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Intégration de la simulation de l'ingénierie des processus et de l'évaluation du cycle de vie pour la modélisation de l'impact environnemental de la filtration membranaire

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Résumé

Le développement des procédés de bioraffinerie nécessite des outils d'éco-conception efficaces afin de démontrer leur réel intérêt écologique tout en minimisant le temps d'acquisition des données permettant l'extrapolation industrielle. L'objet de cet article concerne un de ces outil combinant l'ingénierie des systèmes de procédés (ISP) avec la méthodologie de l'analyse de cycle de vie (ACV) afin d'optimiser un procédé ou une opération unitaire en tenant compte des performances techniques et environnementales. En effet, l'ISP ne permet pas la simulation de toutes les opérations unitaires impliquées dans les procédés de bioraffinerie, comme l'ultrafiltration. Cette étude démontre les avantages du couplage de la simulation de l'ultrafiltration avec l'ACV, en utilisant les logiciel ProsimPlus© et SimaPro©. Un modèle d'ultrafiltration a été développé dans ProsimPlus© afin de simuler l'impact des conditions opératoires sur le flux de perméat en fonction du seuil de coupure des membranes. Les paramètres de ce modèle (α et β) ont été identifiés à partir de résultats expérimentaux de filtration de composés phénoliques avec une membrane comprise entre 5 et 100 kDa, avec une validation montrant une incertitude inférieure à 25%. Ces paramètres ont été utilisés pour modéliser les performances de filtration sous différents types de colmatage, puis la simulation a permis l'évaluation environnementales de chaque condition avec SimaPro©. Les résultats indiquent que réduire l'impact environnemental nécessite de diminuer la surface de filtration. L'ACV identifie également l'étape de nettoyage comme le principal point chaud environnemental et montre qu'en présence de polarisation de concentration, l'impact de la consommation d'énergie est aussi important que celui de la surface de la membrane.

Integration of process engineering simulation and life cycle assessment for modeling the environmental impact of membrane filtration

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Abstract

Developing biorefinery processes requires effective eco-design tools to demonstrate their real ecological interest while minimizing the data acquisition time allowing industrial extrapolation. This article emphasizes the integration of process systems engineering (PSE) with life cycle assessment (LCA) to optimize process conditions and minimize environmental impacts. Indeed, PSE does not allow the simulation of all the unit operations involved in biorefinery processes, such as ultrafiltration. This review focused on developing eco-design tools for the biorefinery process by combining PSE and LCA. It demonstrates the benefits of integrating ultrafiltration simulation with LCA, using ProsimPlus© software to model the effects of operating conditions on permeate flux, with parameters derived from experimental ultrafiltration of phenolic compounds ranging from 5 to 100 KDa. The model's results closely align with experimental data, achieving less than 25% validation uncertainty. Parameters α and β were employed to model different types of fouling, and the simulation was combined with an environmental assessment using SimaPro© software. Results show that reducing environmental impact involves decreasing the filter surface area. Additionally, LCA identifies the cleaning step as the primary environmental hotspot and highlights that, with concentration polarization, the impact of energy consumption becomes as significant as the membrane area.

Introduction

Traditionally, the design of chemical processes is based on technical and profitability criteria. Nevertheless, process optimization must now consider other criteria as climate concerns grow as described by Azapagic in 1999. This transition requires considering the environmental and societal impacts. Consequently, various methodologies have emerged, among which the integration of process engineering and LCA proves to be a promising approach to simulate the process and assess its impacts. Nevertheless, implementing this coupling is challenging for processes developed for biomass fractionation. These processes include unit operations (membrane filtration, chromatography, etc.) that are not as well described as processes like distillation in conventional software such as Aspen© and SuperPro-designer©.

Due to the transition towards sustainable technologies, the chemical and biochemical process industries are progressively adopting bio-based production strategies over traditional chemical methods. Although this change is promising, entirely relying on bio-based production doesn't ensure eco-friendliness in process technologies. The energy-intensive separation and purification of products, which often involve harsh chemicals, is a significant challenge in downstream processes. Membrane filtration technology provides a solution by efficiently separating components without the need for harsh chemicals or heavy energy consumption. The combining of bio-based production membrane filtration process holds great potential for a more sustainable and cleaner future in the chemical sectors as highlighted by Clark and Macquarrie in 2002, and Green and Southard 2018. There are several types of membrane technologies including microfiltration, ultrafiltration, and nanofiltration that are widely used across industries such as chemical, food, pharmaceutical, and dairy sectors. The accumulation of material within or on the membrane, which is attributed to phenomena known as fouling and concentration polarization is the major drawback of the membrane filtration process as highlighted by Chen, Li, and Elimelech in 2004. Fouling involves the accumulation of microorganisms, particles, colloids, or other substances within the pores or on the membrane surfaces. It occurs through several mechanisms, including pore blockage, formation of a cake layer, and adsorption of foulants on the membrane. Concentration polarization appears when substances accumulate near the membrane interface in colloidal or solubilized form as a result of convection forces. It substantially affects the performance of the membrane, resulting in significant filtration resistance and reduced membrane flux as described by Bolton, LaCasse, and Kuriyel in 2006.

Indeed, while membrane modeling has been studied for a long time, the developed models are generally based on the description of mechanisms but are not suitable for simulating the performance of a wide range of membranes. Therefore, in the classical process engineering approach, membrane performance is evaluated based on mass balance and requires experimental data. To address this gap in membrane filtration, it is necessary to have a model capable of simulating results ranging from nanofiltration to microfiltration.

The objective of this study is to present a new approach to facilitate the sustainable design of biomass fractionation processes by integrating process engineering simulation using ProsimPlus© by Process Simulation Software in 1989 and LCA with SimaPro© software by Pre consultant in 1990. This methodology is illustrated through a case study related to the purification of phenolic compounds by ultrafiltration. The work aims to demonstrate the effectiveness of coupling ultrafiltration process modeling with LCA as a valuable tool for eco-optimizing. This is achieved by studying the effect of operational conditions on the process and determining key environmental impact categories. and highlighting the influence on environmental impacts.

Materials and methods

Model

In process engineering, the design and simulation of membrane filtration systems present unique challenges due to the absence of a universally applicable model. Each membrane filtration process is highly specific, influenced by variables such as feed composition, membrane material, operating conditions, and target separation requirements. These factors necessitate tailored approaches to accurately predict performance and optimize design. Consequently, the lack of a truly generic model led to the development of a custom model to ensure precise and efficient filtration system design based on experimental data and specific process requirements. The ProsimPlus© software allows for the simulation and scaling of various chemical engineering processes. Although it is not inherently suited for membrane filtration, it can be adapted by implementing a specific model within a dedicated module.

In this study, a model (Equation 1) describing the performance of ultrafiltration was introduced into ProsimPlus© and used to achieve a comprehensive analysis of technical performance under varying operating conditions. These simulations were linked to SimaPro© to determine the influence of operational conditions on the environmental impact of the unit's operation. The simulation challenges stem from the varying thicknesses across different membrane types and the effects of fouling and concentration polarization layers during filtration. The membrane filtration model was developed based on Darcy's law and modified to account for the effect of concentration polarization and fouling.

$$\mathrm{J} = \frac{\mathrm{L_{p,s}}}{\beta.\mu}\left(1 - \exp\left(\frac{\mathrm{TMP}}{\alpha}\right)\right) \qquad (1)$$

Where J represents the permeate flux (L.m^{-2}.h^{-1}), $L_{p,s}$ the permeability (L.m^{-2}.bar^{-1}), μ the viscosity of the solution (Pa.h), TMP represents the transmembrane pressure (Pa), and β and α are dimensionless parameters that represent the intensity of fouling and the concentration polarization layer, respectively.

The permeability is calculated based on Equation 2:

$$\mathrm{L_{p,s}} = \frac{\epsilon \mathrm{d_p^2}}{32\tau \mathrm{e_m}} \qquad (2)$$

Where ϵ is the membrane porosity (%), d_p is the pore size (m), τ is the tortuosity, and e_m is the membrane thickness (m).

The recommended method for calculating the pore size involves using the MWCO correlation for dextran, as described by Ren, Li, and Wong in 2006.

$$\mathrm{r_p} = 0.33\mathrm{x}10^{-1}(\mathrm{MWCO})^{0.46} \qquad (3)$$

Where r_p is the pore radius in nm and MWCO is the molecular weight cut-off in Da.

The results were compared with those obtained using the Stokes-Einstein equation to validate the correlation.

$$\mathrm{D} = \frac{\mathrm{K_B T}}{6\pi\mu \mathrm{r_p}} \qquad (4)$$

Where D is the diffusion coefficient (m^2.s^{-1}), k_B is Boltzmann’s constant (J.K^{-1}), T is the temperature (K), μ is the viscosity of the solvent (Pa.s), and r is the hydraulic radius of the molecule (m).

Table 1. Comparison of methods for calculating the molecular radius to validate the use of dextran - Pore radius calculated by dextran using equation 3 and Stokes-Einstein correlation using equation 4.

	Dextran correlation		Stokes-Einstein correlation	
Molecules	Molecular weight (g.mol^{-1})	Radius (nm)	Diffusion coefficient at 25°C (m^2.s^{-1}) by Nakao and Kimura in 1981	Radius (nm)
Glucose	180.15	0.359	6.9 10^{-10*}	0.355
Sucrose	342.29	0.483	5.2 10^{-10*}	0.471

The summarized results are presented in the table below. From Table 1, it can be concluded that Equation 3 is effective for simulating and characterizing molecules based on their size.

The model parameters (α and β) are used to describe the limitations arising during filtration with the solution. The shapes of the curves obtained with different values of these parameters allow for defining the ranges where they describe the characteristics of strong fouling, polarization layer, and weak fouling (Table 2).

Table 2. Various ranges of alpha and beta for different types of clogging.

Types	α	β
Strong fouling	≥3	≥3
Polarization	0.6≤ α ≤2	0.9≤ β ≤1.5
Weak fouling	3≤ α ≤5	0.2≤ β <0.4

Application of LCA

LCA is a method used to analyze the environmental impacts of products or services throughout their entire lifecycle from production and consumption to disposal (commonly known as "cradle to grave") by Burgess and Brennan in 2001. The LCA process typically involves four main steps: 1) defining the goal and scope, 2) conducting a life cycle inventory analysis (LCI), 3) performing a life cycle impact assessment (LCIA), and 4) interpreting and improving the life cycle analysis as outlined Pennington et al. in 2004.

LCA was performed using SimaPro©. This software simplifies database management and aids process visualization, adhering to ISO 14044 and 14040 standards. It allows the decision-makers to identify the most environmentally friendly operational scenarios for the process of the treatment units. The most recommended European method used for impact assessment is the EF 3.0 method.

In this study, the LCA was performed using SimaPro© software. This software simplifies database management and helps with process visualization, adhering to the ISO 14040 and 14044 standards. SimaPro© enables decision-makers to identify the most environmentally friendly operational scenarios for treatment unit processes. The EF 3.0 method, the most recommended European method, is used for impact assessment.

The installation is equipped with a feed tank continuously supplied by a feed pump with a flow rate equivalent to the sum of the volumes of retentate and permeate (Fig. 1). The manometric height of this pump is estimated at 10 m, corresponding to a pressure drop estimated at 1 bar upstream of the process. A circulation pump allows the tangential speed to be controlled within the module. The manometric height is estimated as the transmembrane pressure to be applied to the membrane, plus a pressure drop on the circuit is estimated at 1 bar.

Case study

The system’s studied function is to produce a purified extract through continuous withdrawal of permeate and retentate at the same flow rate at the ultrafiltration module outlet. To construct the production inventory of the system, the following functional unit has been chosen: The functional unit is chosen to be 40 L.h^{-1} of permeate flow rate produced with a VCF of 2.

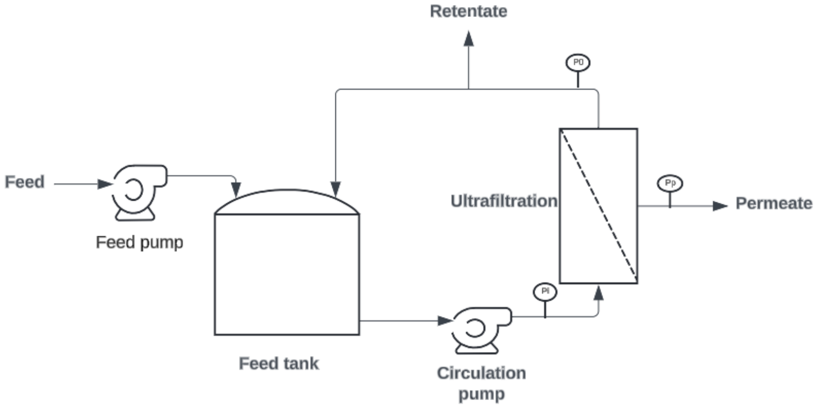

Figure 1. Flowsheet of the UF production process

As illustrated in Fig. 2, the system boundary is divided into two main components: membrane installation and membrane operation, which includes both filtration and cleaning. The figure outlines three stages in the life cycle of the unit process under study: manufacturing of ultrafiltration modules, end-of-life management of the modules, and the operational processes. The system under consideration encompasses the electricity required for operating and cleaning the ultrafiltration module, as well as the production of water and chemicals needed for post-filtration membrane cleaning. The system boundary is defined as gate-to-gate, focusing exclusively on a specific stage within the product's life cycle from raw material acquisition to the completion of the production process. During the membrane manufacturing phase, the system considers the production of materials for the modules, as well as the energy and water requirements for their manufacture, and waste management. In the operational phase, it accounts for the electricity consumption during filtration and cleaning, along with the production of water and chemicals necessary for membrane cleaning post-filtration.

Before conducting the analysis, it is necessary to clarify the assumptions made for the realization of this study:

- The quality of the input extract for ultrafiltration is constant, and its behavior is illustrated by the modeling results.
- It is difficult to find data on the lifespan of ultrafiltration membranes, especially since it is highly dependent on the frequency of membrane cleaning operations. In the field of water treatment, literature estimates the lifespan of membranes to be between 3 and 5 years by Staff 2007. However, in this field, less frequent membrane cleaning helps preserve the lifespan of the modules. Therefore, it seems realistic to consider that, in our case, a module has a lifespan of one year. The lifespan of a membrane is estimated at the production of 40,000 $L.h^{-1}$ of permeate, corresponding to one thousand functional units.
- The interval between two cleanings allows the total installation to produce one hundred functional units.
- The ultrafiltration module in SimaPro© is a hollow fiber, and the membrane material is polyvinyl fluoride.
- The lifespan of feed and circulation pumps is greater than 30 years; therefore, the impact of their manufacture is not considered. The same applies to their maintenance and end-of-life.
- Electricity is derived from the French energy model.
- All pumps used have an efficiency range between 70-80%.

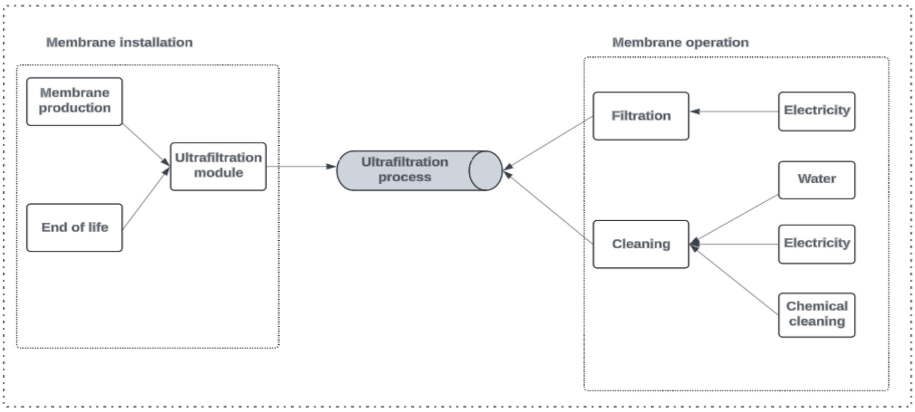

Figure 2. System boundary for the study of the ultrafiltration process

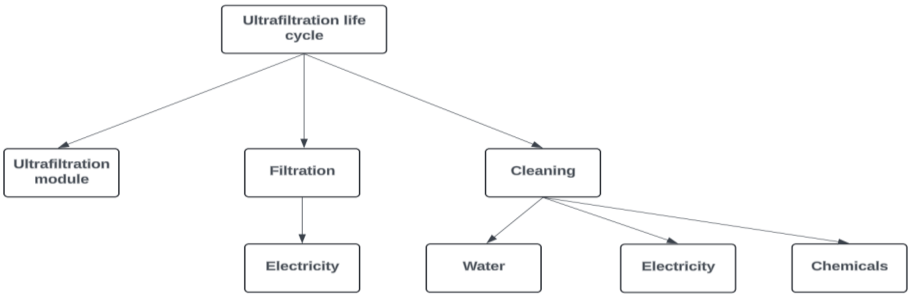

Figure 3. Hierarchical Process tree structure for ultrafiltration scenario

Fig. 3 illustrates the life cycle process tree for the ultrafiltration unit. This study is distinguished by its integration of both a 'cradle to grave' approach and a 'gate to gate' approach. Within the defined system boundaries, it considers various infrastructures, including the manufacturing and end-of-life processes of ultrafiltration modules, while also examining the operational aspects of the unit process. By abandoning the notion of treating the unit process as a black box, the study takes into account operational conditions when constructing the inventory. Subsequently, these stages can be grouped into two broader stages: operation including filtration and cleaning; and installations, including manufacturing and end-of-life of modules. For each of these stages, the process approach is used to list all the inputs and outputs.

The life cycle inventory combines foreground data collected using ProsimPlus© after simulating the ultrafiltration and coding its engineering design equations, and background data from ecoinvent cut-off in SimaPro©. LCIA interprets this data to assess its environmental significance and potential implications. For modeling the life cycles, SimaPro© v9.6.0 was used, with the ecoinvent v3.10 database for background data (refer to Fig. 4).

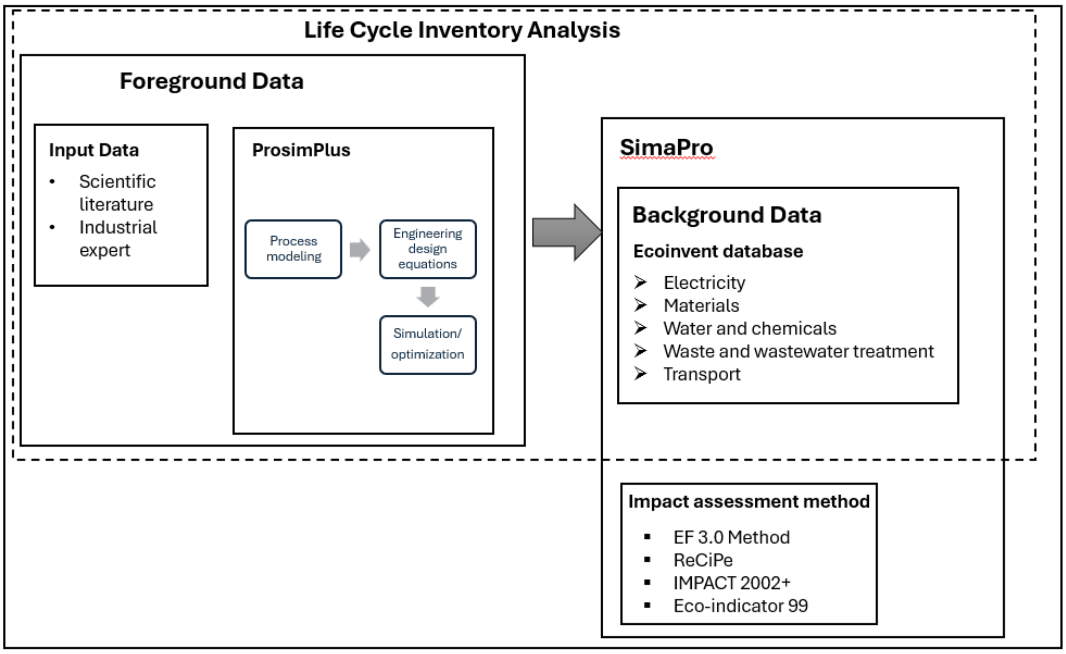

Figure 4. Model framework of LCI for integrating LCA with ultrafiltration process simulation

Results

Ultrafiltration model

The first step in calibrating the model from experimental results is to determine membrane permeability. The ultrafiltration membranes used were hollow fiber polyethersulfone membranes (PES) with an MWCO ranging from 5 kDa to 100 kDa, as identified by Beaufils in 2019. These membranes provided a surface area of 0.085 m² for 5 kDa MWCO and 0.14 m² for other MWCOs in laboratory-scale extractions. Table 2 shows minimal differences in errors between experimental permeability tests and theoretical permeability calculations using the permeability model in Equation 2.

Table 2. Permeability differences of ultrafiltration membrane for various MWCO resulted from experimental study by Beaufils in 2019 and the model (Equation 2) along with values of α and β corresponding to each MWCO.

MWCO (KDa)	$L_{p,s}$ experimental ($L.m^{-2}$)	$L_{p,s}$ theoretical ($L.m^{-2}$)	Error (%)	α	β
5	$1.4\ 10^{-10}$	$1.5\ 10^{-10}$	6.6	3	6
10	$2.2\ 10^{-10}$	$2.2\ 10^{-10}$	0	3.5	4.5
30	$2.4\ 10^{-10}$	$2.4\ 10^{-10}$	0	3	4
50	$3.7\ 10^{-10}$	$3.9\ 10^{-10}$	5.4	5	4
100	$7.5\ 10^{-10}$	$7.4\ 10^{-10}$	1.3	0.6	5

The second step in the modeling process involves evaluating parameters that describe flux limitations during solution filtration (α and β). Experimental results indicate that permeate flux increases nearly linearly with transmembrane pressure (TMP) for membranes ranging from 5 to 50 kDa, suggesting minimal concentration polarization. The water permeabilities measured for each membrane are significantly higher than the slope coefficients of the resulting curve. This suggests that if the filtration occurs below the critical flow, the solvent passage resistance may arise from adsorption on the membrane surface or in the pores and pore blockage. The permeate flux for a MWCO of 100 KDa increases linearly with the TMP, but a drop is observed above 0.8 bar, indicating an intensified limiting phenomenon. At this point, filtration is no longer governed by the TMP but by material transfer, which restricts the flow. Increasing the pressure above 0.8 bar leads to the formation of a viscous layer or gel, causing clogging due to molecule precipitation on the membrane surface, thereby resulting in the appearance of a polarization layer, as highlighted by Beaufils. Consequently, alpha and beta values for MWCOs of 5, 10, 30, and 50 kDa are expected to fall within the fouling behavior range, while the alpha value for MWCO 100 kDa should be within the polarization layer range. Applying equation 1 to calculate permeate flux for MWCOs (5, 10, 30, 50, and 100 kDa) confirms the theoretical model's validation against Beaufils's experimental data (Fig. 5), with discrepancies not exceeding 15%.

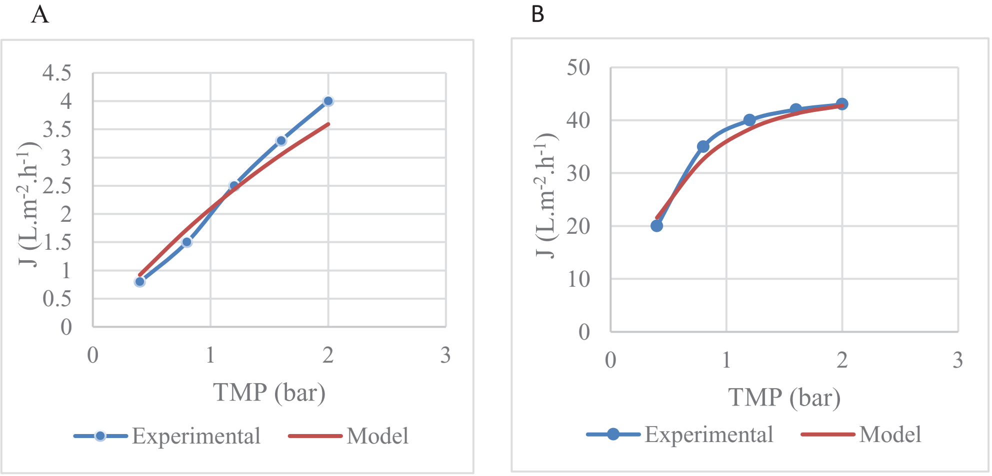

Figure 5. Permeate flux as a function of TMP for the experimental study of phenolic compounds by Beaufils and the generated model applied for various MWCO (A: MWCO 5 kDa; B: MWCO 100 kDa)

Life cycle assessment

The model was employed to assess the environmental impact of ultrafiltration on tannin solutions using a 5 kDa membrane, studying the influence of pressure on permeate flux under three conditions: strong fouling, low fouling, and strong polarization. The normalization method was used to understand the magnitude of the damage for each midpoint impact category, helping to select the most significant ones. The selection was based on the two categories that exhibited the highest impact when simulating the influence of pressure using different α and β values. The potential environmental impacts considered are therefore water use (WU), and resource use fossils (RUF), which have the highest impact category scores.

The strong fouling consistently results in the highest environmental and resource impacts across all categories, whereas the other two conditions have lower impacts as shown in Figure 6. Increasing pressure reduces the environmental impact in all cases, with a particularly marked decrease in the case of strong fouling. For polarization, increasing pressure reduces water consumption but resource consumption remains stable. This increase in pressure also leads to an increase in permeate flux, thus reducing the membrane surface area required to produce the functional unit. In calculating the impacts, a decrease in flux implies the use of a greater number of membrane modules to achieve the required flow rate, which increases energy consumption for filtration and cleaning operations. Furthermore, increased use of water and chemicals for cleaning worsens the overall environmental impact of the process. Conversely, when transitioning from conditions of strong fouling to polarization and eventually to weak fouling, there is a notable improvement in permeate flux. This improvement significantly reduces the number of required membrane modules. For instance, during polarization, the influence of pressure is lower, and the energy consumed mitigates the reduction of the surface.

The findings show in Figure 6 that the environmental impacts are linked to resource use fossil consumption and water deprivation due to the highest impact scores. This is because of the electricity consumption during filtration and cleaning, along with the usage of significant amounts of water and chemicals necessary for membrane cleaning post-filtration. The extraction, processing, and transportation of these fossil-based materials consume energy and generate emissions, contributing to the environmental footprint of the membrane filtration system. Membrane filtration processes can be energy-intensive, especially during the operation of pumps for filtration and cleaning. Since a majority of global energy production still heavily relies on fossil fuels, the energy consumption in membrane filtration systems often results in significant emissions of greenhouse gases and other pollutants.

Among the stages of the ultrafiltration process, the cleaning stage emerges as the hotspot, contributing 67% of the total environmental impact due to substantial water usage. However, the operation stage accounts for 28%, with electricity consumption being the primary contributor. By identifying the major hotspots of the ultrafiltration stages, the optimization should be focused on improving the cleaning efficiency and water usage, as well as reducing electricity consumption during the operation stage to mitigate the total environmental impact of the process.

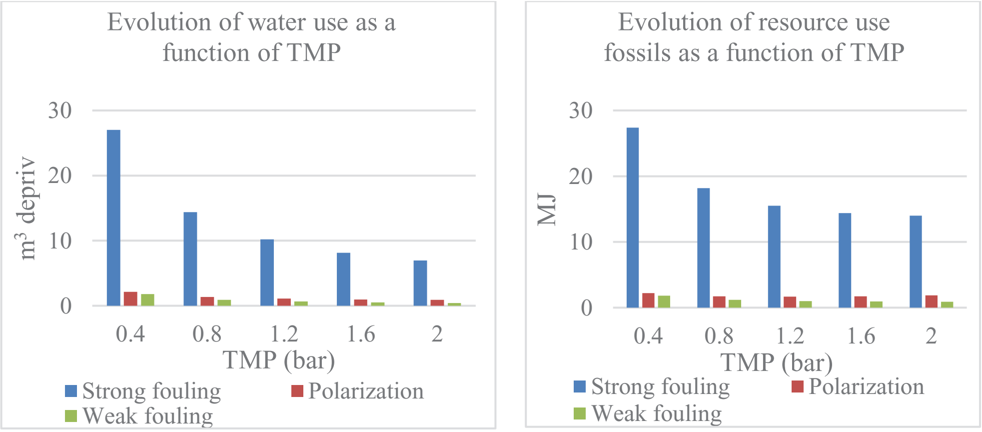

Figure 6. Evolution of midpoint impact categories as a function of Transmembrane pressure for three clogging types (strong fouling, polarization, and weak fouling) with a MWCO of 5 KDa

Conclusion

This study introduces methodological innovation by integrating process modeling with LCA. This approach has been applied to membrane filtration through the development of a model in ProsimPlus© software. The model calculates membrane water permeability based on MWCO and describes different types of fouling using two parameters, α and β. It simulates experimental results with a standard deviation below 25%. By combining pressure influence on permeate fluxes with LCA, the results indicate that under fouling conditions, a lower membrane surface area reduces impact, making higher pressure more advantageous. In contrast, when a polarization layer is present, the situation is more complex, and LCA proves to be an effective tool for determining the optimal conditions. The cleaning stage has been identified as the major hotspot in the ultrafiltration process. To mitigate this impact, increasing the interval between cleanings defined as the number of functional units produced between two cleaning cycles can be an effective strategy.

There are further areas for improvement though the results presented in this article are promising. The model could be extended to describe the tangential velocity's influence on permeate flux, and calculate the retention rate, hence offering a more significant insight into the operational conditions and their environmental implications.

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