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Treatment of cauliflower processing wastewater by nanofiltration and reverse osmosis in view of recycling

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Keywords

Membrane process, food industry, water reuse, effluent treatment, cauliflower, reverse osmosis

Abstract

The vegetable industry is a large consumer of drinking water. This paper investigates the possibilities of Reverse Osmosis (RO) or tight Nanofiltration (NF) for the treatment of cauliflower blanching wastewater with a view to recycling within the production unit. Ultrafiltration at 100 000 g.mol⁻¹ molecular weight cut-off was necessary to decrease turbidity below 1 NTU as required before NF or RO. Three NF (DK, NF270 and SRD3) and one RO (ESPA4) membranes were tested at bench-scale in a crossflow filtration mode. Only RO allowed to reach the desired quality for a reuse purpose, with an acceptable residual COD content of 225 mg O₂.L⁻¹. The Solution-Diffusion model was validated for the transfer of glucose and fructose, for NF270, DK and ESPA4 membranes and their permeability coefficients calculated.

Highlights:

- Ultrafiltration followed by reverse osmosis allows to consider recycling of cauliflower wastewater
- ESPA4 membrane at 19 bar leads to 70 L.h⁻¹.m⁻² permeate flux and 95% COD rejection
- Solution-diffusion model considering concentration polarization was successfully applied
- DK, NF270 and ESPA4 permeabilities for fructose and glucose were determined
- Nanofiltration with 150-300 g.mol⁻¹ molecular weight cut-off is not suitable, due to the transfer of small metabolites

34 1. Introduction

35 The industries that consume a large amount of water are more and more keenly concerned by the necessity to
36 save water resources. The food industry, including the fruit and vegetable transformation sector, is
37 particularly concerned: according to a study of the European Commission (European, 2018), water
38 consumption in the latter ranges from 0.5 to 15 m³/ton of processed raw material. Reuse (recycling without
39 treatment) and reconditioning (recycling after treatment) of these effluents thus become consequential in
40 order to reduce the environmental impact of these industries and restore water quality to an acceptable level.

41 Considered as robust, flexible and “green” (Dewettinck and Le, 2011; Guiga and Lameloise, 2019),
42 membrane processes are becoming favourite technologies for treating wastewater before recycling
43 (Warsinger et al., 2018; Wenten and Khoiruddin, 2016) especially for the food processing industry (Meneses
44 et al., 2017). Among them, reverse osmosis (RO) or tight nanofiltration (NF) ensure the highest water quality
45 and have already proved valuable for wastewater reconditioning in the dairy (Bortoluzzi et al., 2017; Brião et
46 al., 2019; Suárez et al., 2014) and brewery industries (Braeken et al., 2004). They can provide high permeate
47 fluxes and rejections at relatively low transmembrane pressure (*TMP*) provided several issues are considered:
48 first, adequate pre-treatment should be implemented to bring the Silt Density Index (SDI) to below 5 and
49 turbidity to 1 NTU (Sim et al., 2018). Second, membrane operations should be run below the critical flux to
50 avoid irreversible fouling (Aimar et al., 2010).

51 The possibilities of reuse and reconditioning of wastewater in the food industry, and the development of
52 toolboxes to evaluate the impact of these solutions are primary objectives in the French research program
53 MINIMEAU (ANR-17-CE10-0015). A recent study on carrot peeling wastewater highlighted that high-
54 quality water could be obtained through RO or tight NF membranes after microfiltration (MF) (Garnier et al.,
55 2020). In the vegetable-processing industry, one plant usually transforms different vegetables, either
56 simultaneously or successively. Consequently, the present study aimed to check the feasibility of the process
57 developed in Garnier et al. (2020) for wastewater arising from rinsing of carrots after peeling, for treating
58 wastewater from cauliflower blanching.

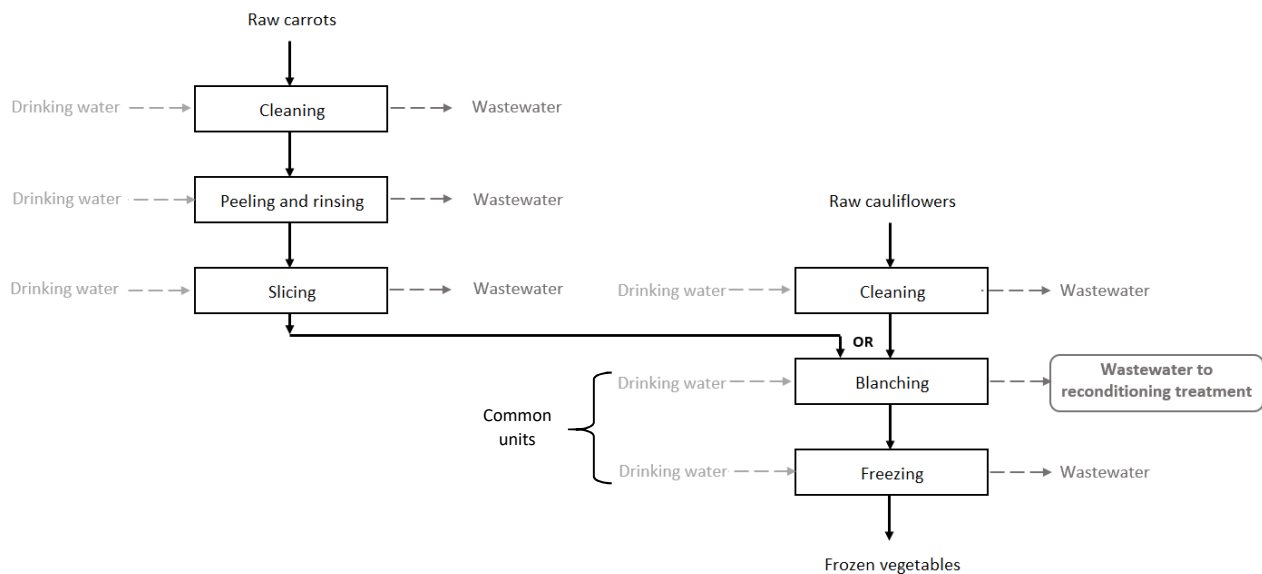
59 In the Drinking Water Standard, lists of parameters are to be respected for both drinking water quality and
60 the water source from which it originates (mainly surface or groundwater), while there is no regulatory
61 context outlining the water quality of recycled industrial wastewater. For food safety, drinking water is
62 usually requested in the food processing industry (Casani et al., 2005). In this work, characterisation of the
63 cauliflower blanching wastewater was made in order to select key parameters to be eliminated. Optimized
64 pre-treatment and membrane treatment were selected with drinking water quality as the objective. Finally,
65 the Solution-Diffusion model, commonly used to represent water and solutes transfer in non-porous
66 membranes (Qasim et al., 2019; Wijmans and Baker, 1995) was applied to obtain water and solutes
67 permeabilities for NF and RO membranes, considering the concentration polarization phenomenon. In this
68 work, it was acquired especially for sugars contained in cauliflower effluent, and compared with data

69 extracted from the literature results. Such database is essential for the design tool developed in the
70 MINIMEAU Project.

71 **2. Material and Methods**

72 *2.1. Wastewater origins*

73 The wastewater was obtained from the French factory already selected by the Technical Center for Food
74 Product Conservation (CTCPA, Paris, France) for the study of Garnier et al. (2020) on carrot processing
75 wastewater. This factory produces several frozen vegetables sometimes on the same production line.
76 Effluents of cauliflower processing are produced at the outlet of several operation units. In particular, a
77 cleaning unit is used only for cauliflower and a blanching and freezing unit alternately for all vegetables
78 (Fig. 1). Blanching consists in a short heat treatment with hot water (80 °C to 100 °C) to inactivate or delay
79 bacterial growth and enzyme action. Cauliflower wastewater collected at the outlet of the blanching
80 operation unit was selected for our study and stored at -18 °C before treatment tests. Drinking water used in
81 the factory was analysed as a reference.



82

83 **Fig. 1.** Schematic representation of vegetable processing operation in the factory under study.

84 *2.2. Analytical methods*

85 The following analyses were performed:

- 86 - Global parameters: Total Suspended Solids (TSS), particulate and dissolved Carbon Oxygen
87 Demand (COD), conductivity, pH, turbidity, Carbonate Hardness (CH), Total Nitrogen (TN), optical
88 density (OD), color,
89 - Dissolved organic pollution: glucose, fructose (accuracy $\pm 4\%$) and sucrose (accuracy $\pm 5\%$),

- Free and total chlorine (accuracy $\pm 0.06 \text{ mg.L}^{-1} \text{ Cl}_2$),
- Ionic composition: chloride, nitrite, nitrate, phosphate, sulphate, sodium, ammonium, potassium, magnesium and calcium (accuracy $\pm 2.5\%$).

Most analytical methods are already described in Garnier *et al.* (2020). High-performance ion-exchange chromatography (HPIC) was used to analyse anions and cations as well as sugars. At the pH of the effluent (4.7) and based on the equilibrium diagram of CO_2 , the concentration of carbonate in the effluent was negligible meaning that Carbonate Hardness (CH) could represent the concentration of bicarbonate.

COD (accuracy $\pm 3\%$), TN, CH (variable accuracy), chlorine (free and total) were determined with rapid test tubes and photometric measurement (Nanocolor vis II - Macherey Nagel, Hoerd, France). Color and turbidity (accuracy $\pm 2\%$) were performed with the same photometric material. Color was established in the CIELAB color space adopted by the International Commission on Illumination (CIE) in 1976, where color is expressed as three values: L^* (lightness from black (0) to white (100)), a^* (from green to red) and b^* (from blue to yellow).

COD being mainly composed of sugars, additional organic matter was quantified through a differential COD ($\text{mg O}_2.\text{L}^{-1}$) defined as:

$$\text{COD}_{\text{diff}} = \text{COD} - \text{COD}_{\text{sugars}} \quad (1)$$

Where $\text{COD}_{\text{sugars}}$ is the COD ($\text{mg O}_2.\text{L}^{-1}$) deduced from sugar concentrations and the stoichiometry of the oxidation reaction (1g.L⁻¹ of fructose and glucose corresponds to a COD of 1.066 $\text{mg O}_2.\text{L}^{-1}$ when 1 g.L⁻¹ of sucrose corresponds to a COD of 1.122 $\text{mg O}_2.\text{L}^{-1}$).

UV absorbance measurement at 216.4 nm ($\text{OD}_{216.4}$) and 264.4 nm ($\text{OD}_{264.4}$) allowed evaluating the presence of amino acids or peptides. Samples were diluted ten times.

2.3. Pre-treatment

Sieving at 169 μm and 79 μm followed by dead-end MF 0.6 μm was first implemented, as in Garnier *et al.* (2020). The obtained turbidity remaining too high to fulfil quality requirements for further NF or RO step, the permeate obtained from MF was further ultra-filtrated. Three UF organic membranes with different Molecular Weight Cut-Off (MWCO), namely 100 000, 10 000 and 5 000 g.mol^{-1} , were tested therefore (Table 1).

2.4. NF and RO membranes

Cauliflower processing wastewater contained sucrose ($\text{MW} = 342 \text{ g.mol}^{-1}$) and mainly glucose and fructose ($\text{MW} = 180.16 \text{ g.mol}^{-1}$). Consequently, three NF membranes with MWCO between 150 and 300 g.mol^{-1} and one RO membrane were selected for further purification (Table 1). New membranes were stored dry at 4 °C.

122 To remove the protective coating or storage solution, they were dipped before experiments in a 0.4 g.L⁻¹
123 KOH solution for 2 h and then in deionized water for 24 h minimum. Prior to experiments, membranes were
124 pre-compacted (20 bar, 15 min) in the filtration device.

125

126 **Table 1**

127 Overview of membranes characteristics according to manufacturer's data

Supplier	Membrane	Type	Rejection	MWCO (g.mol ⁻¹)	Active layer polymer	Maximum temperature (°C)	Maximum pressure (bar)	Pure water permeability (20°C - L.h ⁻¹ .m ⁻² .bar ⁻¹)
Alfa Laval (Elancourt, France)	FS40PP	UF	-	100 000	Fluoro polymer	60	10	78 ^a
Koch Membrane Systems Division (Lyon, France)	HFK-131	UF	-	10 000	Polyethersulfone	55	9.7	53 ^a
Koch Membrane Systems Division (Lyon, France)	HFK-328	UF	-	5 000	Polyethersulfone	55	9.7	33 ^a
GE water & process technologies (Saint-Thibault-des- Vignes, France)	DK	NF	98% 2000 ppm MgSO ₄ (7.6 bar, 25°C)	150–300	Semi-aromatic polypiperazine amide	50	41.4 bar if θ < 35 °C; 30 bar otherwise	4.0 ^b
DOW France (Saint-Denis, France)	NF270	NF	97% 2000 ppm MgSO ₄ (4.8 bar, 25°C)	150–300	Semi-aromatic polypiperazine amide	45	41	14.8 ^b
Koch Membrane Systems Division (Lyon, France)	SR3D	NF	> 99.0% 5000 ppm MgSO ₄ (6.5 bar, 25°C)	200	Proprietary Thin- Film Composite polyamide	50	44.8	7.5 ^b
Hydranautics – Nitto France (Roissy, France)	ESPA4	RO	99.2% (99.0% minimum) 1500 ppm NaCl (10.3 bar, 25°C)	-	Aromatic polyamide Thin- Film Composite	45	40	6.3 ^b
(^a)	This	study;	(^b)	from	Garnier	et	al.,	2020

128

2.5. Membrane setup and operating conditions

Experiments were run using the LabStak M20 filtration device from Alfa Laval described in Garnier *et al.* (2020). It allows testing several flat-sheet membranes simultaneously. The effective area for each membrane was $2 \times 0.018 \text{ m}^2$.

To study water permeability and solutes' rejection, experiments were run in total recirculation mode: deionized water filtration ($< 2 \text{ h}$) for pure water permeability measurement, wastewater filtration ($< 8 \text{ h}$), and deionized water filtration *once more* (after rinsing with deionized water for 10 min minimum). For all experiments, retentate flowrate was set at 300 L.h^{-1} and temperature at 20°C . For UF membranes, two transmembrane pressures (*TMP*) were tested: 3 and 5 bar. For NF and RO membranes, *TMP* was increased from 5 to 25 bar by 5 bar steps and then decreased symmetrically. Sampling and measurements were done after at least 30 min run.

Once UF membrane selection was made, filtration was run in discontinuous mode to produce a sufficient amount of ultra-filtrated permeate: the permeate stream was collected in a distinct tank and the concentrate returned to the feed tank until reaching the desired volume reduction ratio (*VRR*):

$$VRR = \frac{V_i}{V_f} \quad (2)$$

Where V_i is the initial volume in the feed tank and V_f the final volume.

3. Filtration efficiency and solution-diffusion model application

Filtration efficiency was estimated by the permeate flux J_p (m.s^{-1} or $\text{L.h}^{-1}.\text{m}^{-2}$) evolution with *TMP*, as well as by the solutes' rejections Tr_i , calculated for each solute or parameter i (COD, COD_{diff}, total nitrogen, OD, sugars, ions).

$$J_p = \frac{Q_p}{S} \quad (3)$$

$$Tr_i = \frac{C_{r,i} - C_{p,i}}{C_{r,i}} \quad (4)$$

Where Q_p ($\text{m}^3.\text{s}^{-1}$ or L.h^{-1}) is the permeate flow rate, S (m^2) is the effective membrane area and $C_{r,i}$ and $C_{p,i}$ (mol.m^{-3}) are the concentrations of solute i respectively in the retentate and in the permeate.

Experimental solute i flux (J_i ($\text{mol. s}^{-1}.\text{m}^{-2}$)) through the membrane was calculated according to:

$$J_i = C_{p,i} \times J_p \quad (5)$$

The Solution-Diffusion (*SD*) model is commonly used for describing the transport of non-ionic organic solutes through dense membranes such as RO and tight NF ones (Nguyen, D. *et al.*, 2016; Qasim *et al.*, 2019;

Wijmans and Baker, 1995). For diluted solutions and in the absence of irreversible fouling, this model can be simplified to predict J_p and J_i , provided the water and solutes permeabilities are known. When concentration polarization is considered on the retentate side (Aimar et al., 2010), the following equation arises for the permeate flux:

$$J_p = A_w \times [TMP - \Delta\pi] \quad (6)$$

Where A_w ($\text{m.s}^{-1}.\text{Pa}^{-1}$ or $\text{L.h}^{-1}.\text{m}^{-2}.\text{bar}^{-1}$) is the pure water permeability, $TMP = \frac{P_f + P_r}{2} - P_p$ (Pa or bar) is the transmembrane pressure (P_f , P_r and P_p are the pressures in the feed, the retentate and the permeate, respectively (bar)), and $\Delta\pi = \pi_{r,m} - \pi_p$ (Pa or bar) is the osmotic pressure gradient between the membrane interface in the retentate (considering concentration polarization) and the permeate.

A_w could be deduced with Eq. 6 for pure water filtration experiments at different TMP , before and after effluent treatment on the membrane.

With the same SD model, solute i flux is given by:

$$J_i = B_i \times [C_{r,m,i} - C_{p,i}] \quad (7)$$

Where B_i (m.s^{-1}) is the membrane permeability to solute i and $C_{r,m,i}$ (mol.m^{-3}) is its concentration at the membrane interface in the retentate, that can be estimated through the film model theory:

$$C_{r,m,i} = C_{p,i} + [C_{r,i} - C_{p,i}] \times \exp \frac{J_p}{k_i} \quad (8)$$

Where k_i (m.s^{-1}) is the mass transfer coefficient of solute i in the polarization layer.

175

To assess the simplified Solution-Diffusion model and determine k_i and B_i , eq. (5), (7) and (8) were combined to give:

$$\ln \left(\frac{C_{p,i} \times J_p}{C_{r,i} - C_{p,i}} \right) = \ln(B_i) + \frac{J_p}{k_i} \quad (9)$$

Plotting $\ln \left(\frac{C_{p,i} \times J_p}{C_{r,i} - C_{p,i}} \right)$ vs J_p led to the graphical determination of B_i and k_i .

4. Results and Discussion

4.1. Characterisation of raw wastewater

Cauliflower processing wastewater had a particular odour which could be attributed to sulphur and N-bearing molecules, and foaming attested the presence of proteins. Table 2 shows the composition of the blanching wastewater (two samples). The difference between total and dissolved COD was within the accuracy limit.

186 Fructose and glucose represented respectively 72% and 46% of the total COD of the raw wastewater,
187 showing its variability. These proportions increased to 99% and 79% when raw wastewater was micro
188 filtrated, indicating that these are the main dissolved organic substances present. Other organic dissolved
189 substances were estimated by UV spectrophotometry at 216.4 nm (possibly corresponding to peptide bonds)
190 and 264.4 nm (corresponding to aromatic rings), as well as by TN measurement. These may be amino acids
191 or peptides/proteins containing aromatic rings like histidine, phenylalanine, tryptophan and tyrosine, present
192 in cauliflowers.

193 By comparing the composition of wastewater with that of typical cauliflower (Table 2), the transfer of sugars
194 and most minerals (phosphate, sulphate, sodium, potassium, magnesium and calcium) into wastewater during
195 blanching is confirmed. Glucose and fructose are transferred in the same proportion. Sucrose, present in
196 small amounts in cauliflower (Bhandari and Kwak, 2015) is also transferred into the wastewater.

197 As in the study on carrot wastewater (Garnier et al., 2019; Garnier et al., 2020), TSS, COD, conductivity,
198 fructose, glucose and sucrose were selected as key parameters. Concerning the French drinking water
199 standard and regarding ions, only ammonium was out of the range (8–12 mg.L⁻¹ in raw wastewater vs 0.1
200 mg.L⁻¹ in French standard), so it was selected as another key parameter. The other ions were merged as key
201 parameter “conductivity”. As raw wastewater was white with an orange tint, color was also monitored.

202

203

204

205 **Table 2**

206 Characteristics and composition of cauliflower blanching raw wastewater (two samples from [cauliflower](#)
207 blanching), cauliflower and drinking water

Parameter	Raw wastewater	Cauliflower (USDA*)	Drinking water (French standard)	Drinking water (factory)
Temperature (°C)	50 – 80	ni	≤ 25	nd
TSS (mg.L ⁻¹)	150 – 290	ni	-	nd
Total COD (mg O ₂ .L ⁻¹)	7 410 – 10 120	ni	-	3.8
Dissolved COD (mg O ₂ .L ⁻¹)	7 560 – 10 290	ni	-	nd
Total Nitrogen (mg N.L ⁻¹)	190 – 265	ni	-	nd
Conductivity (μS.cm ⁻¹)	2 420 – 2 640	ni	180 – 1 000	261
pH	5.8 – 5.9	ni	6.5 – 9	6.86
Turbidity (NTU)	45 – 125	ni	≤ 0.5	< 0.1
Color	L* = 66 – 90 a* = 11 – 23 b* = 11 – 21	ni	Acceptable to consumers and no abnormal change	nd
UV absorbance	OD _{216.4} = 2 200 – 2 360 OD _{264.4} = 950 – 1 230	ni	-	nd
Carbonate hardness (°f)	24.5 – 23.5	ni	-	3.5
Fructose	2 130 – 2 210 mg.L ⁻¹	0.97 g/100g	-	absence
Glucose	1 930 – 2 190 mg.L ⁻¹	0.94 g/100g	-	absence
Sucrose	170 – 630 mg.L ⁻¹	0 g/100g	-	absence
Cl ⁻ (mg.L ⁻¹)	110 – 140	ni	≤ 250	42
NO ₂ ⁻ (mg.L ⁻¹)	< LOD	ni	≤ 0.5	< LOD
NO ₃ ⁻ (mg.L ⁻¹)	16 – 21	ni	≤ 50	5
SO ₄ ²⁻ (mg.L ⁻¹)	100 – 140	ni	≤ 250	15
PO ₄ ³⁻	80 – 100 mg.L ⁻¹	P: 44 g/100g	-	< LOD
Na ⁺	32 – 40 mg.L ⁻¹	30 g/100g	≤ 200	19
NH ₄ ⁺ (mg.L ⁻¹)	8 – 12	ni	≤ 0.1	< LOD
K ⁺	1 030 – 1 050 mg.L ⁻¹	299 g/100g	-	4
Mg ²⁺	34 – 44 mg.L ⁻¹	15 g/100g	-	6
Ca ²⁺	78 – 92 mg.L ⁻¹	22 g/100g	-	22

208 * <https://fdc.nal.usda.gov/fdc-app.html#/food-details/170393/nutrients> published 4/1/2019.

** ni: not indicated, nd: not determined

The removal efficiency of the sieving-MF pre-treatment was 60% for TSS, 34% for COD, 17% for OD_{216.4}, 39% for OD_{264.4} and null for sugars. Nevertheless, the residual turbidity (average 48 NTU) was too high for feeding a NF or RO process. Additional pre-treatment with UF membrane (MWCO of 100 000, 10 000 or 5 000 g.mol⁻¹) was then experienced on microfiltration permeate. Whatever the pressure (3 and 5 bar) and the membrane, the residual turbidity was below 0.5 NTU, complying with the recommendations of the NF and RO manufacturers. Membrane FS40PP with the highest MWCO (100 000 g.mol⁻¹) and a 3 bar pressure was preferred as it limited permeability loss during the filtration stage (41%, against 68% and 62% with membranes of 10 000 and 5 000 g.mol⁻¹ MWCO, respectively).

Finally, the removal efficiency of this pre-treatment (sieving + microfiltration + ultrafiltration on FS40PP at VRR 3.5) reached 99% for turbidity, 50% for COD, 12% for COD_{diff}, 42% for TN, 40% for conductivity, 26% for OD_{216.4} and 49% for OD_{264.4}. The decrease in sugar concentrations was unexpected: 56% for fructose, 98% for glucose and 100% for sucrose. This result was due to fermentation (Paramithiotis et al., 2010) during storage even if mostly at 4°C, detected by the decrease of pH and chlorine concentration, and confirmed by specific acetate and lactate peaks on HPIC chromatograms.

224 4.3. NF and RO performances

225 4.3.1. Critical flux, concentration polarization and fouling

All the following experiments were performed with pure water or with the pre-treated wastewater produced as described in section 4.2. Results are presented on Fig. 2, from which pure water permeability A_w could be deduced according to Eq. 6 (with $\Delta\pi = 0$) (Table 3). For SR3D and ESPA4 membranes, A_w values before effluent filtration were similar to those in Garnier et al (2020) (Table 1), while they appeared much lower or higher respectively for NF270 (-30%) and DK (+ 45%).

A small A_w decrease was observed after NF or RO treatment proving that fouling had occurred during effluent treatment. For wastewater filtration at the lowest TMP values, the relation between J_P and TMP was linear, showing that no fouling had yet occurred but only a reversible concentration polarization phenomenon (eq. 8) (Aimar, 2006). Above a given flux value, named the critical flux, it was no longer linear meaning that irreversible concentration polarization occurred together with a likely irreversible fouling (Aimar et al., 2010). The critical flux and corresponding pressure obtained graphically (Table 4) show that membranes do not differ from each other on these parameters but rather on A_w level (Table 3). To confirm the fouling phenomenon, J_P was studied over time and compared with initial pure water flux: for each TMP applied up to 15 bar, flux measurements were made after 5 min (initial flux) and 30 min; for 25 bar, it was after 5, 15 and 30 min. TMP was then decreased (20, 15, 10, 5 and 1 bar) and a measurement was made after 10-min run. As shown in Fig. 2, during pressure increase and below the critical flux, the steady state was quickly reached as the permeate flux was almost the same after 5 and 30 min. On the contrary, for NF270 membrane

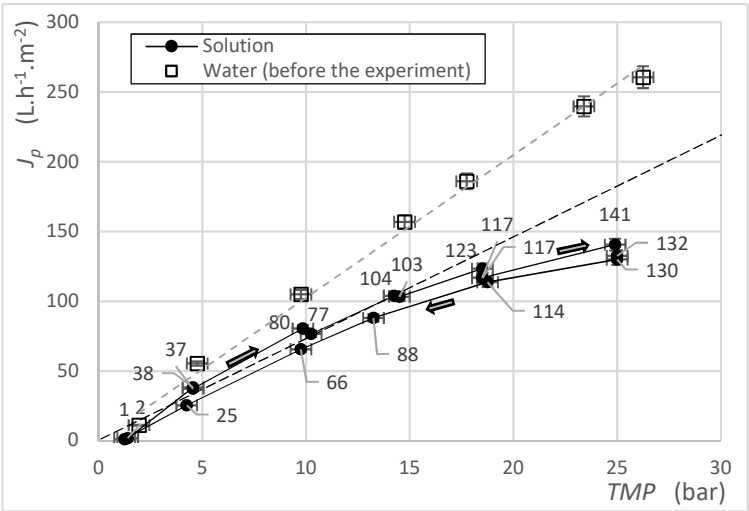
and above the critical flux, a decrease of up to 10% of J_p was observed over time (Fig. 2a). Moreover, hysteresis appeared for all membranes when TMP was decreased, confirming that critical flux had been exceeded and that fouling had developed (Aimar et al., 2010).

Table 3

Pure water permeability A_w before and after NF and RO treatment

Supplier	Membrane	Type	A_w measured at 20°C (L.h ⁻¹ .m ⁻² .bar ⁻¹)	
			Before effluent filtration	After effluent filtration
DOW	NF270	NF	10.4 ± 0.4	8.9 ± 0.3
Koch	SR3D	NF	7.0 ± 0.2	6.4 ± 0.2
GE	DK	NF	5.8 ± 0.2	5.3 ± 0.2
Hydranautics	ESPA4	RO	6.4 ± 0.2	5.3 ± 0.2

a)



b)

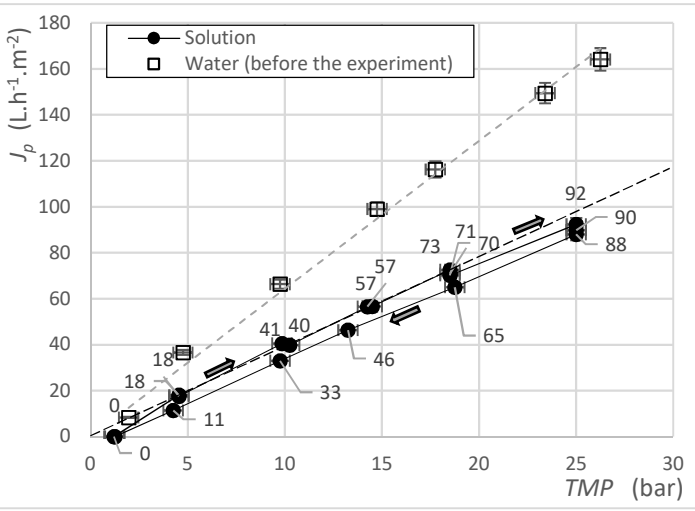


Fig. 2. Permeate flux for pure water and cauliflower pre-treated wastewater, highlighting hysteresis - J_p values obtained over time are indicated directly on the points:

(a) NF270 membrane (b) ESPA4 membrane.

259 **Table 4**

260 Critical flux and corresponding pressure for NF and RO membranes

Membrane	NF270	SR3D	DK	ESPA4
Critical flux (L.h ⁻¹ .m ⁻²)	100	100	90	90
Pressure at critical flux (bar)	15	18	19	24

261 *4.3.2. Solutes rejections*

262 The rejection performances of the membranes were compared before and after the critical flux as both
 263 concentration polarization and fouling might have a beneficial or detrimental impact on membrane
 264 selectivity (Aimar et al., 2010). Rejections of COD, glucose and fructose versus permeate flux are given Fig.
 265 3. After pre-treatment, sucrose concentrations were below the quantification limit and were not considered.
 266 As expected, RO gave the highest rejections. For NF, rejections decreased with MWCO increase (Table 1),
 267 assessing that size exclusion was the major mechanism for NF membranes. DK and NF270 showed
 268 comparable patterns.

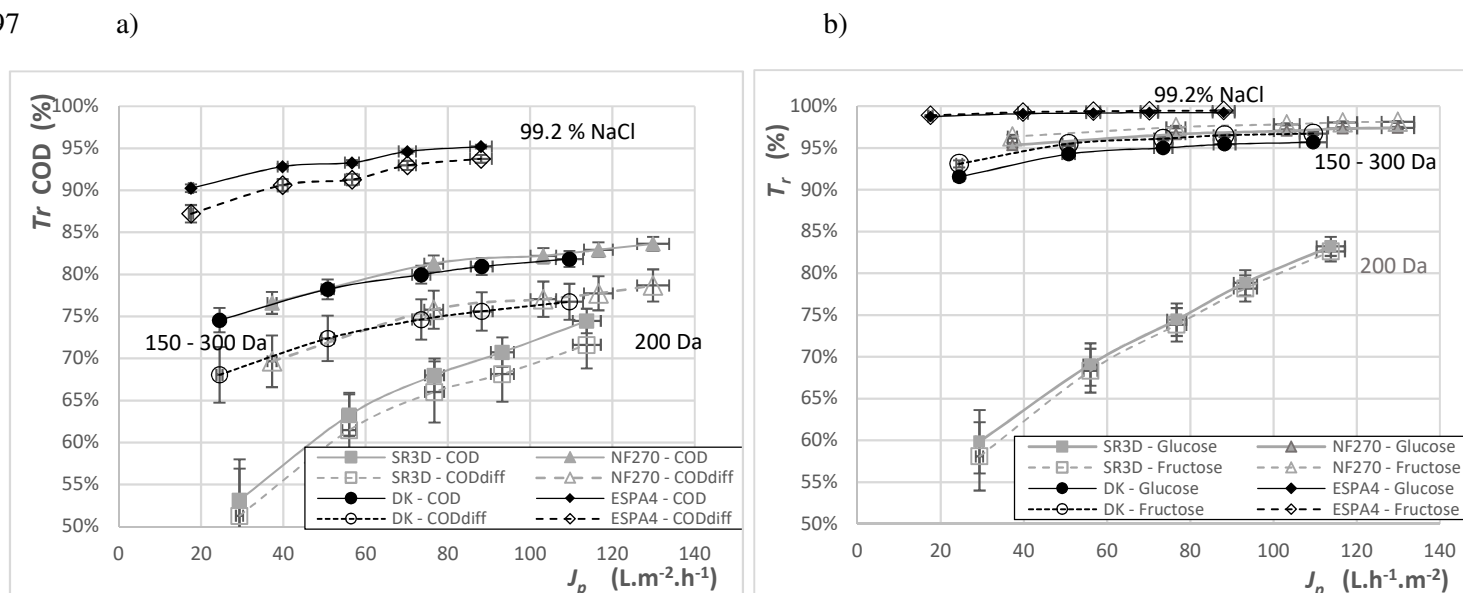
269 The COD rejections for DK (150–300 g.mol⁻¹), NF270 (150–300 g.mol⁻¹), SR3D (200 g.mol⁻¹) and ESPA4
 270 (99.2% NaCl) membranes increased with *TMP* from 74.6% to 81.8%, 76.6% to 83.6%, 53.2% to 74.5% and
 271 90.3% to 95.2%, respectively (Fig. 3-a). At 25 bar, the minimal COD values in the permeate remained high
 272 for NF (above 700 mg O₂.L⁻¹), and much lower for ESPA4 (208 mg O₂.L⁻¹). COD rejections continually
 273 increased but more and more slowly showing that exceeding the critical flux (between 90 and 100 L. h⁻¹.m⁻²)
 274 is not efficient. For fructose and glucose (87–89% and 11–13% of total sugars in the retentate, respectively)
 275 the same membrane ranking was observed (Fig. 3-b). The rejections were at least 95% for NF270, 91% for
 276 DK and 98% for ESPA4 membrane which was consistent with other studies on sugars (Garnier et al., 2020;
 277 Nguyen et al., 2015). Low rejection (58 to 86%) was observed for SR3D confirming a different behaviour, as
 278 already noticed on carrot processing wastewater for protons, amino-acid-type and bicarbonates (Garnier et
 279 al., 2020). COD_{diff} rejection was always lower than the COD rejection (Fig. 3-a) which suggested that
 280 organic non-sugar molecules showed poor rejection. To investigate this, the rejections of TN, OD_{216.4} and
 281 OD_{264.4} were examined and compared with that for COD and COD_{diff} (Fig. 4). Results for NF270 membrane
 282 were not presented, as it behaves like DK membrane.

283 OD_{216.4} rejections were below COD rejections for SR3D (200 g.mol⁻¹), similar for NF270 and DK (150–300
 284 g.mol⁻¹) and above for ESPA4 membrane. This suggests that size exclusion was the main selectivity factor
 285 and that OD_{216.4} represents non-aromatic and non-sugar molecules with molecular weight below 150 g.mol⁻¹
 286 (Fig. 4). For ESPA4 membrane (RO), OD_{216.4}, OD_{264.4} and TN rejections were above COD_{diff} rejection
 287 suggesting that small and undetermined molecules migrate through the membrane (Fig. 4-c). For all
 288 membranes, OD_{264.4} rejections were similar and slightly above TN rejections suggesting that the main part of
 289 nitrogen compounds detected by TN measurements absorb at 264.4 nm and would thus contain aromatic
 290 amino acids identified in cauliflower (Table 5). OD_{264.4} rejections of SR3D, NF270 and DK membranes were

291 respectively between 65.0% and 80.9%, 89% and 91.7% and 87.8% and 90.1%, consistent with MW of those
 292 aromatic amino acids and MWCO of the membranes. They appeared to be better rejected by ESPA4
 293 membrane, with OD_{264.4} rejections between 96.8% and 100%. OD_{216.4} rejections were always below OD_{264.4}
 294 and TN rejections, showing that non-aromatic amino acids partially transfer through NF membranes,
 295 probably due to smaller MW.

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298 **Fig. 3.** COD and sugars rejection versus permeate flux (20°C, feed flow rate = 300 L.h⁻¹):

299 (a) COD and COD_{diff} (b) glucose and fructose.

300 **Table 5**

301 Main amino acids and aromatic amino acids in cauliflowers and their properties

Main Amino acids	Concentration in cauliflower (USDA) (g / 100 g)	Solubility in water at 25°C (g / 100 g)	Molecular weight (g.mol ⁻¹)	Isoelectric point	Net charge at pH = 4.7
Glutamic acid	0.245	0.9	147.1	3.22	Negative
Aspartic acid	0.216	0.5	133.1	2.77	Negative
Leucine	0.107	2.4	131.2	5.98	Positive
Lysine	0.099	0.6	146.2	9.74	Positive
Alanine	0.097	16.7	89.1	6.01	Positive
Serine	0.096	25	105.1	5.68	Positive
Valine	0.092	8.8	117.1	5.96	Positive
Proline	0.079	162.5	115.1	6.30	Positive
Aromatic amino acids (16% w/w of total amino acids in cauliflower)					
Phenylalanine	0.066	2.79	165.2	5.48	positive

Tyrosine	0.04	0.05	181.2	5.66	positive
Histidine	0.037	4.35	155.1	7.59	Positive

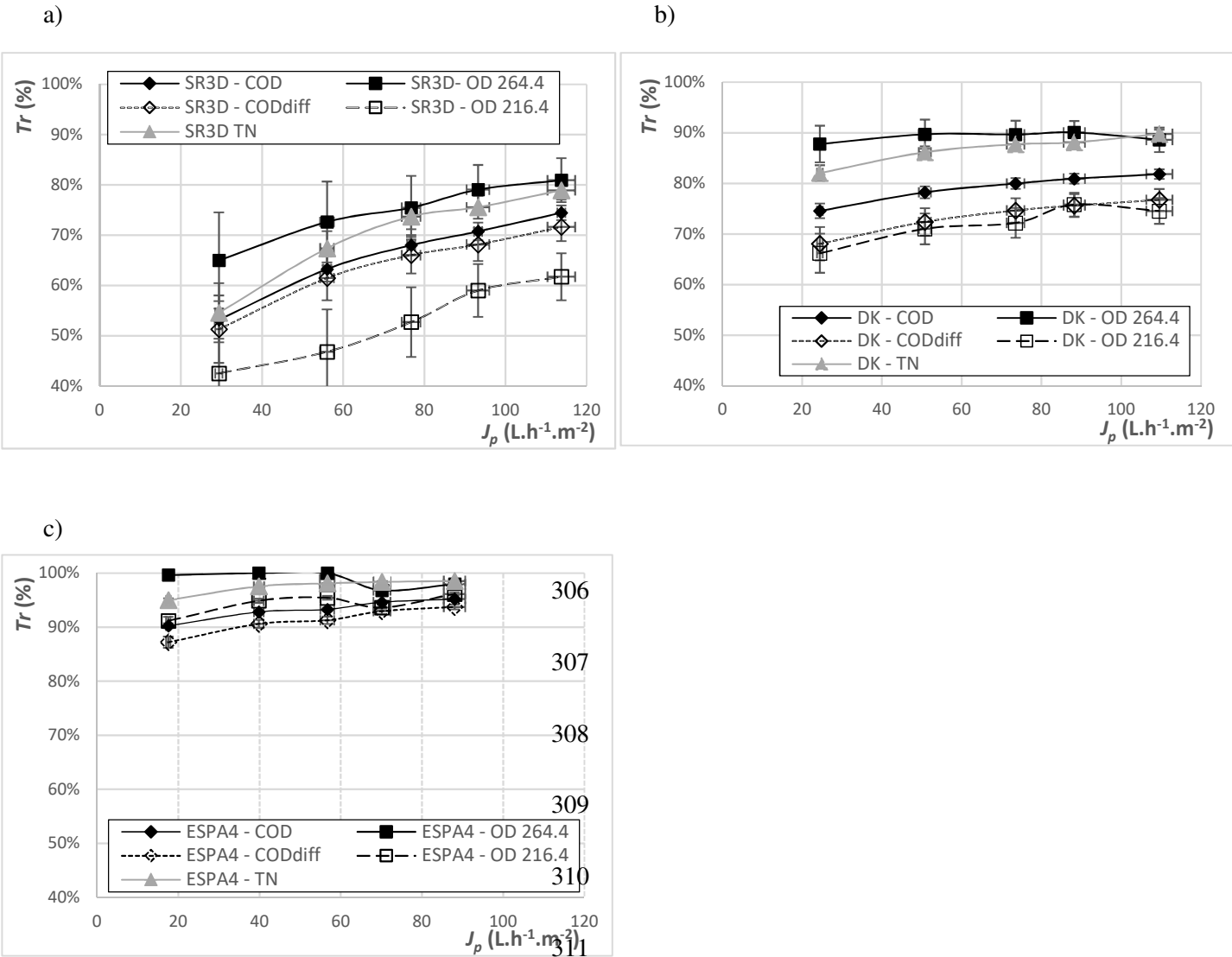


Fig. 4. COD, COD_{diff}, TN, OD_{216.4} and OD_{264.4} rejection versus permeate flux

(20°C, feed flow rate = 300 L.h⁻¹): (a) SR3D membrane (b) DK membrane (c) ESPA4 membrane.

4.3.3. Minerals rejection

Rejections of sulphate and magnesium for NF270 and DK membranes were consistent with manufacturer data (Table 6). Differences can be due to operating concentrations and pressures, or to model solutions (and not complex effluents) used by manufacturers. Again, different behaviour of SR3D was observed with sulphate rejection (69.7% instead of 99%). For all minerals (Table 7), this membrane generally gave lower rejections than other NF membranes (NF270 and DK).

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Table 6

Magnesium (Mg^{2+}) and sulphate (SO_4^{2-}) rejections with NF membranes à 4.6 bar

		NF270	DK	SR3D
This study	Mg^{2+} rejections	96.0 %	96.0 %	64.0 %
	SO_4^{2-} rejections	99.2 %	99.1 %	69.7 %
Manufacturer's data		97% at 4.8 bar 2 000 ppm MgSO_4	98% at 7.6 bar 2 000 ppm MgSO_4	> 99% at 6.5 bar 5 000 ppm MgSO_4

Ionic mass balances were established for the retentate and the permeate for DK and ESPA4 (Table 7), both at 19 bar. As in Garnier et al. (2020), for both membranes the sum of the negative charges was far lower than the positive ones, especially in the retentate and with DK. This difference can be explained by the presence of negatively charged molecules at the pH of the pre-treated effluent (pH 4.7), such as amino acids (Table 5) or organic acids (lactic acid, pKa 3.86) that were detected but not quantified in the effluent. Consequently, cations appeared globally more retained than anions in the case of the DK membrane, which can be an artifact of this proportion of negative ions not quantified in the retentate.

For DK membrane, the main identified compounds that transferred through the membranes were HCO_3^- , Cl^- and K^+ . For ESPA4, the only RO membrane, it was Cl^- and K^+ (but below 5%). ESPA4 exhibited the best rejections ($\geq 95\%$) due to its more selective polyamide layer, much thicker than that of NF membranes (Freger, 2003).

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350 **Table 7**351 **Ionic mass balance and rejection for DK and ESPA4 membranes at 19 bar**352 (Note: with the Labstak pilot Cr is the same for both membranes)

Minerals	C_r (mmol.L ⁻¹)	DK		ESPA4	
		C_p (mmol.L ⁻¹)	Tr (%)	C_p (mmol.L ⁻¹)	Tr (%)
HCO ₃ ⁻	2.27	1.35	40.3	0.00	100.0
Cl ⁻	1.97	0.94	52.0	0.07	96.7
NO ₃ ⁻	0.15	0.09	41.7	0.02	89.3
H ₂ PO ₄ ⁻	0.49	0.02	96.9	0.00	100.0
SO ₄ ²⁻	0.71	0.00	100	0.00	99.8
Sum of negative charges	6.29 meq.L⁻¹	2.43 meq.L⁻¹	61.4	0.08 meq.L⁻¹	98.7
Na ⁺	0.90	0.31	65.7	0.01	99.0
NH ₄ ⁺	0.37	0.21	44.7	0.02	94.4
K ⁺	11.11	4.56	58.9	0.25	97.8
Mg ²⁺	0.82	0.01	98.5	0.00	99.8
Ca ²⁺	1.10	0.03	97.6	0.00	99.7
H ⁺	Negligible	Negligible	-	Negligible	-
Sum of positive charges	16.21 meq.L⁻¹	5.15 meq.L⁻¹	68.2	0.29 meq.L⁻¹	98.2
<u>Negative charges missing</u>	9.91 meq.L ⁻¹	2.72 meq.L ⁻¹		0.21 meq.L ⁻¹	

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355 Rejections of the main **monovalent** (Fig. 5) and **divalent ions** (Fig. 6) are presented separately. Due to their
356 low concentration in the raw wastewater (Table 2), ammonium, sodium and nitrate rejections are not
357 presented. At the pH of the effluent (4.7) and based on its equilibrium diagram, **phosphate was mainly in**
358 **H₂PO₄⁻ form.**

359 **Whichever membrane was used**, rejections of monovalent ions (Na⁺, K⁺, Cl⁻, NO₃⁻, HCO₃⁻) were generally
360 between 20 and 60%, much lower than that of divalent ions (Ca²⁺, Mg²⁺, SO₄²⁻), above 70%. This **is**
361 consistent with the Donnan space charge model (Aimar, 2006), **based on electrostatic repulsions and**
362 **considering the ions' valence.** Moreover, for two ions with the same charge but different radii, the one
363 having the highest charge density would exhibit the highest rejection (Epsztein et al., 2018). This may
364 explain the highest rejection of Cl⁻ as compared to NO₃⁻, or that of Na⁺ as compared to NH₄⁺, due to their
365 respective ionic radii (Lide, 2004; Shannon, 1976). Far more H₂PO₄⁻ is rejected due to its higher molecular
366 weight (MW = 98 g.mol⁻¹).

ESPA4 led to the best rejections, at about 100% for divalent ions and above 95% for monovalent ones provided pressure was above 10 bar (or J_p above 40 L.h⁻¹.m⁻²).

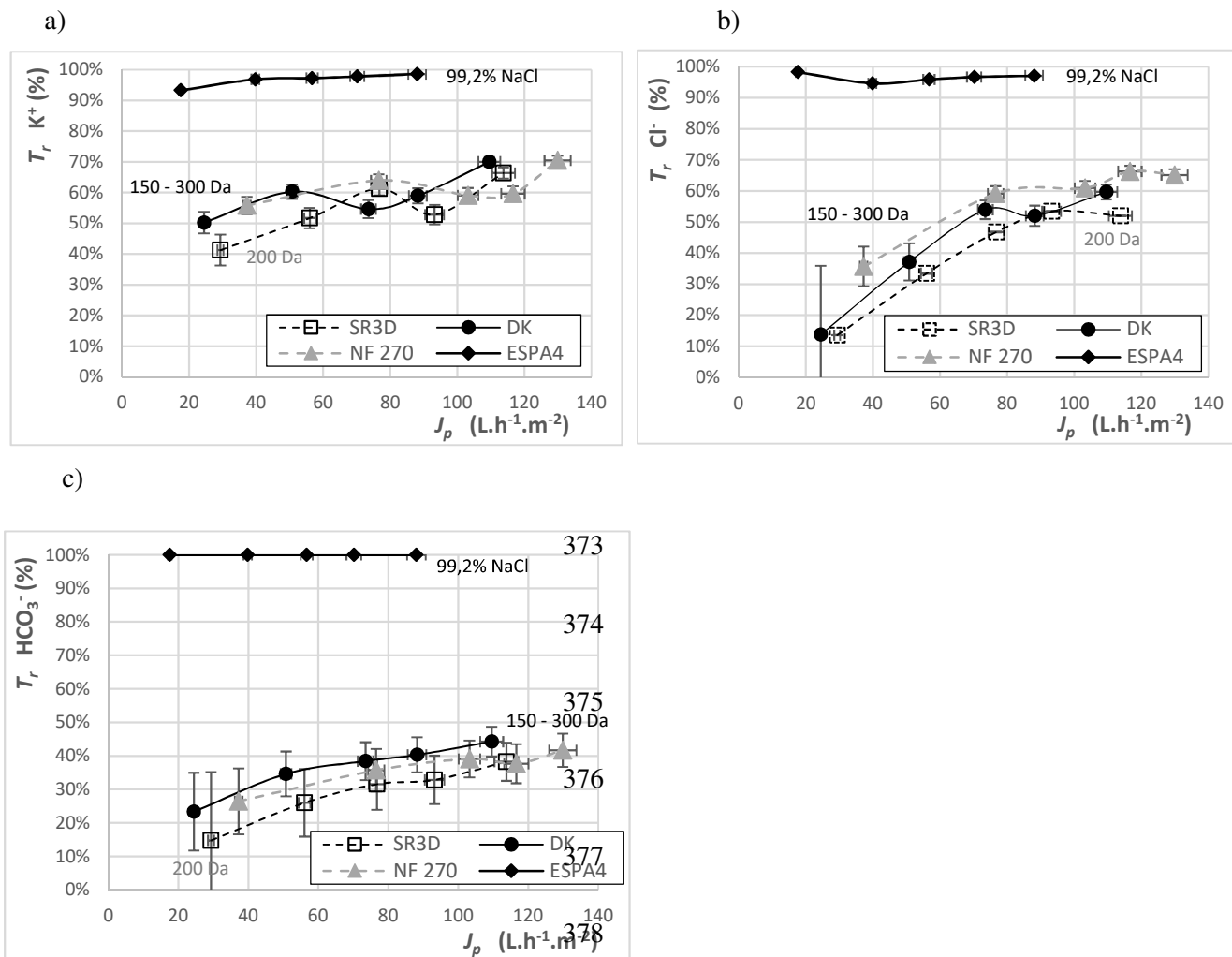


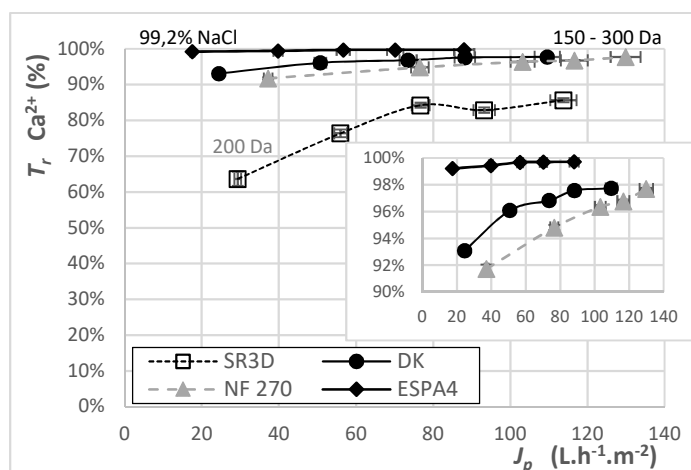
Fig. 5. Monovalent ions rejection versus permeate flux (20°C, feed flow rate = 300 L.h⁻¹):

(a) K⁺ (b) Cl⁻ (c) HCO₃⁻.

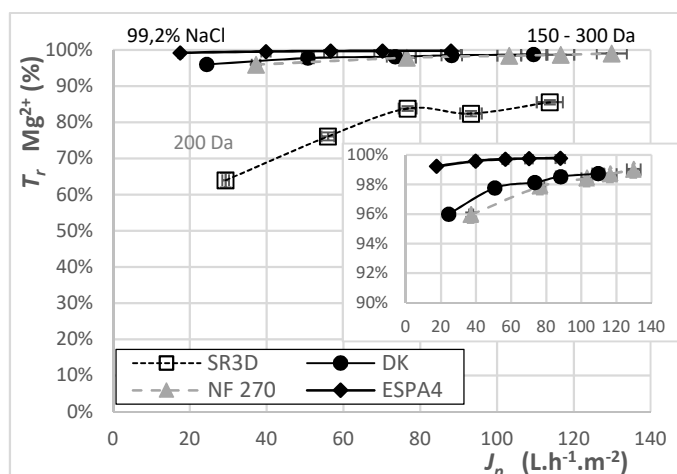
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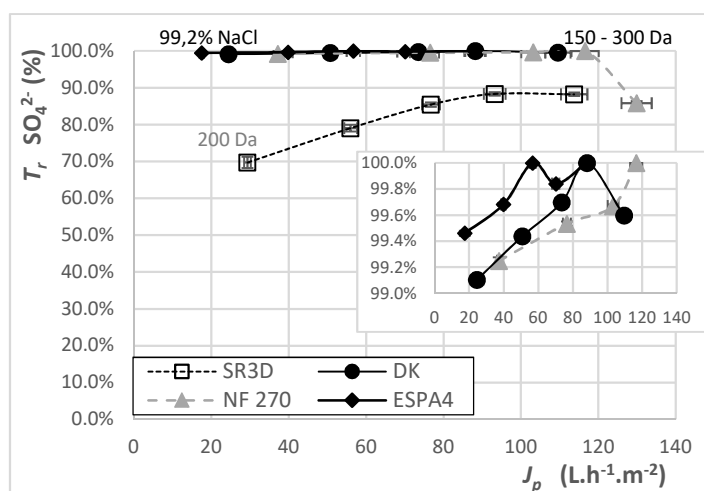
b)



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c)



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Fig. 6. Divalent ions rejection versus permeate flux (20°C, feed flow rate = 300 L.h⁻¹):

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(a) Ca²⁺ (b) Mg²⁺ (c) SO₄²⁻.

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4.3.4 Choice of NF or RO membranes for the reconditioning treatment

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Comparable results were observed with NF270 and DK membranes and lower performance (lower rejections) with SR3D membrane. NF270 at $TMP = 15$ bar and DK at $TMP = 19$ bar (pressure at critical flux) appeared as the best compromises for COD rejection and permeate flux. With RO membrane (ESPA4), the rejections were higher and critical flux corresponded to $TMP = 24$ bar (Table 4). To obtain the best compromise between COD rejection and permeate flux and to ensure a residual COD in the permeate below 400 mg O₂.L⁻¹, ESPA4 membrane was selected at about 19 bar. The permeate quality indicators are summarized in Table 8. For an equivalent permeate flux, the ESPA4 treatment of complex carrot peeling effluent at about 15 bar had allowed a better permeate quality (Garnier et al., 2020). This can be explained by

the much lower organic load of the carrots processing wastewater (Table 9), similar rejections leading to lower concentrations in the permeate. An additional explanation may be a higher fermentation of sugar due to longer storage in the case of cauliflower processing wastewater, leading to an increase in small metabolites content such as acetic or lactic acids, which can permeate through the membrane.

Table 8

Permeate quality for selected membranes and optimized conditions

	NF270 (NF)	DK (NF)	ESPA4 (RO)
Optimum <i>TMP</i> (bar)	15	19	19
J_p (L.h ⁻¹ .m ⁻²)	103	88	70
Total COD (mg O ₂ .L ⁻¹)	733	797	225
TN (mgN.L ⁻¹)	15	17	2
Conductivity (μS.cm ⁻¹)	512	527	83
pH	5.1	4.9	3.8
Carbonate Hardness (°f)	14.2	13.5	< 2
Fructose (mg.L ⁻¹)	18	31	5
Glucose (mg.L ⁻¹)	3	5	1
Sucrose (mg.L ⁻¹)	< 1	< 1	< 1
Cl ⁻ (mg.L ⁻¹)	26	33	2
NO ₃ ⁻ (mg.L ⁻¹)	7	6	1
PO ₄ ³⁻ (mg.L ⁻¹)	< 1	1	< 1
SO ₄ ²⁻ (mg.L ⁻¹)	< 1	< 1	< 1
Na ⁺ (mg.L ⁻¹)	7	7	< 1
NH ₄ ⁺ (mg.L ⁻¹)	3	4	< 1
K ⁺ (mg.L ⁻¹)	177	178	10
Mg ²⁺ (mg.L ⁻¹)	< 1	< 1	< 1
Ca ²⁺ (mg.L ⁻¹)	2	1	< 1
OD _{216.4}	0.445	0.404	0.106
OD _{264.4}	0.031	0.040	0.012
Color	L* = 100.1 a* = 0.0 b* = 0.1 (colorless)	L* = 100.0 a* = 0.0 b* = 0.1 (colorless)	L* = 100.1 a* = 0.0 b* = 0.0 (colorless)

422 **Table 9**

423 Rejection efficiency of RO treatment with ESPA4

	Carrot / $TMP = 15$ bar (from Garnier et al., 2020)			Cauliflower / $TMP = 19$ bar		
	Retentate	Permeate	Tr_i (%)	Retentate	Permeate	Tr_i (%)
COD (mg O ₂ .L ⁻¹)	620	12	98.0	4 179	225	94.6
Sucrose (mg.L ⁻¹)	305	2	99.4	2	< 0.5	> 99.5
Glucose (mg.L ⁻¹)	61	0.5	99.2	114	1	99.2
Fructose (mg.L ⁻¹)	67	0.6	99.2	889	5	99.4

424 4.4. Sugars' transfer modelling

425 For glucose and fructose with the SR3D membrane, $\ln\left(\frac{C_{p,i} \times J_p}{C_{r,i} - C_{p,i}}\right)$ vs J_p plot was not linear (eq. 9),
 426 demonstrating that the Solution-Diffusion model was not applicable and confirming the singularity of this
 427 membrane. On the contrary, for the DK, NF270 and ESPA4 membranes, high R^2 values (0.91 to 0.99) were
 428 obtained. k_i and B_i at 293.15 K and 300 L.h⁻¹ feed flowrate obtained for sugars are summarized in Table 10
 429 and compared with those extracted from results obtained in similar operating conditions with carrot
 430 processing wastewater (Garnier et al., 2020). For both effluents, $B_{i \text{ glucose}}$ was similar to $B_{i \text{ fructose}}$, at about 0.45
 431 x10⁻⁶ m.s⁻¹ for DK and 0.3 x 10⁻⁶ m.s⁻¹ for NF270. As observed in Almazan (2015), concentration of sugars
 432 did not affect B_i . As expected, for dense RO membrane (ESPA4), $B_{i \text{ Glu/Fru}}$ was much lower than for NF
 433 membranes, at $B_{i \text{ Glu/Fru}} = 0.1 \times 10^{-6}$ m.s⁻¹ for cauliflower wastewater, twice that for carrot ($B_{i \text{ Glu/Fru}} = 0.05 \times$
 434 10^{-6} m.s⁻¹). However, it may be underlined that for this membrane, rejection was quite constant with J_p , lying
 435 between 99.0 and 99.5 %, which made inaccurate k_i and B_i determination. Other studies on glucose rejection
 436 with DK membrane allowed $B_{i \text{ glucose}}$ parameter to be extracted (Table 11). They lie between 0.25 and 0.95 x
 437 10⁻⁶ m.s⁻¹, with an average at 0.55 x 10⁻⁶ m.s⁻¹, consistent with the average value of 0.45 x 10⁻⁶ m.s⁻¹ in this
 438 work, despite the diverse compositions of the studied solutions.

439 For NF membranes, k_i values increased with retentate concentrations. It was quite the opposite for RO
 440 membrane: respectively for fructose and glucose, 23 x 10⁻⁶ and 18 x 10⁻⁶ m.s⁻¹ in cauliflower effluent with
 441 higher concentrations compared to 42 x 10⁻⁶ and 40 x 10⁻⁶ m.s⁻¹ in carrot processing effluent (with lower
 442 concentrations).

443 For all the membranes investigated and cauliflower or carrot wastewaters, B_i was far lower than k_i ($40 < k_i/B_i$
 444 < 460) and especially for ESPA4 (k_i/B_i between 360 and 460), showing that the resistance to transfer was
 445 logically mainly due to diffusion inside the membrane, increasingly with RO membranes due to their higher
 446 density. Moreover, $C_{r \text{ m,i}} / C_r$ ratios for glucose and fructose increased with TMP respectively from 1.2 to 2.9
 447 (2.3 at 19 bar) and from 1.3 to 4.0 (3.0 at 19 bar) confirming the polarisation concentration.

448

449 **Table 10**

450 k_i and B_i for fructose and glucose from the simplified Solution-Diffusion model (eq. 9) for cauliflower ([this](#)
 451 [study](#)) and carrot wastewater (from results in Garnier [et al.](#), 2020).

Solute		Fructose		Glucose	
Vegetable of raw wastewater		Cauliflower	Carrot	Cauliflower	Carrot
Concentration range (mg.L ⁻¹)		830 – 926	63 – 69	112 – 124	59 – 63
pH		4.8	7.5	4.8	7.5
DK	k_i (m.s ⁻¹ x 10 ⁻⁶)	33	19	30	18
	B_i (m.s ⁻¹ x 10 ⁻⁶)	0.42	0.45	0.52	0.42
	k_i/B_i	78	42	58	43
NF270	k_i (m.s ⁻¹ x 10 ⁻⁶)	46	12	40	12
	B_i (m.s ⁻¹ x 10 ⁻⁶)	0.33	0.23	0.41	0.22
	k_i/B_i	139	52	98	55
ESPA4	k_i (m.s ⁻¹ x 10 ⁻⁶)	23	42	18	40
	B_i (m.s ⁻¹ x 10 ⁻⁶)	0.05	0.10	0.05	0.09
	k_i/B_i	460	420	360	444

452

453 **Table 11**

454 B_i for glucose deduced from several publications obtained with the simplified Solution-Diffusion model
 455 taking [concentration polarization](#) into account (eq. 9)

Reference		Nguyen, N. et al., 2016	Almazán et al., 2015	Lyu et al., 2016		Mohammad et al., 2010	Wang et al., 2018	Zhou et al., 2013a, b
$C_{glucose}$ (g.L ⁻¹)		10	5-100	20	0.5	3-12	4-20	4-20
DK	B_i (m.s ⁻¹ x 10 ⁻⁶)	0.27	0.95	0.54	0.25	0.37	0.91	0.59

456

457 5. Conclusion

458 A complex cauliflower processing wastewater resulting from blanching was treated using membrane
 459 processes, in order to produce water of a quality high enough to be reused inside the factory. The adopted
 460 pre-treatment consisted in a double sieving step at 169 μm and 79 μm followed by a 100 000 g.mol^{-1} MWCO
 461 ultrafiltration. Its removal efficiency reached 99% for turbidity, 50% for COD and 40% for conductivity
 462 especially. At industrial scale, this pre-treatment could be replaced by a single submerged hollow fibre
 463 ultrafiltration (Nelson et al., 2007). RO treatment with ESPA4 membrane was then necessary to reach the
 464 best permeate quality. It was optimized at 19 bar, leading to a residual COD value in the permeate of 225
 465 $\text{mg O}_2.\text{L}^{-1}$ due to the transfer of small non-aromatic compounds. Solution-Diffusion model and film model
 466 theory were applicable to describe glucose and fructose transfer, for DK, NF270 and ESPA4 membranes.
 467 Permeability coefficient B_i obtained for glucose and fructose was similar ($0.45 \times 10^{-6} \text{ m.s}^{-1}$) and consistent
 468 with values calculated from other studies (0.25 to $0.95 \times 10^{-6} \text{ m.s}^{-1}$) regardless of the concentration of glucose
 469 in the feed solution and its composition.

470 These results, if industrially confirmed, open the possibility of water recycling of cauliflower blanching
 471 wastewater. However, it would be necessary to investigate long-term accumulation of the residual solutes in
 472 the recycled effluent. A Life Cycle Assessment on the plant under study confirmed that this wastewater
 473 recycling through UF plus RO treatment was beneficial. It offers a way to limit the reliance on water
 474 resource and to face water restrictions that in certain regions lead to stop or delay food plants production.

475

476 Nomenclature and units

477	A_w	membrane permeability to pure water ($\text{m.s}^{-1}.\text{Pa}^{-1}$ or $\text{L.h}^{-1}.\text{m}^{-2}.\text{bar}^{-1}$)
478	B_i	membrane permeability to solute i (m.s^{-1})
479	$C_{p,i}, C_{r,i}, C_{r m,i}$	concentration of solute i in the permeate, the retentate and at the membrane interface in the
480		retentate, respectively (mol.m^{-3})
481	CH	Carbonate Hardness ($^{\circ}\text{f}$)
482	COD	Carbon Oxygen Demand ($\text{mg O}_2.\text{L}^{-1}$)
483	COD_{diff}	differential COD = difference between COD and $\text{COD}_{\text{sugars}}$
484	$\text{COD}_{\text{sugars}}$	COD deduced from sugar concentrations ($\text{mg O}_2.\text{L}^{-1}$)
485	$\Delta\pi$	osmotic pressure gradient between the membrane interface in the retentate and the permeate
486		(Pa or bar)
487	J_i	flux of solute i through the membrane ($\text{mol. s}^{-1}.\text{m}^{-2}$)
488	J_p	permeate flux (m.s^{-1} , usually expressed in $\text{L.h}^{-1}.\text{m}^{-2}$)

489	k_i	mass transfer coefficient of solute i in the polarization layer (m.s^{-1})
490	OD	Optical Density (-)
491	P_f, P_r, P_p	pressure in the feed, the retentate and the permeate, respectively (Pa or bar)
492	$\pi_p, \pi_{r m}$	osmotic pressure in the permeate and at the membrane interface in the retentate, respectively
493		(Pa or bar)
494	Q_p	permeate flow rate ($\text{m}^3.\text{s}^{-1}$ or L.h^{-1})
495	S	effective membrane area (m^2)
496	TMP	TransMembrane Pressure (Pa or bar)
497	TN	Total Nitrogen (mg N.L^{-1})
498	Tr_i	rejection rate of solute i (-)
499	TSS	Total Suspended Solids (mg.L^{-1})
500	V_i, V_f	initial and final volume of solution in the feed tank for a discontinuous filtration run (m^3)
501	VRR	Volume Reduction Ratio (-)

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