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- 1 Unraveling the role of feed temperature and cross-flow velocity on organic fouling in
- 2 membrane distillation using response surface methodology
- 3
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14 Highlights

- Feed temperature governs the fouling behaviour more than cross-flow velocity
- Higher feed temperature increases the fouling accumulation
- Higher cross-flow velocity decreases the fouling accumulation
- Feed temperature and cross-flow velocity were statistically significant for the RSM
- RSM is a powerful tool to assess performance and fouling behaviour in MD

20 Abstract

21 Understanding the role of operating condition on fouling development in membrane distillation (MD) is critical for the further optimization of MD technology. In this study, organic fouling 22 23 development in MD was investigated with a synthetic model solution of humic acid varying the feed inlet temperature from 35 to 65 °C and the cross-flow velocity from 0.21 to 0.42 m/s. For 24 each experiment, the final fouling layer thickness was estimated using optical coherence 25 tomography, a non-invasive imaging technique. The set of experiments was mined to model the 26 initial flux decline, the final flux, and the final foulant thickness responses by central composite 27 design, a useful response surface methodology (RSM) tool. A strong influence on the initial flux 28 29 was observed by varying feed inlet temperature. The results indicated a linear increment of the fouling thickness by increasing the feed inlet temperatures. Overall, the feed inlet temperature 30 governed both the initial flux decline and the fouling deposition rate. A more complex behaviour 31 32 was observed by varying the cross-flow velocity. To this extent, higher cross-flow velocities showed a positive effect on the initial flux, which however translated in larger values of the 33 initial flux decline rate. On the other hand, the higher shear stress contributed to a decrease of the 34 final fouling layer thickness. The proposed approach was proven to be a valuable tool to assess 35 the role of the operating conditions on fouling and process performance in MD. 36

37

38 Keywords: Direct contact membrane distillation (DCMD); Membrane fouling; Optical
39 coherence tomography (OCT); Response surface methodology (RSM);

40 1 Introduction

Membrane distillation is a thermal-based desalination technology which has gained an 41 exponential interest during the last decades [1, 2]. Among all the possible membrane distillation 42 configurations, direct contact membrane distillation (DCMD) is the most compact. Due to its 43 simplicity, this process has been extensively studied at laboratory scale to approach scale-up 44 45 applications in MD [3-5]. In fact, DCMD does not require an external condenser and it is more 46 suitable for water-based applications than air gap, vacuum, or sweep gas membrane distillation. In DCMD, the hot feed and the cold permeate solutions are in contact with a hydrophobic 47 membrane. Under ideal working conditions, only water vapor passage is allowed through this 48 49 microporous membrane [6, 7]. However, several operational challenges might cause decrease in productivity or even process failure [8]. According to the type of treated feed solution, three 50 51 main drawbacks observed in the operational DCMD phase are: (i) pore wetting, (ii) mineral 52 scaling, and (iii) membrane fouling. Wetting mainly occurs when membrane hydrophobicity is reduced, together with the liquid entry pressure, to the point which allows liquid passage through 53 the pores [9, 10]. Wetting is easily induced by amphiphilic molecules, such as surfactants, and it 54 leads to process failure even in a preliminary recovery stage [11, 12]. Mineral scaling is due to 55 56 crystal formation of salts at the solid membrane interface, initiating a rapid and severe flux 57 decline which can also translate into pore wetting and membrane damage [13]. Membrane fouling leads to flux reduction over time due by accumulation of feed contaminants on the 58 membrane surface [14]. 59

With the increasing interest in MD, the number of possible applications has been also expanded.
As a thermal-based process, MD has been largely used for desalination to produce high-quality
water while concentrating the feed above typical reverse osmosis limits [15]. Recently, DCMD

has been also employed for the treatment of challenging wastewater, such as produced water, 63 textile, and pharmaceutical wastewater [16-18]. Within this range of possible applications, recent 64 studies demonstrated how effective pre-treatment strategies and process optimization could 65 highly reduce pore-wetting and mineral scaling propensity [16, 19]. In this context, membrane 66 fouling is still considered one of the main bottlenecks of MD operations [20, 21]. Among 67 68 different foulant species, humic substances showed particularly high fouling propensity in lowpressure processes due to high adhesion capacity of these compounds on the membranes [22]. 69 Humic acids are also the major constituents of natural organic matter, as well as widely present 70 71 constituents in surface water, groundwater, and seawater [23]. In this study, organic fouling in DCMD was investigated by using humic acid as model compound under accelerated fouling 72 conditions. 73

74 Optical coherence tomography (OCT) has been recently demonstrated as an effective and 75 versatile tool for fouling characterization. This non-destructive technique enables monitoring the filtration system under continuous operation, providing real-time information of the fouling layer 76 [24, 25]. OCT allows acquiring non-invasively 2D cross-sectional and 3D volumetric images 77 with micron-level resolution without interfering with the membrane operation. Recently, the use 78 79 of OCT has been employed for studying the fouling behavior in MD when treating textile, 80 pharmaceutical wastewater, and concentrated brines [17, 18, 26]. In these studies, OCT results were efficiently linked to process performance data allowing an in-depth understanding on how 81 fouling and scaling impact the water flux during the DCMD process. However, these literature 82 83 studies were often limited to narrow ranges of operative conditions in MD. To extend the 84 understanding of fouling under a wider range of temperatures and cross-flow velocities, response 85 surface modeling (RSM) was implemented in this study through Design Expert software [27-29].

86 One of the way to implement RSM is by using central composite design (CCD), an array whereby investigated parameters are efficiently distributed to allow a second-order generalized 87 regression between the experimental results [30, 31]. Recently, RSM combined with CCD has 88 gained a growing interest in membrane process optimization as it is able to consider several 89 variables at the same time with easy evaluation of the generated responses. An interesting 90 application of RSM in MD was developed by Shokrollahi et al., who effectively modeled flux 91 and thermal efficiency for a wide range of interacting parameters [32]. There, numerical 92 modeling with CCD method for flux optimization showed that temperature and module length 93 94 have the most important influence on MD productivity.

95 In this study, central composite design is implemented to guide the design of MD experiments conducted with different combinations of feed inlet temperature and cross-flow velocity. The 96 97 experiments are performed with a synthetic model solution of humic acid, where the distillate flux is monitored as a function feed volume concentration factor. Additionally, OCT in-situ 98 monitoring is employed to characterize the fouling layer developed at the end of each MD test. 99 The flux performance and fouling data are discussed and critically examined also to assess a 100 valuable experimental based modeling. Therefore, (i) the initial flux decline rate, (ii) the final 101 flux, (iii) the total flux decline, and (iv) the final fouling thickness are applied as responses 102 103 (dependent variables) in the RSM analysis to investigate the mechanism of fouling and to identify the most suitable DCMD operating conditions. The investigation assesses the role of 104 105 process parameters and governing factors on fouling in MD, and it proposes the rational 106 deployment of RSM as a tool to move toward scale-up applications.

107 2 Materials and Methods

108

109 2.1 Membrane and Feed composition

Accelerated fouling conditions were employed in this study using a synthetic feed solution with 110 an initial humic acid (HA) concentration of 500 mg/L in deionized (DI) water. To enhance the 111 fouling deposition, 20 mM of calcium chloride, CaCl₂, was also added to the feed solution [33]. 112 HA and CaCl₂ were purchased from Sigma-Aldrich. The organic compound was received in 113 powder form. The stock solution was prepared by dissolving the chemicals in 600 mL of DI 114 115 water. The stock was then added, prior to flux stabilization, to the remaining 400 mL of 116 deionized water used as initial feed. Initial volumes of 1 L were thus used for both the feed and 117 permeate solutions, the latter consisting of DI water.

A commercially available hydrophobic polytetrafluoroethylene with a polypropylene support (PP-PTFE) membrane (Membrane Solutions corp., US) was used for all the experiments. The membrane characteristics, provided by the manufacturer or obtained in the lab, are listed in Table 1. The membrane permeability coefficient was calculated by dividing the experimental water flux by the vapor pressure difference across the membrane (see calculated angular coefficient from Fig S.1).

124 **Table 1**. Porous PP-PTFE membrane characteristics

Data source	Parameter	Units	Value
	Thickness	μm	174 - 245
Provided by the	Mean pore size	μm	0.22
	Bubble point	psi	16.0-20.3
From experiments	Membrane permeability coefficient	kg m ⁻² h ⁻¹ bar ⁻¹	143.8

126 2.2 MD lab setups

127 All the MD tests were performed in direct contact membrane distillation (DCMD) configuration with a lab-scale batch system. In this process, the feed and permeate streams were circulated 128 129 counter-currently on their respective sides of the hydrophobic membrane, not enabling liquid 130 passage through the pores. Thanks to the applied thermal gradient, the colder liquid is enriched 131 by the water vapor extracted from the feed stream during the process. Ranges of feed 132 temperature of 35 - 65 °C and feed cross-flow velocity of 0.21 - 0.42 m/s (deriving from a crossflow rate ranging from 25 to 50 L/h) were investigated in this study. To clearly assess the role of 133 134 feed parameters on fouling deposition, a constant temperature and cross-flow velocity of 20 °C and 0.1 m/s were maintained in the permeate side. For the same reason, no feed spacer was used. 135 The temperatures in the feed and permeate inlet of the flow cell were maintained constant 136 throughout the experiment using a thermostatic water bath and a chiller (Corio-CD, Julabo, 137 Germany). The heat exchangers were accurately controlled by the temperature sensors integrated 138 in the conductivity meters (TetraCon 325, Xylem Analytics, Germany) located just before the 139 inlet of the flow cell. On the permeate side, purified water with electrical conductivity below 20 140 141 μ S/cm was used, whereas the initial feed conductivity was 4.2 \pm 0.2 mS/cm. For each experiment, the permeate conductivity was continuously monitored to ensure no liquid passage 142 during the tests, i.e., no pore wetting. Cross-flow velocity and outlet-temperature were measured 143 by digital cross-flow meters located in proximity of the flow cell outlet. The flux across the 144 membrane was calculated by recording the change in weight of the permeate tank in time 145 through a computer-interfaced balance. All the instruments were digitally connected and 146 controlled by Lab View software. The DCMD flow cell in polymethyl methacrylate was 147

148 customized to allow *in-situ* characterization with OCT. The flow cell was had dimensions of 10.0 149 $\times 3.3 \times 0.1$ cm (length \times width \times height) for a total active membrane area of 33 cm².

150 2.3 Design of experiments and statistical analysis

Design Expert software was used to setup and analyze the response surface methodology (RSM) 151 for DCMD experiments. Central composite design (CCD) was applied to define the number of 152 runs needed for the optimization of the variables and responses. Feed inlet temperature and 153 cross-flow velocity were selected as operating factors, while the initial flux decline rate, the final 154 flux, and the final thickness of the fouling layer were selected as responses after a preliminary 155 phase investigation of experimental results. The Supplementary Material appendix presents 156 further details of the applied CCD method and analyses. The selected ranges of investigation for 157 158 the various factors are reported in Table 2, together with the coded experimental values extrapolated by Design Expert software. The CCD method generated a suggestion for nine total 159 160 runs, each with a specific combination of values of T_f and CFV. This procedure allowed 161 weighted probing of the entire multidimensional space. The experimental results were used as 162 input data to generate the model for each response according to the best fit. ANOVA was used 163 for the statistical analysis of the results to evaluate the quality of the model.

Table 2. Experimental design of the selected of operating conditions, representing the range of
 experimental variables used in the RSM model

Factors	Unit	Minimum	Maximum	Coded low	Coded high	Mean
Temperature	(C°)	35.0	65.0	40.0	60.6	50.0
Cross-flow velocity	(m/s)	0.21	0.42	0.24	0.39	0.31

166 2.4 Filtration experiments protocol

Fouling experiments consisted of two phases: (i) a stable flux phase and (ii) a fouling phase. The 167 flux was first stabilized using DI water only as feed, without organic foulants (J_0) . This stage 168 allowed achievement of the hydrodynamic equilibrium. The fouling phase then started at time 169 zero, when the appropriate amount of organic foulant stock solution was added into the feed 170 171 tank. This second phase was run until a volume concentration factor of 2.5 was reached, which was always associated with sufficient operational time to obtain a near stable flux and fouling 172 layer thickness. The increment of CaCl₂ concentration during experiments can be considered 173 174 negligible for any possible effect in the reduction of the feed vapor tension value. For this reason, the flux decrement observed during the fouling tests can be predominantly attributed to foulant 175 deposition. 176

177 2.5 Optical coherence tomography (OCT) analysis

A spectral-domain optical coherence tomography (SD-OCT) system Ganymede II from 178 Thorlabs, GmbH (Germany) was used to assess the fouling deposition on the membrane surface 179 under accelerated fouling conditions. The OCT was equipped with a scan lens (LSM 03BB). The 180 OCT probe was positioned on top of middle point of the DCMD module to characterize the 181 182 fouling layer thickness at the end of each experiment. 3D cross-sectional OCT scans (666 pixel × 666 pixel \times 1022 pixel) corresponded to 4.0 mm \times 4.0 mm \times 2.25 mm (width \times length \times depth). 183 The OCT scans were processed with the FiJi software. Images were filtered to reduce the noise, 184 then the contrast and brightness were adjusted. 3D scans were then visualized by AVIZO (Field 185 186 Electron and Ion Company, Hillsboro, OR, USA) software and modified for visualization purpose. The fouling layer thickness was calculated using a customized MATLAB code. 187

188 **3** Results and discussion

189

190 3.1 Beyond the RSM: An overall picture of the process parameters in MD organic fouling

191 **3.1.1** The Effect of temperature and cross-flow velocity on the experimental flux

This section describes the experimental results while providing an in-depth understanding of the selected responses (dependent variables) of the RSM model. In this study, Design Expert was employed as a statistical tool to create the experiment plan aimed at studying the impact of operating condition on the fouling behavior in DCMD. The list of experiments is reported in Table S.1 and in the legend of Fig 1, where experimental results of water production in DCMD at different operating conditions are also shown.

In DCMD process, the initial flux, J_0 is related to the feed temperature and cross-flow velocity 198 [34, 35]. As expected, in this study the inlet feed temperature was found to govern J₀. By 199 increasing T_f from 35 to 65 °C, the J₀ increased from 3 to 22.5 kg m⁻²h⁻¹, while increasing the 200 cross-flow velocity from 0.21 to 0.42 m/s at fixed T_f 50 °C led to an increase of only 2 kg 201 $m^{-2}h^{-1}$. This result can be attributed to the nature of the driving force, namely, the vapor tension 202 difference between the feed and the permeate, which can be easily determined through Antoine 203 204 equation [36]. On the other hand, the cross-flow velocity can contribute to the flux increment by reducing the temperature polarization effects [37]. 205



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Figure 1. Results of fouling experiments performed with the synthetic feed water in the presence of 500 mg/L humic acid and 20 mM of calcium chloride at different initial permeate flux, J_0 , obtained by changing the applied feed temperature and cross-flow velocity in DCMD. Water fluxes (J_w) were investigated until a volume concentration factor (VCF) of 2.5 was reached, at which a value of near-stable final flux was observed for all the experiments. Dash lines represent the best linear fit of the first 1.25 volume concentration factor (VCF) of the initial flux decline.

In all the tests, the water flux decreased almost linearly in the initial phase, to then reach an approximate flux stabilization over time when the nominal driving force was counterbalanced by resistances due to fouling accumulation to yield a constant effective driving force [38]. As fouling deposition is proportional to the water transport across the membrane, high accumulation typically occurs in the initial phases of operation, contributing to the formation of a cake layer during this initial stage [39, 40]. The initial flux decline rates were estimated from the best linear 221 fit (see dash lines in Fig 1a) of the water flux measured between 1 and 1.25 VCF. The values are reported as a function of the initial flux J_0 (Fig 2a) and of the inlet feed temperature (Fig 2b). The 222 results suggest a smooth and gradually incrementing correlation of the initial flux decline when 223 224 increasing J_0 . The proportional effect of J_0 on the initial flux decline has been also widely investigated in osmotically and pressure-driven membrane processes [41-43]. As expected, an 225 analogous behavior was observed when looking at the data as a function of the inlet feed 226 temperature (Fig 2b), as the driving force is closely related to this parameter. The data also allow 227 assessment of the role of the cross-flow velocity, whose increment seems to slightly affect the 228 229 initial flux decline, as an indirect effect of slightly larger values of J₀ observed when increasing CFV. The values of near stable flux at the end of the tests, $J_{\rm w},$ and the ratio $J_{\rm w}$ / J_0 were also 230 extrapolated from the flux decline data for each tested condition. 231





Figure 2. Plot of the initial flux decline rate as a function of (a) the initial permeate flux J_{0} , and (b) the inlet feed temperature (T_f). The arrows indicate the increment of the cross-flow velocity (CFV). Data were extrapolated from dash lines rates reported in Fig 1.

A few interesting observations may be made by analyzing the data presented in Figs 1, 2. While 236 utilizing an inlet feed temperature set at 65 °C produced an initial flux (22.5 kg m⁻²h⁻¹) that is 237 nine times higher the flux observed with a temperature equal to 35 °C (2.5 kg m⁻²h⁻¹), the flux at 238 the end of the tests (J_w) was only 3 times larger, reaching roughly 7.5 kg m⁻²h⁻¹ for the former 239 condition, whereas no significant decline in flux was observed at the lower feed temperature. 240 These results give reasons for operating at low-medium feed temperatures, namely, at or below 241 50 °C for water streams with high fouling potential and if membrane cleanings are not frequently 242 operated. In such cases, the long-term productivity may be similar within a wide range of bulk 243 feed temperature and working at lower temperature would result in savings in terms of energy 244 demand. This may in turn translate, e.g., into cheaper solar fields with smaller footprints if the 245

energy is harvested from the sun, or anyway into a higher energy efficiency and gain outputration (GOR) value for the overall process.

248 **3.1.2** OCT results for the fouling layer thickness

Water flux data were linked to non-invasive direct fouling characterization performed with OCT 249 to characterize the fouling layer developed on the membrane surface by scanning the central 250 251 positions of the cell. Please note that the homogeneity of the fouling layer along the membrane length was confirmed by preliminarily evaluating the thickness growth at different positions of 252 the cell. As no spatial gradient was observed, the middle position was selected as a representative 253 location. The 3D OCT rendering images (Fig 3) show the fouling deposition obtained at T_f of 35, 254 50, 65 °C, thus covering the whole range of T_f investigated. The results highlight the increase of 255 256 the foulant deposition by incrementing the inlet feed temperature. In these three examples, the same cross-flow velocity of 0.31 m/s was applied, representing the central point suggested by 257 Design Expert within the explored CFV range (see Table S.1). In general, a slight increment of 258 259 the foulant roughness was observed by increasing T_f, as nodule-like and valley-like structures 260 became more pronounced. This phenomenon was also discussed by Laqbaqbi et al. when testing 261 DCMD fouling at a temperature close to 70 °C [44].



Figure 3. 3D OCT rendered scans (4mm \times 4mm) of the final steady-state foulant layer thickness from the experiments conducted at cross-flow velocity of 0.31 m/s and feed inlet temperature of 35 °C, 50 °C, 65 °C in (a), (b), (c), respectively. The frame color of the OCT images corresponds to the color of the associated data points reported in (d). Here, the final thickness is plotted against the feed inlet temperature for all the tests. The number indicated close to each data point represents the cross-flow velocity associated with the respective test and expressed in m/s.

270 For all the experiments, fouling layer thickness measured with the OCT after 2.5 of volume concentration factor (VCF) is reported in Fig. 3d. In general, a near-linear increment of the 271 thickness was observed as a function of T_f, for the entire investigated range of temperatures. 272 Thus, the thickest deposition was observed for the experiment performed at 65 °C, achieving a 273 layer thickness of almost 1200 µm at the end of the test. A much smaller layer of roughly 200 274 µm was observed with T_f of 35 °C. This last result is in agreement with previous DCMD studies 275 reporting negligible organic fouling with a feed temperature below 40 °C [45]. As opposed to the 276 effect of T_f, higher CFV values were beneficial for reducing the fouling layer development in 277 278 MD. The arrows in Fig 3d indicate data points associated with different CFV values. As reported in the literature, the increment of the shear stress thwarted foulant accumulation by lowering the 279 boundary layer thickness [46, 47]. 280

The results presented imply a strong cause-consequence relation between the operating 281 parameters in MD and fouling development, but without considering how fouling deposition can 282 affect the overall driving force, i.e., the thermal balance during the process. Although the 283 reciprocal influence between the driving force and fouling deposition has been widely 284 investigated for pressure-driven and osmotically-driven processes [43, 48, 49], further research 285 286 efforts are required to evaluate the interaction between governing factors and fouling in MD. In 287 summary, the *in-situ* observation performed in this study confirmed the link between feed temperature and fouling propensity. The fouling thickness was found to (i) increase with feed 288 inlet temperatures T_f, while (ii) slightly decreasing with cross-flow velocity. The thickness of the 289 290 fouling layer may thus also be used as a robust response parameter for the RSM analysis discussed below. 291

3.2 Modeling of organic fouling in DCMD through response surface methodology (RSM)

294

295 **3.2.1** Significance of operating parameters

296 Organic fouling in DCMD was investigated under different feed inlet temperatures and crossflow velocities by performing nine DCMD filtration experiments, with combinations of 297 operating parameters suggested by the central composite design approach. Based on the results 298 described above, four parameters were selected as potentially valuable responses for the response 299 surface analysis: (i) initial flux decline rate, (ii) J_w/J_0 value at the end of the test indicating the 300 relative loss of productivity due to fouling, (iii) near-stable flux, (iv) final foulant layer thickness. 301 Experimental results for these parameters were used as input data (responses) to generate the 302 relative model function. According to Design Expert, all these responses were statistically 303 significant to both T_f and CFV, i.e., low p-value. Table 3 summarizes the p-values obtained from 304 ANOVA. Specifically, T_f was found to be considerably more significant than CFV. Each 305 response was fitted by a different model function. The initial flux decline was described by a 306 307 quadratic model while all the other responses were adequately described by a linear model, as can be seen by the absence of cross-correlation terms in Table 3. Within the three linear 308 responses, CFV was not highly significant, as the p-value was > 0.1. However, CFV was 309 included in the model to respect the hierarchy of the statistical method and to improve the fit 310 [50]. For each response, the final equation calculated by Design Expert and relating operating 311 parameters with fouling outcomes (responses) is reported in the Supplementary Material 312 appendix (see Table S.2.). 313

Table 3 Summary of response significance values estimated by ANOVA statistical analysis.

Source	Initial flux decline	Final J _w /J ₀	Final flux	Final thickness
	p-value	p-value	p-value	p-value

Model	0.0001	0.0004	< 0.0001	0.0133
A-Cross-flow velocity	0.0835	0.4156	0.2145	0.2191
B-Feed temperature	< 0.0001	0.0001	< 0.0001	0.0059
AB	0.0812			
A ²	0.1153			
B ²	0.0017			

316 To provide a detailed statistical analysis, the diagnostic plots of the initial flux decline, the final flux, and the final J_w/J_0 values are shown in Fig S.2, S.3 and S.4, respectively. Figure 4 displays 317 the diagnostic plots for the final foulant layer thickness. Please note that the following discussion 318 319 relates directly to layer thickness, but the conclusions and implications are also valid for the other selected responses. Figure 4a reports the normal probability plot of residuals (error terms), 320 a graphical tool for comparing a data set with the normal distribution: if the data can be 321 adequately described with a normal distribution, characterized by a mean and a variance, then a 322 plot of the theoretical percentiles of the normal distribution versus the observed sample 323 324 percentiles should be approximately linear. In Fig 4a, the red linear line represents the theoretical normal distribution while the ten dots represent the observed samples (10 runs, Table S1). For all 325 the responses, normal probability plot of the residuals fell on a straight line, which implies that 326 327 error terms had a normal distribution [51, 52].

Figure 4b shows that all points are scattered around the 0 y-axis (variance or standard deviation) reflecting equal or similar variances of collected data. In fact, nine points out of ten lie within two standard deviations, meaning that 95% of values are included in this range (empirical rule). In this case, the variance of residuals can be considered as a constant (homoscedasticity). Homoscedasticity is an important assumption of parametric statistical tests. In Fig. 4c, residuals vs. data points do not follow a specific pattern, which suggests that responses are not dependent on the order of runs. Lastly, Fig. 4d illustrates that predicted values vs. experimental values lay 335 on a straight 45 degree line, an indication of high-quality modeling outcome. In conclusion, the 336 diagnostic plots of all the responses indicate the robustness of the statistical analysis, which 337 enable to assess the impact of the operating parameters on the organic fouling behavior in MD.



Figure 4. Diagnostic plots for the foulant layer thickness response: (a) normal % probability vs.
residuals; (b) residuals vs. predicted; (c) residuals vs. run order; (d) predicted vs. actual.

341

342 3.2.2 Single responses evaluation

Fig 5 shows the outcome of the RSM model in terms of effects of operating parameters, i.e., 343 temperature and cross-flow velocity, on the fouling behavior, namely, initial flux decline rate, 344 final J_w/J_0 , the near-stable flux, and the final layer thickness. This discussion aims at providing 345 an effective view of fouling behavior in the whole range of investigated conditions of T_f and 346 CFV and to facilitate any direct comparison among the selected fouling parameters. In 347 accordance with the description in the section above, all the responses were mainly governed by 348 the feed temperature. Specifically, Fig 5b shows how contour values decrease from a $J_{\rm w}$ / J_0 of 349 0.7 to below 0.3 when the T_f increases from 40 to 65 °C, thus only roughly 1 kg m⁻²h⁻¹ of stable 350 flux is gained for each 5 °C-step in ΔT_f (see Fig 5c). As illustrated in Fig 5c, a net increment of 351 the fouling deposition can be observed by increasing T_f, as a twofold increase of layer thickness 352 is associated to an increase of the temperature from 40 to 65 °C. 353



354

Figure 5. 2D surface response plots as a function of the feed inlet temperature and cross-flow velocity for the (a) initial flux decline rate, (b) final to initial flux ration, (c) near-stable flux, and (d) final foulant layer thickness. The near-stable flux is the flux at volume concentration factor of 2.5 for the experiments reported in Fig 1. The magnitude for each response increases from blue to red and is also indicated by numeric values for each contour line.

361 In contrast to feed inlet temperature, different behaviors were observed by varying the cross-flow velocity. The initial flux decline rate was more affected by the CFV at higher T_f values (see Fig 362 5a). This result can be explained by the proportionality between T_f and temperature polarization 363 (TP) [52]. Faster CFVs thwart TP and this effect is more pronounced when TP tends to be of 364 greater magnitude, that is, at higher values of T_f. This translates into larger J₀ (see Fig 1) and the 365 ensuing steeper flux decline, as observed for CFV values above 0.3 m/s [53, 54]. On the other 366 hand, increasing CFV yielded a positive outcome in terms of productivity when considering the 367 magnitude of the near-stable flux (see Fig 5b, c), likely due to lower fouling deposition 368 369 associated with smaller boundary layers, as observed in Fig 5c. In fact, the OCT scans highlighted a reduction in fouling layer thickness by increasing the CFV. Interestingly, the CFV 370 was more impactful in decreasing the foulant thickness rather than increasing the overall flux 371 decline J_w / J₀, implying that foulant thickness and flux loss are not directly correlated but that a 372 complex mechanism is in play. This result might be rationalized with the fact that these two 373 parameters are not independent, for example, J_w may not simply intensify the likelihood of 374 foulant deposition but simultaneously cause enhanced compactness of the resulting layer [55]. 375 Both thickness and compactness of the foulant layer play a role in mass and heat transport, and 376 377 thicker but more porous layer may be less detrimental than thin dense layers that would produce a larger variation in diffusion coefficients and thermal conductivity with respect to the bulk 378 solution. 379

In conclusion, the RSM analysis well depicted the link between fouling propensity and feed temperature, as fouling behavior worsened with the increase of the feed inlet temperature, thereby negatively affecting the flux decline, J_w/J_0 , while a positive but gradually more marginal enhancement of productivity (near-stable flux) was observed when increasing the feed inlet temperature. Moreover, it was found that the increase in cross-flow velocity led to a slight decrease of the fouling thickness deposited on the membrane, while keeping an overall benefit in terms of productivity.

387 Conclusions

Organic fouling in MD process was investigated using humic acid and calcium chloride in 388 the feed solution. The goal of this study was to assess the role of the feed temperature and 389 cross-flow velocity on fouling behaviour in DCMD. The different operating conditions of the 390 experiments were selected through Design Expert software aiming to build the model 391 function of the selected responses. The four process performance parameters selected as 392 responses (dependent variables) for the RSM were: (i) the initial flux decline rate, (ii) the 393 near-stable flux measured at the end of the tests, (iii) the stable to initial flux ration, J_w / J_0 , 394 and (iv) the final fouling layer thickness. 395

Higher influence of feed inlet temperature than cross-flow velocity on loss of productivity was 396 397 observed experimentally and then confirmed by robust statistical analysis, due to the major role of flux in the development of organic fouling in DCMD. In detail, a sharp increment in 398 399 the overall flux decline, J_w/J_0 occurred at higher feed inlet temperatures, making the case for 400 the need to select an appropriately transmembrane temperature difference that guarantees feasible fluxes but also minimizes loss of driving force and energy demand. The benefits in 401 402 water productivity obtained by increasing the feed temperature were always offset by higher 403 fouling deposition.

Another interesting trade-off between more rapid initial flux decline and thinner layer thickness was observed by increasing cross-flow velocity above 0.3 m/s. Layer thickness is only one of many aspects of the foulant layer that relates to productivity loss, others may include density, pore structure and thermal conductivity, which can directly influence mass and heat transport through this unmixed layer. Optical coherence tomography (OCT) was used in this study to assess layer thickness, but further efforts are needed to deepen investigations

on foulant deposition and on how layer characteristics relate to deposition mode and then flux loss. Overall, working at relatively high cross-flow velocity may be beneficial at high values of the nominal driving force, i.e., transmembrane temperature difference, while the results suggest that the effect of channel feed flow velocity may not play a significant role when the flux is below a certain level, approximately 10 kg m⁻²h⁻¹.

Finally, the proposed approach is not limited to this application but was proven to be a valuable tool to assess the role of the process parameters and governing factors on fouling and process performance in membrane distillation (MD). The results of this study highlight the effectiveness of combining flux data, OCT characterization, and response surface methodology (RSM) to advance the understanding of fouling in MD and open future perspective related to this crucial topic to making MD feasible at commercial scale.

421	Supplementary material
422	Assessing the effect of feed temperature and cross-flow velocity on organic
423	fouling in membrane distillation using response surface methodology
424	
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Figure S.1. Linear correlation between the obtained water vapor flux as a function of the
applied vapor tension difference for the PP-PTFE membrane. The permeability, *b*, is
reported.

Table S.1. List of the experiments suggested by Design Expert software for different
combinations of cross-flow velocity and inlet feed temperature. Related experimental results
for each response are also listed from the third column. C.P indicates the central point, a
repetition of the 4th run, required by the software to retrieve a better model fitting.

Run	CFV	T _f	Initial flux decline	Final flux	Final thickness	J _w /J ₀
-	m/s	°C	Kg m ⁻² h ⁻¹ /[-]	Kg m ⁻² h ⁻¹	μm	-
1	0.42	50.0	24.7	5.8	888	0.48
2	0.39	39.4	3.0	3.6	375	0.79
3	0.31	35.0	1.1	2.4	273	0.80
4	0.31	50.0	16.9	5.4	940	0.52
5	0.39	60.6	59.7	7.4	633	0.39
6	0.24	60.6	51.0	7.6	1066	0.42
7	0.31	65.0	67.2	7.6	1183	0.34
8	0.24	39.4	6.5	3.2	510	0.64
9	0.21	50.0	16.3	5.2	1000	0.47
C.P	0.31	50.0	17.6	5.1	920	0.57



447 Figure S.2. Diagnostic plots for initial flux decline rate as response: (a) normal %
448 probability vs. residuals; (b) residuals vs. predicted; (c) residuals vs. run order; (d) predicted
449 vs. actual.



Figure S.3. Diagnostic plots for near-stable flux at the end of the test as response: (a) normal
% probability vs. residuals; (b) residuals vs. predicted; (c) residuals vs. run order; (d)
predicted vs. actual.



456

457 **Figure S.4.** Diagnostic plots for J_w/J_0 as response: (a) normal % probability vs. residuals; (b) 458 residuals vs. predicted; (c) residuals vs. run order; (d) predicted vs. actual.

460 Tab S.2. Final equations computed by the statistical analysis and relating operating

461 parameters to fouling behaviour. The equation can be used to make predictions about each462 response.

		Initial Flux decline	Final Flux	Final Thickness	J _w /J ₀
		+206.19420	+5.37	+780.80	+1.21292
A-Cross flow velocity	*	-452.03539	+0.1409	-90.80	+0.243943
B-Feed temperature	*	-7.19720	+1.95	+262.62	-0.015014
AB	*	+3.90112			

A ²	*	+453.12607		
B ²	*	+0.082623		

464 Appendix

The model is generated for the four responses and is based on experimental data collected in the lab fitting a linear model for the final flux, final thickness, and final J_w/J_0 responses and a fitting a quadratic model for the initial flux decline rate response. The most general equation is reported here below:

469
$$y = \beta_0 + \sum k_i = 1 \beta_i x_i + \sum k_i = 1 \beta_{ii} x_i^2 + \sum k_i = 1 \sum k_j = i + 1 \beta_{ij} x_i x_j + \varepsilon$$

where y is the predicted response, x represents the factors, k is the number of factors, β_0 is the 471 constant coefficient, and β_i , β_{ii} , and β_{ij} are the regression coefficients of linear, quadratic, and 472 interaction terms, respectively. To select the amount of experimental data to be collected, 473 Central Composite Design (CCD) was applied. This design defines 2k corner points, 2k axial 474 points, where k is the number of independent variables (or factors) selected, and a central 475 point. In this study, two factors (Feed inlet temperature and feed cross-flow velocity) were 476 selected. The number of experiments was directly calculated by the software according to the 477 equation $n = 2^k \cdot 2k + Cp$, resulting in a total of 10 experiments, one of them represented by 478 the central point (Cp). These test are a combination of different factors levels defined by the 479 coded values calculated by applying the formulas in Table S2. The coded value associated 480 with a is representative of the rotatability of the model which suggested Practical alpha due to 481 k < 6 and equal to 1,41, which represents the distance. 482

483

484

(Eq S.2)

Coded value	Un-coded value
-α	X _{min}
-1	$\frac{(\alpha-1)X_{max}+(\alpha+1)X_{min}}{2\alpha}$
0	$\frac{X_{max} + X_{min}}{2}$
+1	$\frac{(\alpha-1)X_{min}+(\alpha+1)X_{max}}{2\alpha}$
$+\alpha$	X _{max}

Table S2. Coded and un-coded values for CCD

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