



Assessment of the environmental impact of polymeric membrane production

Pooja Yadav^{a,1}, Norafiqah Ismail^{b,1}, Mohamed Essalhi^b, Mats Tysklind^b,
Dimitris Athanassiadis^{a,**}, Naser Tavajohi^{b,*}

^a Department of Forest Biomaterials and Technology, Swedish University of Agricultural Sciences, SE-90183, Umeå, Sweden

^b Department of Chemistry, Umeå University, SE-90187, Umeå, Sweden

ARTICLE INFO

Keywords:

Polymeric membranes
Hollow fiber Membrane
Life cycle assessment
Environmental impact
Environmental cost
Sustainability

ABSTRACT

Polymeric membranes are important in advanced separation technologies because of their high efficiency and low environmental impact. However, procedures for membrane production are far from sustainable and environmentally friendly. This work presents a life cycle assessment of the environmental impact of fabricating 1000 m² of hollow fiber polymeric membranes. Membrane materials considered include the most popular fossil- and bio-based polymers in current use, i.e., polysulfones, polyvinylidene fluoride, and cellulose acetate. Solvents considered for use in polymer dope solution included polar aprotic solvents (*N*-Methyl-2-pyrrolidone, *N*, *N*-dimethylacetamide, and dimethylformamide) that are widely used in industry and an alternative green solvent (ethylene carbonate). The impacts of membrane production on global warming, marine ecotoxicity, human carcinogenic and non-carcinogenic toxicity, land use potential, and fossil resource scarcity were analyzed. Additionally, the impact on the sustainability and environmental cost of membrane production resulting from replacing fossil-based polymers with bio-based polymers or substituting toxic solvents with a green alternative was investigated. Hot spots in the membrane production process were identified, and measures to reduce the environmental impact of membrane production were proposed.

1. Introduction

Membranes are semipermeable barriers that control the transport of substances between two adjunct phases. As such, membranes play key roles in many advanced separation technologies. The earliest studies on membranes were conducted with natural membranes such as pig bladders, animal guts, or rubber bands. However, the golden age of membrane technology began with the invention of reverse osmosis by Reid and Breton in 1959 [1], and the development of the asymmetric cellulose acetate (CA) membrane by Loeb and Sourirajan in 1962 [2,3]. Many new applications of membranes were subsequently developed; they now find use in molecular separation methods (including reverse osmosis, nanofiltration, ultrafiltration, and microfiltration) [4,5], promoting chemical transformations (in catalytic membranes [6,7], membrane reactors [8,9], and membrane bioreactors [10,11]), energy storage [12], and green and blue energy production [13–18]. Membranes are also important in various medical and biological applications including

dialysis, drug release, and cell culture [19]. The rapid progress of membrane science and technology can be attributed to the simplicity, low energy consumption, ease of control and scale-up, flexibility, and environmental friendliness of many membrane-based technologies [20].

Modern membranes are typically prepared from organic and/or inorganic materials, and may have different configurations (notably, flat sheets or hollow fibers). Polymeric membranes have attracted particular industrial interest due to their comparatively small footprints, cost-effectiveness and ease of production [21]. Of the known methods for polymeric membrane fabrication (which include sintering, track-etching, stretching, and phase inversion), phase inversion is the most widely used in both industry and academia [22].

The first step in phase inversion is to prepare a homogeneous polymer solution by dissolving the polymer in a suitable solvent. Phase inversion is then induced using an external or internal factor. External factor methods include nonsolvent-induced precipitation, precipitation from the vapor phase, and thermally-induced phase separation. The main internal factor method is evaporation-induced phase inversion.

* Corresponding author.

** Corresponding author.

E-mail addresses: Dimitris.Athanassiadis@slu.se (D. Athanassiadis), naser.tavajohi@outlook.com (N. Tavajohi).

¹ Contributed equally.

Nomenclature

CA	Cellulose acetate
CV	Coefficient of variation
DMAC	Dimethylacetamide
DMF	Dimethylformamide
DMSO	Dimethyl sulfoxide
EC	Ethylene carbonate
FRSP	Fossil resource scarcity
FU	Functional unit
GLO	Global electricity mix
GWP	Global warming potential
HCTP	Human carcinogenic toxicity potential
HNCTP	Human non-carcinogenic toxicity potential

IRP	Ionizing radiation potential
LCA	Life cycle assessment
LCI	Life cycle inventory
LUP	Land use potential
MEP	Marine ecotoxicity potential
NIPs	Non-solvent induced phase separation
NMP	N-methyl-2-pyrrolidone
PSF	Polysulfone
PVDF	Polyvinylidene fluoride
RER	European electricity mix
TEP	Triethyl phosphate
TEGDA	Triethylene glycol diacetate
THF	Tetrahydrofuran

Industrial-scale production of flat sheet and hollow fiber membranes mainly relies on nonsolvent-induced or thermal phase separation [23].

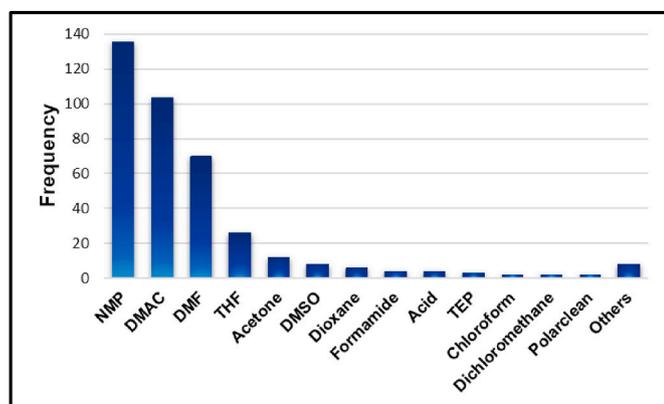
Fig. 1 lists polymers and solvents that are commonly used to fabricate membranes by phase inversion. Unfortunately, most of the solvents used in membrane fabrication are highly toxic and their industrial use is strictly regulated. For instance, in May 2020 the European Union restricted the use of *N*-Methyl-2-pyrrolidone (NMP), which is the most popular solvent for membrane fabrication [24]. The severity of this problem is amplified by the way in which solvent wastes from membrane production are currently managed. The majority of the used

solvent is released during the phase inversion process in the coagulation/quenching bath. Residual solvents are then extracted in the washing bath (known as external coagulation bath) due to their adverse impact on membrane performance. Consequently, membrane fabrication is estimated to cause the release of over 50 billion liters of contaminated water annually [25]. According to a survey, around 70% of industrial membrane fabricators flush generated waste without purification or dilute wastewater with excess water to bring the solvent concentration below the legally mandated upper limit [25].

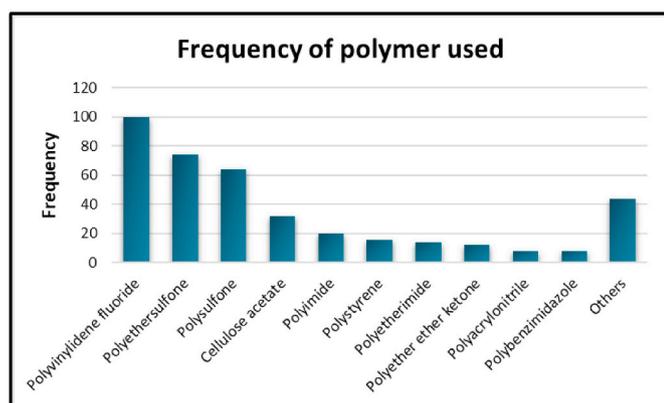
In the last few years, several so-called green solvents, i.e. a solvent that is environmentally friendly or biosolvent, have been used for membrane fabrication. Examples include tributyl *O*-acetyl citrate [27], triethylene glycol diacetate (TEGDA) [28], Methyl-5-(dimethylamino)-2-methyl-5-oxopentanoate (Rhodiasolv[®] PolarClean) [29], Cyrene[™] [30], and organic carbonates [31]. In 2014, Figoli et al. [32] reviewed the use of green solvents in membrane fabrication. However, one issue that has not been addressed to date is the environmental impact of solvent production. If a green solvent is prepared in a very toxic manner, any effort invested into developing sustainable membrane production using that solvent may be wasted. Therefore, it is critical to consider the upstream emissions of the solvents used in membrane fabrication rather than the final product. Another important point that is rarely considered is the source of the polymers. While many studies on membrane fabrication have been based on natural polymers such as cellulose-based polymers, fossil-based polymers are widely used in industry due to their thermal, chemical, and mechanical stability. It is currently not known the effect of using such polymers (i.e., bio-based polymers) in membrane fabrication or how replacing fossil-based polymers with bio-based alternatives might affect the sustainability of membrane production.

To better assess sustainability in membrane production, it is critical to quantitatively evaluate the impact of the entire process from different perspectives, considering impacts on global warming, human carcinogenic toxicity, human non-carcinogenic toxicity, fossil resource scarcity, and marine ecotoxicity. Life cycle assessment (LCA) has emerged as a key framework for evaluating the environmental impact of products and manufacturing processes [33,34]. The LCA approach can be used to study product manufacturing processes with multiple interacting parts, and to obtain a better understanding of the potential environmental impacts of both the process and the final product.

The aim of this study was to clarify the environmental impact of polymeric membrane production under various conditions. Specifically, the impact of replacing toxic solvents such as *N*-Methyl-2-pyrrolidone (NMP), *N*, *N*-dimethylacetamide (DMAC), and dimethylformamide (DMF) with a green solvent (ethylene carbonate) was investigated, along with the impact of substituting popular fossil-based polymers such as PVDF and PSF with a bio-based alternative, in order to identify hotspots in the membrane fabrication process. Hollow fiber membranes are



(a)



(b)

Fig. 1. Frequencies of use for (a) solvents and (b) polymers in membrane preparation by NIPs from 2014 to 2019 [26]. *N*-Methyl-2-pyrrolidone (NMP), *N*, *N*-dimethylacetamide (DMAC), and dimethylformamide (DMF), Tetrahydrofuran (THF), Dimethyl sulfoxide (DMSO), Triethyl phosphate (TEP).

generally preferred to flat sheets because of their high packing density, self-standing nature, and spacer-free design. Therefore, the fabrication of hollow fiber membranes (1000 m²) was taken as the target process for the analysis. The reliability of the results obtained was demonstrated by an uncertainty analysis performed using the Monte Carlo analysis function of the Simapro 9 software package. Effects on all environmental impact categories due to hollow fiber membrane production were evaluated in detail and measures to increase the environmental friendliness of membrane production are suggested.

2. Methodology

The LCA study was carried out in accordance with the ISO 14040 standards, which state that an LCA should include four key elements: goal and scope definition, life cycle inventory analysis, environmental impact assessment, and interpretation of results [35,36].

2.1. Goal and scope definition

The study's goal was to assess the environmental impact of the hollow fiber membrane preparation. The production of 1000 m² of the hollow fiber membrane was taken as the functional unit (FU) of the analysis; all inputs (material and energy) and outputs (emissions) were evaluated on a per-FU basis. Two of the most widely used fossil-based polymers and one popular bio-based polymer were considered as membrane materials. Further, three polar aprotic solvents widely used in the membrane industry and one green solvent were considered for use in the production process (Fig. 2). In total, seven different polymer/solvent combinations were considered to evaluate the environmental impact resulting from the choice of solvent (green or toxic) and polymer (fossil- or bio-based), and the source of the energy used in the production process. The polymer/solvent systems considered here are shown in Fig. 2. Table S1 presents a list of published papers about hollow fiber membrane by using the investigated polymer/solvent systems in this study.

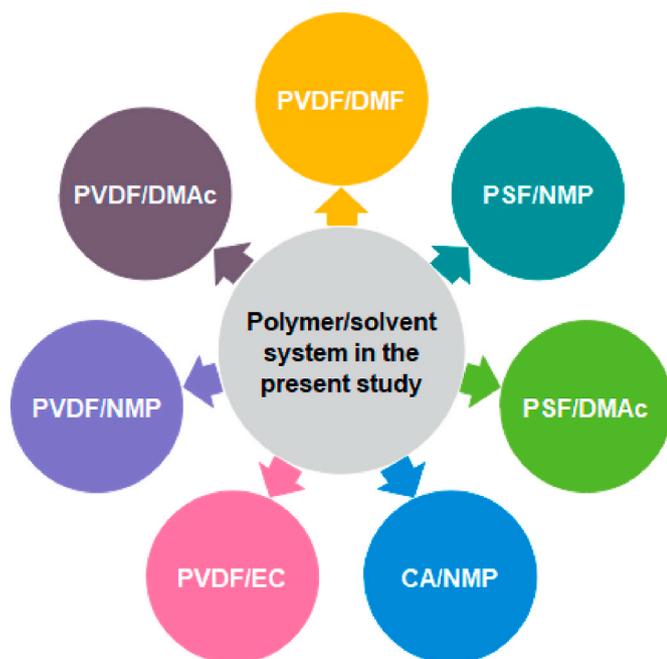


Fig. 2. Combinations of solvents (NMP, ethylene carbonate (EC), DMAc, or DMF) and polymers (PSF, polyvinylidene fluoride (PVDF), or CA) considered in this work.

2.1.1. System boundary

A "cradle to gate" analysis was performed, with the system boundary of the study shown in Fig. 3. Aspects of the membrane preparation process considered in the analysis included (i) the preparation of the dope solution by mixing the polymer and the solvent, and (ii) the hollow fiber membrane spinning process (Fig. 4). All of the relevant raw materials, energy, utilities (e.g., electricity and water), chemicals, and emissions involved at each stage were considered to be within the system boundary. Environmental impacts due to the production of machinery and the plant for hollow fiber membrane fabrication were not considered. The substitution of the traditional solvent with a green alternative was examined on a theoretical basis. It was assumed that all electricity used in the fabrication process was sourced from a grid whose generation mix was identical to Sweden's. Wastewater emissions were considered, and it was assumed that each plant had its own onsite closed-circuit wastewater treatment system.

2.2. Life-cycle inventory

Data for the life cycle inventory (LCI) were used on a per-FU basis. Primary data were collected from experimental trials conducted in Umeå University; other data were retrieved from the literature and the Ecoinvent 3.5 database.

To conduct this analysis, we assumed that: 1) the minimum required amount of nonsolvent is ten times more than the required solvent; 2) the nitrogen pressure is constant at 1 bar in the entire spinning; 3) for the upscaling of the membrane production process from 1 m² to 1000 m² all inputs and outputs were multiplied according to laboratory values except electricity; electricity consumption is lower in industrial level compared to laboratory level, and that is the motivation that electricity consumption was taken from Prézéus et al. [37] in which the authors have measured the industrial level electricity consumption of hollow fiber membrane production, 4) each membrane production plant have its own onsite closed-circuit wastewater treatment system, so wastewater emissions were not considered in the analysis and 5) Swedish mix electricity was used and the composition is shown in Table S2. Furthermore, the impact of additives was neglected in the analysis because of the wide variety of additives that may be used in membrane fabrication and their wide range of concentration in different polymer/solvent system. Table S3 shows all inputs adjusted for the production of 1000 m² membrane production.

2.2.1. Hollow fiber membranes preparation

Hollow fiber membranes were fabricated as follows. First, a homogeneous dope solution containing 20 wt% polymer and 80 wt% solvent

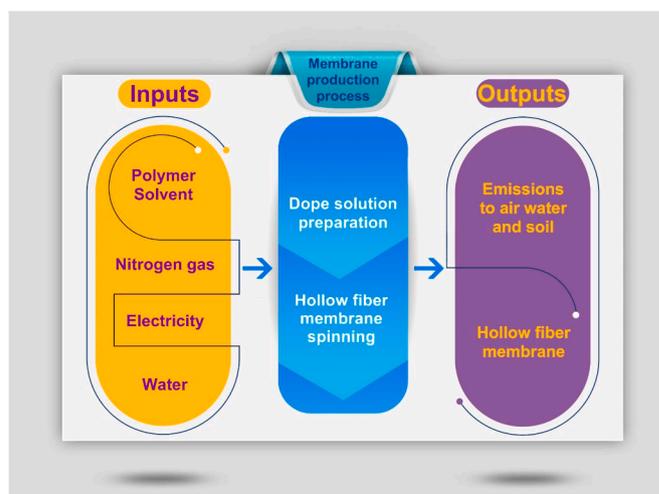


Fig. 3. System boundaries for hollow fiber membrane fabrication.

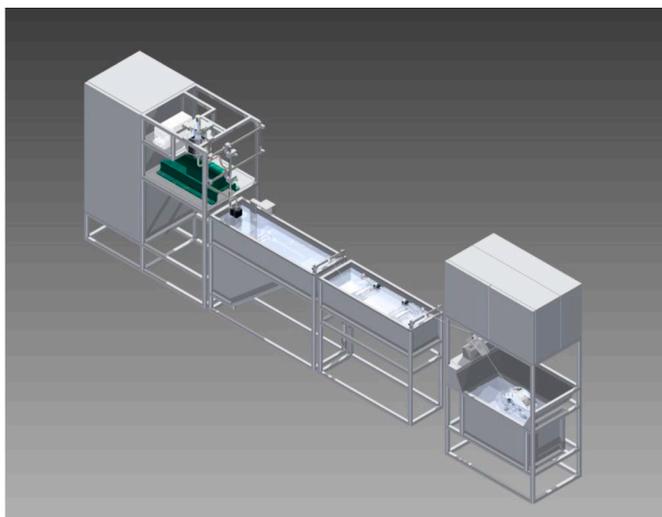


Fig. 4. Schematic depiction of a hollow fiber membrane fabrication system.

was prepared in the laboratory. After degassing, the solution was transferred to a reservoir for use in membrane fabrication. Nitrogen gas (1 bar) was used to purge the dope solution from the reservoir through a spinneret, and a gear pump was used to control its flow rate, which depended on the solution's viscosity. Water was used as a bore fluid, and was transported to the inner lumen of the spinneret to form the hollow fiber shape. When the solution entered the coagulation bath, solvent-nonsolvent (water) exchange occurred, resulting in phase inversion. The membrane began to solidify as water entered the solution and the solvent was extracted. The membrane remained immersed in water for at least 24 h to remove all residual solvent. The impact of additives was neglected in the analysis because of the wide variety of additives that may be used in membrane fabrication and their wide range of concentration in different polymer/solvent system. The results obtained during the laboratory-scale fabrication of hollow fiber membranes were then scaled up to estimate the process requirements for fabricating a membrane area of 1000 m² (which would require around 317,965 fibers each around 1 m long) with a thickness of 0.8 mm. The fabrication of such a membrane using the process outlined above would require around 3000 L of tap water, 80 kg of polymer (PSF, PVDF or CA), 320 kg of solvent (NMP, DMAc, DMF or EC), 0.561 L of nitrogen, 2832 kWh of electricity, and a temperature of 25–30 °C. For all scenarios, secondary data (such as production of PSF (Table S4), NMP (Table S5), DMF (Table S6), DMAc (Table S7) and EC (Table S8)) on the membrane preparation process were retrieved from the Ecoinvent 3.5 database. Input/output data for PVDF (Table S9) and CA (Table S10) production were taken from Abbasi [38] and Cerdas et al. [39], respectively.

2.3. Life cycle environmental impact assessment

The environmental impact assessment was conducted using the characterization factors specified for the ReCiPe 2016 (World-H) midpoint method [40]. The potential impact categories assessed were global warming potential (GWP; kg CO₂ eq.), ionizing radiation potential (IRP; kBq Co-60 eq.), marine ecotoxicity potential (MEP; kg 1,4-DCB eq.), human carcinogenic toxicity potential (HCTP; kg 1,4-DCB eq.), human non-carcinogenic toxicity potential (HNCTP; kg 1,4-DCB eq.), land use potential (LUP; m²a crop eq.), and fossil resource scarcity (FRSP; kg oil eq.).

2.4. Sensitivity analysis

The LCA results for the polymer/solvent systems were sensitive to the source of the electricity modeling parameter. A sensitivity analysis

was therefore conducted by simulating the use of alternative electricity sources, namely the European electricity mix (denoted as “medium voltage RER market” in the Ecoinvent 3.5 database) and the global electricity mix (denoted as “medium voltage GLO market” in the Ecoinvent 3.5 database) in place of the Swedish generation mix (denoted as “medium voltage SE market” in the Ecoinvent 3.5 database) [41].

2.5. Uncertainty analysis

Uncertainty analysis was conducted by performing Monte Carlo simulations in SimaPro 9.0 with a 95% confidence level using the inventory data (PSF, PVDF, CA, electricity, water, nitrogen, NMP, EC, DMAc and DMF) for fabrication of a 1000 m² membrane area. A semi-quantitative approach using the Ecoinvent 3.5 database was adopted, in which ratings were assigned for five data quality indicators: reliability, completeness, temporal correlation, geographical correlation, and further technological correlation. A score between 1 (highest data quality) and 5 (lowest data quality) was assigned for each indicator. One thousand iterations were performed to derive measures of uncertainty for the chosen environmental impact categories.

2.6. Environmental cost

Environmental cost is the social cost of pollution, expressed in €/kg of pollutants. It thus measures the loss of economic well-being that occurs when an additional kg of pollutant enters the environment [42]. Pollutant-level costs provide information on the cost of each environmental impact, and are therefore frequently used in environmental impact assessments. Environmental costs were calculated by multiplying the environmental impact for each category by its externality costs, which were 0.056 €/kg CO₂ eq. for GWP, 0.046 €/kBq Co-60 eq. for IRP, 0.0073 €/kg 1,4-DCB eq. for MEP, 0.099 €/kg 1,4-DCB eq. for HCTP, 0.099 €/1,4-DCB eq. for HNCTP, and 0.0845 €/m² crop eq. for LUP. Prices for each environmental impact category were taken from the EU28 Environmental Prices Handbook [42]. Due to the unavailability of environmental costs for the FRSP impact category, impacts in this category were not assessed in terms of cost.

3. Results and discussions

3.1. The impact of water and nitrogen

In all polymer/solvent systems (Fig. 5 and Tables S11 to 17), the use of water and nitrogen had a negligible effect on all environmental impact categories. As stated previously, it was assumed that each plant had its own onsite closed-circuit wastewater treatment system, and wastewater emissions were not considered in the analysis.

3.2. The environmental impact of polymer choice

Three different polymer/solvent systems were considered, as shown in Fig. 6. The PSF/NMP and PVDF/NMP systems use fossil-based polymers, while the CA/NMP system uses a bio-based polymer. The most common solvent in the membrane industry (i.e., NMP) was used in all three cases, and all other parameters were kept constant. Based on the calculations, changing the membrane material from PSF to the fluorinated polymer PVDF had a strong negative impact on environmental performance, causing significant increases in HNCTP, GWP, MEP and IRP. This can be attributed to the impact of industrial PVDF production, the inputs for which are acetylene, hydrogen fluoride, lime (hydrated), and zinc. Hydrogen fluoride production in turn consumes fluor spar (97% purity) and sulfuric acid, and causes high emissions of sulfur dioxide into the air, leading to a strong impact on human toxicity.

One potentially interesting way to reduce the environmental impact of membrane fabrication is to replace fossil-based polymers with bio-based alternatives. In order to evaluate the impact of making such a

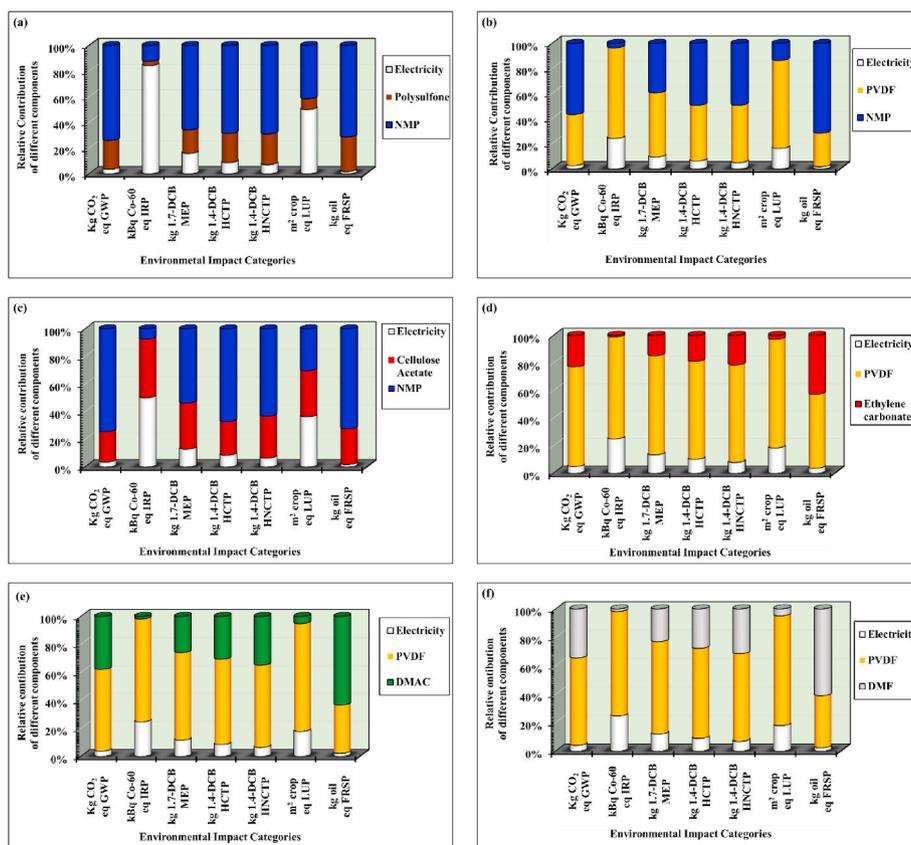


Fig. 5. Relative contributions of the energy source and the choice of polymer and solvent to the chosen environmental impact categories. a) PSF/NMP, b) PVDF/NMP, c) CA/NMP, d) PVDF/EC, e) PVDF/DMAC and f) PVDF/DMF. The impact of water and nitrogen was negligible, so the corresponding results are not shown. Definitions of symbols and abbreviations are given in section 2.3.

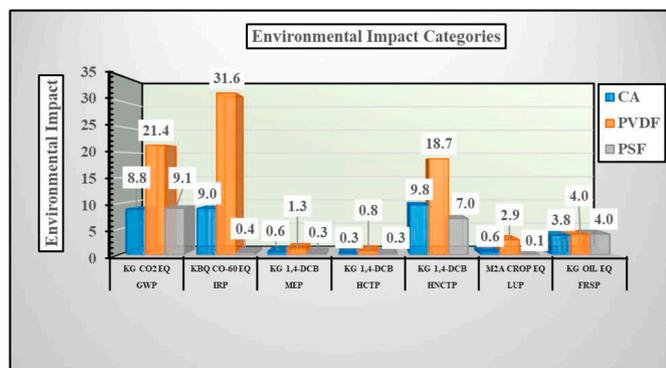


Fig. 6. Environmental impacts of producing 1 kg of the polymers CA, PVDF, and PSF.

switch, the substitution of PVDF or PSF with a cellulose-derived polymer (i.e., CA) was investigated. Surprisingly, changing the membrane material from PSF to CA had only a small effect on the studied impact categories, although there were slight reductions in GWP and FRSP. Cellulose is abundant, inexpensive, biodegradable, and can be obtained from natural resources. One of the main sources of cellulose-based polymer environmental impact is the insolubility of cellulose in organic solvents. Due to the difficulty of dissolving cellulose in organic solvents, cellulose derivatives such as CA are commonly used in membrane production. The conversion of cellulose into CA or other cellulose derivatives reduces its mechanical and chemical stability; in addition, the chemicals used in this process and the waste and byproducts that it generates all have adverse environmental impacts. CA is produced using

(among other things) cellulose fibers, acetic anhydride and acetic acid. The acetic acid production process in particular made a substantial contribution to multiple environmental impact categories. The environmental impact of CA production could be reduced by using green acetylation [43] and different kinds of agricultural waste material as feedstocks.

3.3. The environmental impact of solvent choice

Modern membrane fabrication methods rely heavily on toxic solvents, which is the main factor limiting the sustainability of membrane production. Fig. 5a, b, and c show that the use of toxic solvents is the main source of GWP, HCTP, HNCTP, and FRSP impacts due to membrane production. In recent years, several green solvents have been introduced in the membrane industry. However, there is no universal green solvent that has enough solvating power and stability as well as a low enough cost to compete with traditional polar aprotic solvents.

One way to circumvent this issue would be to use a less toxic solvent to reduce the negative impact of membrane production when no suitable green solvent is available. For instance, one of the polar aprotic solvents DMF or DMAC could be used instead of NMP. To investigate this hypothesis, three systems (PVDF/NMP, PVDF/DMAC, and PVDF/DMF) were considered as dope solutions for producing 1000 m² hollow fiber membranes. All other parameters were kept constant (Fig. 5b, e and 5f). The analysis showed that NMP is the worst of these three solvents from an environmental impact perspective; its negative impact on human health and the environment is significantly worse than that of DMF or DMAC. Therefore, replacing NMP with less toxic polar aprotic solvents such as DMAC or DMF could represent a short-term step towards more sustainable membrane production.

A larger step towards sustainable membrane production would be

made by using green solvents synthesized in a green manner, such as an organic carbonate [31]. The PVDF/EC system was therefore evaluated to analyze the impact of using a green solvent. As shown in Fig. 5d, the use of this system significantly reduced the environmental impact of membrane production when all other parameters were kept constant.

However, it is important to also consider the impact of producing the green solvent; a green solvent produced via a toxic procedure cannot be expected to greatly increase the sustainability of membrane production. To investigate this issue, the environmental impact of producing 1 kg of solvents commonly used in membrane fabrication was calculated and compared to that of ethylene carbonate (Fig. 7). This analysis revealed that replacing NMP with EC significantly reduced overall environmental impact.

3.4. The impact of the source of electricity

The results of the sensitivity analysis of the membrane preparation process are tabulated in Table 2. GWP increased by as little as 28% (RER) for the PVDF/NMP system and as much as 92% (GLO) for the PVDF/EC system. The main sources of energy in Sweden are nuclear power (42%) and hydropower (41%) [44]. Additionally, 17% of Sweden's electricity is generated from other renewable sources such as wind, solar, and biomass (Table S2). In all polymer/solvent systems, electricity had the greatest impact on IRP (due to nuclear generation) and LUP (due to the high land use involved in cultivating biomass for energy generation). Conversely, 25% of the electricity in the European electricity mix originates from coal, lignite, oil, and other fuels, 29% from renewables, 26% from nuclear energy, and 20% from natural and derived gas [45]. GLO is dominated by fossil fuel and showed high GWP in compare to SE and RER. The source of electricity production is the main determinant of GWP impact because nuclear and hydropower produce much lower CO₂ emissions than fossil fuel combustion. However, all impact categories were influenced by changes in the electricity source. Electricity generation using methods that produce high CO₂ emissions, particularly the burning of fossil fuels (coal and natural gas), was the main source of environmental impact. Hydropower and nuclear energy generate no greenhouse gases and thus have much lesser environmental impacts. However, the effects on the MEP and HNCCTP impact categories caused by switching from the European electricity mix (RER) to the global electricity mix (GLO) were much weaker than those for the other impact categories because these two categories are more sensitive to the chemical substances used during the synthesis. The changes in environmental impact due to the use of the RER and GLO electricity mixes for different solvent and polymer systems are presented in the supplementary material (Fig. S1).

3.5. Uncertainty analysis

For each environmental impact category, the coefficient of variation

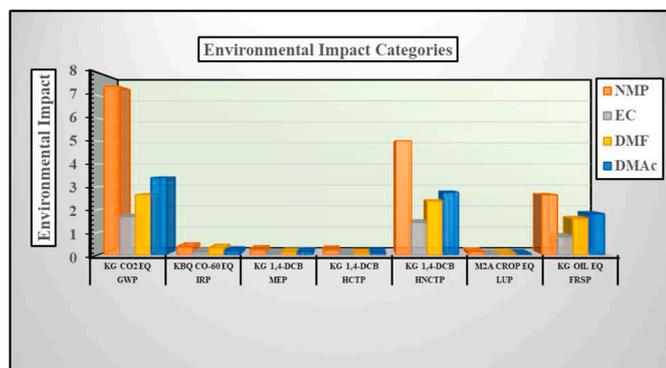


Fig. 7. Environmental impact of producing 1 kg of each solvent considered in this work.

Table 1 Effects of different electricity generation mixes on the environmental impact of membrane preparation (1000 m²) using seven polymer/solvent combinations, and the uncertainty in the results (expressed in terms of the CV) ^a.

Category/Impact (unit)	PSE/NMP			PVDF/NMP			CA/NMP			PVDF/EC			PVDF/DMAc			PVDF/DMF			PSE/DMAc		
	SE (CV)	RER	GLO	SE (CV)	RER	GLO	SE (CV)	RER	GLO	SE (CV)	RER	GLO	SE (CV)	RER	GLO	SE (CV)	RER	GLO	SE (CV)	RER	GLO
GWP (kg CO ₂ eq)	3224 (12.3)	4399	5392	4209 (10.9)	5384	6377	3207 (12.01)	4381	5375	2365 (7.6)	3539	4532	2931 (7.1)	4105	5099	2784 (6.8)	3958	4952	1946 (6.9)	3120	4114
IRP (kBq Co-60 eq)	1010 (130.4)	795	418	3502 (125.3)	3287	2909	1697 (159.40)	1482	1105	3417 (149.6)	3202	2824	3438 (136.9)	3223	2846	3439 (158.5)	3225	2847	946 (129.5)	731	354
MEP (kg 1,4-CB)	118 (52.6)	161	161	198 (47.2)	241	240	145 (55.45)	188	187	141 (44.3)	184	183	162 (55.6)	205	204	156 (46.8)	199	198	82 (52.3)	125	125
HCTP (kg 1,4-DCB)	106 (136.7)	167	177	147 (106.6)	208	219	108 (96.84)	169	180	92 (174.2)	153	164	107 (84.9)	168	179	103 (85.2)	164	175	66 (72.3)	127	138
HNCCTP (kg 1,4-DCB)	2349 (69.3)	3210	3184	3285 (73.7)	4146	4120	2567 (83.31)	3428	3402	2122 (73.3)	2983	2957	2555 (91.7)	3416	3390	2429 (77.2)	3290	3264	1619 (69.3)	2480	2454
LUP (m ² a crop eq)	108 (16.9)	101	79	330 (15.6)	323	301	148 (15.96)	141	119	293 (15.5)	287	265	300 (14.6)	294	272	299 (15.3)	293	271	78 (16.9)	72	50
FRSP (kg oil eq)	1192 (11.8)	1492	1702	1189 (13.2)	1489	1699	1174 (12.56)	1474	1685	592 (8.1)	892	1103	923 (10.3)	1223	1434	870 (9.8)	1170	1381	926 (9.6)	1226	1437

^a Abbreviation: Polysulfone (PSP), N-methyl-2-pyrrolidone (NMP), Polyvinylidene fluoride (PVDF), Cellulose acetate (CA), Ethylene carbonate (EC), Dimethylacetamide (DMAc), Dimethylformamide (DMF), Global warming potential (GWP), Ionizing radiation potential (IRP), Marine ecotoxicity potential (MEP), Human carcinogenic toxicity potential (HCTP), Human non-carcinogenic toxicity potential (HNCCTP), Land use potential (LUP), Fossil resource scarcity (FRSP), Sweden electricity mix (SE), European electricity mix (RER), Global electricity mix (GLO).

Table 2
Environmental costs of different polymer/solvent combinations^a.

Impact category (Unit)	PSF/NMP	PVDF/NMP	CA/NMP	PVDF/EC	PVDF/DMAc	PVDF/DMF	PSF/DMAc
GWP (kg CO ₂ eq)	181	236	187	132	164	156	109
IRP (kBq Co-60 eq)	46	161	53	157	158	158	44
MEP (kg 1,4-DCB)	1	1	1	1	1	1	1
HCTP (kg 1,4-DCB)	10	15	11	9	11	10	7
HNCTP (kg 1,4-DCB)	233	325	233	210	253	240	160
LUP (m ² a crop eq)	9	28	10	25	25	25	7
FRSP (kg oil eq)	N/A						
TOTAL (Euro/FU)	480	766	494	535	612	591	327

^a Abbreviation: Polysulfone (PSF), N-methyl-2-pyrrolidone (NMP), Polyvinylidene fluoride (PVDF), Cellulose acetate (CA), Ethylene carbonate (EC), Dimethylacetamide (DMAc), Dimethylformamide (DMF), Global warming potential (GWP), Ionizing radiation potential (IRP), Marine ecotoxicity potential (MEP), Human carcinogenic toxicity potential (HCTP), Human non-carcinogenic toxicity potential (HNCTP), Land use potential (LUP), Fossil resource scarcity (FRSP).

(CV) for the fabrication of a 1000 m² membrane area was estimated (Table 1). The low CVs for GWP, FRSP, and LUP for all polymer/solvent systems indicate that there is little uncertainty regarding the results for these impact categories. However, the CVs for the HNCTP, HCTP and IRP categories were higher (Table 2), indicating that the results in these categories are uncertain because of data variability issues within the Ecoinvent 3.5 database.

3.6. Environmental cost of membrane preparation process

Table 2 shows the environmental cost of membrane fabrication under different scenarios. The highest cost was due to the HNCTP impact category under all scenarios, and the second highest was due to the GWP impact category. The overall environmental cost is proportional to the impact in each category. The system with the highest total environmental cost was PVDF/NMP (766 €/FU), and that with the lowest was PSF/DMAc (327 €/FU). The use of PVDF was associated with high IRP impact costs. Additionally, the PVDF/DMAc and PVDF/DMF polymer/solvent systems had high environmental costs of 612 and 591 €/FU, respectively.

4. Conclusions

A comparative life cycle assessment of hollow fiber membrane fabrication (1000 m²) using fossil-based polymers (PVDF, PSF), a bio-based polymer (CA), toxic solvents (NMP, DMAc, and DMF), and a green solvent (EC) was performed to determine the scope for reducing the environmental impact of membrane production and improving industrial scalability. The choice of solvent and polymer and the source of electricity were identified as the major determinants of environmental impact and cost. Hollow fiber membrane fabrication using the PVDF/NMP system had the highest environmental cost (766 €/FU), largely due to adverse global warming and human toxicity impacts. Toxic solvents such as NMP are mainly responsible for the negative environmental impact of membrane production, accounting for 40–60% of the total impact. The use of a green solvent such as EC can significantly reduce environmental costs and impacts, by up to 35%. Alternatively, if no suitable green solvent is available, a less toxic solvent can be used. For example, replacing NMP with the less toxic DMAc reduced environmental impact by up to 15% and environmental cost by up to 154 €/FU. Substituting a fossil-based polymer (PVDF or PSF) with a bio-based alternative (CA) can have either a positive (PVDF vs. CA) or negative (PSF vs. CA) environmental impact that depends on the production process of CA; if the production of the bio-based polymer involves environmentally unfriendly steps, substitution may increase environmental costs. Using electricity obtained from renewable sources such as hydropower can further reduce the environmental impact of membrane production.

Author statement

Pooja Yadav: Simulation, Conceptualization, Methodology, Validation, Investigation, Writing - original draft. **Norafiqah Ismail:** Data collection, Conceptualization, Methodology, Validation, Investigation, Writing - original draft, **Mohamed Essalhi:** Conceptualization, Methodology, Validation, Investigation, Writing - original draft, **Mats Tysklind:** Investigation, Writing - original draft, Funding acquisition, **Dimitris Athanassiadis:** Supervision, Visualization, Project administration, Funding acquisition, Writing - review & editing. **Naser Tavajohi:** Supervision, Visualization, Project administration, Funding acquisition, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

The authors acknowledge financial support from the Bio4energy research platform (www.bio4energy.se) and Kempe foundation.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.memsci.2020.118987>.

References

- [1] C. Reid, E. Breton, Water and ion flow across cellulosic membranes, *J. Appl. Polym. Sci.* 1 (1959) 133–143.
- [2] S. Loeb, Sea Water Demineralization by Means of a Semipermeable Membrane, UCLA Dept. of Engineering Report, 1960.
- [3] S. Loeb, S. Sourirajan, High Flow Porous Membranes for Separating Water from Saline Solutions, 1964, 3133132.
- [4] M. Gündođdu, Y.A. Jarma, N. Kabay, T.Ö. Pek, M. Yüksel, Integration of MBR with NF/RO processes for industrial wastewater reclamation and water reuse-effect of membrane type on product water quality, *J. Water Process Eng.* 29 (2019), 100574.
- [5] S. Al Aani, T.N. Mustafa, N. Hilal, Ultrafiltration membranes for wastewater and water process engineering: a comprehensive statistical review over the past decade, *J. Water Process Eng.* 35 (2020), 101241.
- [6] J. Wang, H. Guo, Z. Yang, Y. Mei, C.Y. Tang, Gravity-driven catalytic nanofibrous membranes prepared using a green template, *J. Membr. Sci.* 525 (2017) 298–303.
- [7] L.-P. Zhang, Z. Liu, Y. Faraj, Y. Zhao, R. Zhuang, R. Xie, X.-J. Ju, W. Wang, L.-Y. Chu, High-flux efficient catalytic membranes incorporated with iron-based Fenton-like catalysts for degradation of organic pollutants, *J. Membr. Sci.* 573 (2019) 493–503.
- [8] C. Hu, M.-S. Wang, C.-H. Chen, Y.-R. Chen, P.-H. Huang, K.-L. Tung, Phosphorus-doped g-C₃N₄ integrated photocatalytic membrane reactor for wastewater treatment, *J. Membr. Sci.* 580 (2019) 1–11.
- [9] E. Palo, A. Salladini, B. Morico, V. Palma, A. Ricca, G. Iaquaniello, Application of Pd-based membrane reactors: an industrial perspective, *Membranes* 8 (2018) 101.

- [10] M.B. Asif, J. Hou, W.E. Price, V. Chen, F.I. Hai, Removal of trace organic contaminants by enzymatic membrane bioreactors: role of membrane retention and biodegradation, *J. Membr. Sci.* (2020), 118345.
- [11] P. Krzeminski, L. Leverette, S. Malamis, E. Katsou, Membrane bioreactors – a review on recent developments in energy reduction, fouling control, novel configurations, LCA and market prospects, *J. Membr. Sci.* 527 (2017) 207–227.
- [12] M. Shin, W.-J. Song, H.B. Son, S. Yoo, S. Kim, G. Song, N.-S. Choi, S. Park, Highly stretchable separator membrane for deformable energy-storage devices, *Adv. Energy Mater.* 8 (2018), 1801025.
- [13] B. Smitha, S. Sridhar, A.A. Khan, Solid polymer electrolyte membranes for fuel cell applications—a review, *J. Membr. Sci.* 259 (2005) 10–26.
- [14] H. Zhang, P.K. Shen, Recent development of polymer electrolyte membranes for fuel cells, *Chem. Rev.* 112 (2012) 2780–2832.
- [15] T. Huang, G. He, J. Xue, O. Otoo, X. He, H. Jiang, J. Zhang, Y. Yin, Z. Jiang, J. C. Douglin, D.R. Dekel, M.D. Guiver, Self-crosslinked blend alkaline anion exchange membranes with bi-continuous phase separated morphology to enhance ion conductivity, *J. Membr. Sci.* 597 (2020), 117769.
- [16] L. Zhu, X. Dong, M. Xu, F. Yang, M.D. Guiver, Y. Dong, Fabrication of mullite ceramic-supported carbon nanotube composite membranes with enhanced performance in direct separation of high-temperature emulsified oil droplets, *J. Membr. Sci.* 582 (2019) 140–150.
- [17] R.A. Tufa, E. Rugiero, D. Chanda, J. Hnàt, W. van Baak, J. Veerman, E. Fontananova, G. Di Profio, E. Drioli, K. Bouzek, E. Curcio, Salinity gradient power-reverse electrodialysis and alkaline polymer electrolyte water electrolysis for hydrogen production, *J. Membr. Sci.* 514 (2016) 155–164.
- [18] Q.-Y. Wu, C. Wang, R. Wang, C. Chen, J. Gao, J. Dai, D. Liu, Z. Lin, L. Hu, Salinity-Gradient power generation with ionized wood membranes, *Adv. Energy Mater.* 10 (2020), 1902590.
- [19] D.F. Stamatialis, B.J. Papenburg, M. Gironés, S. Saiful, S.N.M. Bettahalli, S. Schmitmeier, M. Wessling, Medical applications of membranes: drug delivery, artificial organs and tissue engineering, *J. Membr. Sci.* 308 (2008) 1–34.
- [20] Enrico Drioli, E. Lidieta Giorno, Fontananova, *Comprehensive Membrane Science and Engineering*, second ed., Elsevier Science, 2017.
- [21] E.M. Hoek, V.V. Tarabara, H. Bum Park, Gas separation membranes. *Encyclopedia of Membrane Science and Technology*, 2013, pp. 1–32.
- [22] N. Ismail, A. Venault, J.-P. Mikkola, D. Bouyer, E. Drioli, N. Tavajohi Hassan Kiadeh, Investigating the potential of membranes formed by the vapor induced phase separation process, *J. Membr. Sci.* 597 (2020) 117601.
- [23] T. Marino, F. Russo, A. Criscuoli, A. Figoli, TamiSolve® NxG as novel solvent for polymeric membrane preparation, *J. Membr. Sci.* 542 (2017) 418–429.
- [24] J. Sherwood, T.J. Farmer, J.H. Clark, Catalyst: possible consequences of the N-methyl pyrrolidone REACH restriction, *Inside Chem.* 4 (2018) 2010–2012.
- [25] M. Razali, J.F. Kim, M. Attfield, P.M. Budd, E. Drioli, Y.M. Lee, G. Szekely, Sustainable wastewater treatment and recycling in membrane manufacturing, *Green Chem.* 17 (2015) 5196–5205.
- [26] H.H. Wang, J.T. Jung, J.F. Kim, S. Kim, E. Drioli, Y.M. Lee, A novel green solvent alternative for polymeric membrane preparation via nonsolvent-induced phase separation (NIPS), *J. Membr. Sci.* 574 (2019) 44–54.
- [27] Z. Cui, N.T. Hassankiadeh, S.Y. Lee, J.M. Lee, K.T. Woo, A. Sanguineti, V. Arcella, Y.M. Lee, E. Drioli, Poly(vinylidene fluoride) membrane preparation with an environmental diluent via thermally induced phase separation, *J. Membr. Sci.* 444 (2013) 223–236.
- [28] Z. Cui, N.T. Hassankiadeh, S.Y. Lee, K.T. Woo, J.M. Lee, A. Sanguineti, V. Arcella, Y.M. Lee, E. Drioli, Tailoring novel fibrillar morphologies in poly(vinylidene fluoride) membranes using a low toxic triethylene glycol diacetate (TEGDA) diluent, *J. Membr. Sci.* 473 (2015) 128–136.
- [29] N.T. Hassankiadeh, Z. Cui, J.H. Kim, D.W. Shin, S.Y. Lee, A. Sanguineti, V. Arcella, Y.M. Lee, E. Drioli, Microporous poly(vinylidene fluoride) hollow fiber membranes fabricated with PolarClean as water-soluble green diluent and additives, *J. Membr. Sci.* 479 (2015) 204–212.
- [30] T. Marino, F. Galiano, A. Molino, A. Figoli, New frontiers in sustainable membrane preparation: Cyrene™ as green bioderived solvent, *J. Membr. Sci.* 580 (2019) 224–234.
- [31] M.A. Rasool, P.P. Pescarmona, I.F.J. Vankelecom, Applicability of organic carbonates as green solvents for membrane preparation, *ACS Sustain. Chem. Eng.* 7 (2019) 13774–13785.
- [32] A. Figoli, T. Marino, S. Simone, E. Di Nicolò, X.M. Li, T. He, S. Tornaghi, E. Drioli, Towards non-toxic solvents for membrane preparation: a review, *Green Chem.* 16 (2014) 4034–4059.
- [33] N. Wrisberg, H.A.U. De Haes, W. Klöpffer, Analytical tools for environmental design and management in a systems perspective, *Int. J. Life Cycle Assess.* 9 (2004) 137.
- [34] M.C. McManus, C.M. Taylor, The changing nature of life cycle assessment, *Biomass Bioenergy* 82 (2015) 13–26.
- [35] P. Yadav, S.R. Samadder, A critical review of the life cycle assessment studies on solid waste management in Asian countries, *J. Clean. Prod.* 185 (2018) 492–515.
- [36] P. Yadav, S.R. Samadder, A global prospective of income distribution and its effect on life cycle assessment of municipal solid waste management: a review, *Environ. Sci. Pollut. Res. Int.* 24 (2017) 9123–9141.
- [37] F. Prézéus, L. Tiruta-Barna, C. Guigui, J.-C. Remigy, A generic process modelling – LCA approach for UF membrane fabrication: application to cellulose acetate membranes, *J. Membr. Sci.* 618 (2021), 118594.
- [38] A.S. A., Exergetic Life Cycle Assessment of Electrospun Polyvinylidene Fluoride Nanofibers, Ph.D. thesis, University of South Florida, 2014.
- [39] F. Cerdas, M. Juraschek, S. Thiede, C. Herrmann, Life cycle assessment of 3D printed products in a distributed manufacturing system, *J. Ind. Ecol.* 21 (2017) S80–S93.
- [40] M.A.J. Huijbregts, Z.J.N. Steinmann, P.M.F. Elshout, G. Stam, F. Verones, M. Vieira, M. Zijp, A. Hollander, R. van Zelm, ReCiPe2016: a harmonised life cycle impact assessment method at midpoint and endpoint level, *Int. J. Life Cycle Assess.* 22 (2017) 138–147.
- [41] Ecoinvent 3.6 Database, 2019.
- [42] S. Bruyn, M. Bijleveld, L.d. Graaff, E. Schep, A. Schrotten, R. Vergeer, S. Ahdour, *Environmental Prices Handbook EU28 Version, Methods and Numbers for Valuation of Environmental Impacts*, CE Delft, Delft, 2018.
- [43] D. Araújo, M.C.R. Castro, A. Figueiredo, M. Vilarinho, A. Machado, Green synthesis of cellulose acetate from corncob: physicochemical properties and assessment of environmental impacts, *J. Clean. Prod.* 260 (2020), 120865.
- [44] K. Shanmugam, V. Gadhamshetty, P. Yadav, D. Athanassiadis, M. Tysklynd, V.K. K. Upadhyayula, Advanced high-strength steel and carbon fiber reinforced polymer composite body in white for passenger cars: environmental performance and sustainable return on investment under different propulsion modes, *ACS Sustain. Chem. Eng.* 7 (2019) 4951–4963.
- [45] EEA (European Environment Agency), Overview of electricity production and use in Europe. <https://www.eea.europa.eu/data-and-maps/indicators/overview-of-the-electricity-production-2>, 2019.