HAL open science

Filtration-compression step as downstream process for flavonoids extraction from citrus peels: Performances and flavonoids dispersion state in the filtrate

N. Zarate Vilet, E. Gué, A. Servent, M. Delalonde, C. Wisniewski

▶ **To cite this version:**

N. Zarate Vilet, E. Gué, A. Servent, M. Delalonde, C. Wisniewski. Filtration-compression step as downstream process for flavonoids extraction from citrus peels: Performances and flavonoids dispersion state in the filtrate. Food and Bioproducts Processing, 2020, 120, pp.104 - 113. 10.1016/j.fbp.2020.01.001 . hal-03489895

HAL Id: hal-03489895

https://hal.science/hal-03489895v1

Submitted on 21 Jul 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

Version of Record: https://www.sciencedirect.com/science/article/pii/S0960308519310223
Manuscript_e51e5969240d7b925a9d7d7840da504c

Filtration-compression step as downstream process for flavonoids extraction from citrus peels: performances and flavonoids dispersion state in the filtrate

N. Zarate Vilet*[1], E. Gué[1], A. Servent[2], M. Delalonde[1] and C. Wisniewski[1]

[1] QualiSud, Univ Montpellier, CIRAD, Montpellier SupAgro, Univ Avignon, Univ La Réunion, Montpellier, France

[2] CIRAD, UMR QualiSud, F-34398 Montpellier, France

Keywords: downstream process; filtration-compression; enzymatic treatment; grapefruit peels; flavonoids

Highlights:

- Filtration-compression for downstream processing of flavonoid extraction from peel
- Identification of the maximum extractable liquid of peel by filtration-compression
- Cell-wall degrading enzymes favour naringin and narirutin extraction from peel
- Identification of the dispersion state of flavonoids for a relevant post-extraction

*corresponding author:

Nuria Zarate Vilet[1]
CIRAD
UMR 95 Qualisud
TA-B95/15
73 rue Jean-François Breton
34398 Montpellier cedex 5
nuria.zarate_vilet@cirad.fr
+33-4-67-59-38-30

© 2020 published by Elsevier. This manuscript is made available under the CC BY NC user license
https://creativecommons.org/licenses/by-nc/4.0/

Abstract

Initiative of waste treatment has to be based on user-friendly technologies, using robust, cost-effective and low-energy consuming processes. The global objective of this study was to reduce technical steps for the extraction of flavonoids (naringin and narirutin) from citrus peel (grapefruit peel). After a first fresh peel grinding, the relevance of a simple filtration-compression, as a first downstream process, of the obtained slurry was studied. An optimization of this solid-liquid separation was proposed and the impact of a pectinolytic enzymatic treatment, resulting potentially in a larger release of flavonoids, was investigated. The results demonstrated that a preliminary step of filtration-compression, directly realized on fresh grinded peels as downstream processing for flavonoids extraction could be pertinent and that the enzymatic treatment improved the slurry filterability. An optimal separation was obtained with a transmembrane pressure of 5 bar, leading to highest extractable liquid phase volume and to an extraction around 80% of naringin and narirutin. A modelling of the filtration step, essential for the scaling of a filtration-compression process on site, was proposed. An originality of this work was to identify the dispersion state of the flavonoids within the liquid phase, capital identification for a relevant choice of the subsequent extraction step of these compounds.

Abbreviations and nomenclature

d_{50}	Mean diameter (µm)
J	Filtration rate ($m.s^{-1}$)
J_0	Initial filtration rate ($m.s^{-1}$)
K_b	Blocking constant for complete blocking law (m^{-3})
K_c	Blocking constant for cake filtration law (m^{-3})
K_i	Blocking constant for intermediate blocking law (m^{-3})
K_s	Blocking constant for standard blocking law (m^{-3})
M	Filtrate mass (g)
$M_{add.}$	Mass of water added for the enzymatic treatment (g)
M_{DM}	Mass of dry matter in slurry (g)
$M_{ext.liq.}$	Mass of extracted intrinsic liquid
M_F	Maximum filtrate mass (g)
$M_{liq.}$	Total liquid mass of the grinded peel (g)
t	Filtration time (s)
TMP	Transmembrane pressure (bar)
V	Filtrate volume (m^3)
w	Enzymatic solution/slurry mass ratio (g/g)

Greek letters

η	Theoretical filtration efficiency (%)
η*	Experimental filtration efficiency (%)
Ω	Filtration surface (m^2)

Index

DM	Dry matter
ET	Enzyme treated slurry
H	Heating (50°C, 2 hours)
NT	Non-treated slurry
W	Water addition in a ratio 1/1
W+H	Water addition in a ratio 1/1 + Heating (50°C, 2 hours)

1. Introduction

Citrus genus (orange, mandarin/tangerine, grapefruit and lemon/lime) is among the largest cultivated fruit crops in tropical and subtropical regions worldwide, with a global production of 92.088 million metric tons in 2017/18, and is expected to increase further in the future [1]. About a quarter of the harvested fruit is transformed into various commercial products such as juice, jam, marmalades and flavouring agents, using only about 50% of the total fruit. Hence, these transformations generate yearly 11 million metric tons of wastes and by-products, which are considered harmful to soil and water bodies due to their low pH (3-4), high water (around 80-90%) and organic matter (95% of total solids) contents [2–4]. Yet today waste management is at the heart of the concerns with the sustainable food systems development aiming at a « 0 waste » production but also with the biorefinery tendency of the food industry [3,5,6]. For Southern citrus industries (ensuring 70-80% of the total citrus production [1,7]), due to their insufficient infrastructure, little or no valorisation are ensured. Thus, peels, pulp and seeds are generally discarded on adjacent land leading to the generation of putrefying waste. Sometimes, theses by-products are valorised as cattle feedstock but with a low economic interest as this valorisation requires a previous drying which is a high-energy consuming operation.

These practices are particularly unfortunate, as *Citrus* by-products can be considered as a cheap source of high-added value components such as vitamins, minerals, essential oils, fibres and bioactive compounds mainly carotenoids, and polyphenols [2–4,8,9]. Flavonoids are polyphenolic compounds with a wide spectrum of beneficial effects; this includes anticancer, antiatherosclerotic, cardiovascular, anti-inflammatory, neuroprotective, hypolipidemic, antidiabetic, hepatoprotective activities and applications in the treatment of bone disorders [10–13].

In the literature, one of the most studied valorisation ways for Citrus by-products is the recovery of flavonoids from peels. This recovery purpose requires not only efficient extraction technology

but also various preliminary treatments of the peel, facilitating first its storage before extraction and secondly the solid/liquid separation step. Generally, a safe storage is ensured by drying (hot air, freeze-drying….). Then, with an objective to facilitate the mass transfer during extraction, grinding eventually followed by sifting is carried out to reduce and homogenize the particle size [14]. Extraction of the target compounds is generally combined with treatments or technologies facilitating the flavonoids transfer from the solid to the liquid phase. This step has attracted considerable scientific interest and thus several traditional or emerging extraction technologies are proposed in the literature: solvent extraction [15], hot-water extraction [16], ultrasound-assisted [15,17,18], microwave-assisted [19–21], enzyme-assisted [22], pulsed electric fields [23,24], high voltage electrical discharge [25] subcritical water [26–28], and supercritical CO_2 [29–32].

However, in Southern countries, the sustainability of waste treatment is threatened by the lack of financial resources and technical support. Moreover, most of the aforementioned processes, mostly studied at laboratory scale, require a high investment or high-energy consumption making these technologies non-suitable for those countries. As the success of valorisation initiative is based on the user's acceptance toward easy and user-friendly technologies, it seems pertinent to think about a high-added-value compounds recovery chain using optimized green, simple, robust, cost-effective and low-energy consuming processes. Our study fell within this context, with the aim to avoid a maximum of preliminary stages, especially the highly energy consuming drying step, before the extraction of flavonoids from Grapefruit (*C. paradisi*), rich source of naringin and narirutin [33].

Thus, the objective of this work was to study the relevance of a simple filtration-compression step directly done on the fresh grinded peels as a first downstream process for flavonoids extraction. The optimization of the solid-liquid separation was based on the estimation of the mass of the mechanically extractable liquid phase from the slurry under different transmembrane

131 pressures, combined with the quantification of the flavonoids (narirutin and naringin) content in
132 the filtrate. Before the filtration in a pressurized filtration-compression cell, fresh peels were
133 grinded to form a slurry with an objective to favour mass transfer by an increase of the exchange
134 surface and a disruption of plant tissues, flavonoids being considered to be largely stocked within
135 the cell vacuoles [24]. The potential impact on the solid-liquid separation of a pectinolytic
136 enzymatic treatment, causing a cell wall degradation resulting potentially in a larger release of
137 flavonoids, was investigated. An originality of this work was also to identify the dispersion state
138 of the flavonoids within the mechanically extractable aqueous phase obtained after this
139 preliminary solid-liquid separation. This last investigation was supposed to be capital for a
140 relevant choice of the following extraction of these compounds.

141

142 **2. Materials and Methods**

143 *2.1. Grapefruit peel supply and storage*

144 Grapefruits (*Citrus paradisi*, Star ruby from Turkey, provided by Terreazur, Saint-Jean-de-
145 Védas, France) were first bleached. Flavedo, which can be valorised through the extraction of
146 essential oils [4], was then manually removed from the peel, the target flavonoids being mostly
147 contained in the albedo [34–38]. Then, juice was extracted using an electric juice extractor
148 (ZX7000, Krups). Albedo and pulp rests (core, juice sacs and segment rests), representing in our
149 experiments about 35% of the weight of the total fruit, were then manually cut into small pieces
150 (around 10 x 10 mm) and stored at -25°C.

151

152 *2. 2. Experimental methodology*

153 *2.2.1 Grinding*

154 After defrosting at room temperature (45 min to 1 hour), peels were subjected to grinding. Grinding
155 was ensured using a hand blender (Moulinex DD873D10 Infiny Force ultimate, France) for

156 1.5 minutes at 1 kW (corresponding to a 9.10^5 J·kg^{-1} of fresh peel). After grinding, a thick
157 suspension, i.e. a slurry, was obtained.
158
159 *2.2.2. Enzymatic treatment*
160 The enzymatic treatment was based on the use of Peclyve® PR (Soufflet biotechnologies®), a
161 commercial pectinolytic enzyme preparation with two main pectinase activities, Pectin-methyl-
162 esterase (EC3.1.1.11; 150 U/g) and Pectin-lyase (EC4.2.2.10; 1100 U/g). An enzymatic solution
163 was prepared with the objective to provide 0.3 g of enzyme per kg of slurry, with a mass ratio
164 solution/slurry (*w*) of approximatively 1/1. The enzymatic treatment consisted in the mixing of
165 the slurry and the enzymatic solution during 2 h at 50°C. These operating conditions were
166 recommended by Soufflet biotechnologies® for an optimal enzyme activity.
167
168 *2.2.3. Filtration-compression*
169 The liquid phase of the raw (NT) or enzyme treated (ET) slurry was extracted by filtration-
170 compression.
171 Filtration-compression experiments were carried out with a pressurized cylindrical cell of 70 mm
172 internal diameter and 0.60 L volume (Figure 1). A perforated disk was located at the bottom of
173 the cylinder to support a plane cotton cellulose membrane of 6 µm in pore size (Whatman,
174 Maidstone, UK); the pore size of 6 µm was supposed to retain the great majority of the suspended
175 solids, while allowing acceptable filtration rate. Transmembrane pressure (TMP) was applied by
176 a pneumatically driven-piston dragged by nitrogen gas.
177 With an objective to compare and discuss flavonoids extraction efficiencies, filtrations were
178 operated from same mass of dry matter (around 3.5 g), whatever the type of slurry (ET or NT);
179 thus the initial slurry mass (or volume) to be filtered was different for NT and ET slurry (20 g
180 and 40 g respectively, according to their different water content).

Filtration-compression experiments were carried out at different transmembrane pressures (1, 3, 5, 7 and 10 bar).

The filtrate mass (M) was monitored during time until its stabilization; M_F, maximum filtrate mass, was considered as the maximum mass of the liquid phase that could be extracted from slurry by filtration-compression at the defined operating condition (i.e. TMP).

At the end of the filtration, filter cake and filtrate were recovered and characterized.

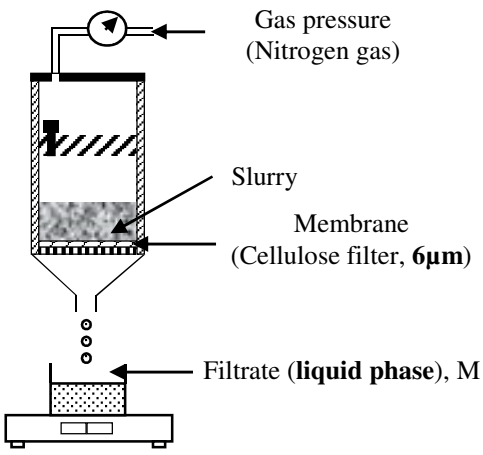


Figure 1. Filtration-compression cell

The filtration efficiency, in terms of extracted liquid volume, was estimated for the two slurries, i.e. the raw (NT) one and the enzyme treated (ET) one.

The NT slurry filtration efficiency η_{NT} (given by Eq.(1)) was defined by the ratio between the extracted liquid mass, $M_{ext.liq.NT}$, and the total liquid mass of the grinded fresh peel $M_{liq.}$.

$$\eta_{NT} = \frac{M_{ext.liq.NT}}{M_{liq.}} \quad (1)$$

Regarding ET slurry filtration, the total liquid mass corresponding to the liquid mass present in the grinded fresh peel $M_{liq.}$ plus the added water ($M_{add.}$ due to addition of the enzymatic solution), η_{ET} was given by Eq.(2):

$$\eta_{ET} = \frac{M_{ext.liq.ET}}{M_{liq.} + M_{add.}} \quad (2)$$

With the suppositions that (i) the enzymatic treatment did not modify the part of the mechanically removable liquid of the slurry and (ii) all the water added ($M_{add.}$) for the enzymatic treatment is easily removed by filtration, then, the theoretical ET slurry filtration efficiency η_{ET} can be given by Eq.(3):

$$\eta_{ET} = \frac{M_{ext.liq.NT} + M_{add.}}{M_{liq.} + M_{add.}} \quad (3)$$

Considering the enzymatic treatment protocol (with $M_{add.} = w(M_{DM} + M_{liq.})$, where M_{DM} is the mass of dry matter in slurry), η_{ET} can be deducted from η_{NT} by Eq.(4):

$$\eta_{ET} = \frac{\eta_{NT} + w\,(1 + \frac{M_{DM}}{M_{liq.}})}{1 + w\,(1 + \frac{M_{DM}}{M_{liq.}})} \quad (4)$$

2.3. Physicochemical and biochemical characterization

2.3.1. Physicochemical characterization

Dry matter content determination: Slurry, filter cake and filtrate dry matter content was determined according to the AOAC method with some modifications [39]. Samples were homogenously distributed on aluminium dishes and kept in a vacuum oven (Heraeus RVT 360, Hanau, Germany) at 70°C for 48h. Water content was determined from the weight difference before and after drying. Each measure was done in triplicate.

Particle size distribution: Particle size distribution (in percentage of volume density) and mean diameter (d_{50}) of NT and ET slurries were determined by Laser diffraction using a Malvern Mastersizer (Mastersizer 3000, Malvern Instruments Limited, Worcestershire, UK) equipped

214 with a HydroMV® cell. Refractive indices of 1.73 and 1.33 were used for cloud particles and
215 dispersion phase and 0.1 was used as the absorption index of cloud particles [40]. Samples were
216 introduced into the volume presentation unit, which already contained deionized water; in this
217 unit, the diluted sample was gently stirred and pumped through the optical cell.
218 *Flavonoids dispersion state:* Filtrate was considered as an aqueous solution in which colloidal
219 and supra-colloidal particles were suspended. These insoluble particles were supposed to be
220 remaining vegetable tissue and cellular rests and therefore to be susceptible of containing
221 flavonoids. In order to identify the dispersion state of naringin and narirutin within the filtrate,
222 centrifugation of the filtrate for 4 hours at 20 000 g was proposed; this centrifugation operating
223 condition was chosen with the objective of separating the soluble fraction from the rest of the
224 suspension. According to Stokes law, a cut-off size of approximately 100 nm was supposed to
225 be reached with this chosen centrifugation time and acceleration; the separation selectivity has
226 been subsequently confirmed by means of particles-size measurements (Dynamic Light
227 Scattering). Flavonoids (naringin and narirutin) present in the centrifugation pellet, supposed on
228 crystallized form or bound to some larger solids, were considered as *insoluble* flavonoids.
229 Flavonoids present in the supernatant, supposed solubilized or potentially bound with some
230 colloids or macromolecules, were considered as *soluble* ones.
231
232 *2.3.2. Flavonoids biochemical characterization*
233 Naringin and narirutin content of the slurry and filtrates was estimated by High Performance
234 Liquid Chromatography (HPLC) analyses. A solvent extraction was performed using a modified
235 method based on [41] to estimate the total content of flavonoids in the samples.
236 A mass of slurry (corresponding to 250 mg of dry matter) or a volume of 500 µl of filtrate was
237 withdrawn and diluted with 5 ml of dimethylformamide (DMF) and 5 ml of ammonium oxalate
238 (0.05 M). Afterwards, samples were placed in a hot oil bath (90°C) for 10 minutes, cooled and

239 adjusted to 18 ml for filtrates and 25 ml for slurries. Solutions were then filtered through 0.45 µm
240 Whatman cellulose acetate syringe filters (Sigma-Aldrich, St Louis, MO) before injection.
241 Naringin and narirutin content was quantified using an Agilent 1200 series HPLC system equipped
242 with a degasser, a quaternary pump, an auto-sampler, a thermostated column compartment, and
243 a diode array detector DAD. Separation of flavanones was performed using a C18 column
244 (Uptisphere HDO 5 µm, 250 mm × 4.6 mm, Interchim, Montluçon, France). The mobile phase
245 consisted of an isocratic solvent system combining solvent A (water/acetic acid, 99/1, v/v),
246 solvent B (acetonitrile), and solvent C (tetrahydrofuran) - A/B/C 80/16/4, v/v/v. The flow was
247 set at 1 mL/min and the column was heated at 25°C. UV detector was set at 280 nm, an aliquot
248 of 20 µL was injected.
249
250
251 **3. Results and Discussion**
252 *3.1. Slurry characterization*
253 The physicochemical and biochemical properties of fresh peel and slurries with (ET) and without
254 an enzymatic treatment (NT) are shown in Table 1.
255 The fresh peel and the NT slurry presented similar characteristics in terms of dry matter (DM)
256 and water content, around 17 and 83 g per 100 g of wet matter (i.e. fresh peel or slurries)
257 respectively; these values were coherent with the ones available in the literature [42,43]. The
258 enzymatic treatment, requiring a water addition, increased the water content of the ET slurry until
259 91 g per 100 g of wet matter, coherent value considering the addition of an enzymatic aqueous
260 solution close to a 1/1 ratio.
261
262
263

264 **Table 1**
265 Physicochemical properties of fresh peel and slurries
266

	Fresh Peel	NT slurry	ET slurry
Dry matter			
g/100 g wet matter	17.3 ± 0.9	17.3 ± 0.5	8.6 ± 0.2
Water content			
g/100 g wet matter	82.6 ± 0.9	82.7 ± 0.5	91.4 ± 0.2
d_{50}			
µm	-	716 ± 19	575 ± 17
Naringin			
g/100 g dry matter		5.1 ± 0.4	
g/100 g wet matter		0.9 ± 0.1	
Narirutin			
g/100 g dry matter		0.57 ± 0.02	
g/100 g wet matter		0.1 ± 0.01	

267
268
269
270
271
272
273

274 The particle size distributions of NT and ET slurries are given in Figure 2. The NT slurry was
275 composed of particles ranging from approximately 25 to 3500 µm, with a relatively important
276 volume percentage of particles around 1000 µm; the d_{50} was about 716 µm. If the enzymatic
277 treatment did not significantly modify the size distribution shape, it was however noted that the
278 amount of particles with size from 300 to 2000 µm was reduced while the percentage of particles
279 with size from 10 to 300 µm increased. These results suggested that a part of the larger particles
280 were broken under the effect of the enzymatic treatment. Pectin-methyl esterase and pectin-lyase
281 had almost certainly catalysed the demethylation and the depolymerisation of pectin in the albedo
282 [44], leading to the weakening of cell structures and a consequent particle size reduction [45]. As
283 a consequence of this size distribution modification, a decrease of the d_{50} was observed after the
284 enzymatic treatment.

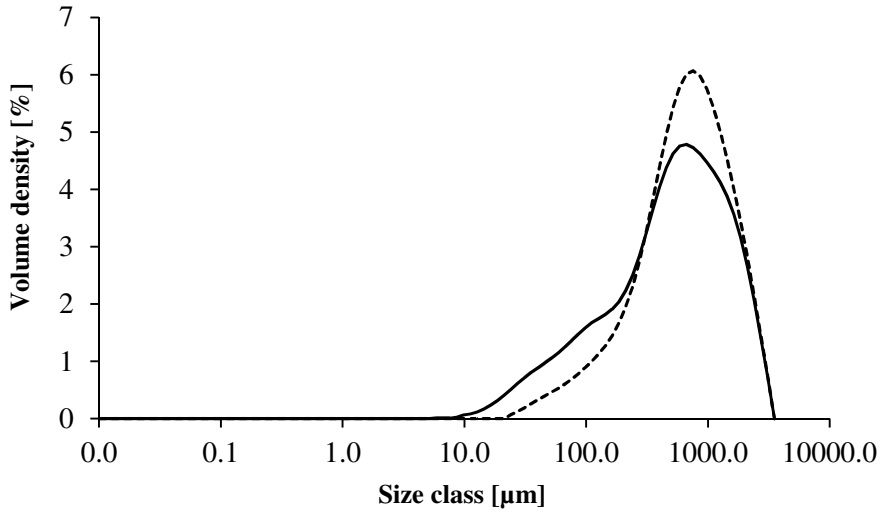


Figure 2. Particle Size distribution of: – ET slurry and --- NT slurry

The total concentration of naringin and narirutin in NT slurry, estimated per 100 g of dry matter and wet matter, are given in Table 1. The concentration of naringin was about 10 times higher than the narirutin one. Grapefruit naringin and narirutin concentrations found in the literature for the different parts of the fruit are reported in Table 2. This bibliographic review indicated that the measured concentrations were in the same range of those found in literature for albedo and confirmed the large presence of naringin and narirutin in peel, and specifically in albedo. The significant variability observed in the values reported in Table 2 could be easily explained by various factors (e.g. fruit origin, fruit variety, manufacturing procedures, storage conditions…) but also variation in the degree of maturity of the fruit [35,36,46–48].

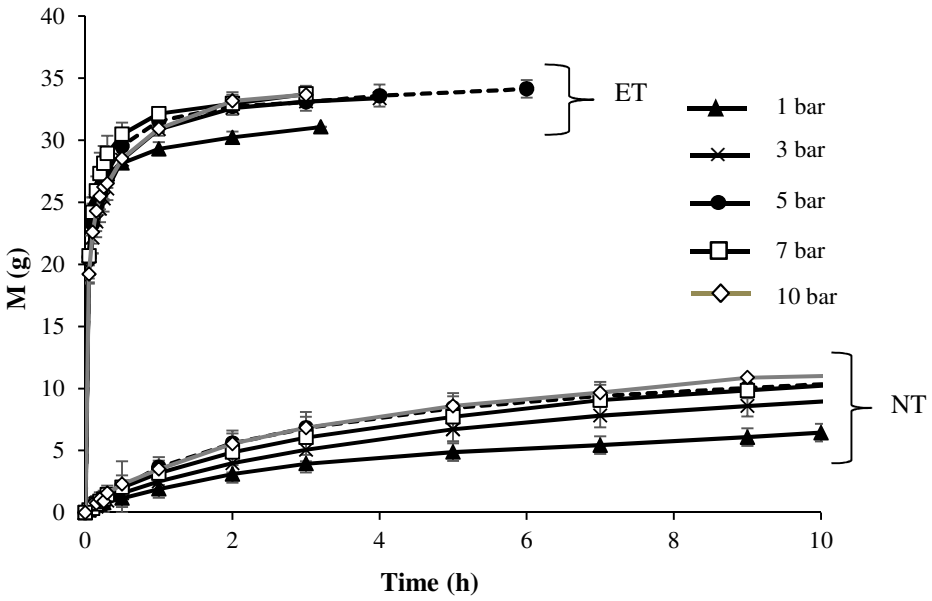
Table 2
Naringin and Narirutin content in grapefruit (*C. paradisi*)
[a] *g/100g juice*
[b] *g/100g wet matter*

	Juice	Flavedo	Albedo
Naringin	0.006 – 0.087[a,[36-38]]	0.35 – 0.99[b,[34,35]]	0.62 – 2.7[b,[34,35]]
Narirutin	0.009 – 0.012[a,[4]]	0.11[b,[2]]	0.23[b,[2]]

305 *3.2. Filtration-compression of the slurry*

306 Figure 3 presents the evolution of the filtrate mass (*M*) versus time, for NT and ET slurries during
307 filtration at different transmembrane pressures.

308 As it can be observed, the applied transmembrane pressure influenced the quantity of collected
309 liquid at the end of each filtration, i.e. M_F. Recovered filtrate mass was higher for ET slurry than
310 for NT one, which was coherent with the filtration protocol (filtration of the same mass of dry
311 matter, whatever the slurries), leading to the filtration of a higher mass of ET slurry than of the
312 NT slurry (considering the addition of the enzymatic solution and consequently of water to the
313 slurry).



314 **Figure 3.** M_F versus filtration time at different transmembrane pressures, for NT and ET slurries

315

316 The influence of pressure and of the enzymatic treatment on the solid/liquid separation efficiency
317 of filtrations is discussed below. The description of the evolution of the filtrate mass, through the
318 study of the fouling mechanisms, is also proposed with an objective to provide some indication
319 about the fouling propensity of NT and ET slurries.

320

3.2.1. Solid/liquid separation efficiency

Figure 4 represents the maximum recovered filtrate mass (M_F), corresponding to the steady filtrate mass, versus each applied transmembrane pressure.

An increase of the transmembrane pressure TMP, from 1 to 5 bar, induced an increase of M_F. For NT and ET slurries, all the curves tend towards an asymptote corresponding to a M_F value closed to 12 and 34 g respectively.

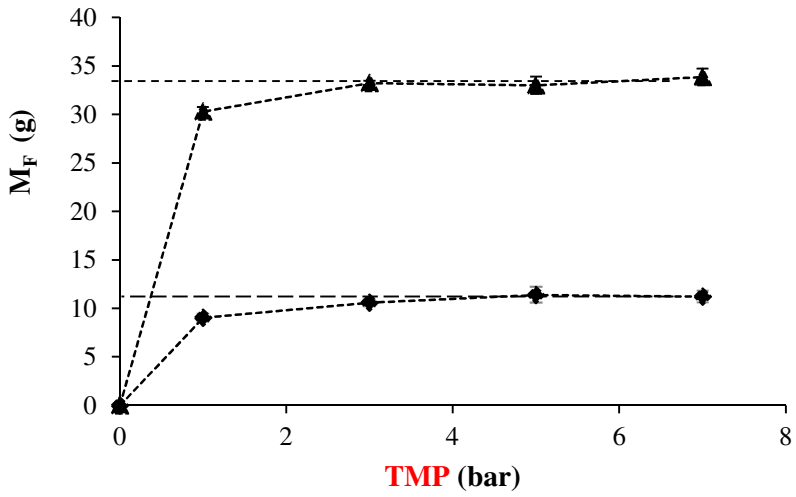


Figure 4. M_F value versus TMP (NT ♦, ET ▲)

Figure 4 shows that, in the presence or absence of an enzymatic treatment, a transmembrane pressure of 5 bar appeared to be sufficient to reach the asymptote and thus to extract all the mechanically extractable liquid part of the slurry. The nature of the interactions between the liquid (aqueous solution) and solid (e.g. pieces of vegetable tissues as cell wall fragments, proteins, pectin and fibres …) phases of the slurry could explain this result. In a complex medium, as slurry, the solid/liquid interactions can be low or high, in relation with the liquid binding forces [49–52]; at least two types of water can be defined: free and bounded water, respectively concerned by low and high interactions with solids. The experimental distinction between these two types of water can be based on water activity [53], freezable and non-freezable ability [54]

or extractability by mechanical means [55]. This last definition, according to our experimental strategy and the obtained results, led us to admit that mechanical action imposed by pressure in a filtration-compression cell promoted the release of the slurry free liquid. Thus, results revealed that, for grinded grapefruit peels, with or without enzymatic treatment, a pressure of 5 bar appeared to be sufficient to extract all the easily removable liquid, i.e. free water. For ET slurry, the extracted liquid phase corresponded not only to the free liquid of the fresh grinded peel, but also included the water added for the enzymatic treatment, water considered as free water and therefore very easily extractable by filtration-compression.

However, with an objective to verify if enzyme presence and action were susceptible to modify the interactions between solids and liquid, i.e. if enzymes were susceptible to modify the quantity of the mechanically removable liquid part of the slurry, the estimation of the theoretical filtration efficiency η_{ET} (estimated from η_{NT}, through Eq. 4) was compared to the experimental one, η^*_{ET}, estimated through the quantification of dry matter and water content of the filtrate and filter cake. Table 3 presents the characteristics of the slurry, filter cake and filtrate in terms of dry matter and water content and extraction efficiencies, from 5 bars-trials (i.e. for pressure conditions allowing to extract the maximum of the liquid phase). Mass balance was approximately verified, considering the losses of matter (between approximately 10 and 20%, i.e. stagnant water in filtrate tube, arduous separation of filter cake from filtration membrane, deposit on the walls of the filtration-compression cell…).

Table 3

Slurry, filter cake and filtrate characterization

	Slurry (g)				Filter cake (g)			Filtrate (g)			η
	Total mass	Added water mass	DM mass	Water mass	Total mass	DM mass	Water mass	Total mass	DM mass	Water mass	
NT	20.1 ± 0.0	0.0	3.5 ± 0.1	16.6 ± 0.1	5.21 ± 0.3	1.97 ± 0.17	3.05 ± 0.20	11.6 ± 0.3	1.2 ± 0.0	10.4 ± 0.3	**63 ± 2%**
ET	39.9 ± 0.3	20.0 ± 0.1	3.5 ± 0.1	36.5 ± 0.2	3.14 ± 1.3	0.94 ± 0.09	2.20 ±1.06	33.5 ± 1.3	2.2 ± 0.1	31.3 ± 1.2	**86 ± 2%**

360 The enzymatic action was first observed through the difference between ET and NT filtrate dry
361 matter content, as well as ET and NT filter cake one (Table 3). The evolution of the slurry particle
362 size distribution after enzymatic treatment (Figure 2) had induced a reduction of solids retained
363 by the membrane (pore size around 6 µm) and inversely an increase of solids in the filtrate
364 (greater presence of finer fragments).

365 Concerning the comparison between the experimental and theoretical (i.e. calculated) filtration
366 efficiency, an experimental efficiency of 86% was obtained while the theoretical one, that was
367 based on the fact that the enzymatic treatment did not modify the mechanically removable part
368 of liquid, was equal to 83% (according to η_{NT} and Eq. 4). This comparison demonstrated that, in
369 modifying the cell structures and solids physicochemical characteristics, the enzymatic treatment
370 could slightly increase the mechanically removable part of liquid of the grinding fresh peel
371 (ANOVA, $p < 0.01$).

372

373 *3.2.2. Filtration modelling and fouling propensity of the slurry*

374 We proposed to describe the filtrate mass (M) evolution during filtration time through classical
375 filtration modelling at constant pressure, i.e. Hermia models. The Hermia models consist of four
376 different filtration mechanisms: complete blocking, standard blocking, intermediate blocking,
377 and cake formation. The cake formation model, initially established by [56,57], considers that
378 during filtration, fouling is due to a cake formation on the surface of the filter medium,
379 corresponding to the deposit of particles of larger size than the membrane pore size. Both
380 complete and intermediate blocking laws describe the pore plugging due to foulants reaching the
381 top surfaces of pores. In contrast, the standard blocking law deals with the pore constriction
382 caused by the deposition of foulants onto the pore wall. Table 4 presents mathematical equations
383 giving the relation between the filtrate volume V and the filtration time t, for each filtration law

at constant pressure condition [58]. As proposed by [58], the predominant blocking filtration laws describing fouling pattern were determined from plots based on linear expressions of the law (Table 4), allowing a rapid identification of the relevant fouling mechanisms. If standard blocking and cake filtration models could describe experimental pattern for NT slurry, standard blocking and intermediate blocking were more suitable to describe the fouling evolution of ET slurry.

Table 4
Blocking filtration equations for constant pressure filtration [55]

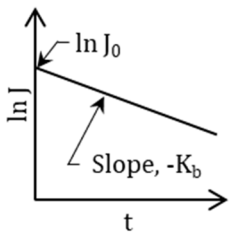
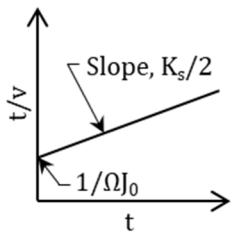
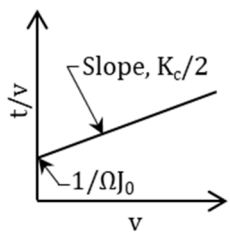
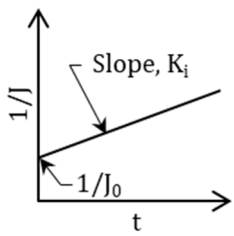
Function	Complete blocking	Standard blocking	Intermediate blocking	Cake filtration
$v = f(t)$	$v = \frac{J_0}{K_b}\{1 - exp(-K_b t)\}$	$\frac{t}{v} = \frac{K_s}{2}t + \frac{1}{J_0\Omega}$	$K_i v = ln(1 + K_i J_0 t)$	$\frac{t}{v} = \frac{K_c}{2}v + \frac{1}{J_0\Omega}$
$J = f(t)$	$J = J_0\, exp(-K_b t)$	$J = \frac{J_0}{\left(\frac{K_s J_0}{2}t + 1\right)^2}$	$K_i t = \frac{1}{J} - \frac{1}{J_0}$	$J = \frac{J_0}{\left(1 + 2K_c {J_0}^2\right)^{1/2}}$
Linear graphical representation				

Figure 5 presents the theoretical evolution associated to the models providing the better description of the experimental one for 5 bar experiments; similar results were obtained for the other filtration pressures.

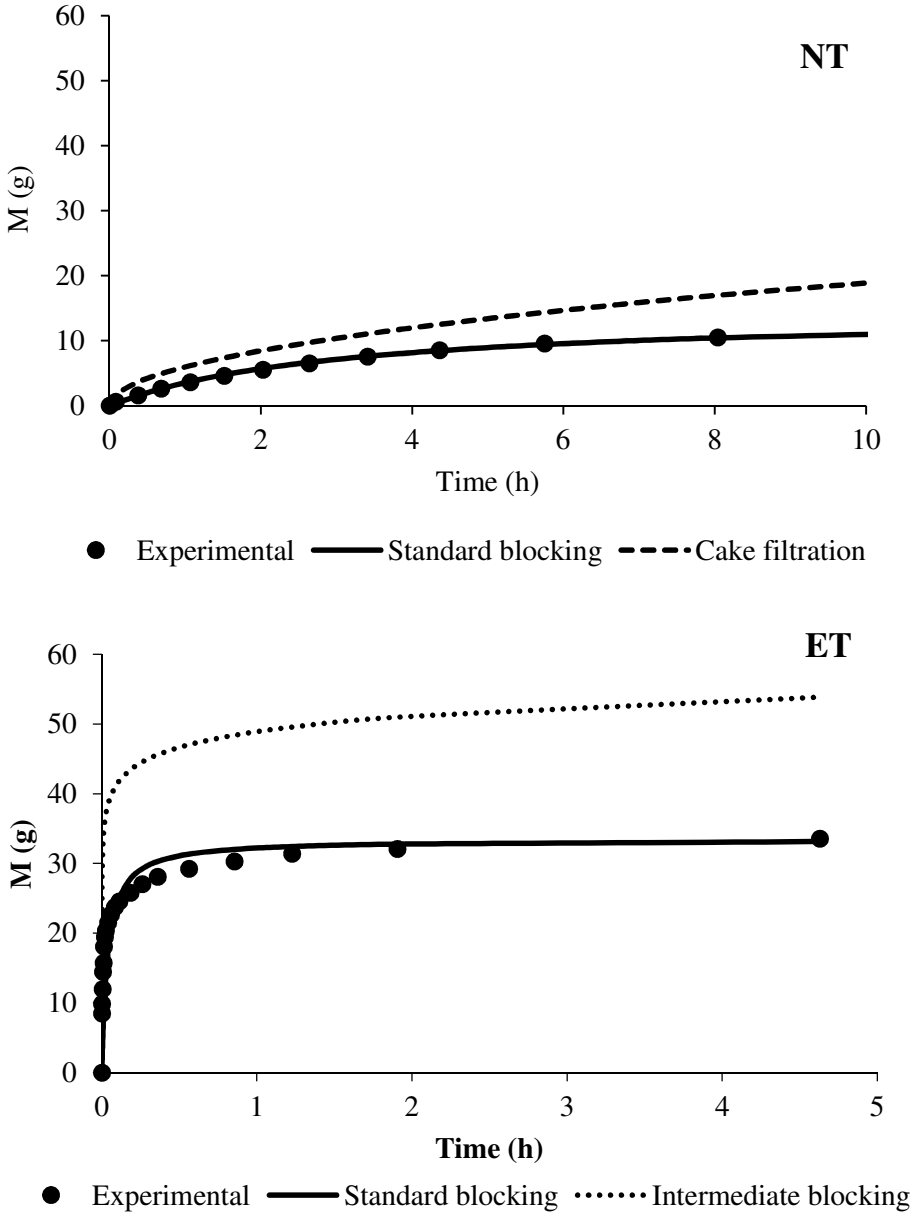


Figure 5. Theoretical and experimental evolution of M versus time (5 bar, filtrate density of 1040 kg.m^{-3})

As it can be observed in Figure 5, the standard blocking law is the only law describing properly the evolution of M versus time, demonstrating a possible membrane pore constriction caused by the deposition of foulants into the pore wall, and this, independently of the slurry modification induced by the enzymatic treatment. Thus, filtration models demonstrated that if the filtration kinetic of NT and ET slurries was different, the filtration behaviour of these two suspensions were comparable in terms of fouling mechanisms whatever the applied pressure.

Table 5 presents the value of K_s, blocking constant for standard blocking law, in m^{-3}, for the two types of slurry at 5 bar. Considering that lower is K_s, higher is the filtrate volume V for a filtration time t, the enzymatic treatment was supposed to be favourable to a weak fouling and consequently a rapid filtration. This result was consistent with the use of pectin-methyl esterase and pectin-lyase to enhance juice filterability notably through a reduction of the juice viscosity [59]. This modelling and the estimation of the K_S constant could be very useful for the implementation of a filtration-compression process, with an objective of providing and achieving a sustainable productivity.

Table 5
Standard blocking constant (5 bar, filtrate density of 1000 $kg.m^{-3}$)

Standard blocking model	$\frac{t}{v} = \frac{K_s}{2}t + \frac{1}{J_0\Omega}$	
	NT	**ET**
Ω (m^2) x 10^4	38	38
J_0 ($m.s^{-1}$) x 10^7	3 ± 0.2	1 210 ± 800
K_s (m^{-3})	**139 000 ± 1400**	**70 000 ± 10 000**

3.3. Flavonoids extraction efficiency

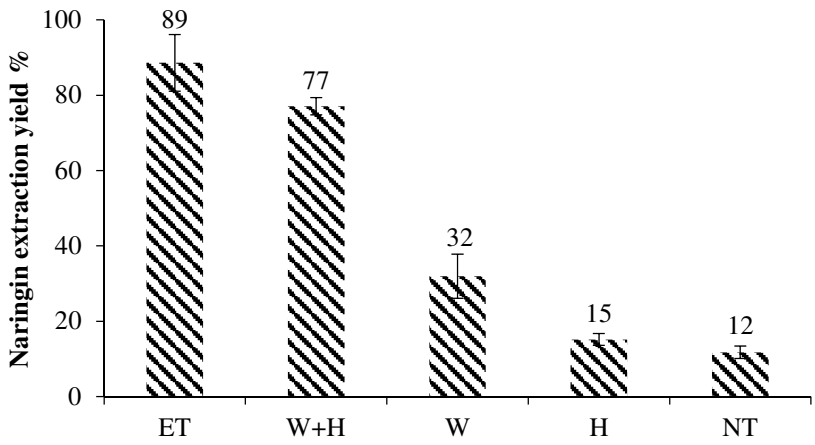
3.3.1. Identification of the role of the operating conditions

Figure 6 and Figure 7 show the extraction yields of naringin and narirutin for filtration at 5 bar (i.e. the lower transmembrane pressure inducing the maximum liquid extraction).

For ET slurry, approximately 89% (4.5 g per 100 g of peel dry matter) of the naringin and 92% (0.5 g per 100 g of peel dry matter) of narirutin were recovered in the filtrate; lower yields were obtained without enzymatic treatment, i.e. 12% and 61% respectively. Similar data were reported by [60] and [15] with recovery of 4.1 g (hot methanol extraction) and 3.6 g (ultrasound-assisted extraction) of naringin per 100 g of peel dry matter respectively.

429 The high recovery of naringin and narirutin obtained for the ET slurry could be explained by the
430 hydrolytic effect of the enzyme on pectin, inducing cell wall disruption and consequently
431 enhancing the release of these flavonoids into the extracellular liquid phase recovered by
432 filtration.

433 However, with the objective to discriminate the effect of the added enzymes from that of the
434 operating conditions associated with the enzymatic treatment (heating and water addition),
435 complementary experiments were carried out: (i) heating of the NT slurry in the same conditions
436 as for the enzymatic treatment (50°C for 2 hours) (H), (ii) addition of water to the NT slurry in
437 the same ratio (w) as for the enzymatic treatment (W) and (iii) heating and water addition in the
438 same conditions as for enzymatic treatment (50°C for 2 hours, same w) (W+H). This
439 complementary study could also be pertinent, with an objective to propose other pre-treatments
440 than an enzymatic one to enhance flavonoids extraction by filtration-compression.



441
442 **Figure 6.** Naringin extraction yield at 5 bar
443 *Enzymatically treated **ET**, raw **NT**, water addition with heating **W+H**, water addition **W**, heating **H***
444

445 As it is observed on Figure 6, heating the slurry (H) did not lead to a significant increase of
446 naringin recovery in comparison with raw slurry. Even if high temperatures are susceptible of
447 increasing the diffusion rates of flavonoids, the low water content limit the transfert phenomena
448 of the flavonoids towards the mechanically extractable liquid. By adding water to the slurry (W),

449 a marked increase on the recovered naringin was noticed; an extraction twice as efficient as the
450 one obtained with heating was observed. The addition of water, which diluted the medium,
451 facilitated the mobility of flavonoids from the solid matrix towards the free extracellular liquid,
452 through an increase of the concentration gradient at the solid/liquid interface and a possible
453 increase of the diffusion coefficient by a decrease of the viscosity of the medium.
454 The twinning effect of heating and water addition (W+H) led to an important increase of the
455 naringin extraction resulting in an efficiency close to 78%. The addition of pectinases, which
456 hydrolysised pectins of the cell walls, increased the naringin extraction efficiency from 78 to
457 89%.
458 These results demonstrated that, if the enzymatic treatment enhanced the naringin extraction,
459 water addition and heating, due to their synergistic effect, could be simple and low-cost slurry
460 pre-treatments to enhance naringin extraction. Thus, the benefit of an enzymatic treatment on the
461 naringin recovery has to be considered, given the enzyme cost.

462 Regarding narirutin recovery (Figure 7), the lower concentration in peel residues combined with
463 its higher solubility allowed a higher release of this flavonoid (between 61% and 92 % depending
464 on the treatment) into the aqueous phase. In this case, the most impacting factor is the water
465 quantity available for rapid narirutin diffusion and dissolution; thus, a single water addition could
466 recover the same quantity of narirutin than an enzymatic treatment.

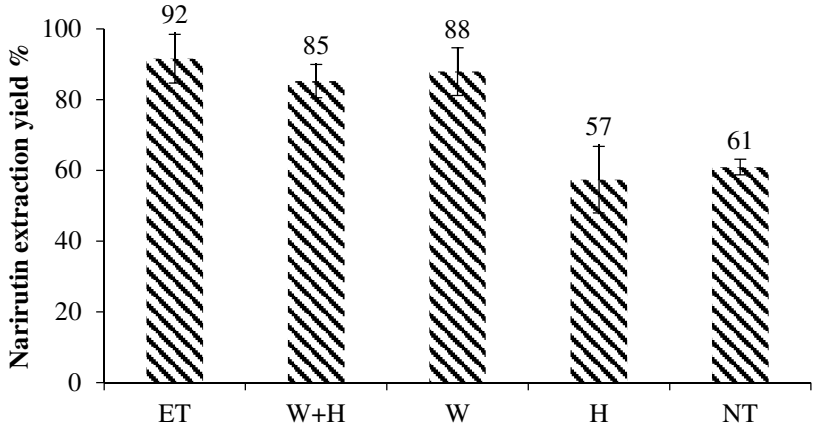


Figure 7. Narirutin extraction yield at 5 bar
*Enzymatically treated **ET**, raw **NT**, water addition with heating **W+H**, water addition **W**, heating **H***

3.3.2. Identification of the dispersion state of extracted naringin and narirutin

With an objective to identify the dispersion state of the extracted naringin and narirutin according to the different pre-treatments, centrifugations of the different filtrates were realized to separate the *soluble* and *insoluble* part of naringin and narirutin (see methodologies in 2.3.1).

Figure 8 presents the total naringin content compared to the *soluble* one. While naringin was only on a *soluble* state in H and NT filtrates, *insoluble* naringin appeared in the filtrates obtained with other pre-treatments.

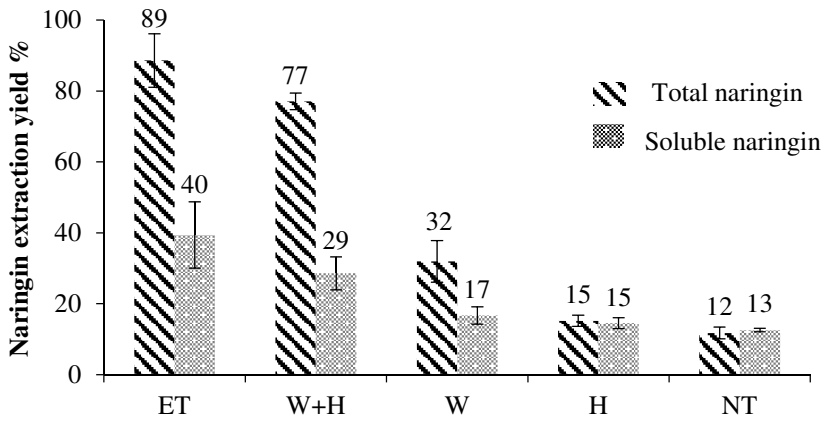


Figure 8. Naringin total and soluble extraction yields at 5 bar for different slurries
*Enzymatically treated **ET**, raw **NT**, water addition with heating **W+H**, water addition **W**, heating **H***

The presence of *insoluble* naringin was directly linked to the enhancement of the total naringin extraction. Water addition, alone or combined with heat treatment, had favoured the diffusional transfer and consequently the extraction of a higher part of naringin within the filtrate; the enzymatic treatment had led to a weakening of vegetable cell wall due to pectinases, facilitating naringin diffusion and thus extraction [22]. The results seemed to establish that these released flavonoids, initially mainly on *soluble* state in the filtrate, could rapidly form large aggregates, explaining the increase of an *insoluble* fraction. Thus, about half of the total naringin was on insoluble state after these different pre-treatments. It was supposed that naringin could form colloidal structures with macromolecules (e.g. proteins, phospholipids…) that could be present in large quantity and with a great diversity in the supernatant after such pre-treatment (because of the facility of the diffusional transfer of other compounds or of the enzymatic action on the vegetable tissue). Additional specific tests need to be performed to complete these hypotheses, and notably to quantify the naringin solubility in such complex medium.

Concerning narirutin, the supernatant analysis of the filtrates showed that this flavonoid was entirely present under its *soluble* form probably due to its higher solubility and lower concentration within the peel residues (Data not shown).

4. Conclusion

In order to promote circular and sustainable food systems, various strategies are proposed to valorise wastes. In the case of Citrus transformation industry, grapefruits flavonoids (naringin and narirutin), presenting interesting pharmaceutical properties, could be extracted from peels, constituting a source of income for Southern countries. Most of the extraction processes require high investment or high-energy consumption making these technologies non-suitable for the Southern countries. Thus, it seems pertinent to think about a high-added-value compounds

recovery chain using optimized robust, cost-effective and low-energy consuming processes. The objective of this work was to study the relevance of a simple filtration-compression step directly done on the fresh grinded peels as a first downstream process for flavonoids extraction. The potential impact on the solid-liquid separation of a pectinolytic enzymatic treatment was investigated.

The results demonstrated that a preliminary step of filtration-compression, directly realized on fresh grinded peels, as downstream processing for flavonoids extraction could be pertinent. The optimization of solid/liquid separation was obtained with a transmembrane pressure of 5 bar, providing the removal of the maximum of the extractable liquid phase from peel. Filtration modelling demonstrated that the standard blocking law properly described the evolution of the filtrate mass versus time, whatever the operating conditions (pressure, enzymatic treatment or none…). If the fouling mechanisms were similar for enzymatic treated slurry and no-treated one, the enzymatic treatment improved significantly the filterability of the slurry. The blocking constant of the standard blocking law, indispensable for the scaling of a filtration-compression process on site, was, under 5 bar, two times higher for non-treated than enzymatic treated peel. The quantification of naringin and narirutin content in this removable liquid phase (i.e. filtrate) demonstrated that the enzymatic treatment or a hot-water addition were suitable to enhance the naringin and narirutin extraction yield, which could reach 80 and 90% respectively. The dispersion state of the flavonoids within the mechanically extractable aqueous phase obtained after this preliminary solid-liquid separation was identified; the part of the *insoluble* (crystallized or bound to some larger solids) naringin could represent half of the total extracted naringin. These last results require further investigations, with an objective to provide pertinent elements to guide the choice of the subsequent extraction step.

Acknowledgements

This research was partially supported by the National Council on Science and Technology (CONACYT, Mexico). We are thankful to our colleagues, Bénédicte Marion and Céline Roques, who provided expertise in analytical measurements. We are also immensely grateful to Soufflet Biotechnologies ® for providing enzymes and operating conditions choice guidance.

Declaration of interest

The authors report no declarations of interest.

References

[1] USDA, Citrus World Markets and Trade, (2018).
[2] K. Sharma, N. Mahato, M.H. Cho, Y.R. Lee, Converting citrus wastes into value-added products: Economic and environmently friendly approaches, Nutrition. 34 (2017) 29–46. https://doi.org/10.1016/j.nut.2016.09.006.
[3] B. Satari, J. Palhed, K. Karimi, M. Lundin, M.J. Taherzadeh, A. Zamani, Process Optimization for Citrus Waste Biorefinery via Simultaneous Pectin Extraction and Pretreatment, BioResources. 12 (2017) 1706–1722. https://doi.org/10.15376/biores.12.1.1706-1722.
[4] D.A. Zema, P.S. Calabrò, A. Folino, V. Tamburino, G. Zappia, S.M. Zimbone, Valorisation of citrus processing waste: A review, Waste Manag. 80 (2018) 252–273. https://doi.org/10.1016/j.wasman.2018.09.024.
[5] S. Talekar, A.F. Patti, R. Vijayraghavan, A. Arora, An integrated green biorefinery approach towards simultaneous recovery of pectin and polyphenols coupled with bioethanol production from waste pomegranate peels, Bioresour. Technol. 266 (2018) 322–334. https://doi.org/10.1016/j.biortech.2018.06.072.
[6] M. Boukroufa, C. Boutekedjiret, L. Petigny, N. Rakotomanomana, F. Chemat, Bio-refinery of orange peels waste: A new concept based on integrated green and solvent free extraction processes using ultrasound and microwave techniques to obtain essential oil, polyphenols and pectin, Ultrason. Sonochem. 24 (2015) 72–79. https://doi.org/10.1016/j.ultsonch.2014.11.015.
[7] FAO, Citrus fruit fresh and processed - Statistical Bulletin, Food and Agriculture Organization of the United Nations, Rome, 2017.
[8] D. Mamma, P. Christakopoulos, Biotransformation of Citrus By-Products into Value Added Products, Waste Biomass Valorization. 5 (2014) 529–549. https://doi.org/10.1007/s12649-013-9250-y.
[9] M. Russo, I. Bonaccorsi, V. Inferrera, P. Dugo, L. Mondello, Underestimated sources of flavonoids, limonoids and dietary fiber: Availability in orange's by-products, J. Funct. Foods. 12 (2015) 150–157. https://doi.org/10.1016/j.jff.2014.11.008.
[10] D. Barreca, G. Gattuso, E. Bellocco, A. Calderaro, D. Trombetta, A. Smeriglio, G. Laganà, M. Daglia, S. Meneghini, S.M. Nabavi, Flavanones: Citrus phytochemical with

570 health-promoting properties: Citrus phytochemical with health-promoting properties,
571 BioFactors. 43 (2017) 495–506. https://doi.org/10.1002/biof.1363.
572 [11] O. Benavente-García, J. Castillo, Update on Uses and Properties of Citrus Flavonoids:
573 New Findings in Anticancer, Cardiovascular, and Anti-inflammatory Activity, J. Agric.
574 Food Chem. 56 (2008) 6185–6205. https://doi.org/10.1021/jf8006568.
575 [12] P. Lavrador, V.M. Gaspar, J.F. Mano, Bioinspired bone therapies using naringin:
576 applications and advances, Drug Discov. Today. 23 (2018) 1293–1304.
577 https://doi.org/10.1016/j.drudis.2018.05.012.
578 [13] N. Mahato, K. Sharma, M. Sinha, M.H. Cho, Citrus waste derived nutra-/pharmaceuticals
579 for health benefits: Current trends and future perspectives, J. Funct. Foods. 40 (2018)
580 307–316. https://doi.org/10.1016/j.jff.2017.11.015.
581 [14] C. Zhong, Y. Zu, X. Zhao, Y. Li, Y. Ge, W. Wu, Y. Zhang, Y. Li, D. Guo, Effect of
582 superfine grinding on physicochemical and antioxidant properties of pomegranate peel,
583 Int. J. Food Sci. Technol. 51 (2016) 212–221. https://doi.org/10.1111/ijfs.12982.
584 [15] E.M. Garcia-Castello, A.D. Rodriguez-Lopez, L. Mayor, R. Ballesteros, C. Conidi, A.
585 Cassano, Optimization of conventional and ultrasound assisted extraction of flavonoids
586 from grapefruit (Citrus paradisi L.) solid wastes, LWT - Food Sci. Technol. 64 (2015)
587 1114–1122. https://doi.org/10.1016/j.lwt.2015.07.024.
588 [16] K. Sudto, S. Pornpakakul, S. Wanichwecharungruang, An efficient method for the large
589 scale isolation of naringin from pomelo (Citrus grandis) peel, Int. J. Food Sci. Technol. 44
590 (2009) 1737–1742. https://doi.org/10.1111/j.1365-2621.2009.01989.x.
591 [17] M.K. Khan, M. Abert-Vian, A.-S. Fabiano-Tixier, O. Dangles, F. Chemat, Ultrasound-
592 assisted extraction of polyphenols (flavanone glycosides) from orange (Citrus sinensis L.)
593 peel, Food Chem. 119 (2010) 851–858. https://doi.org/10.1016/j.foodchem.2009.08.046.
594 [18] S. Nipornram, W. Tochampa, P. Rattanatraiwong, R. Singanusong, Optimization of low
595 power ultrasound-assisted extraction of phenolic compounds from mandarin (Citrus
596 reticulata Blanco cv. Sainampueng) peel, Food Chem. 241 (2018) 338–345.
597 https://doi.org/10.1016/j.foodchem.2017.08.114.
598 [19] N. M'hiri, I. Ioannou, N.M. Boudhrioua, M. Ghoul, Effect of different operating
599 conditions on the extraction of phenolic compounds in orange peel, Food Bioprod.
600 Process. 96 (2015) 161–170. https://doi.org/10.1016/j.fbp.2015.07.010.
601 [20] B. Nayak, F. Dahmoune, K. Moussi, H. Remini, S. Dairi, O. Aoun, M. Khodir,
602 Comparison of microwave, ultrasound and accelerated-assisted solvent extraction for
603 recovery of polyphenols from Citrus sinensis peels, Food Chem. 187 (2015) 507–516.
604 https://doi.org/10.1016/j.foodchem.2015.04.081.
605 [21] Z. Liu, L. Qiao, H. Gu, F. Yang, L. Yang, Development of Brönsted acidic ionic liquid
606 based microwave assisted method for simultaneous extraction of pectin and naringin from
607 pomelo peels, Sep. Purif. Technol. 172 (2017) 326–337.
608 https://doi.org/10.1016/j.seppur.2016.08.026.
609 [22] B.B. Li, B. Smith, Md.M. Hossain, Extraction of phenolics from citrus peels: II. Enzyme-
610 assisted extraction method, Sep. Purif. Technol. 48 (2006) 189–196.
611 https://doi.org/10.1016/j.seppur.2005.07.019.
612 [23] S.E. El Kantar, N. Boussetta, N. Lebovka, F. Foucart, H.N. Rajha, R.G. Maroun, N.
613 Louka, E. Vorobiev, Pulsed electric field treatment of citrus fruits: Improvement of juice
614 and polyphenols extraction, Innov. Food Sci. Emerg. Technol. (2018).
615 https://doi.org/10.1016/j.ifset.2017.09.024.
616 [24] E. Luengo, I. Álvarez, J. Raso, Improving the pressing extraction of polyphenols of
617 orange peel by pulsed electric fields, Innov. Food Sci. Emerg. Technol. 17 (2013) 79–84.
618 https://doi.org/10.1016/j.ifset.2012.10.005.

[25] S. El Kantar, N. Boussetta, H.N. Rajha, R.G. Maroun, N. Louka, E. Vorobiev, High voltage electrical discharges combined with enzymatic hydrolysis for extraction of polyphenols and fermentable sugars from orange peels, Food Res. Int. 107 (2018) 755–762. https://doi.org/10.1016/j.foodres.2018.01.070.

[26] C.-I. Cheigh, E.-Y. Chung, M.-S. Chung, Enhanced extraction of flavanones hesperidin and narirutin from Citrus unshiu peel using subcritical water, J. Food Eng. 110 (2012) 472–477. https://doi.org/10.1016/j.jfoodeng.2011.12.019.

[27] M.-J. Ko, H.-L. Kwon, M.-S. Chung, Pilot-scale subcritical water extraction of flavonoids from satsuma mandarin (Citrus unshiu Markovich) peel, Innov. Food Sci. Emerg. Technol. 38 (2016) 175–181. https://doi.org/10.1016/j.ifset.2016.10.008.

[28] D. Lachos-Perez, A.M. Baseggio, P.C. Mayanga-Torres, M.R. Maróstica, M.A. Rostagno, J. Martínez, T. Forster-Carneiro, Subcritical water extraction of flavanones from defatted orange peel, J. Supercrit. Fluids. 138 (2018) 7–16. https://doi.org/10.1016/j.supflu.2018.03.015.

[29] A.N. Giannuzzo, H.J. Boggetti, M.A. Nazareno, H.T. Mishima, Supercritical fluid extraction of naringin from the peel of Citrus paradisi, Phytochem. Anal. 14 (2003) 221–223. https://doi.org/10.1002/pca.706.

[30] J.-Z. He, P. Shao, J.-H. Liu, Q.-M. Ru, Supercritical Carbon Dioxide Extraction of Flavonoids from Pomelo (Citrus grandis (L.) Osbeck) Peel and Their Antioxidant Activity, Int. J. Mol. Sci. 13 (2012) 13065–13078. https://doi.org/10.3390/ijms131013065.

[31] A.R. Toledo-Guillén, I. Higuera-Ciapara, G. García-Navarrete, J.C. de la Fuente, Extraction of Bioactive Flavonoid Compounds from Orange (Citrus sinensis) Peel Using Supercritical CO2, J. Biotechnol. 150 (2010) 313–314. https://doi.org/10.1016/j.jbiotec.2010.09.292.

[32] C. Oba, M. Ota, K. Nomura, H. Fujiwara, J. Takito, Y. Sato, Y. Ohizumi, H. Inomata, Extraction of nobiletin from Citrus Unshiu peels by supercritical fluid and its CRE-mediated transcriptional activity, Phytomedicine. 27 (2017) 33–38. https://doi.org/10.1016/j.phymed.2017.01.014.

[33] M.K. Khan, Zill-E-Huma, O. Dangles, A comprehensive review on flavanones, the major citrus polyphenols, J. Food Compos. Anal. 33 (2014) 85–104. https://doi.org/10.1016/j.jfca.2013.11.004.

[34] P.S. Jourdan, E.W. Weiler, R.L. Mansell, Radioimmunoassay for naringin and related flavanone 7-neohesperidosides using a tritiated tracer, J. Agric. Food Chem. 31 (1983) 1249–1255. https://doi.org/10.1021/jf00120a026.

[35] Y. Nogata, K. Sakamoto, H. Shiratsuchi, T. Ishii, M. Yano, H. Ohta, Flavonoid Composition of Fruit Tissues of Citrus Species, Biosci. Biotechnol. Biochem. 70 (2006) 178–192. https://doi.org/10.1271/bbb.70.178.

[36] W.V. De Castro, S. Mertens-Talcott, A. Rubner, V. Butterweck, H. Derendorf, Variation of Flavonoids and Furanocoumarins in Grapefruit Juices: A Potential Source of Variability in Grapefruit Juice–Drug Interaction Studies, J. Agric. Food Chem. 54 (2006) 249–255. https://doi.org/10.1021/jf0516944.

[37] J. Vanamala, L. Reddivari, K.S. Yoo, L.M. Pike, B.S. Patil, Variation in the content of bioactive flavonoids in different brands of orange and grapefruit juices, J. Food Compos. Anal. 19 (2006) 157–166. https://doi.org/10.1016/j.jfca.2005.06.002.

[38] C.-T. Ho, Phenolic Compounds in Food: An Overview, in: C.-T. Ho, C.Y. Lee, M.-T. Huang (Eds.), Phenolic Compd. Food Their Eff. Health I, American Chemical Society, Washington, DC, 1992: pp. 2–7. https://doi.org/10.1021/bk-1992-0506.ch001.

[39] AOAC, Official methods of analysis of the Association of Official Analytical Chemists, 15th ed., Association of Official Analytical Chemists, Arlington, VA, 1990.

[40] L. Dahdouh, M. Delalonde, J. Ricci, E. Ruiz, C. Wisnewski, Influence of high shear rate on particles size, rheological behavior and fouling propensity of fruit juices during crossflow microfiltration: Case of orange juice, Innov. Food Sci. Emerg. Technol. 48 (2018) 304–312. https://doi.org/10.1016/j.ifset.2018.07.006.

[41] P. Mouly, E.M. Gaydou, J. Estienne, Column liquid chromatographic determination of flavanone glycosides in Citrus: Application to grapefruit and sour orange juice adulterations, J. Chromatogr. A. 634 (1993) 129–134.

[42] M. Cháfer, C. González-Martínez, A. Chiralt, P. Fito, Microstructure and vacuum impregnation response of citrus peels, Food Res. Int. 36 (2003) 35–41. https://doi.org/10.1016/S0963-9969(02)00105-9.

[43] B.B. Li, B. Smith, Md.M. Hossain, Extraction of phenolics from citrus peels, Sep. Purif. Technol. 48 (2006) 182–188. https://doi.org/10.1016/j.seppur.2005.07.005.

[44] D.N. Sila, S. Van Buggenhout, T. Duvetter, I. Fraeye, A. De Roeck, A. Van Loey, M. Hendrickx, Pectins in Processed Fruits and Vegetables: Part II-Structure-Function Relationships, Compr. Rev. Food Sci. Food Saf. 8 (2009) 86–104. https://doi.org/10.1111/j.1541-4337.2009.00071.x.

[45] M. Puri, D. Sharma, C.J. Barrow, Enzyme-assisted extraction of bioactives from plants, Trends Biotechnol. 30 (2012) 37–44. https://doi.org/10.1016/j.tibtech.2011.06.014.

[46] J.A. Del Río, M.D. Fuster, F. Sabater, I. Porras, A. García-Lidón, A. Ortuño, Selection of citrus varieties highly productive for the neohesperidin dihydrochalcone precursor, Food Chem. 59 (1997) 433–437. https://doi.org/10.1016/S0308-8146(96)00303-2.

[47] P.C. Ho, D.J. Saville, P.F. Coville, S. Wanwimolruk, Content of CYP3A4 inhibitors, naringin, naringenin and bergapten in grapefruit and grapefruit juice productsq, Pharm. Acta Helv. 74 (2000) 379–385.

[48] J. Zhang, Flavonoids in Grapefruit and Commercial Grapefruit Juices: Concentration, Distribution, and Potential Health Benefi ts, Proc. Fla. State Hortic. Soc. 120 (2007) 288–294.

[49] P.A. Vesilind, Treatment and disposal of wastewater sludges, Ann Arbor Science Publishers, 1979.

[50] M. Smollen, Moisture retention characteristics and volume reduction of municipal sludges, Water SA. 14 (1988) 25–28.

[51] U.K. Möller, Water Binding, in: J.B. Carberry, A.J. Englande (Eds.), Sludge Charact. Behav., Springer Netherlands, Dordrecht, 1983: pp. 182–194. https://doi.org/10.1007/978-94-009-6860-8_7.

[52] C.E. Arundel, The role of floc density measurements in analyzing sludge dewatering characteristics, Thesis, Virginia Tech, 1986. https://vtechworks.lib.vt.edu/handle/10919/41559 (accessed April 18, 2019).

[53] M.-A. Lazouk, R. Savoire, A. Kaddour, J. Castello, J.-L. Lanoisellé, E. Van Hecke, B. Thomasset, Oilseeds sorption isoterms, mechanical properties and pressing: Global view of water impact, J. Food Eng. 153 (2015) 73–80. https://doi.org/10.1016/j.jfoodeng.2014.12.008.

[54] K.D. Ross, Differential Scanning Calorimetry of Nonfreezable Water in Solute—Macromolecule—Water Systems, J. Food Sci. 43 (1978) 1812–1815. https://doi.org/10.1111/j.1365-2621.1978.tb07420.x.

[55] D.J. Lee, Y.H. Hsu, Measurement of Bound Water in Sludges: A Comparative Study, Water Environ. Res. 67 (1995) 310–317.

[56] B.F. Ruth, Studies in Filtration III. Derivation of General Filtration Equations, Ind. Eng. Chem. 27 (1935) 708–723. https://doi.org/10.1021/ie50306a024.

[57] B.F. Ruth, Correlating Filtration Theory with Industrial Practice., Ind. Eng. Chem. 38 (1946) 564–571. https://doi.org/10.1021/ie50438a010.

[58] E. Iritani, N. Katagiri, Developments of Blocking Filtration Model in Membrane Filtration, KONA Powder Part. J. 33 (2016) 179–202. https://doi.org/10.14356/kona.2016024.
[59] A. Tapre, R.K. Jain, Pectinases: Enzymes for fruit processing industry, Int. Food Res. J. 21 (2014) 447–453.
[60] M.M. Victor, J.M. David, M.C.K. Sakukuma, E.L. França, A.V.J. Nunes, A simple and efficient process for the extraction of naringin from grapefruit peel waste, Green Process. Synth. 7 (2018) 524–529. https://doi.org/10.1515/gps-2017-0112.

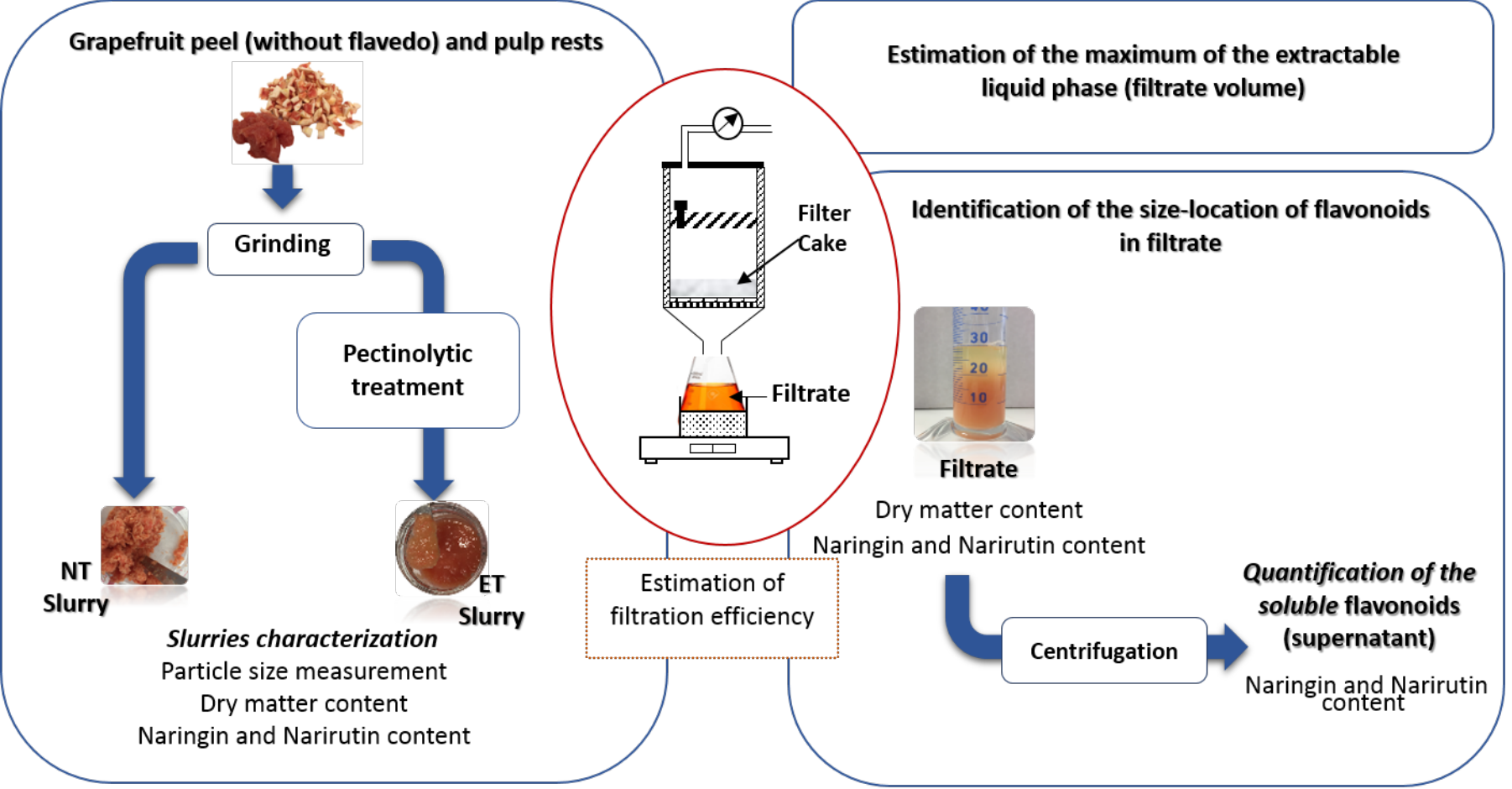
Suggested Referees

1. I. Ioannou
Université de Lorraine, ENSAIA - Laboratoire d'Ingénierie de biomolécules (LIBio), 2 avenue de la Forêt de Haye, TSA 40602, Vandoeuvre Cedex 54518, France
irina.ioannou@univ-lorraine.fr

2. Alfredo Cassano
Institute on Membrane Technology, ITM-CNR, c/o University of Calabria, via P. Bucci, 17/C, I-87030 Rende (Cosenza), Italy
a.cassano@itm.cnr.it

3. Noelia Rosales-Conrado
Departamento de Química Analítica, Facultad de Ciencias Químicas, Universidad Complutense de Madrid, Madrid 28040, Spain
nrosales@ucm.es

4. Rosângela Bergamasco
State University of Maringa, Department of Chemical Engineering, Maringa 87020-900, Parana, Brazil
rosangela@deq.uem.br


Grapefruit peel (without flavedo) and pulp rests
Grinding
Pectinolytic treatment
NT Slurry
ET Slurry
Slurries characterization
Particle size measurement
Dry matter content
Naringin and Narirutin content
Filter Cake
Filtrate
Estimation of filtration efficiency
Estimation of the maximum of the extractable liquid phase (filtrate volume)
Identification of the size-location of flavonoids in filtrate
Filtrate
Dry matter content
Naringin and Narirutin content
Centrifugation
Quantification of the soluble flavonoids (supernatant)
Naringin and Narirutin content