. The values of flux and flux-steps duration were chosen  
162 according to values reported in the literature [43–45].

163 Based on the experimental results, the fouling rate increase was evaluated through the determination of  
164  $d\text{TMP}/dt$  values for each constant flux-step, representing the TMP increase during the last 5 minutes of each  
165 flux-step. A threshold value of  $d\text{TMP}/dt = 1.0 \times 10^{-5} \text{ bar.s}^{-1}$  was chosen in accordance with values used in the  
166 literature [43–46]. Thus, when  $d\text{TMP}/dt$  remained lower than  $1.0 \times 10^{-5} \text{ bar.s}^{-1}$ , fouling was considered as  
167 being low. On the opposite, fouling increase was considered significant when  $d\text{TMP}/dt$  values were higher  
168 than  $1.0 \times 10^{-5} \text{ bar.s}^{-1}$ . Therefore, the threshold flux value was defined as being the highest flux at which  
169  $d\text{TMP}/dt$  remained lower than  $1.0 \times 10^{-5} \text{ bar.s}^{-1}$ .

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### 171 **2.2.3. Immersed membrane filtration performances**

172 Once the threshold flux was identified for each suspension, filtration experiments under batch concentration  
173 mode (until Volume Reduction Ratio of 2) were performed at constant flux below or equal to the identified  
174 threshold flux (sub-critical conditions).

175 The performances of immersed membranes configuration in terms of *membrane fouling* were estimated  
176 through the monitoring of the TMP during the filtration running.

177 The performances of immersed membranes configuration in terms of *selectivity* were evaluated by  
178 performing sampling on the initial feed suspensions and on the permeates resulting from their filtration  
179 under batch concentration mode. Depending on the suspension, various separation objectives were expected  
180 from microfiltration experiments (table 2).

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**Table 2:** Separation objectives related to the microfiltration of grapefruit juice, Red and Seed Extracts

Suspension	Grapefruit juice	Red Extract	Seed Extract
Separation objectives	Clarified permeate rich in total phenolic compounds  Retentate concentrated in carotenoids ( <i>lycopene</i> and <i>β-carotene</i> )	Clarified permeate rich in total phenolic compounds and anthocyanins	Clarified permeate rich in total phenolic compounds

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The ability of immersed membranes filtration to produce a clarified permeate was measured through the analysis of turbidity loss, evaluated through the clarification ratio  $C_{clarif}$  (%), calculated as follows (equation 2):

$$C_{clarif} = 1 - \frac{NTU_p}{NTU_i} \times 100 \quad (2)$$

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The selectivity of the filtration towards valuable compounds was characterized through the retention ratio  $C_{ret}$  (%) representing the percentage of targeted compound retained by the membrane (equation 3):

$$C_{ret} = 1 - \frac{C_p}{C_i} \times 100 \quad (3)$$

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With  $NTU_p$  and  $C_p$  the turbidity and the concentration of targeted compounds in the permeate  
 $NTU_i$  and  $C_i$  the turbidity and the concentration of targeted compounds in the initial feed suspension.

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In the light of the above, various physicochemical and biochemical analyses were carried out for each suspension and related permeate.

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**Turbidity measurements** were performed on water diluted extracts to fall in the turbidimeter precision domain ranging between 0 and 50 NTU (Hanna LP 2000, Hanna instruments, Szeged, Hungary).

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**Total phenolic content** was measured by spectrophotometry, according to a modified Folin Ciocalteu method. Firstly, the suspensions were prepared in an ethanol/distilled water (25:75, v/v) solution. After the addition of 184 μL of distilled water, 24 μL of sample, 12 μL of Folin Ciocalteu reagent and 30 μL of 20



205 % (w/v) Na<sub>2</sub>CO<sub>3</sub> solution in a 96-well microplate (MultiSkan Spectrum, Thermo Scientific), the resulting  
206 mixture was incubated at 25 °C in the darkness for 1h. The absorbance was then measured at 765 nm. Gallic  
207 acid was used as a standard for calibration. Results were expressed as milligrams gallic acid equivalent  
208 (GAE) per liter of sample.

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210 **Total monomeric anthocyanins content** was determined using the pH differential method [47]. The sample  
211 absorbance was measured at pH 1.0 and 4.5 at 510 nm (the wavelength of maximum absorbance) and at 700  
212 nm to correct haze. Measurements were performed using a spectrophotometer (UV 2450, Shimadzu, Kyoto,  
213 Japan). Total monomeric anthocyanins were expressed as follows, as malvidin-3-glucoside equivalent  
214 (mg.L<sup>-1</sup>) [48] (equation 4):

$$C = \frac{A \times MW \times DF}{\epsilon \times l} \times 10^3 \quad (4)$$

215  
216 With *MW* the molecular weight (463.3 g.mol<sup>-1</sup>) and  $\epsilon$  the molar extinction coefficient  
217 (28 000 L.mol<sup>-1</sup>.cm<sup>-1</sup>) of malvidin-3-glucoside, respectively

218 *DF* the dilution factor

219 *l* the path length (1.0 cm)

220 *A* the sample absorbance, calculated as follows (equation 5):

$$A = (A_{510} - A_{700})_{pH1.0} - (A_{510} - A_{700})_{pH4.5} \quad (5)$$

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223 **Carotenoids (lycopene and  $\beta$ -carotene) content** was evaluated thanks to an extraction step followed by  
224 HPLC analysis. Carotenoids were firstly extracted from the samples through two successive extraction steps  
225 using ethanol/hexane (4/3 (v/v) containing 0.1% of BHT as antioxidant) as extraction solvent, under stirring  
226 [49,50]. At each step, residue was separated from the liquid phase by filtration using an n°2 porosity filter  
227 funnel. Ethanol and hexane were successively used to wash the residue. Organic phases were transferred to  
228 a separating funnel and successively washed with 10% sodium chloride and distilled water. The aqueous  
229 layer was removed and the hexanic extract was collected and dried with a rotary evaporator at 30 °C. The  
230 dried carotenoids extracts were then dissolved in 1 mL of dichloromethane/methyl *tert*-butyl ether/methanol  
231 solution (50/40/10 (v/v/v)). Secondly, HPLC analysis of carotenoids were conducted as described by  
232 Polidori *et al.* (2018) using an Agilent 1100 liquid chromatograph (Massy, France) equipped with a  
233 photodiode array detector and a C<sub>30</sub> separation column (250 × 4.6 mm i.d., 5  $\mu$ m YMC, EUROP GmbH,  
234 Germany) [50].

235

## 236 **3. Results**

### 237 **3.1. Feed suspensions characterization**

238 The main physicochemical and chemical characteristics (dry matter (DM), turbidity (NTU), pH, Brix degree  
239 (°Brix), dynamic viscosity ( $\mu_s$ ) and suspended insoluble solids (SIS)) of the three fruit-based suspensions  
240 are given in table 3.

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242  
243 *Table 3: Physicochemical characteristics of the studied suspensions.*

	<b>Grapefruit juice</b>	<b>Red Extract</b>	<b>Seed Extract</b>
<b>DM (g.L<sup>-1</sup>)</b>	105.2 ± 0.2	34.5 ± 0.2	22.6 ± 0.3
<b>Turbidity (NTU)</b>	3720 ± 230	1000 ± 70	1560 ± 250
<b>pH (/)</b>	3.1 ± 0.1	3.6 ± 0.1	4.3 ± 0.1
<b>°Brix (g/100 g)</b>	9.9 ± 0.1	3.6 ± 0.1	3.1 ± 0.1
<b><math>\mu_s</math> (mPa.s)</b>	1.7 ± 0.1	1.4 ± 0.1	1.5 ± 0.1
<b>SIS (g.L<sup>-1</sup>)</b>	1.5 ± 0.1	1.6 ± 0.1	1.5 ± 0.1

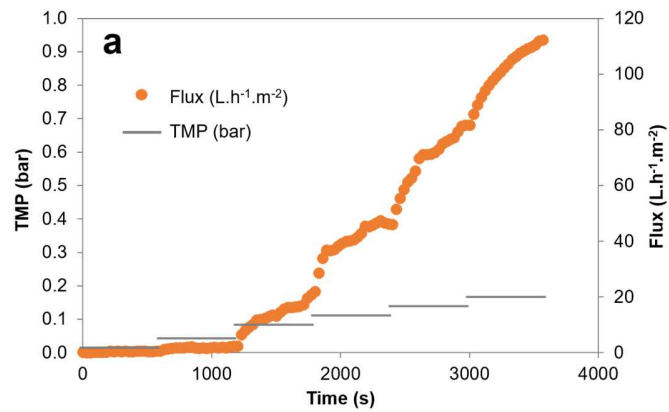
245 *All assays were performed at 25 ± 2 °C and values provided are the average of three replicates.*

246  
247 Among all studied products, grapefruit juice appeared to be the most different suspension with high  
248 turbidity, DM and Brix values. The two winery byproducts were quite similar, apart from a slightly lower  
249 DM value for Seed Extract and a higher turbidity value for Red Extract. These observations are consistent  
250 with the different raw materials and manufacturing processes related to each suspension. For the three  
251 suspensions, the SIS represented a very small part of the total dry matter with low SIS/DM ratios. It is a  
252 coherent observation, since a great part of their dry matter consists in sugar, estimated through the °Brix.

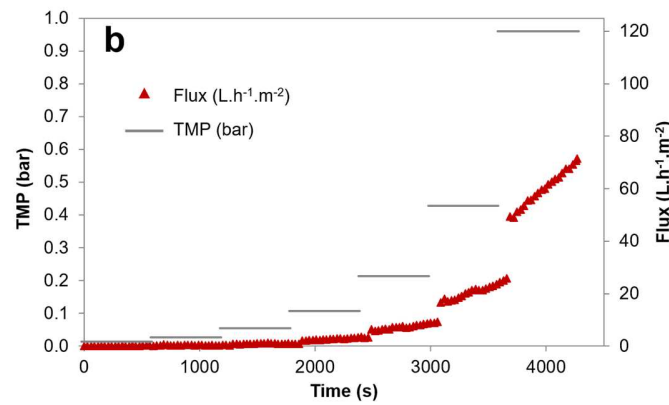
### 254 **3.2. Definition of the optimal operating domain: flux-stepping experiments**

255 Figure 2 presents the TMP evolution as function of time during flux-stepping experiments performed on  
256 grapefruit juice (a), Red (b) and Seed Extracts (c).

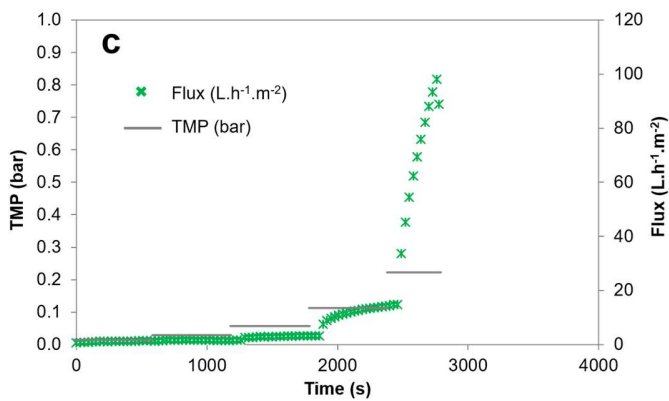
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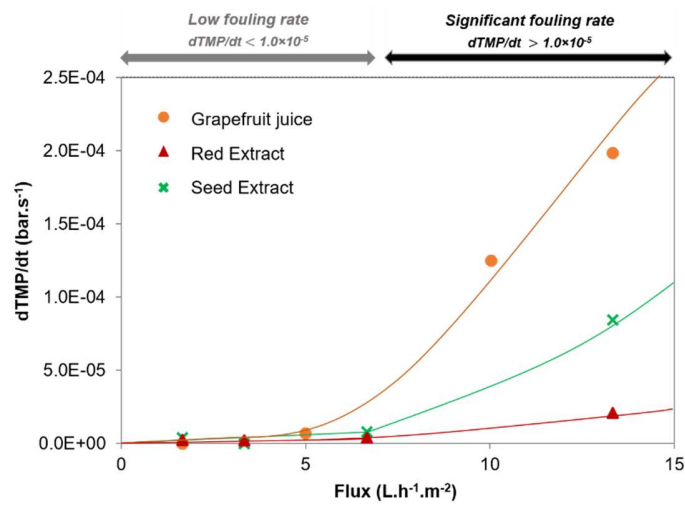


260 **Figure 2:** TMP versus time during flux-stepping experiments of grapefruit juice (a), Red Extract (b) and Seed Extract (c) using 0.1  
261  $\mu\text{m}$  PES hollow-fiber membranes.

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263 For the three suspensions, it can be noticed that the TMP progressively increased in greater or lesser degree  
264 for each flux-step, showing that fouling evolved differently depending on the imposed operating flux. Based  
265 on these experimental results, the increase of the fouling rate was evaluated through the determination of  
266  $d\text{TMP}/dt$  values for each constant flux-step and each suspension (figure 3).

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269 **Figure 3:** *dTMP/dt versus flux during flux-stepping experiments of grapefruit juice, Red Extract and Seed Extract using 0.1 μm PES*  
270 *hollow-fiber membranes. Lines drawn to guide the eye.*

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273 For low operating fluxes, a similar fouling behavior was observed for the three suspensions with values of  
274  $dTMP/dt$  lower than the threshold value of  $1.0 \times 10^{-5} \text{ bar.s}^{-1}$  (low fouling rate). Contrariwise, once specific  
275 values of fluxes were exceeded, different fouling behaviors were identified. Concerning winery byproducts,  
276 a significant increase of the fouling rate was observed between 7 and 13  $\text{L.h}^{-1}.\text{m}^{-2}$ , with  $dTMP/dt$  values  
277 reaching  $2.1 \times 10^{-5}$  and  $8.5 \times 10^{-5} \text{ bar.s}^{-1}$  for Red and Seed Extracts, respectively. Therefore, the threshold flux  
278 was defined as being around 7  $\text{L.h}^{-1}.\text{m}^{-2}$  for both winery byproducts in the studied operating conditions.  
279 Concerning grapefruit juice, the significant increase of the fouling rate was observed between 5 and 10  $\text{L.h}^{-1}.$   
280  $\text{m}^{-2}$  fluxes, with a  $dTMP/dt$  increase reaching  $1.3 \times 10^{-4} \text{ bar.s}^{-1}$ . Therefore, a threshold flux of around 5  $\text{L.h}^{-1}.$   
281  $\text{m}^{-2}$  was identified for this suspension.

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283 It can be noticed that the threshold flux related to grapefruit juice seemed slightly lower than the ones  
284 identified for Red and Seed Extracts. Moreover, the fouling rate increased differently depending on the  
285 suspension. Above threshold flux,  $dTMP/dt$  values were substantially higher for grapefruit juice than for  
286 Seed Extract or Red Extract. These differences are not surprising since each suspension is characterized by  
287 specific biochemical and physicochemical properties. This result highlights the important impact of the  
288 suspension characteristics on membrane fouling and fouling mechanisms. However, despite these  
289 differences, the threshold fluxes of the three studied suspensions were in line with the threshold values  
290 reported for immersed membranes filtration in other fields. Indeed, threshold fluxes ranging between 5 and

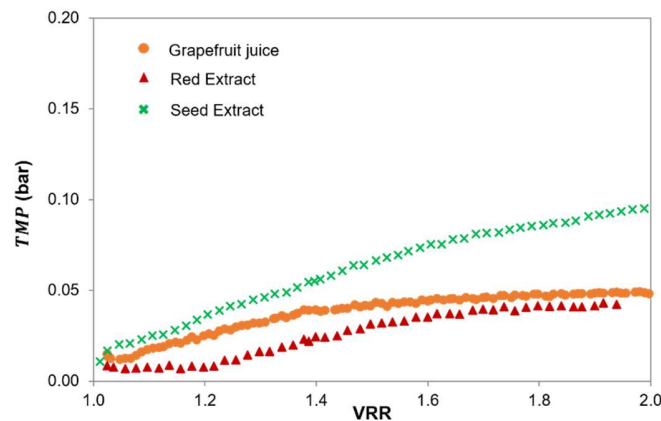
291 10 L.h<sup>-1</sup>.m<sup>-2</sup> were reported during immersed membranes filtration of milk protein concentrate solutions [43],  
292 activated sludge [44], alginate, yeast and bentonite solutions [45].

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### 294 3.3. Immersed membrane filtration performances

#### 295 3.3.1. Membrane fouling

296 For all suspensions, the membrane fouling evolution in immersed membranes configuration was evaluated  
297 in sub-critical conditions, sustainable conditions for long-time filtration running. With a view to comparing  
298 the fouling behavior of the three suspensions, a same constant flux of 5 L.h<sup>-1</sup>.m<sup>-2</sup> was chosen as operating  
299 flux. Figure 4 presents the evolution of TMP as function of VRR during filtration of grapefruit juice, Red  
300 and Seed Extracts.



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302 *Figure 4: TMP as function of VRR during filtration of grapefruit juice, Red extract and Seed Extract (batch concentration mode).*

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304 For the three suspensions, a continuous increase of TMP was observed between VRR 1 and VRR 2,  
305 reflecting an increase in membrane fouling.

306 Different fouling behaviors can be distinguished for the three suspensions. At the beginning of the filtration  
307 (between VRR 1 and 1.2), TMP remained almost constant for Red Extract with TMP surrounding  $8.0 \times 10^{-3}$   
308 bar and  $dTMP/dt$  close to zero. Contrariwise, an important and immediate fouling was observed for Seed  
309 Extract and grapefruit juice, with TMP increasing from  $1.0 \times 10^{-2}$  to  $3.7 \times 10^{-2}$  bar and from  $1.5 \times 10^{-2}$  to  $2.5 \times 10^{-2}$   
310 bar, respectively. Starting VRR 1.2, fouling increased for Red Extract while it stabilized for grapefruit  
311 juice leading to a final TMP of around  $4.0 \times 10^{-2}$  bar at VRR 2 for both suspensions. Concerning Seed  
312 filtration, a continuous fouling was observed throughout the remainder of the filtration operation, with TMP  
313 reaching around  $1.0 \times 10^{-1}$  bar at VRR 2.

314 These differences of fouling behavior might be related to the physicochemical characteristics of each  
315 suspension (table 3). Indeed, the three suspensions presented different dynamic viscosities, pH, DM, Brix  
316 and turbidity and these parameters are known for having a significant impact on membrane fouling [40].  
317 However, considering the complexity of these biological suspensions, these physicochemical properties are  
318 not sufficient to explain the different fouling behaviors of the studied suspensions. Indeed, membrane  
319 fouling results from complex interactions between the suspension compounds and the membrane and  
320 between the suspension compounds themselves [51,52]. These phenomena are governed not only by the  
321 operating conditions of the filtration operation and the physicochemical characteristics of the suspension,  
322 but also by the biochemical nature of the suspension compounds. Hence, the differences of fouling behavior  
323 between the studied suspensions might also be strongly related to the specific biochemical properties of  
324 each suspension.

325  
326 In order to estimate the importance of the fouling rate increase during a running operation,  $dTMP/dt$  were  
327 calculated for each suspension. Between VRR 1 and VRR 2,  $dTMP/dt$  values remained lower than the  
328 threshold value for the three suspensions, with  $dTMP/dt$  of  $5.3 \times 10^{-6}$ ,  $5.5 \times 10^{-6}$  and  $5.9 \times 10^{-6}$   $\text{bar} \cdot \text{s}^{-1}$  for  
329 grapefruit juice, Red and Seed Extracts, respectively. In the light of this, it seems that the fouling rates  
330 remained sustainable throughout the filtration operation, in batch concentration mode. This observation  
331 supports the interest of using the critical or threshold flux concept as pre-filtration test in order to define the  
332 optimal operating conditions prior to effective filtration experiments.

333

### 334 **3.3.2. Membrane selectivity**

335 The selectivity of immersed membranes was evaluated through the characterization of turbidity loss  
336 (clarification), the retention of total phenolic compounds for the three suspensions, and the retention of  
337 carotenoids for grapefruit juice.

338 Concerning clarification efficiency,  $C_{clarif}$  of 99.9 %, 99.5% and 99.3 % were obtained for grapefruit juice,  
339 Red and Seed Extracts, respectively, with permeates turbidities lower than 5 NTU for the three suspensions.  
340 These results are in accordance with values reported in the literature during microfiltration of melon and  
341 pomegranate juices [2,12]. For the three suspensions, immersed membranes filtration allowed decreasing  
342 significantly the initial turbidity of the suspension leading to a clarified permeate.

343

344 Concerning phenolic compounds, initial feed suspensions presented very different concentrations of total  
 345 phenolic compounds, with 145, 4076 and 5517 mg GAE per liter of grapefruit, Red Extract and Seed Extract,  
 346 respectively. These quantitative differences between grapefruit juice and winery byproducts are consistent  
 347 since winery byproduct are known for their particular richness in phenolic compounds [38,39]. As  
 348 mentioned before, these phenolic compounds include, among others, naringin and narirutin for grapefruit  
 349 juice, tannins for Red and Seed Extracts and anthocyanins for Red Extract. Anthocyanins were also  
 350 quantified in Red Extract and there concentration was around 560 mg malvidin-3-glucoside.L<sup>-1</sup> which is in  
 351 the range of what is generally reported in the literature (from 300 to 900 mg malvidin-3-glucoside.L<sup>-1</sup>)  
 352 [48,53]. These differences concerning at the same time the type and the concentration of bioactive  
 353 compounds in each suspension could have an impact on the membrane selectivity during the filtration  
 354 operation. Table 4 presents the  $C_{ret}$  regarding bioactive compounds specific to each studied suspension.

355

356 **Table 4:** Retention ratios (%) regarding total phenolic compounds, anthocyanins,  $\beta$ -carotene, lycopene during grapefruit juice,  
 357 Red and Seed Extracts microfiltration.

358

	Grapefruit juice	Red Extract	Seed Extract
Total phenolic compounds	8	14	3
Anthocyanins	/	17	/
$\beta$ -carotene	96	/	/
Lycopene	96	/	/

359 Regardless of the suspension, phenolic compounds (including anthocyanins) were mainly recovered in the  
 360 permeate with low  $C_{ret}$  ranging between 3 and 17 %. Similar results have been reported during  
 361 microfiltration of pineapple juice (retention of 7 % of total phenolic content [14]), pomegranate juice  
 362 (retention of 16.5 % of total polyphenols and 11.7 % of anthocyanins [54]), red raspberry juice (retention of  
 363 16% of anthocyanins [17]). This is a consistent observation since most of phenolic compounds are smaller  
 364 than the membrane pore size (0.1  $\mu$ m) with molecular weight generally ranging between 300 and 3000 Da  
 365 [55], even though some highly polymerized tannins (condensed tannins) have been reported to reach 20,000  
 366 Da [56]. However, membrane selectivity to phenolic compounds cannot be discussed only in terms of their  
 367 nominal molecular weight. Indeed, in the case of winery byproducts, low molecular weight phenolic  
 368 compounds can interact with each other or with other compounds, leading to the formation of complexes  
 369 with higher average size (e.g. tannin-anthocyanin, anthocyanin–anthocyanin, tannin-proteins complexes)  
 370 [8]. Thus, the higher  $C_{ret}$  observed for Red Extract phenolic compounds (14 %) and anthocyanins (17 %)  
 371 could be explained by complexation phenomena, leading to the formation of compounds larger than the

372 membrane pore size. The partial retention of phenolic compounds and anthocyanins could also be related  
373 their adsorption on/in the membrane layer [51].

374  
375 Concerning carotenoids ( $\beta$ -carotene and lycopene), the concentration of lycopene was higher than that of  
376 beta-carotene in grapefruit, with respective concentrations of 1.75 et 0.18 mg.L<sup>-1</sup>. These differences of  
377 concentrations did not have an influence on  $C_{ret}$  ratios, which were greater than 96 % for both compounds.  
378 Despite their relatively low molecular weight (536.87 g.mol<sup>-1</sup>), almost all carotenoids were retained by the  
379 membrane. This phenomenon has been reported in many studied and is explained by the strong association  
380 of carotenoids with the cell fragments membranes (i.e. pulp) due to their hydrophobicity [2]. Therefore,  
381 carotenoids are mainly concentrated in the retentate during fruit juice microfiltration [2,50].

382  
383 In the light of the above, immersed membranes filtration allowed producing (i) a permeate rich in phenolic  
384 compounds and clarified for the three suspensions and (ii) a retentate concentrated in carotenoids for  
385 grapefruit juice.

#### 386 387 **3.4. Comparison with conventional cross-flow filtration using side-stream membranes**

388 The previous analysis allowed characterizing the performances of immersed membranes configuration in  
389 terms of productivity and selectivity. In order to validate the efficiency of immersed membranes  
390 configuration for fruit-based suspensions microfiltration, previous productivity and selectivity results were  
391 compared with the performances of cross-flow filtration using side-stream tubular membranes. Indeed, this  
392 filtration configuration remains the most used for fruit juices microfiltration [2,16–20]. Cross-flow filtration  
393 experiments were performed under batch mode concentration (until VRR 2), using operating conditions  
394 generally used when dealing with fruit-based suspensions microfiltration (0.1  $\mu$ m inorganic tubular  
395 membranes, constant pressure of 1.5 bar [2,16,18,57]). The cross-flow velocity in each membrane and in  
396 the loop tubes (with diameter close to the membrane one) was around 5 m.s<sup>-1</sup>, corresponding to a Reynolds  
397 number and a membrane shear rate of 7 300 and 22 000 s<sup>-1</sup>, respectively (turbulent flow) ([57]).

398 Steady-state fluxes (flux decay was observed for the three suspensions, followed by a pseudo-equilibrium  
399 of the flux) of 80 L.h<sup>-1</sup>.m<sup>-2</sup>, 30 L.h<sup>-1</sup>.m<sup>-2</sup> and 24 L.h<sup>-1</sup>.m<sup>-2</sup> were obtained for grapefruit juice, Red and Seed  
400 Extracts, respectively. These different flux values are in line with permeate flux values reported during  
401 microfiltration of citrus fruit juices (20 – 80 L.h<sup>-1</sup>.m<sup>-2</sup> [57,58]) and winery byproducts (35 L.h<sup>-1</sup>.m<sup>-2</sup> [38,59]).  
402 However, it is interesting to note that the ranking of the fouling potential obtained during cross-flow  
403 filtration experiments was slightly different than the one observed during immersed membranes filtration  
404 experiments. While Red Extract presented a low fouling potential close to that of grapefruit juice during



405 immersed membranes filtration experiments, it was one of the most foulant suspension during cross-flow  
406 filtration experiments, with low flux ( $30 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$ ) compared to grapefruit juice. These differences of fouling  
407 behavior might be related to the different hydrodynamic conditions of each studied filtration configuration.  
408 However, an extensive characterization of the fouling behaviors of these suspensions (fouling mechanisms,  
409 involved particles, etc.) while using immersed membranes filtration or cross-flow filtration is necessary to  
410 go further on this path.

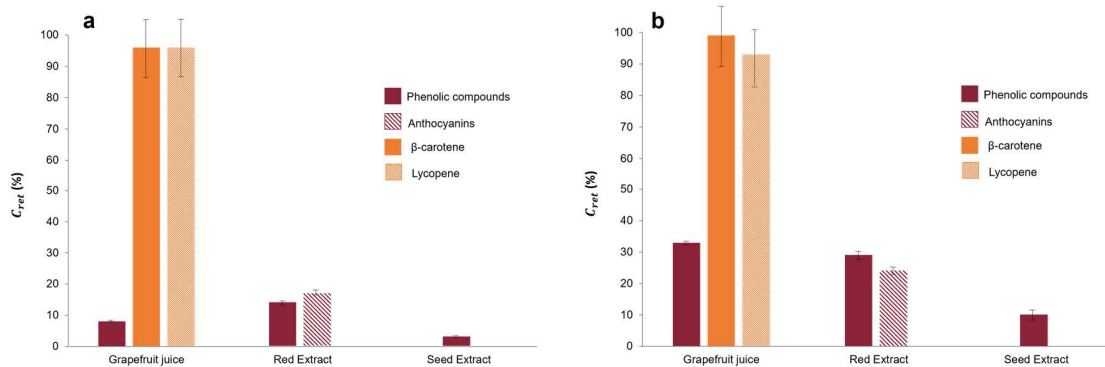
411 Concerning productivity, fluxes obtained for the three suspensions during cross-flow filtration were much  
412 higher (5 to 16 times greater) than the threshold flux of  $5 \text{ L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$  identified during immersed membranes  
413 filtration. These differences of order of magnitude are in line with the fluxes generally reported for these  
414 two configurations. Xue *et al.* (2015) compared the performances of an immersed membranes system with  
415 a side-stream one (cross-flow) during waste leachate treatment and reported fluxes almost 20 times higher  
416 when using cross-flow filtration [60]. Indeed, cross-flow filtration is an intensive process that works with  
417 high shear rates to ensure a high productivity but requires high energy consumption and investment cost.  
418 On the opposite, immersed membranes system can be considered as an extensive process (gentle operating  
419 conditions, low fluxes) for which productivity is generally improved by increasing the membrane surface  
420 thanks to low investment costs and limited energy consumption [61,62]. Despite its relatively low  
421 productivity, immersed membranes configuration could offer interesting applications in fruit juices and  
422 agro-food byproducts processing. Indeed, this simple processing system might be a convenient filtration  
423 configuration for small agro-food producers with limited investment capacities or for the valorization  
424 byproducts with low added-value. However, further analysis are needed to confirm these suggestions,  
425 notably concerning the process selectivity.

426

427 In order to compare the selectivity of both configurations,  $C_{clarif}$  and  $C_{ret}$  were calculated for cross-flow  
428 filtration experiments, based on analysis performed on the permeates collected at the end of the previous  
429 filtration experiments (batch concentration mode) and on the feed suspensions.  $C_{clarif}$  similar to the ones  
430 obtained for immersed membranes filtration were observed at the end of cross-flow filtration experiments  
431 (99.8 %, 96.4 % and 97.3 % for grapefruit juice, Red and Seed Extracts, respectively). Concerning  $C_{ret}$ ,  
432 slight differences were observed between immersed membranes filtration and cross-flow filtration (figure  
433 5).

434

435



436  
437 **Figure 5: Comparison of immersed membranes filtration (a) and cross-flow filtration (b) selectivity.**

438  
439 Concerning grapefruit juice carotenoids, both configurations presented very close  $C_{ret}$  (93 – 99 %). As  
440 during immersed membranes filtration, most of carotenoids were concentrated in the retentate during cross-  
441 flow filtration experiments.

442 When comparing phenolic compounds for the three suspensions and anthocyanins for Red Extract, cross-  
443 flow filtration presented higher  $C_{ret}$  (10 – 33 %) than immersed membranes filtration (3 – 17 %).

444 These differences could be explained by adsorption mechanisms occurring on the mineral tubular membrane  
445 used for cross-flow filtration experiments, as reported by Vladisavljević *et al.* (2013) [17]. Moreover, the  
446 differences of  $C_{ret}$  could also be related to the different hydrodynamic conditions specific to each  
447 configuration. Indeed, the high cross-flow velocities and high TMP specific to cross-flow filtration might  
448 enhance the interactions between the phenolic compounds and between phenolic compounds and other Red  
449 Extract compounds, leading to the formation of complexes larger than the membrane average pore diameter  
450 [63,64]. Considering the many differences between both configurations, it is difficult to state which  
451 phenomenon is responsible of  $C_{ret}$  differences.

452 To sum up, immersed membranes filtration allowed producing retentate and permeate of quality as least as  
453 good as the one obtained with conventional cross-flow filtration. Concerning the process productivity, cross-  
454 flow filtration was the most interesting filtration configuration in terms of flux values for the three studied  
455 suspensions. However, the productivity of immersed membranes configuration could be easily increased by  
456 increasing the membrane surface (low cost of the membranes and great packing density) and thanks to the  
457 low energy consumption of this system. Moreover, immersed membranes system could find interesting  
458 application for small production units thanks to its compactness, easy handling and mobility.

#### 459 **4. Conclusion**

460 The aim of this work was to investigate the possibility of using an immersed membranes configuration for  
461 the microfiltration of fruit-based suspensions. The study focused on three different agro-food suspensions,  
462 for which microfiltration is widely used for clarification and concentration purpose, a grapefruit juice and  
463 two winery byproducts. The performances of a pilot-scale immersed membranes system were characterized  
464 in terms of membrane permeability (membrane fouling, permeate flux) and selectivity (clarification,  
465 concentration of targeted compounds).

466 Flux-stepping experiments performed under total recycle mode followed by filtration experiments under  
467 batch concentration mode allowed defining and validating the optimal operating domains of the studied  
468 system. The optimal permeate flux of immersed membranes configuration was found to be around 5 – 7  
469  $L \cdot h^{-1} \cdot m^{-2}$  for the three studied suspensions, which is in line with permeate fluxes reported in other fields  
470 where immersed configurations have been widely and successfully used. Selectivity analysis showed that  
471 immersed membranes filtration allowed reaching the selectivity goals specific to each suspension by  
472 producing (i) a clarified permeate rich in phenolic compounds and (ii) a retentate concentrated in carotenoids  
473 for grapefruit juice. Finally, immersed membranes filtration productivity and selectivity results were  
474 compared with performances of conventionally used cross-flow filtration with tubular membranes. The  
475 main outcomes were that immersed membranes filtration allowed producing retentate and permeate with  
476 quality as least as good as the one related to conventional cross-flow filtration. Concerning the process  
477 productivity, cross-flow filtration allowed reaching high fluxes compared to immersed membranes  
478 filtration. These differences were in line with the respective order of magnitude of fluxes reported for both  
479 configurations. However, thanks to its high compactness, easy handling and mobility, low investment and  
480 operational costs, immersed membranes configuration could find interesting applications in small  
481 production units of fruit juices and industries dealing with the valorization of byproducts with low added-  
482 value. Immersed membranes configuration could be an affordable and simple process to perform the *in situ*  
483 clarification/concentration of fruit-based suspensions, limiting loss and wastage due to processing delay.

484 All these findings offer new prospects for immersed membranes configuration applied to fruit-based  
485 suspensions microfiltration. Further investigations would be of great interest to get more insight into the  
486 involved fouling mechanisms occurring during immersed membranes filtration of fruit-based suspensions.

487

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