Does substituting reprotoxic solvents during ultrafiltration membrane fabrication really mitigate environmental impacts? Focus on drinking water production

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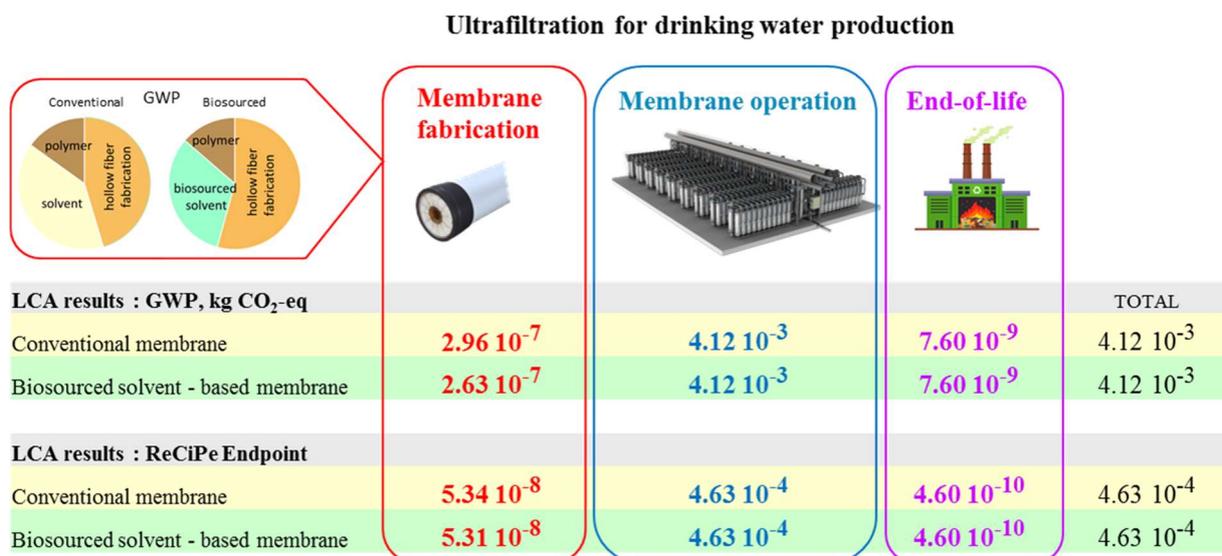Flavie Prézélus[a,b], Jean-Christophe Remigy[b], Christelle Guigui[a], Ligia Tiruta-Barna[a,*]

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Graphical abstract

Abstract

The purpose of this research is to fill knowledge gaps on the relevance of solvent substitution from a life cycle perspective and to put forth relevant operating conditions for possible process optimisation and eco-design. Solvents commonly used to prepare ultrafiltration membranes for drinking water applications are indeed identified by the European REACH regulation for their potential reproductive toxicity and are the subject of research as regards their substitution. Coupling Life Cycle Assessment (LCA) with process modelling allowed to compare conventional membranes (cellulose triacetate as polymer, NMP as solvent) to biosourced membranes (cellulose diacetate as polymer, methyl lactate as solvent). Although methyl lactate fabrication leads to lower environmental scores than NMP fabrication (32% and 10% less GWP100 and ReCiPe Endpoint, respectively), LCA scores for membrane fabrication are similar (1.9 points ReCiPe for $1m^2$ membrane) or slightly different, i.e. 11 kg CO_2-eq and 9.4 kg CO_2-eq for $1m^2$ of conventional and biosourced membrane, respectively. From an environmental point of view, solvent substitution has little influence on impacts. Rather, environmental hotspots lie in the operation stage, with impact scores about 10,000 times higher than those of membrane fabrication, which holds out considerable prospects for the optimisation of operating conditions on drinking water plants.

Highlights

Solvent substitution does not bring about any significant environmental mitigation

Coupled process modelling-LCA is an agile tool for process evaluation and eco-design

Lower environmental impacts of methyl lactate fabrication compared to NMP solvent

Extensive filtration operation mitigates overall impacts

Keywords (max 6)

solvent substitution; membrane filtration; process modelling; Life Cycle Assessment; hollow fiber; water treatment; process ecodesign

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1. Introduction

Pressure-driven membrane filtration is used in a myriad of industrial sectors, be it for juice purification in the agro-food industry or for water treatment. Operating membranes for the physical separation of specific compounds from fluids not only offers a high selectivity but also an efficient management of waste and energy. Other interesting assets are the ease and simplicity of its operation, maintenance, scale-up and control. As regards drinking water production from ground and surface waters for which particulate and pathogens are to be removed, ultrafiltration (UF) has superseded the conventional coagulation-flocculation-settlement treatment thanks to the high quality of the treated water, the limited use of chemicals and the compacity of its facilities (Bersillon et al., 1989; Cabassud et al., 1991; Clark and Heneghan, 1991; Jacangelo et al., 1997).

1.1. The challenge of green-solvents in membrane fabrication

For a variety of reasons including low price, compacity and an appropriate balance between permeability and selectivity, UF membranes in drinking water plants are primarily self-supported hollow fibers based on a porous organic structure (Figoli and Criscuoli, 2017; Pearce, 2011). These membranes are mainly fabricated by non-solvent induced phase separation (NIPS), whereby a polymer initially dissolved in a solvent precipitates upon contact with a non-solvent (typically water) (Cabasso et al., 1976; Koenhen et al., 1977; Strathmann et al., 1975). The final membrane structure and the pores correspond, respectively, to the polymer rich phase and polymer lean phase during the demixing process. Common solvents for NIPS are N-methyl-2-pyrrolidone (NMP), N,N-dimethylacetamide (DMA) and N,N-dimethylformamide (DMF), all of which are of very high concern according to the European REACH regulation due to their potential reproductive toxicity (Baker, 2012; ECHA). Whilst the current use of these solvents requires precautionary health and safety measures to protect exposed workers, their future use is compromised since regulatory restrictions aim at substitution by less toxic alternatives.

Within this regulatory context, research in membrane science has been ongoing to substitute these solvents and obtain membranes as effective as the ones currently commercialized. Examples of novel solvents for membrane preparation are methyl lactate (Medina-Gonzalez et al., 2011), Cyrene™ (Marino et al., 2019), triethylphosphate (Abed et al, 2012), Rhodiasolv® Polarclean (Dong et al., 2018) and TamiSolve® NxG (Marino et al., 2017). Figoli et al.'s (2014) review specifically addresses the implications of replacing reprotoxic solvents on the NIPS process and on final membrane performances (i.e. selectivity, permeability). The term "greenness" is moreover frequently employed to describe the degree to which the involved solvents or membranes reduce environmental impacts. Indeed, solvent toxicity for humans is not the only issue at stake. Other issues relate for example to

toxicity for the environment, energy needed for solvent manufacture or issues of recycling and disposal. Figoli et al. (2014) recommend using Life Cycle Assessment (LCA) to give a quantifiable meaning to the concept of "greenness", although this holistic approach has not been retained in his review. This standardized environmental assessment tool compiles and evaluates the inputs, outputs and the potential environmental impacts of a given system throughout its life cycle (ISO, 2006). An LCA study follows four interdependent phases: goal and scope definition, inventory analysis, impact assessment and results interpretation.

In order to obtain a comprehensive picture of the environmental performance, the LCA method must be coupled with relevant, parameterized models of the studied process. The process modelling – LCA approach, although not very widespread, has already been used for process evaluation and eco-design in previous researches (e.g. Méry et al., 2013; Bissinella de Faria et al., 2015; Ahmadi et al., 2019; Besson et al., 2021) dedicated to water treatment. The advantage of this approach is the possibility to investigate the system for different operating conditions, process steps, raw materials used, and to propose eco-designed solutions.

1.2. Aims and novelty of this work

To the best of our knowledge, no previous work has been carried out on the use of LCA to evaluate the environmental relevance of solvent substitution in membrane fabrication, which could thereby offer a life cycle perspective to membrane science research. In our previous work, process modelling has been coupled with LCA to assess the contribution of material and energy to environmental impacts, as well as the influence of operating conditions. This approach has been first applied to membrane fabrication (Prézélus et al., 2020a; Prézélus et al., 2020b), then to membrane filtration in drinking water plants (Prézélus et al., 2021). In another previous work, an experimental approach has been undertaken to prepare biosourced UF membranes with a non-reprotoxic biosourced solvent (methyl lactate) instead of NMP (Prézélus et al., 2019). In the present study, the mentioned biosourced membranes are compared with conventional ones using the process modelling–LCA tools previously developed. The aims are (1) to fill knowledge gaps on the relevance of solvent substitution from a life cycle perspective and (2) to put forth relevant operating conditions for possible process optimisation and eco-design practices. The following sections describe: i) the life cycle stages of UF membranes for drinking water production considered in this study, as well as the LCA methodology; ii) LCA comparative results considering the membrane life cycle, i.e. membrane fabrication, filtration process and membrane end of life, and for the conventional and new biosourced membrane.

2. Material and methods

2.1. Methodology used

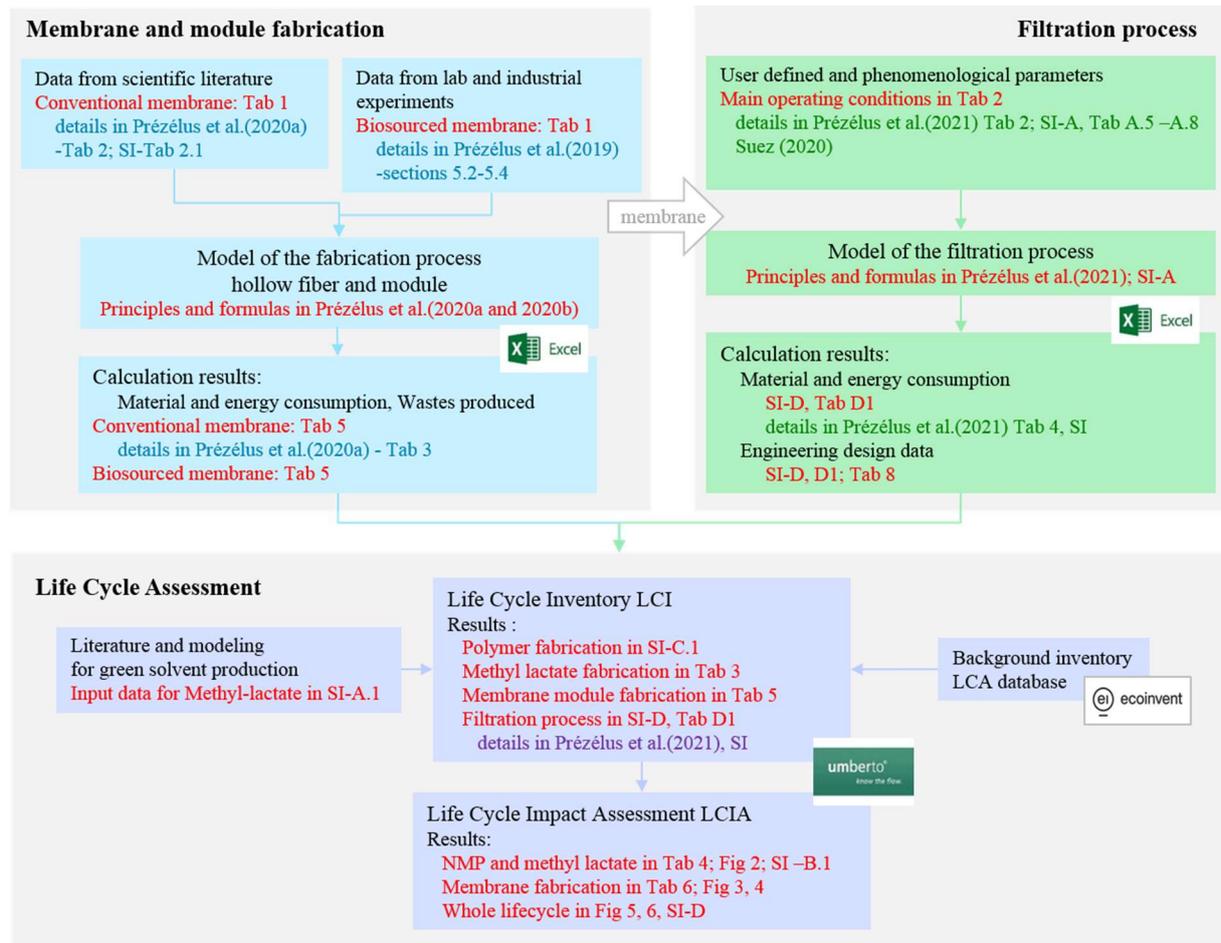The modelling methodology used in this work is schematized in figure 1.

Figure 1. Flowchart of the modelling methodology, data used and results obtained. Notations: SI- supplementary information document, Tab- table; Fig- figure. Colour legend: text in red - information available in this article; text in blue, green, purple - other sources.

Manufacturing of UF membrane and modules, together with the filtration process, represent the foreground part of the life cycle system investigated. In order to build the Life Cycle Inventory (LCI, i.e. the inputs and outputs of the studied life cycle system), material and energy balances need to be calculated based on the selected processes. A process model was previously developed for the membrane fabrication and another one for the filtration process, at industrial scale (Prézélus et al., 2020a, 2020b, 2021). The first model uses data from literature, from lab-scale experiments and industrial sites measurements (source of data mentioned on Figure 1 in red and blue colours), to

calculate, for a produced membrane surface (or number of modules), the material and energy consumption and waste produced (results presented in section 3, mentioned in red and blue on the figure). The second one uses data from literature, industrial sites, and imposed parameters by the user (data source in red and green text), in order to calculate, for a given UF water production, the material and energy consumptionand additional engineering design parameters (e.g. lifetime of membranes) (results available in section 3 and in supplementary information document). Thus, the foreground inventory is obtained. It is then completed with background data from LCA databases and used for environmental impact calculation in a LCA software. The generic background data were completed with own calculations for the missing information, especially for the biosourced solvent used (LCI and LCIA results are displayed and discussed in section 3, as mentioned in red in Figure 1). The method was applied for a conventional and for a fully biosourced membrane, as described in the followings.

2.2. Module description

The case study is centred on two UF modules with a filter surface area of 55 m^2. What differ between the two modules are the hollow fibers within. The conventional membranes are made of cellulose triacetate (CTA), and NMP is the fossil-based solvent used to fabricate them. The new membranes stem from the solvent substitution methodology undertaken in a previous study (Prézélus et al., 2019); these membranes are made of cellulose diacetate (CDA), and methyl lactate is the bio-based solvent. The module configuration is based on a commercialized model for which industrial data has been obtained (Suez, 2020). Graphic information on module components can be found in Prézélus et al. (2020a).

In what follows, the new membrane will be referred to as "biosourced membrane" to distinguish it from the conventional membrane whose production requires a fossil-based solvent.

2.3. Module fabrication process

A generic model for NIPS membrane fabrication has been previously developed to obtain the inventory of material and energy flows as a function of operating conditions (Prézélus et al., 2020a; Prézélus et al., 2020b). The same process is considered in this work, whereby five main steps occur in a sequential order: polymer solution preparation, hollow fiber preparation by NIPS, module preparation, testing and shipment preparation. During the extrusion stage of hollow fiber preparation, a bore liquid is used to obtain the cylindrical shape of membranes. After coagulation and rinsing, these membranes are sprayed with a viscous pre-conditioning liquid to ensure filled pores during the glueing stage. Assumptions and model equations related to mass and energy

balances of each unit operation are detailed in Prézélus et al. (2020b), as well as operating conditions for conventional membrane fabrication (reported in Table 1).

The model was used here with specific operating conditions for biosourced membrane fabrication (see Table 1) determined in the lab-scale experimental study presented in Prézélus et al. (2019).

Table 1 Module fabrication: the main operating conditions

Operating condition	Membrane	
	Conventional[1]	Biosourced[2]
Hollow fiber inner and outer diameters (mm)	0.93, 1.67	
Module inner diameter and length (m)	0.3, 1.3	
Polymer solution composition (wt%)	CTA : NMP : LiCl[3] 20 : 78 : 2	CDA : methyl lactate : LiCl[3] 15 : 82 : 3
Bore liquid (wt%)	NMP : water 5 : 95	methyl lactate : water 16 : 84
Solvent, polymer solution and bore liquid temperature (°C)	60	80
Non-solvent	Water	
Pre-conditioning liquid	Glycerol	

[1] Prézélus et al., 2020a ; Prézélus et al., 2020b

[2] Prézélus et al., 2019

[3] LiCl: lithium chloride, used as porogen

2.4. Membrane filtration process

The filtration process considered in this work is based on a previous study (Prézélus et al., 2021), whereby the fabricated UF modules are included in a membrane unit operating in a dead-end configuration to deliver a defined daily output water flow rate. Membrane operation is carried out under stable state with the alternation of filtration, backwash (BW) and clean-in-place (CIP) cycles. To summarize the previously undertaken modelling of the process, material and energy flows are calculated based on the determination of relative permeability variations during the modules' operating life. These variations namely depend on three operating conditions (fouling rate, BW and CIP efficiencies) and thresholds set by the quality of the input water.

Table 2 summarizes the main operating conditions used in this work and which are valid for both modules. These parameters and data are explained in (Prézélus et al., 2021, Table 2). It is

considered that both membranes have similar performance: a previous work on membrane preparation (Prézélus et al., 2019) reported that physical properties (permeability, porosity structure) of the new membrane are similar to those of ultrafiltration membranes. However, the complete similarity between the two membranes is a strong assumption, and will be discussed in section 3.4. A high-quality input water (1 mg TOC L^{-1}, 1 NTU) is filtered at 100 L h^{-1} m^{-2} with hollow fibers having an initial permeability of 230 L h^{-1} m^{-2} bar^{-1}. This high quality warrants the low fouling rate (-7.9 10^{-3} m^{-1}) and high BW and CIP efficiencies (95% each).

Table 2 Module filtration: the main operating conditions (from Prézélus et al.(2021) – table 2)

Category	Operating condition	Value
Output flow rate and module characteristics	Output flow rate (m^3 day^{-1})	36,000
	Initial permeability (L h^{-1} m^2 bar^{-1})	230
Input water quality	Turbidity (NTU)	1
	Total Organic Content (mg L^{-1})	1
Filtration operating conditions	Flux (L h^{-1} m^{-2})	100
	Cycle duration (min)	60
Fouling rate and remediation efficiencies	Fouling rate (m^{-1})	-7.9 10^{-3}
	BW efficiency (%)	95
	CIP efficiency (%)	95
End-of-life conditions	Operating life (year)	15
	Final permeability (L h^{-1} m^2 bar^{-1})	100

2.5. End-of-life process

Based on industrial practices, incineration is considered as the end-of-life option for modules. Unlike for the module fabrication and filtration processes, module incineration has not been modelled since this process is not specific to membranes. Generic processes for incineration exist in common LCA databases.

2.6. Life Cycle Assessment (LCA)

2.6.1. Goal and scope

The objective of the LCA is to evaluate environmental performances of the entire life cycle of membrane modules. The relevance of solvent substitution can thereby be determined from an environmental point of view. The functional unit is 1 m^3 of ultrafiltered water.

The system boundaries encompass the entire life cycle of membrane modules: extraction of raw materials, fabrication, transportation, operation and incineration. Modules are fabricated, operated and incinerated in France. Raw materials either originate from France or, by default, from Europe. The construction of the concerned plants is taken into account, as well as the treatment of any waste.

2.6.2. Life cycle inventory (LCI)

The LCI for the foreground processes, i.e. membrane fabrication and filtration process, is calculated using the process models developed in (Prézélus et al., 2020a, 2020b, 2021), and applied for the present work to conventional and biosourced membranes.

Inventory data of background processes (*e.g.* energy and chemical production, waste treatment, transportation) in the life cycle system stem from ecoinvent 3.4 database used with the allocation method 'allocation at the point of substitution' in the background part of the life cycle system. This choice was made because of the use of multiple chemicals whose production is accompanied by other chemicals as valuable wastes/by-products.

Four chemicals involved in module fabrication are not present in ecoinvent database: adhesive hardener, methyl lactate, and CTA and CDA polymers. The adhesive hardener has been approximated to a generic organic chemical. The inventory of CTA fabrication has been calculated based on Manda et al.'s (2014) study and can be found in Prézélus et al.'s work (2020a) with the underlying assumptions. The inventory of CDA fabrication is derived from that of CTA by adapting the degree of substitution of acetate in the molecule (see supplementary document, Appendix C). The inventory of methyl lactate fabrication has been calculated based on the process description in the literature. The corresponding process flowsheet and assumptions are given in supplementary document Appendix A.

2.6.3. Life cycle impact assessment (LCIA)

Umberto NXT software was used for impact assessment with two internationally recognized evaluation methods: (1) IPCC's GWP100 for climate change (Global Warming Potential at 100 years) and (2) ReCiPe endpoint method with a hierarchist cultural perspective. The latter evaluates 17 impact categories, which can be grouped in 3 areas of protection (human health, ecosystem quality and resource scarcity). Category scores can further be aggregated into a unique score. ReCiPe endpoint method provides results in points (unidimensional) and considers climate change at the level of damages on human health and ecosystem health , thus the IPCC method was chosen for complement the results with the popular GWP100 expressed in kg $CO_2 - eq$.

3. Results and discussion

In this section, LCA comparative results between the conventional and biosourced systems are presented. First, the fabrication of methyl lactate is compared to that of NMP, on the basis of 1 kg solvent. Then, results for CTA and CDA polymer fabrication are compared on the basis of 1 kg of polymer. Conventional and biosourced membrane fabrication are investigated with results expressed for 1 m² of membrane. Finally, filtration operation is added in the life cycle model and the entire life cycle of these two systems is analysed for the production of 1 m³ of ultrafiltered water. The input data sources and the results obtained at each step are mentioned on Figure 1.

3.1. Fabrication of conventional versus non-reprotoxic biosourced solvent

NMP, the conventional petrochemical solvent used for membrane fabrication in this study, is known for its reprotoxic properties, hence the use of methyl lactate as a non-reprotoxic and biosourced substitute (ECHA). The inventory of the former exists in ecoinvent and is replicated in the supplementary document –Appendix A. It is based on average industry data. On the contrary, methyl lactate's inventory has been calculated from literature data (Miller et al., 2011; details in supplementary document Appendix A) specifically for this study (see Table 3). Separation (purification) processes account for 94.5% of the direct energy use, the remaining 5.5% being for lactic acid fermentation and esterification. The value for direct energy use (i.e. 6.32 kWh, equivalent to 22.8 MJ kg⁻¹ methyl lactate) is in line with the 30.3 MJ-eq needed to produce 1 kg of polylactide, which is also obtained by lactic acid fermentation (Vink, 2007). Cumulated energy demand (total energy consumption over the life cycle of solvent fabrication) of 133 MJ-eq kg⁻¹ methyl lactate is in the range of cumulative energy demand calculated by Capello et al. (2007) for petrochemical solvents (between 40 and 270 MJ-eq kg⁻¹ solvent).

The GWP100 and ReCiPe endpoint scores for methyl lactate fabrication are respectively 32% and 10% less than for NMP fabrication (see Table 4). Obtained values of major ReCiPe endpoint impact categories such as "fossil depletion", "climate change for ecosystems" and "climate change for human health" (table 4) are indeed lower for methyl lactate production (36%, 32% and 32% lower respectively) but it should not be overlooked that impacts of 11 out of 17 categories are actually greater for methyl lactate fabrication (see Figure 2 and Appendix B). In particular, there is a 10-fold increase of impacts related to "agricultural land occupation", mainly accountable to sugar beet cultivation. Sugar is the main raw material for methyl lactate production. Contributions of cluster process inputs and waste to total environmental scores of both solvents' fabrication are detailed in Appendix B.

This first comparison highlights that reprotoxicity is not the only criteria that can discriminate two solvents. Reprotoxicity is of course a major health concern, especially for workers who are exposed to the concerned solvents, and its regulatory recognition at European level through REACH is certainly

a driving force in research efforts on solvent substitution in the field of membrane fabrication. However, other criteria can also be put forward to prefer methyl lactate over NMP, albeit non-regulatory ones for the moment. In the context of global warming, the use of fossil-based materials and emissions of greenhouse gases are examples of topical criteria.

Table 3 Methyl lactate fabrication – Inventory

Process inputs and waste		Quantity (per kg ML)	Ecoinvent v3.4 unit process
Factory	Factory (unit)	$1.92\ 10^{-10}$	Market for chemical factory, organics [GLO]
Sugar	Sugar (kg)	0.865	Market for sugar, from sugar beet [GLO]
Yeast	Yeast extract (kg)	0.109	Yeast paste, from whey, at fermentation [CH]
Calcium hydroxide	Calcium hydroxide (kg)	0.356	Market for lime, hydrated, loose weight [RoW]
Methanol	Methanol (kg)	0.923	Market for methanol [GLO]
	Methanol recovered (kg)	0.616	-
Sulphuric acid	Sulphuric acid (kg)	0.502	Market for sulfuric acid [GLO]
Water	Water (kg)	8.00	Market group for tap water [RER]
	Deionised water (kg)	1.67	Market for water, deionised, from tap water, at user [Europe without Switzerland]
	Deionised water recovered (kg)	1.67	-
Membrane & cleaning agents	Titanium oxide (kg)	$6.05\ 10^{-5}$	Market for titanium dioxide [RER]
	Water (kg)	1.16	Market group for tap water [RER]
	Sodium hydroxide (kg)	$2.90\ 10^{-3}$	Market for sodium hydroxide, without water, in 50% solution state [GLO]
Activated carbon & regeneration	Activated carbon (kg)	$6.28\ 10^{-5}$	Market for activated carbon, granular [GLO]
	Regenerated activated carbon (kg)	$9.30\ 10^{-5}$	Treatment of spent activated carbon, granular from hard coal, reactivation [RER]
Resin, conditioning & regeneration	Resin (kg)	$4.00\ 10^{-4}$	Anionic resin production [RoW]
	Sulphuric acid (kg)	2.90	Market for sulfuric acid [GLO]
	Sodium hydroxide (kg)	$1.44\ 10^{-4}$	Market for sodium hydroxide, without water, in 50% solution state [GLO]
	Deionised water (kg)	29.5	Market for water, deionised, from tap water, at user [Europe without Switzerland]
Waste	Liquid waste (m^3)	$4.21\ 10^{-5}$	Treatment of wastewater, average, capacity 4.7E10 l/year [CH]
	Calcium sulphate waste (kg)	0.654	Treatment of waste gypsum, sanitary landfill [Europe without Switzerland]

	Activated carbon waste (kg)	$6.28\ 10^{-5}$	Treatment of inert waste, sanitary landfill [Europe without Switzerland]
	Titanium oxide waste (kg)	$6.04\ 10^{-5}$	Treatment of inert waste, sanitary landfill [Europe without Switzerland]
	Resin waste (kg)	$4.00\ 10^{-4}$	Market for spent anion exchange resin from potable water production [GLO]
Electricity	Electricity (kWh)	6.32	Market group for electricity, medium voltage [RER]

Table 4 NMP vs methyl lactate – Environmental impact scores

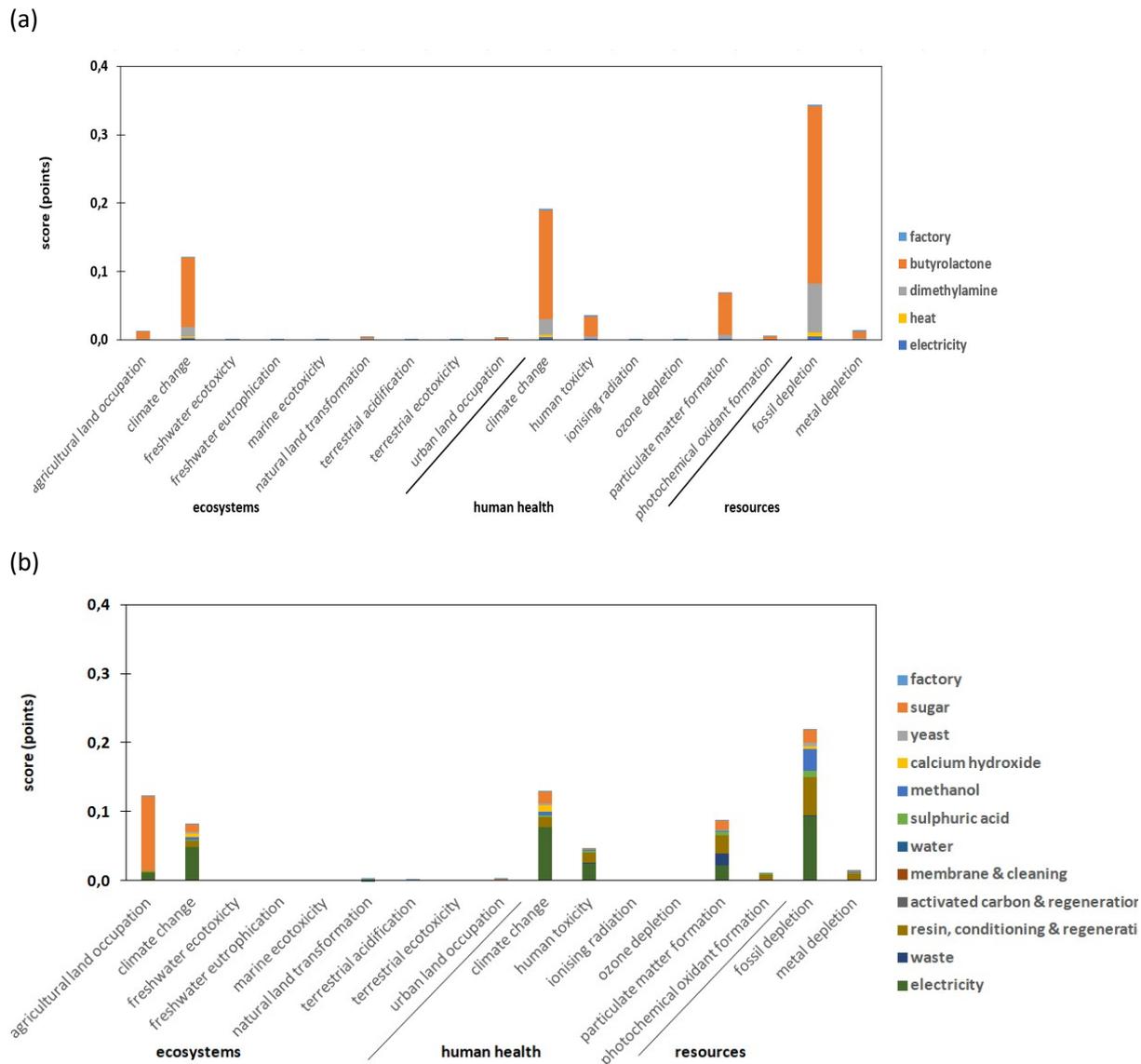| Evaluation method	Score (per kg solvent)		Percentage change (%) compared to NMP
	NMP	Methyl lactate	
GWP100 (kg CO$_2$-eq)	**6.9**	**4.7**	**-32**
ReCiPe endpoint (pts)	**0.80**	**0.72**	**-10**
Ecosystem quality (pts)	0.14	0.21	51
Human health (pts)	0.30	0.28	-9
Resources (pts)	0.36	0.24	-35

(a)

(b)

Figure 2 Contribution of cluster process inputs and waste treatment to ReCiPe endpoint categories for (a) NMP and (b) methyl lactate fabrication (per 1 kg solvent)

3.2. Fabrication of conventional versus alternative polymer

As a reminder, conventional membranes considered here are obtained by first dissolving CTA in NMP, whereas CDA and methyl lactate are used for biosourced membranes due to limitations of methyl lactate to dissolve CTA (as described in previous work (Prézélus et al., 2019)).

Before looking into membrane fabrication, the respective polymers need to be considered. Both CTA and CDA are prepared from pre-treated cellulose by acetylation with acetic anhydride. Quenching of

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the reaction is carried out by adding water. More water is added to obtain the desired degree of substitution of hydroxyl groups (2.8 for CTA and 2.4 for CDA). In both cases, cellulose acetate is precipitated in powder form with an acetic acid solution, washed and filtered from acids and salts, centrifuges and dried to further reduce water content. The inventories obtained by process modelling and the corresponding environmental scores of CTA and CDA fabrication are detailed in Appendix C. Slight differences in consumptions and waste production between CTA and CDA fabrication exist (in the range 6.0-37.4%). This can firstly be explained by the difference in respective polymer molecular weight: for a given input cellulose quantity, the CTA mass produced is 6.0% greater than for CDA. The remaining difference comes from input consumptions of acetic acid, water and sulphuric acid that are adapted during hydrolysis to the desired degree of substitution. Environmental impacts globally increase for the two evaluation methods for the fabrication of CDA by 6.5-6.6% (see Table C2). This increase is in line with the increase in consumptions and waste observed in Table C1. The higher percentage change of waste, sulphuric acid and sodium acetate consumptions (15.6-37.4%) compared to other inputs (6.0-9.2%) does not significantly affect environmental scores since, as for CTA fabrication, their respective contributions remain below 1%. Note should indeed be taken that the relative contributions of process inputs and waste to environmental scores of CTA and CDA fabrication are identical (maximum 1% change).

3.3. Fabrication of conventional versus biosourced membrane

The differences between conventional and biosourced membranes do not limit themselves to a solvent substitution and a polymer adaptation with all other parameters being held constant. Operating conditions such as the composition of the polymer solution and spinning temperatures vary according to choices of solvent and polymer (see Table 1). As a result, different inventories for the fabrication of the two membranes are obtained (see Table 5). Changes in mass compositions of polymer solution mechanically induce changes in additive and polymer consumptions: respectively 56% increase and 22% decrease. Methyl lactate consumption is 18% greater than NMP consumption, which is not entirely accounted for by its use as bore fluid since this represents only 7% of its total consumption. The two other main reasons are the greater solvent mass percentage in the polymer solution (5% greater) and the greater biosourced polymer solution's density (4% greater). Glycerol consumption remains unchanged (0.1%). It is not used as a bore fluid in biosourced membrane fabrication but this only represented 2% of its total consumption. This decline is compensated by the greater quantity needed during pre-conditioning to fill pores of biosourced membranes, which are more porous than

conventional membranes. Increasing mixing and spinning temperatures to 80°C leads to a 29% increase in natural gas consumption. On the other hand, no change in consumptions is observed for module components other than the hollow fibers (i.e. membrane housing, auxiliary equipment, adhesive, conditioning liquid).

Table 5 Inventories for module fabrication for biosourced and conventional membranes

Category	Cluster	Process inputs and waste	Quantity (per m² membrane)		Percentage change (%) compared to conventional membrane
			Conventional	Biosourced	
Membrane fabrication	**Lithium chloride**	Lithium chloride (kg)	0.0151	0.0235	56
	Glycerol	Glycerol (kg)	0.780	0.781	0.1
	Sodium bisulfite	Sodium bisulfite (kg)	0.0818	0.0818	0.0
	Water	Tap water (kg)	117	119	2.3
		Softened water (kg)	26.6	27.1	2.2
	Adhesives	Epoxy resin (kg)	0.127	0.127	0.0
		Hardener (kg)	0.0677	0.0677	0.0
		Calcium carbonate (kg)	0.0428	0.0428	0.0
	Module components	Glass fibre (kg)	0.255	0.255	0.0
		Polyethylene (kg)	0.0391	0.391	0.0
		Polyvinylchloride (kg)	$3.64\ 10^{-4}$	$3.64\ 10^{-4}$	0.0
		Noryl (kg)	$6.36\ 10^{-3}$	$6.36\ 10^{-3}$	0.0
	Waste	Liquid waste (m³)	0.143	0.147	2.3
		Solid waste (kg)	0.287	0.284	-0.9
	Electricity	Electricity (kWh)	3.35	3.40	1.6
	Natural gas	Natural gas (m³)	0.166	0.212	29
	Factory	Factory (unit)[a]	$7.02\ 10^{-14}$	$5.46\ 10^{-14}$	-22
Solvent fabrication	-	NMP or methyl lactate (kg)	0.588	0.691	18
Polymer fabrication	-	CTA or CDA (kg)	0.151	0.117	-22

[a] Both membrane fabrication factories are approximated to a chemical factory as indexed in the ecoinvent database. The two inventory values are proportional to the membrane's mass (not to the surface), hence their difference.

Comparing environmental impacts of conventional and biosourced membrane fabrication produces mixed results. As shown in Table 6, biosourced membrane fabrication gives lower GWP100 scores (-11%). The larger part (89-99%) of these percentage changes are due to variations of impacts related to solvent fabrication. The same trend in evaluation scores has actually been observed for the fabrication of 1 kg of the two solvents with a 32% decrease in GWP100 for methyl lactate. Despite the 18% increase in solvent consumption in biosourced membrane fabrication, the GWP100 score remains lower than that of conventional membrane fabrication; a 52% increase of solvent consumption would be necessary to give identical GWP100 scores between the two fabrication processes. Similar total ReCiPe endpoint scores (-0.5% percentage change) are obtained between the two fabrication processes, although discrepancies exist between the three endpoint categories (5.7, -0.5 and -13% respectively for ecosystem quality, human health and resources).

Table 6 Biosourced vs conventional membrane fabrication – Environmental impact scores

Evaluation method	Score (per m² membrane)		Percentage change (%) compared to conventional membrane
	Conventional membrane	Biosourced membrane	
GWP100 (kg CO$_2$-eq)	11	9.4	-11
ReCiPe endpoint (pts)	1.9	1.9	-0.5
Ecosystem quality (pts)	0.97	1.0	5.7
Human health (pts)	0.47	0.47	-0.5
Resources (pts)	0.47	0.41	-13

Further discrepancies appear between ReCiPe endpoint categories. As can be seen in Figure , there is an almost complete burden shift from "fossil depletion" and "climate change for human health" to "agricultural land occupation" and "particulate matter formation"; the increase of the latter two categories equals 95% of the decrease of the former two categories. The lower scores of "fossil depletion" and "climate change for human health" for biosourced membrane fabrication are entirely due to lower impacts of solvent fabrication; greater impacts of membrane fabrication is indeed offset by lower impacts of polymer fabrication (see Figure 4 (a) and (b)). This is true to a lesser extent for "agricultural land occupation" and "particulate matter formation"; solvent fabrication contributes to 95 and 86% respectively to the overall increase of the two categories' scores (see Figure 4 (c) and (d)).

Impacts of membrane fabrication only (excluding solvent and polymer fabrication) are indeed greater for biosourced membrane fabrication; percentage changes of four main ReCiPe endpoint categories vary between 1 and 11%. The greatest contributor to these increases is by far bore liquid and preconditioning liquid consumption (61 to 99%), which is accounted for by the increase in glycerol consumption during preconditioning due to greater membrane porosity. The increase in natural gas consumption for biosourced membranes, due to higher temperatures during polymer solution preparation, contributes to one third of the increase in fossil depletion. The increase in lithium chloride consumption due to a greater mass percentage in the polymer solution (from 2 to 3 wt%) impacts on climate change, agricultural land occupation and particulate matter formation with varying degrees from 1 to 12%.
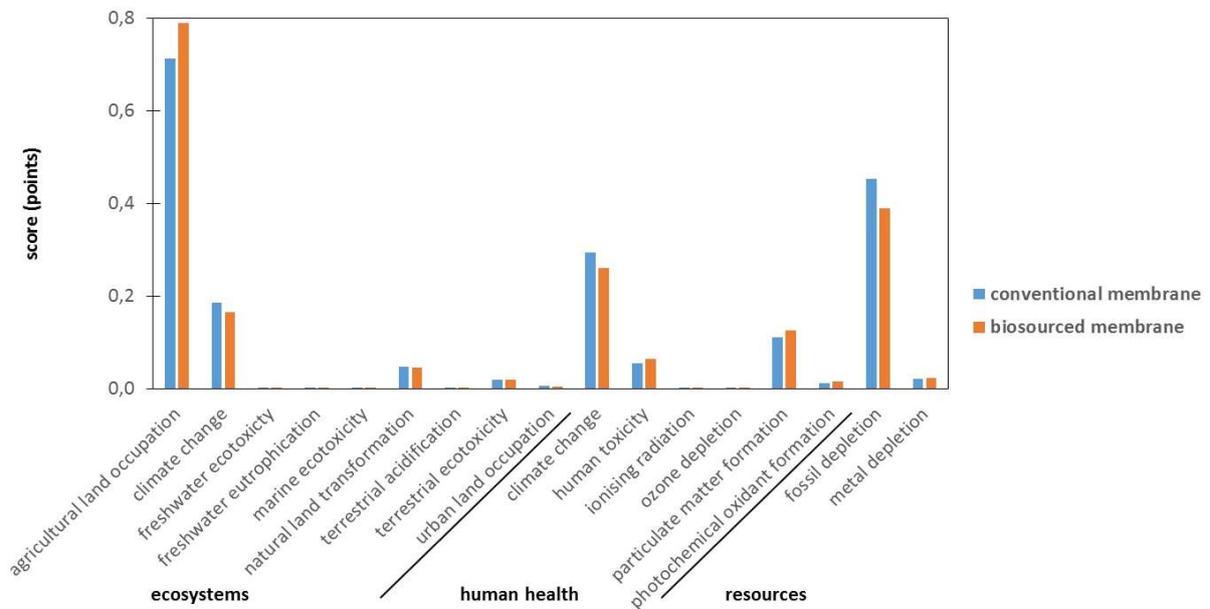

Figure 3 Biosourced vs conventional membrane fabrication – ReCiPe endpoint results (per 1 m^2 membrane)

(a)

(b)

(c)

(d)

polymer fabrication
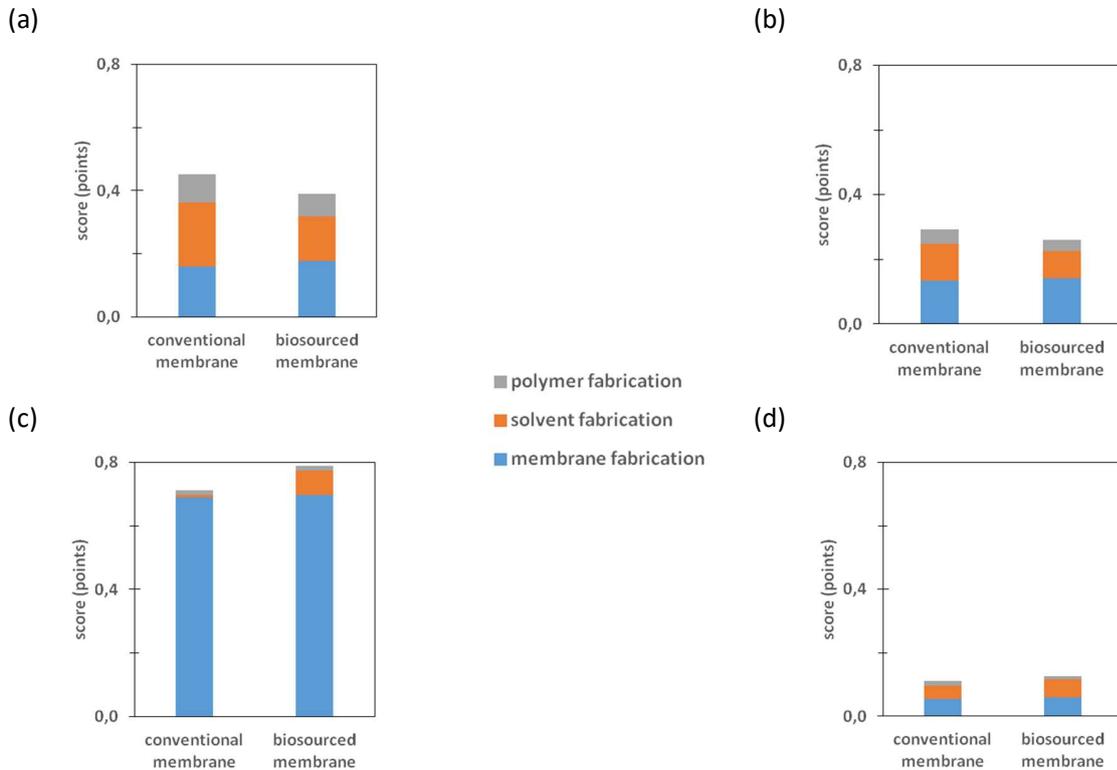solvent fabrication
membrane fabrication

Figure 4 Biosourced vs conventional membrane fabrication – Contribution of categorized process inputs and waste to ReCiPe endpoint category (a) fossil depletion, (b) climate change for human health, (c) agricultural land occupation for ecosystems, and (d) particulate matter formation for human health (results per 1 m^2 membrane)

These results hold for the specific conditions of this case study, in particular as regards choices made of solvent, polymer and geographical context. Here, the relevance of solvent substitution for environmental mitigation has shown to be moderate (up to 11% fall in impacts) but it should be stressed that the environmental evaluation should be updated for every new case. The production route of each solvent is for example unique, notably in terms of number of reactions and separation processes involved. Capello et al. (2007) observed that the fabrication of solvents derived from petrochemical basic products in few process steps (e.g. methanol from natural gas, or hexane and heptane from naphta) have a lower cumulative energy demand compared to more elaborate solvents (*e.g.* tetrahydrofuran, cyclohexanone). Lower environmental impacts may thus be obtained by substituting elaborate solvents by more basic ones, independently of their origin (petrochemical or biosourced). This room for

manoeuver is also made possible by the relatively weak contribution (6-15%) of polymer fabrication for both conventional and biosourced membranes. This is true for CTA and CDA, as well as for another polymer commonly used, i.e. polysulfone, of which the fabrication inventory is present in ecoinvent. Under the hypothesis that polysulfone substitutes CTA and CDA with all other operating conditions kept constant, the solvent substitution of NMP to methyl lactate gives similar results as for this case study: 1.5 and 12% decrease in GWP100 and ReCiPe endpoint scores, respectively, compared to 0.5 and 11% for cellulose acetate polymers. Along the same line, industrial practices (e.g. nature of the pre-conditioning liquid) and the geographical context (e.g. electricity mix) influence the contributions of other process inputs and waste. If the pre-conditioning liquid were for example obtained from waste cooking oil instead of conventionally from oil seed crops for both conventional and biosourced membranes, a decrease in GWP100 and ReCiPe endpoint scores of 3 and 15% would be observed (instead of 0.5 and 11%).

3.4. Life cycle of conventional versus biosourced module (fabrication – filtration – end of life)

It is interesting to ponder on the relevance of solvent substitution from a life cycle perspective, that is to say by taking into account all stages: fabrication, operation and end of life. Given that filtration performances of the biosourced membrane with real water are unknown, the same as those of the conventional membrane are taken. This is a reasonable assumption since both membranes are designed for ultrafiltration and, as such, have permeability and selectivity in the same order of magnitude. Furthermore, similar fouling behaviour can be assumed due to the chemical similarity of their cellulose acetate material.

Simulations performed with the model presented in Prézélus et al. (2021), allowed to obtain the energy and chemical consumption for the filtration stage, expressed for $1m^3$ of produced water: electricity 0.034 kWh m^{-3}, citric acid 2.9 10^{-6} kg m^{-3}, ammonia 5.2 10^{-7} kg m^{-3} and sodium hydroxide 6.0 10^{-7} kg m^{-3} (Appendix D, Table D1). This inventory was then used for LCIA step.

Figures 5 (a) and (b) show that there is no significant difference in GWP100 and ReCiPe endpoint scores between the life cycles of conventional and biosourced membranes: percentage differences are less than 0.001%. From a life cycle perspective, solvent substitution brings about no environmental mitigation; the operation stage predominates and changes in the fabrication stage (i.e. solvent, polymer solution composition, temperature…) have no significant influence on environmental scores. Regardless of the considered membrane or evaluation method, membrane operation indeed contributes to over 99.98% of scores related to the membrane's life cycle. Contributions of process inputs and waste at the

level of the life cycle are thus also identical to those at the level of the membrane operation stage (see Prézélus et al. (2021) for these contributions).
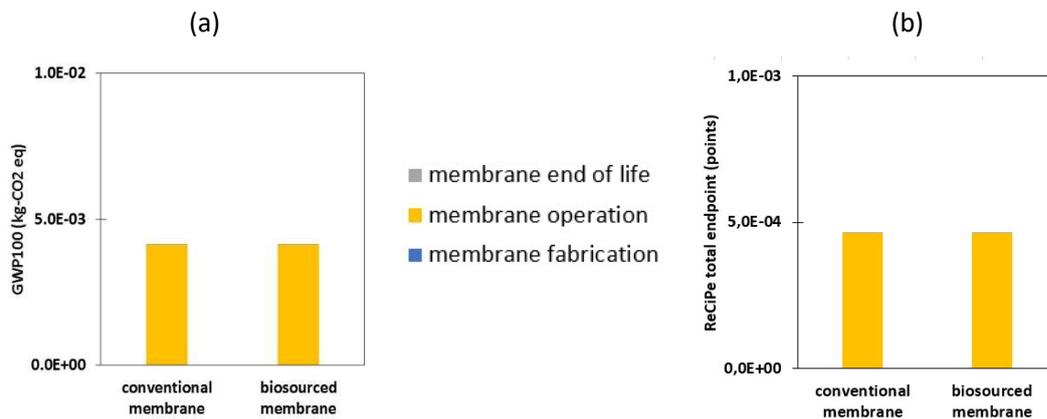

Figure 5 Biosourced vs conventional membrane life cycle results for (a) GWP100 and (b) ReCiPe total endpoint scores (for 1m^3 ultrafiltered water produced)

The predominance of the operation stage over the fabrication and end-of-life stages is logical given the lifetime of several years of UF membranes for potable water production. In particular, relative GWP100 scores between the operation and fabrication stages equal 13,919 and 15,696 for conventional and biosourced-solvent membranes, respectively (see Figure 6 (a) and (b)). These ratios equal 8,663 and 8,704 for ReCiPe endpoint scores. When considering specific ReCiPe impact categories, "terrestrial ecotoxicity" and "agricultural land occupation" are the only categories with ratios below 1,000 (257 and 256 for the former, 637 and 609 for the latter). GWP100 and ReCiPe (for all impact categories) scores are all above 13,000 when comparing the operation to the end-of-life stage. All ratios between stages can be found in Appendix D. It can be concluded that the contributions of fabrication and end-of-life stages to GWP100 and ReCiPe impact categories are negligible compared to the operation stage.
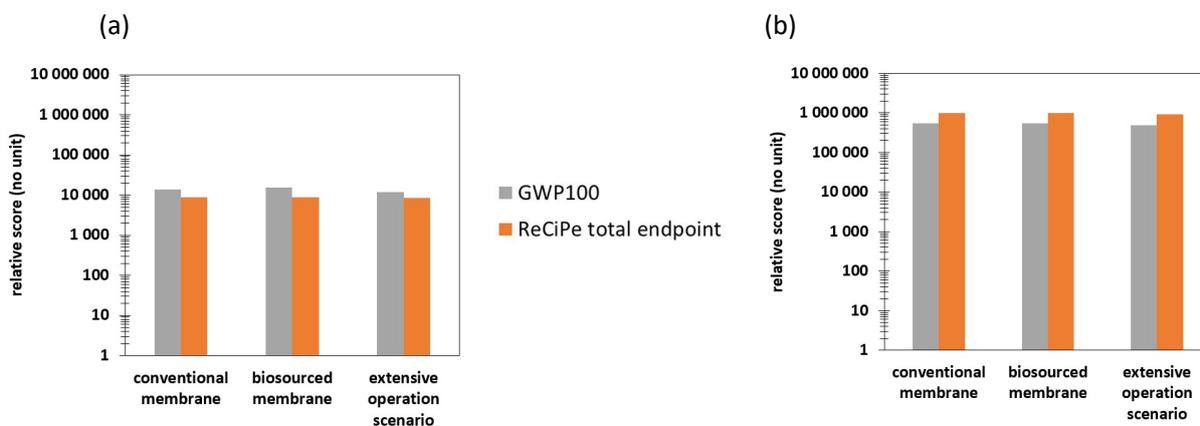
Figure 6 Relative environmental scores of the membrane operation stage to (a) membrane fabrication and (b) membrane end of life

An extensive operation scenario can be derived from the conventional membrane scenario to analyse the sensitivity of the results, especially to validate the dominance of the operation stage against the very low contribution of fabrication and end of life stages. In other words, the extensive operation scenario is defined as an environmentally favourable situation for membrane operation (i.e. minimal impact) and unfavourable situations for both membrane fabrication and end of life (i.e. maximum impact). Here, a realistic and industrially applicable scenario has been chosen. Membrane modules are assumed to be fabricated in Western India, since the change in electricity mix (i.e. 82% from hard coal) has shown to increase GWP100 and ReCiPe endpoint scores by 50% and 26%, respectively, compared to the nuclear-based French context (Prézélus et al., 2020a). The end of life by incineration of the conventional membrane scenario is kept unchanged because it is already an unfavourable situation. Indeed, incinerating solid waste is more impactful than disposing it in a sanitary landfill or recycling (ecoinvent v3.4). As for membrane operation, a previous study has shown that environmental impacts are highly sensitive to filtration flux (Prézélus et al., 2021): lower filtration fluxes mitigates the impacts. To minimize environmental impacts in the extensive operation scenario, a 30% decrease in filtration flux is taken (i.e. 70 instead of 100 L h^{-1} m^{-2}), which remains realistic at an industrial level. The filtration duration is risen accordingly from 60 to 86 min to maintain constant the deposited mass during a filtration cycle. In other words, constant deposited mass means the product "filtration duration" times "filtration flux" remains constant. The lower filtration flux also directly implies a greater membrane

surface needed, whence the name of "extensive operation". The parameters that differ between the conventional membrane and extensive operation scenarios are summarized in Table 8.

Table 8 Differences between the reference and extensive operation scenario

Stage	Operating condition	Conventional membrane	Extensive operation scenario
Fabrication	Geographical electricity mix	France	Western India
	Geographical waste treatment	France	Rest of world
Filtration	Filtration flux (L h^{-1} m^{-2})	100	70
	Filtration duration (min)	60	86
	Number of modules (-)	252	352

As shown in Figure 6 (a) and (b), relative environmental scores of the extensive operation scenario do not significantly change compared to that of the conventional membrane scenario. Relative scores between the operation and fabrication stages change from 13,919 to 11,677 for GWP100, and from 8,663 to 8,591 for ReCiPe endpoint. When comparing the operation to the end-of-life stage, ratios are always greater than 12,000 (instead of 13,000 for a conventional membrane). Other relative scores between stages can be found in Appendix D. To summarize, membrane operation in the extensive operation scenario still contributes to over 99.98% of scores related to the membrane's life cycle.

For a deeper analysis, a hypothetical scenario is considered for which cumulative environmental impacts of the fabrication and end of life equal those of the operation stage. It is assumed that conditions are the same as those for the conventional membrane scenario (see Table 1 and Table 2). Simulations shown that the operation stage would contribute to 50% of life cycle scores if the membrane life span is between 9 and 16 hours (instead of 15 years in conventional membrane scenario). Such a short operating life rather corresponds to single-use membranes, such as used as artificial lungs (i.e. oxygenators) or artificial kidneys (i.e. hemodialyzers). Beyond environmental considerations, replacing membranes as often is an economic and logistical burden; membranes have an economical cost and maintenance time would need to be allocated to changing membranes. From these different perspectives, such disposable membranes are undesirable unless the application justifies it.

Finally, the simulated scenarios show that UF membranes with slightly different performances will not significantly change the share of impacts between fabrication and operation stages. Supposing the new

membrane has lower filtration flux (lower performance), then a higher surface will be necessary to achieve the same productivity. In this case, the system is actually represented by the "extensive operation scenario" with the results explained above.

3.5. Putting into perspective

The ensemble of the presented results puts forth that the operation stage will remain the hotspot for further environmental mitigation of UF membranes' life cycle, in particular when considering fabrication by NIPS, dead-end ultrafiltration and incineration as the end-of-life option. It can be argued that the relatively limited environmental footprint of the fabrication stage gives a significant room for manoeuvre for innovation in this field. The fabrication stage may indeed be considered as a lever to prepare high-performance membranes. Research endeavours in membrane material or phase inversion, for example, seek to act upon membrane performances crucial during the operation stage (i.e. permeability, fouling propensity…). Obtaining novel membranes that are both highly permeable and low-fouling would allow to limit energy consumption due to rising transmembrane pressure and control chemical consumption for permeability remediation. However, ongoing research efforts face technical challenges linked to the complexity of phase inversion and the difficulty of modelling the sensitivity of membrane performances to fabrication operating conditions (Amirilargani et al., 2010; Saljoughi and Mohammadi, 2009). Implementing innovation and research in such a context is a tedious and long-term task with uncertain outcomes.

On the contrary, mitigation strategies identified for UF membrane operation are both technically feasible and quickly deployable (Prézélus et al., 2021). Given that the two main contributors are electricity and BW chemical production, one strategy is to adjust BW operating conditions (i.e. concentration, injection time) to limit chemical overconsumption due to precautionary measures (Vroman et al., 2021). Another strategy is to operate in an extensive manner at lower filtration fluxes, as opposed to an intensive operation at high filtration fluxes. Besides lowering average transmembrane pressure and thus electrical consumption, such an extensive operation leads to greater membrane surface being needed to produce the daily water flowrate. A fall in operational expenditure due to energy use would supposedly be accompanied by additional capital expenditure due to both the cost of membranes and the physical footprint of the UF unit. This holds for new units as well as for existing ones that should potentially be expanded. Despite these legitimate concerns, it should be kept in mind that adjusting both BW and filtration operating conditions has one major asset compared to innovating in membrane fabrication: immediate environmental mitigation. While awaiting the development of novel

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membranes, improving the environmental performance of existing and operational membranes can already give appreciable outcomes.

As demonstrated in this work, the so named "green membranes", obtained from biosourced raw materials, are not as green as expected compared to conventional UF membranes, when their whole life cycle (fabrication, utilization and end-of-life) is considered. In the field of UF for water treatment applications, the effort to replace conventional solvents is of little interest from this point of view. The NMP's toxicity is an immediate issue at the factory level, but a green solvent can also have hazardous properties. For instance, the flammability of methyl-lactate is higher than that of NMP. On the other hand, the change of solvent implies the adaptation of the whole manufacturing process to obtain high performance membranes, and requires additional investments.

Finally, switching to biobased raw materials in UF membrane fabrication is a multicriteria decision issue, including technical performance, on-site risk assessment, and LCA global impacts, as well as economics.

4. Conclusion

This work has sought to ponder on the environmental relevance of solvent substitution in membrane fabrication by phase inversion, thereby going beyond membrane science considerations only. Although the fabrication of the studied non-reprotoxic biosourced solvent (methyl lactate) shows lower total environmental scores than the fabrication of the studied conventional solvent (NMP), scores are similar between the fabrication of conventional and biosourced membranes. When considering the entire life cycle of these cellulose acetate membranes, the fabrication stage does not have any significant contribution to environmental scores, as opposed to the operation stage. Environmental hotspots lies not so much in the solvent used during membrane fabrication as in the choice of operating conditions on a drinking water plant.

These results highlight the relevance of coupling process modelling and LCA as an evaluation tool to clearly identify the hotspots on technical proposals. It becomes even more relevant when evaluating operating conditions as variables. The process modelling–LCA approach as applied in this work reveals that innovation in membrane fabrication (high permeability, high separation efficiency) could have a potential to bring about environmental mitigation, which is of greater certainty as regards the operation stage on a drinking water plant. This work, which builds on previous developments (the modelling tools developed are available on demand), can be continued by exploring other membrane materials as well

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as analysing the economic implications of extensive membrane operation at low filtration fluxes as compared to intensive operation.

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