Investigations on the impact of membrane cleaning on the structure of a fouling layer from biorefining

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Abstract

Membrane cleaning became crucial to remove foulants and recover membrane performance. Therefore, the aim of this work is to investigate the influence of the parameters temperature, duration and concentration of an alkaline cleaning agent on the cleaning success. This study was conducted on polysulfone membranes fouled with thermomechanical pulping process water by ultrafiltration. Lastly, the membrane surface and the fouling layer were analysed with contact angle measurements, Fourier transform infrared spectroscopy and Brunauer-Emmett-Teller analysis.

From the Design of Experiment evaluation, it was revealed that concentration of the cleaning agent is the most relevant cleaning parameter and that the interaction between concentration and temperature displays a considerable influence in the cleaning efficiency as well. Membrane analysis indicated that the main foulants attached to the membrane surface are polysaccharides, in the form of hemicelluloses, and they seem to be eliminated with some success by cleaning with an alkaline solution. It was also detected an extensive development of pore blocking on the fouled membranes.

Keywords: lignocellulosic biorefineries, thermomechanical pulping, ultrafiltration, membrane fouling, membrane cleaning, polysaccharides

1. Introducion

1.1. Pulp and Paper Mills

The manufacturing of paper consists of a two-step operation: first fibrous raw material is separated into a pulp and then the pulp is transformed to produce paper. Pulp can be produced by chemical or mechanical processes **[1]**. This work focusses on thermomechanical pulping, where pulp is shortly preheated and then it is conducted to steam pressurized refiners **[2]**.

1.2. Lignocellulosic Biorefineries

About 95% of wood biomass is used to produce paper in

thermomechanical pulp mills, while the remaining 5% are discharged and wasted **[3]**. Consequently, pulp and paper mills have started to consider other options to increase profitability and competitiveness and the intention of converting existing pulp and paper mills into integrated forest biorefineries (IFBR) emerged.

1.3. Lignocellulosic Biomass

Extractives, cellulose, hemicelluloses and lignin are the main components of lignocellulosic biomass. Cellulose and hemicelluloses are both polysaccharides that provide support in the plant cell wall. Galactoglucomannan (GGM) represents the major hemicellulose in softwood, and it has a great potential as a raw material for bio-based products **[4]**.

In a thermomechanical pulp and paper mill, cellulose is used in the production of pulp and paper, while hemicelluloses are underutilized, instead of being converted into value-added products, including hydrogels, oxygen barrier films in food packaging and emulsion stabilizers. Lignin has a great potential regarding biofuels production due to its heat value **[5]**.

1.4. Membrane Separation Processes

Membrane filtration is a promising method to put the concept of an IFBR into action, since it provides an efficient separation and fractionation **[5]**. Membrane technologies demand low energy when compared to conventional separation technologies, such as centrifugation, drying and evaporation **[6]**.

In pressure driven membrane processes, the transmembrane pressure (TMP) is the driving force. These processes can be classified into four types, according to working pressure and membrane pore size **[12]**: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Each process has different pressure requirements due to the difference in membrane pore size **[7]**. UF technology has a wide range of applications due to its outstanding selectivity. It allows to efficiently recover and purify valuable compounds from pulp and paper mills waste streams, such as hemicelluloses and lignin **[5]**

1.5. Membrane Fouling

Membrane fouling is defined as an agglomeration of undesirable matter (foulants) on the membrane surface and into the membrane pores.

Membrane fouling is what prevents the adoption of membrane technology in a large scale in biorefineries **[5]**. Fouling leads to a decrease in membrane lifetime and reduction in filtration capacity. Furthermore, it leads to higher operation costs due to membrane replacements **[8]**.

Membrane fouling can be classified as removable, irremovable and irreversible. In case of removable fouling, loosely foulants are attached on the membrane surface, which can be removed by physical cleaning by e.g. backflushing. Irremovable fouling can only be removed by chemical cleaning. Irreversible fouling is not possible to be removed by any kind of cleaning **[9]**.

It has been reported that wood extractives from pulp and paper mill waste streams like fatty acids and resin cause membrane fouling **[10]**. It has also been shown that polysaccharides are one of the main foulants in pulp mill process streams **[9]**. Fouling mechanisms can be classified into three types: adsorption, pore blocking and cake layer formation (**Figure 1**). Pore blocking occurs when the size of the solute molecules is similar to the size of the membrane pores **[6]**. Cake layer formation takes place if the solute molecules are larger than the membrane pores. Adsorption occurs if the solute molecules are smaller than the membrane pores instead**[11]**.

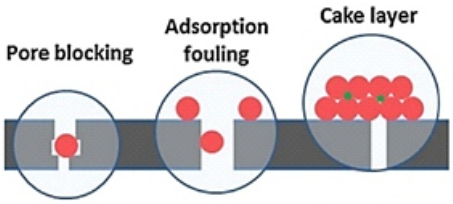

Figure 1 - Illustration of the three types of membrane fouling. Adapted from [19].

1.6. Membrane Cleaning

Due to the negative effects of membrane fouling on the filtration process over time, regularly membrane cleaning becomes a necessary procedure **[9]**. It is imperative to implement regular cleanings into membrane plants to avoid irreversible fouling **[13]**.

Temperature and duration along with concentration and

type of cleaning agent represent the most important cleaning variables **[15]**.

Drawbacks of chemical cleaning are originating pollution **[11]** and damaging membrane materials, if the cleaning conditions are too harsh **[8]**.

Cleaning agents used for chemical membrane cleaning are mainly acids, alkalis, detergents **[5],** or enzymes.

An adequate cleaning agent should demonstrate a good performance regarding dissolving and retaining fouling particles in dispersion, without damaging the membrane and the system.

1.7. Membrane Analysis

In this work, contact angle measurements, Fourier Transform Infrared Spectroscopy (FTIR) and Brunauer-Emmett-Teller (BET) analysis have been applied to further characterise membrane fouling caused by thermomechanical pulping process water **[16]**.

1.8. Design of Experiment

Design of Experiment (DoE) provides an organized approach to study the interaction of multiple factors on which an experiment depends on, representing a vital tool to optimize processes in scientific field **[17]**.

With DoE it is possible to collect only relevant data performing the minimum experimental runs, reducing time and costs **[8]**, by establishing a correlation between factors (independent variables) and responses (dependent variables) with mathematical models **[18]**.

DoE has been used to study and optimize membrane cleaning efficiency of UF membranes. It is an efficient method to investigate several parameters at the same time and identify the main factors and their interactions **[13,14]**.

1.9. Objectives

The main purpose of this thesis is to contribute to the progress of the state of the art with a better understanding about the impact of the parameters temperature, duration and concentration of a commonly used cleaning agent on the cleaning performance of membranes fouled with process water from thermomechanical pulping. This work aims to provide an optimized cleaning procedure plus a comprehensive understanding on the structure of a fouling layer caused by thermomechanical pulping process water.

2. Materials and Methods

Diluted retentate after MF of process water from thermomechanical pulping (Stora Enso, Sweden) was used as a feed solution to foul the membranes. Suspended solids were removed with a drum filter directly at the pulp mill.

The feed solution together with the permeate and the retentate obtain from UF were analysed based on procedures stated in **[21]**. The results of the analysis are given in **Tables 1 and 2**.

Table 1 - Composition of the feed solution (diluted retentate after MF of process water from thermomechanical pulping) and the permeate and retentate after UF of the feed solution.

Solution	Turbidity (NTU)	Dry Solids (mg/g)	Ash (mg/g)	Total Lignin (g/L)	Acidic Lignin (g/L)	Klason Lignin (mg/g)
Feed	409.3	2.52	0.00	0.34	0.02	0.33
Permeate	1.0	0.10	0.04	0.02	0.01	0.07
Retentate	319.7	5.12	0.05	0.37	0.02	0.39

Table 2 - Concentration of monosugars of the feed solution (diluted retentate after MF of process water from thermomechanical pulping) and the permeate and retentate after UF of the feed solution.

Solution	Mannose (g/L)	Galactose (g/L)	Glucose (mg/g)	Xylose (g/L)	Arabinose (g/L)
Feed	0.29	0.14	0.16	0.00	0.02
Permeate	0.1	0.00	0.01	0.00	0.00
Retentate	0.69	0.32	0.35	0.00	0.08

The membranes used in this study were commercial PSU UF membranes of the type UFX5-pHt with a nominal molecular weight cut-off of 5000 g/mole. The membranes are permanently hydrophilized.

The membranes used in the first 11 experiments (**Table 4**) were flat-sheet membranes from one batch. The ones used in the last 8 experiments (**Table 5**) were originated from a spiral-wound module which was opened and cut into several flat sheets. All the experiments were performed in a cross-flow module with three membrane samples in parallel (**Figure 2**). The effective membrane area of each sample was 1960 mm^2.

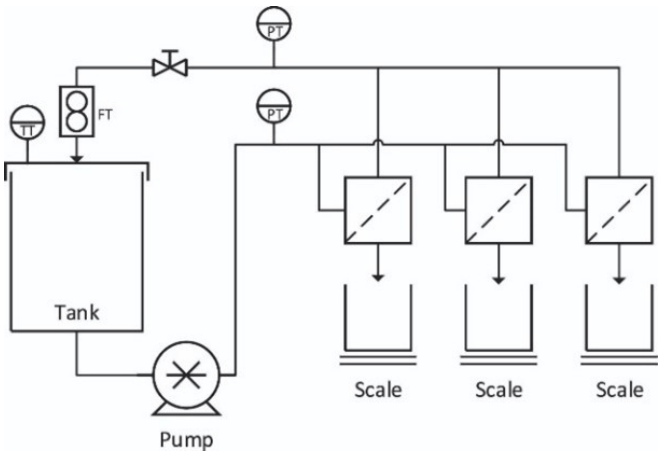

Figure 2 - Schematic of a cross-flow mode membrane filtration set-up [22]. FT = flow meter, PT = pressure transmitter, TT = temperature transmitter.

Before each fouling and cleaning experiment, the membranes were conditioned to remove storage chemicals, such as glycerine. The commonly used alkaline cleaning agent, Ultrasil 10, was used for this. Conditioning was performed with 5 L of a 1% (w/w) solution of Ultrasil 10 at 50 ˚C for 1 hour, with a TMP of 2 bar and a CFV of 0.28 m/s and recirculating both the permeate and the retentate. Then, the system was rinsed with pure water and the rinsing procedures were based on membrane filtration procedures that can be found in **[22]**. In this work, transmembrane pressure (TMP) was calculated by the following equation **[11]**:

$$TPM = \frac{p_{feed} + p_{retentate}}{2} - p_{permeate} \qquad \textbf{(1)}$$

The conditioned membranes were fouled by UF of 5 L the feed solution at 70 ˚C. The feed tank was covered with a lid, to avoid evaporation of the liquid inside the tank. UF with recirculation was conducted for 26 hours, at 2 bar and the CFV at 0.28 m/s. The detailed fouling procedures were based on membrane filtration procedures that can be found in **[22]**.

For the membranes from the flat-sheet batch, after 26 hours the recirculation was stopped and a concentration was initiated, where the permeate was collected for 1 hour. This was done to intensify the fouling further. For the membranes originated from the spiral-wound module, after the 26 hours of recirculation, no concentration was conducted as fouling was already severe enough. Finally, in order to remove all the fouling solution inside the system, the same rinsing protocol used as after membrane conditioning was followed.

Three cleaning parameters were studied in this project: temperature, concentration, and duration. The values of these parameters for each cleaning experiment are displayed on **Tables 4 and 5**. They were arranged based on the requirements showed on **Table 3**. For all cleaning experiments, the TMP was kept constant at 2 bar and the CFV at 0.28 m/s. Subsequently to the cleaning, the same rinsing protocol used as for membrane conditioning was followed.

Fouling and cleaning efficiency were assessed based on pure water flux (PWF) measurements. The PWF was determined after membrane conditioning, membrane fouling, and membrane cleaning, respectively, for the three membranes assembled on the UF equipment. For this, the permeate was collected with beakers and continuously weighed with an electronic balance. The PWF was measured at 30 ˚C, 0.28 m/s, and at four TMPs: 0.5, 1, 1.5, and 2 bar. The flux (J) was calculated based on the following equation:

$$J = \frac{m_{permeate}}{A_{membrane} \times t \times \rho} \qquad \textbf{(1)}$$

The mass of permeate ($m_{permeate}$) was recorded during each measurement, and its density (ρ) was assumed to be 1000 g/L. The time (t) of each measurement was 5 minutes and the effective membrane area of each sample ($A_{membrane}$) was 1960 mm^2.

The average of the flux data from the three membranes assembled on the UF equipment was calculated for each pressure point. A linear regression of the correlation between the average flux and the TMP (0.5, 1, 1.5, and 2 bar) was determined. The obtained slope corresponds to the permeability (P).

The fouling factor (FF) was calculated by the following equation:

$$FF = \frac{P_{conditioned\ membrane} - P_{fouled\ membrane}}{P_{conditioned\ membrane}} \qquad \textbf{(2)}$$

The cleaning success (CS) was calculated by the following equation:

$$CS = \frac{P_{cleaned\ membrane}}{P_{conditioned\ membrane}} \qquad \textbf{(3)}$$

Central composite face-centered (CCF) was the

selected design to implement DoE. In CCF, there is a cuboidal design space with points centred on each face of the cube, on each vertex and on the centre (**Figure 3A**). This design requires three levels of each factor: high level, low level and average (**Table 3**). Three cleaning parameters (factors) were investigated: temperature, concentration of the cleaning agent (Ultrasil 10), and duration. These three factors are represented by three axes (**Figure 3B**) and each level varies between -1 and 1.

The CCF method was executed with the software MODDE 13 Pro. This model has the purpose of studying and optimizing the relationship between multiple input factors (the three investigated cleaning parameters) and one output response. The input factors and their range of values were set up according to **Tables 4 and 5**. For the output response (cleaning success), a range between 0.3 and 1.3 and a target of 1 was chosen.

Two DoE studies were carried out, one for the membranes from the flat-sheet batch and another one for the membranes originated from the spiral-wound module.

Table 3 - Leverage of the three cleaning parameters for the DoE setup.

Design Variable	Low Level	Average	High Level
Temperature (˚C)	30	50	70
Concentration (%*)	0	0.25	0.5
Duration (min)	10	35	60

*% (w/w) Ultrasil 10

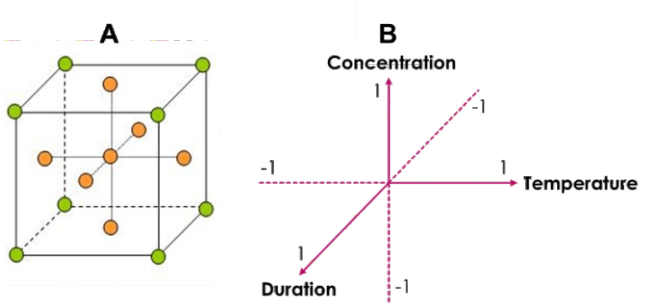

Figure 3 - Cuboidal space design with the data points (A) and the three cleaning factors, represented by three axes (B).

All membrane samples were stored and dried in an oven at 30 ˚C until they were further analysed. Investigated were the hydrophobicity of the membrane surfaces by the water contact angle measurements, the chemical composition of the membrane surfaces by attenuated FTIR, and the inner area and volume of the membrane samples by BET analysis.

Contact angle analysis were performed according to the sessile drop technique on the three membrane samples of each experiment with a drop and bubble shape tensiometer PAT-1M and the software PAT-1M. A water droplet with a volume of 5-6 mm^3 was dropped onto a membrane sample with an approximated area of 100 mm^2. The measurements were conducted at room temperature for 60 seconds. The right and the left angle of each sample were determined by the average of the last 20 data points of each measurement. Then, the contact angle per sample was calculated by the average of the right and the left angle. Finally, the contact angle per experiment was obtained by the average of the contact angles of the three membrane samples.

FTIR analysis were performed on the three membrane samples (approximated area of 100 mm^2) of each experiment to characterize the chemical composition of the sample surfaces. The measurements were executed with a spectrometer ALPHA II and with the software OPUS . The absorbance for each sample was measured within the wavenumber range from 400 to 4000 cm^{-1} and 72 scans were recorded with a resolution of 2 cm^{-1}. It was measured a background signal that was subtracted from the sample spectra. The spectra were base line corrected, and the absorbance data was then converted to transmittance for better comparison.

BET analysis were conducted with a 3Flex surface characterization analyser. Before the analysis, the membranes were cut into small pieces and degassed at 50 ˚C for 12 hours in a smart VacPrep 067 degassing unit. Adsorption and desorption isotherms of nitrogen gas were recorded at -196 ˚C (boiling point of nitrogen). The data was evaluated with the software 3Flex. The presented data was originated from the desorption branch of the obtained isotherms.

3. Results and Discussion

3.1. Ultrafiltration Solutions Analysis

Analysis performed on the feed solution and on the permeate and retentate obtained from the UF are presented in **Table 1**. The majority of lignin after UF was

detected in the retentate.

Table 2 displays the concentration of monosaccharides measured in the feed solution, UF permeate, and UF retentate. Mannose, glucose and galactose were the monosaccharides detected in the feed solution and in the retentate. This was expected, as GGM is the main hemicellulose in softwood, and it is composed of those three monomers. Most of the GGM was retained in the UF retentate, since only negligible amounts of mannose and glucose were in measured in UF permeate.

3.2. Membrane Cleaning

Tables 4 and 5 show an overview of the results from the fouling and cleaning experiments. The fouling factor and the cleaning success for each experiment were calculated according to **Equations 2 and 3**. The fouling factor was an important indicator of the fouling efficiency.

Table 4 - Experimental parameters, fouling factor and cleaning success calculated with Equations 3 and 4 of the cleaning experiments performed on the membranes from the flat-sheet batch.

Experiment	Cleaning Temperature (˚C)	Cleaning Concentration (%)	Cleaning Duration (min)	Fouling Factor (-)	Cleaning Success (-)
1	50	0.25	60	0.266	1.144
2	70	0.5	60	0.090	1.721
3	50	0.5	35	0.135	1.191
4	30	0.5	10	0.114	1.331
5	70	0	60	0.225	0.961
6	30	0.5	60	0.158	1.229
7	30	0	10	0.115	0.952
8	50	0.25	35	0.108	1.280
9	50	0	35	0.124	0.972
10	50	0.25	35	0.215	1.237
11	30	0.25	35	0.160	1.311

Table 5 - Experimental parameters, fouling factor and cleaning success calculated with Equations 3 and 4 of the cleaning experiments performed on the membranes originated from the spiral-wound module.

Experiment	Cleaning Temperature (˚C)	Cleaning Concentration (%)	Cleaning Duration (min)	Fouling Factor (-)	Cleaning Success (-)
12	70	0.5	10	0.928	0.625
13	30	0	60	0.912	0.106
14	70	0	10	0.933	0.102
15	70	0.25	35	0.920	0.364
16	70	0.5	60	0.933	0.493
17	50	0.25	35	0.903	0.190
18	30	0	10	0.916	0.064
19	30	0.5	60	0.933	0.145

3.3. Design of Experiment

The CCF model was implemented with the software MODDE 13 Pro based on the cleaning parameters (inputs) and the cleaning success (output) from **Tables 4 and 5**.

Figures 4 and 5 present the observed versus predicted values for the cleaning parameters. In both figures, the points are close to a straight line, meaning that the model is well suited.

For the membranes from the flat-sheet batch, R^2 is 0.883 and for the membranes originated from the spiral-wound module, R^2 is 0.975. The value of R^2 is higher for the membranes originated from the spiral-wound module, indicating a better fit for the selected model. In both cases, R^2 is higher than 0.8, showing that the models have a high significance.

Q^2 gives an indication future prediction precision of the model. For the membranes originated from the spiral-wound module Q^2 is higher than 0.5, indicating that the model is rather good. For the membranes from the flat-sheet batch Q^2 is negative, denoting that the model does not have a predictive relevance.

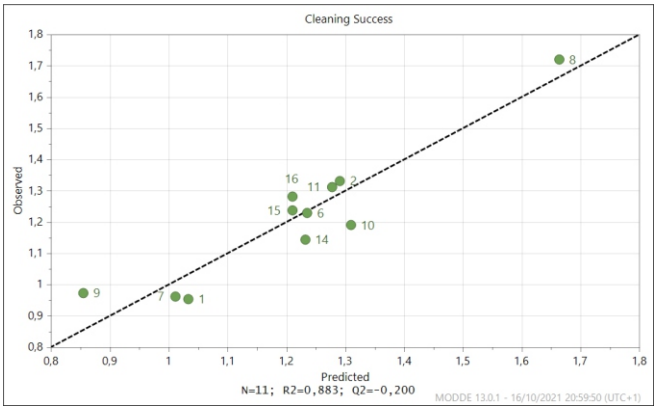

Figure 4 - Observed vs predicted plot of the membranes from the flat-sheet batch.

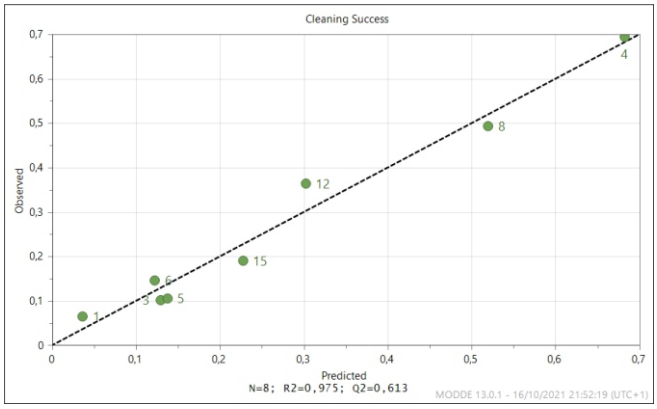

Figure 5 - Observed vs predicted plot of the membranes originated from the spiral-wound module.

Figures 6 and 7 present the coefficient plots that evaluate the significance of the model terms. Even though it is more evident for the membranes from the flat-sheet batch, the cleaning agent concentration is the

parameter that has the stronger impact on the cleaning success in both studies. The impact is positive and it means that an increase in concentration would likely result in an increase in cleaning success.

Duration accused a rather low impact on cleaning success in both studies. The negative impact of the cleaning duration for the membranes originated from the spiral-wound module is not reasonable but cannot be explained at the moment.

Coefficient plots (**Figures 6 and 7**) access the interaction between the factors as well. For the membrane from both batches, the interaction between concentration and temperature revealed a remarkable influence in the cleaning success as well.

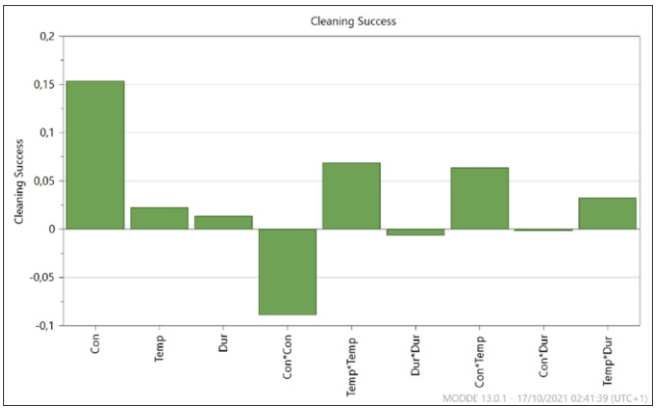

Figure 6 - Coefficient plot of the membranes from the flat sheet-batch.

Figure 7 - Coefficient plot of the membranes originated from the spiral-wound module.

Figures 8 and 9 express the design space that estimates the probability of success according to the response specifications and provides an area of operability. Inside the green area (area of operability), the probability of failure is less than 0.5%. For the membranes from the flat-sheet batch, a response within that area is attainable even for the lowest values of the three combined cleaning parameters (**Figure 8**). Using values higher than the ones illustrated by the design space would result in a waste of resources while cleaning. However, higher values of temperature and concentration are required when cleaning the membranes originated from the spiral-wound module, as seen in **Figure 9**. For those membranes, the green area is wider for the duration of 10 min, supporting the negative impact of this parameter on the cleaning success observed on **Figure 7**.

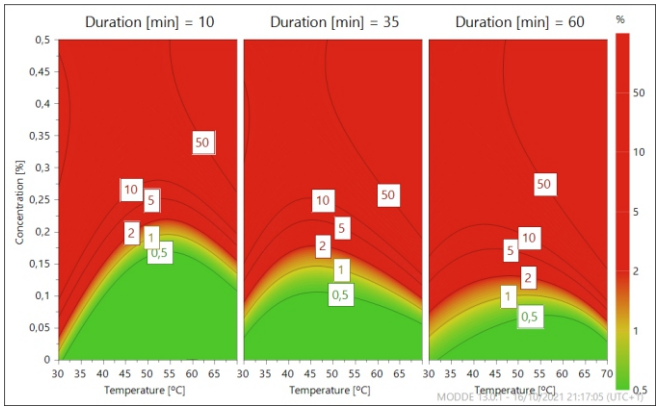

Figure 8 - Design space plot of the membranes from the flat sheet-batch.

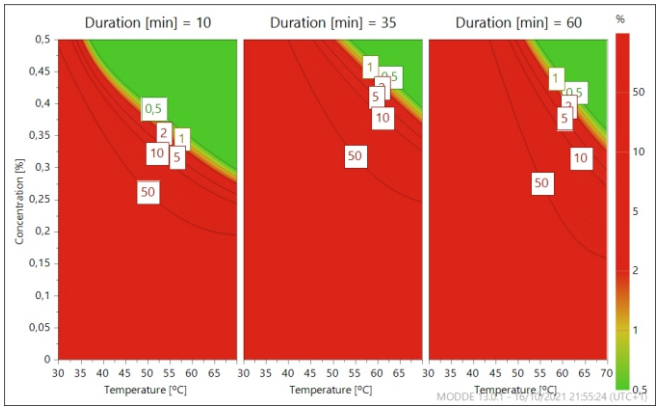

Figure 9 - Design space plot of the membranes originated from the spiral-wound module.

3.4. Membrane Analysis

Contact angles of the membranes from all cleaning experiments were determined. For comparison reasons, also the contact angles of fouled and conditioned membranes were determined. A lower contact angle implies a higher hydrophilicity of the membrane, while a higher contact angle indicates a higher hydrophobicity of the membrane.

For the membranes from both batches, the contact angles of the fouled membranes (**Pictures 10A and 11A**) are lower than the contact angles of the conditioned membranes (**Pictures 10B and 11B**), meaning that the fouled membranes are more hydrophilic than the conditioned ones. The main foulants present in

thermomechanical pulping process water are polysaccharides (hydrophilic) and extractives (hydrophobic). For that reason, it is possible that the foulants remaining on membrane surface are mostly polysaccharides.

Figure 10 - Contact angle comparison between a fouled membrane (**A**) and a conditioned membrane (**B**), both from the flat-sheet batch.

Figure 11 - Contact angle comparison between a fouled membrane (**A**) and a conditioned membrane (**B**), both from the spiral-wound module.

For the membranes from the flat sheet batch, the contact angles of the membranes that were cleaned with pure water (0% of Ultrasil 10) (**Figure 12A**) are lower than the ones measured on the membranes used in the other experiments (**Figure 12B**). The contact angles of those membranes (**Figure 13A**) are also quite similar to ones measured on the fouled membranes (**Figure 13B**). This suggests that cleaning with pure water barely changes the hydrophilicity of the fouled membranes, whereas cleaning with Ultrasil 10 increases the hydrophobicity of the fouled membranes. A possible explanation is that by using Ultrasil 10, polysaccharides were removed in a larger scale than the extractives.

Figure 12 - Contact angle comparison between a membrane cleaned with pure water (**A**) and a membrane cleaned with Ultrasil 10 (**B**), both from the flat-sheet batch.

Figure 13 - Contact angle comparison between a membrane cleaned with pure water (**A**) and a fouled membrane (**B**), both from the flat-sheet batch.

3.5. Fourier Transform Infrared Spectroscopy

FTIR analysis were performed on the membranes from all cleaning experiments, and additionally on fouled and conditioned membranes.

The results obtained on the membranes from the flat-sheet batch are presented in **Figure 14**, which shows the FTIR spectra of a fouled, a conditioned and a cleaned membrane. Overall, the spectrum of the conditioned membrane is less accentuated than the other two spectra. The spectrum of the fouled membrane is quite similar to the spectrum of the cleaned membrane, although it is possible do detect some differences between the two.

There is a slight attenuation in peak intensity in the cleaned membrane compared to the fouled membrane at 1106, 1150, 1168, and 1653 cm^{-1}, and it can be interpretated as a signal of polysaccharides. The attenuation in band intensity in the cleaned membrane compared to the fouled membrane from 3000 cm^{-1} to 3400 cm^{-1} can be assign to polysaccharides as well. The observation above suggests that the presence of polysaccharides on the fouled membrane surface were attenuated after cleaning. Finally, there are small variations in transmittance between the peaks at 1014 and 1080 cm^{-1} which can result from both polysaccharides and PSU, the polymer the membranes used in this project are made of. Those peaks were attenuated in consequence of cleaning and for that reason, they are probably due to polysaccharides that were removed, since the amount of PSU on the membrane surface should not change after cleaning.

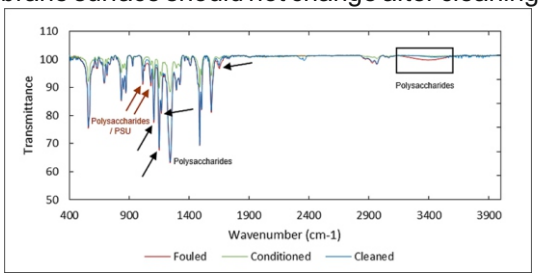

Figure 14 - FTIR spectra of a conditioned membrane, a fouled membrane and a cleaned membrane, all from the flat-sheet batch.

FTIR spectra of three cleaned membranes were analysed: the ones that were exposed to the harshest cleaning, the mildest cleaning, and an average cleaning, but the difference between the spectra intensity is too small to allow an interpretation of the varying impact of the cleaning procedure on the fouling removal.

3.6. Brunauer-Emmett-Teller Analysis

BET analysis were conducted on a conditioned, a fouled, and three cleaned membranes. The latter membranes were treated with the harshest cleaning, the mildest cleaning and an average cleaning.

The pore area and pore volume as a function of pore diameter obtained on the membranes from the flat-sheet batch are presented in **Figures 15 and 16**. **Figure 15** shows the pore area distribution of the membrane samples and **Figure 16** displays the pore volume distribution of the membrane samples.

When comparing the distribution of the fouled membrane with the distribution of the conditioned membrane in both figures, a strong reduction in pore area can be observed and that comes along with a decrease in pore volume for all the pore widths. This could be due to considerable fouling caused by pore blocking. In both figures, it can be seen that the distributions of the three cleaned membranes are rather similar.

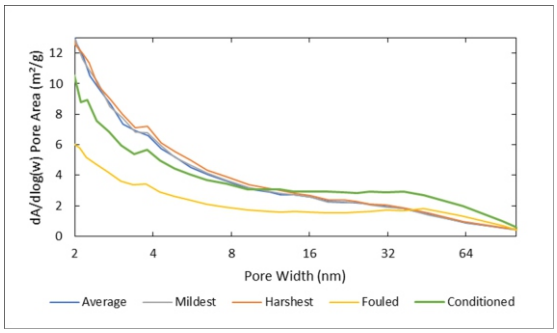

Figure 15 - BJH pore area as a function of pore diameter of a conditioned membrane, a fouled membrane and three cleaned membranes after the harshest cleaning, the mildest cleaning and an average cleaning, all from the flat-sheet batch.

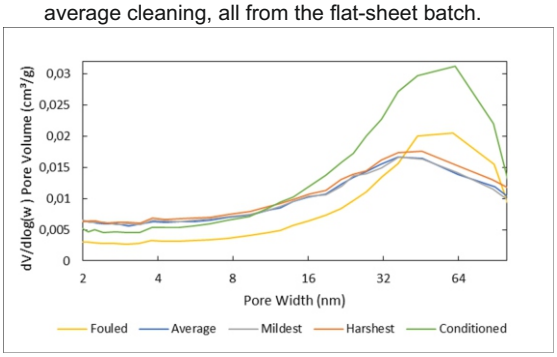

Figure 16 - BJH pore volume as a function of pore diameter of a conditioned membrane, a fouled membrane and three cleaned membranes after the harshest cleaning, the mildest cleaning and an average cleaning, all from the flat-sheet batch.

4. Conclusions

DoE evaluation was quite optimist regarding the analysis and predictions for the membranes from the flat-sheet batch when compared to the membranes originated from the spiral-wound module. Both models showed a high significance, but the selected model manifested a lack of predictive relevance for the data of membranes from the flat-sheet batch. On the other hand, the negative impact of the cleaning duration observed on study of the membranes originated from the spiral-wound module is not reasonable but cannot be explained for now. Besides, it seems unreasonable to pursue only the maximized cleaning success. It is crucial to find a balance between a good cleaning success and a sustainable cost-effective cleaning protocol.

Overall, concentration of the cleaning agent proved to the be the most relevant cleaning parameter, while duration demonstrated a rather low impact on the cleaning success. The interaction between concentration and temperature appears to have a considerable effect in the cleaning success as well.

Contact angle analysis suggests that the main foulants attached to the membrane surface are polysaccharides, in the form of hemicelluloses and they seem to be removed to some extent by cleaning with an alkaline solution. Cleaning with pure water has not expressed any changes regarding the hydrophilicity of the membrane, indicating that its cleaning efficiency was quite low.

FTIR analysis emphasize the findings from the contact angle analysis, since polysaccharides appear to be the main foulants on the membrane surface and based on the observation of the differences in transmittance, they are partly eliminated by cleaning.

BET analysis only accused fouling in the form of pore blocking, perhaps caused by polysaccharides molecules, based on the conclusions provided by the previous analysis.

These membrane analyses combined proved to be effective techniques to supply a deeper understanding regarding the fouling layer composition and its inner area and volume.

5. References

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