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Environmental Impact Improvement of Chitosan-Based Mixed-Matrix Membranes Manufacture for CO_2 Gas Separation by Life Cycle Assessment

The environmental impacts of the manufacture of chitosan (CS) and polymeric poly(1-trimethylsilyl-1-propyne) (PTMSP) mixed-matrix membranes (MMMs) for CO_2 separation by life cycle assessment (LCA) are compared. An ionic liquid of non-reported toxicity is used in CS membranes to enhance the mechanical strength, and different fillers are used to increase mechanical and functional properties: ETS-10, ZIF-8, HKUST-1, and Zeolite A. Results with the same CO_2 permeation flux indicate that ETS-10/IL-CS is the membrane manufacture with highest impacts due to its lower permeability. When comparing impacts with same permeation areas, the polymeric one is the membrane with highest impacts. Biopolymer and polymer manufacture are the components with highest contribution to the total environmental impacts of each membrane. To decrease all their impacts below fossil polymer membrane for the same CO_2 permeation flux, CS membranes permeabilities should be improved by a numerical factor of 1000, 100, and 2 for the ETS-10, ZIF-8, and HKUST-1/IL-CS MMMs, respectively.

Supporting Information available online

Keywords: Biobased mixed-matrix membranes, Chitosan, CO_2 separation, Sustainable membrane manufacture

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1 Introduction

Over the last decades, the concentration of greenhouse gases in the atmosphere has increased producing a disruption of the natural carbon cycle and raising the global warming. Among all the gases generated mainly from the combustion of fossil resources to produce energy that are responsible for this, carbon dioxide (CO_2) is the main one, due to the higher concentration levels. Despite the urgent necessity to mitigate the global warming problem, it is still not possible to cover the global demand of energy without fossil fuels, so the improvement of methods to reduce CO_2 emissions are now essential [1].

Even though absorption is a largely applied and well-established technology for post-combustion CO_2 gas separation in industries, this method requires a large amount of energy and is not very economical. Among absorption, different post-combustion technologies as adsorption, chemical reactions, and membrane technologies have been studied during the last years with good results [2]. The latter shows advantages over the other methods, such as lower energy requirement, low operating cost, and simplicity of performance and scale-up [3, 4]. In addition, the environmental emissions are lower than by absorption, although these emissions depend strongly on the membrane type [5].

The ability of a membrane to trap more or less CO_2 depends on the materials it is made of [6]. Although polymeric and inorganic membranes have been synthesized at laboratory scale to separate CO_2 from N_2 and CH_4, their viability as CO_2 systems separators at industrial scale is limited due to their permeabilities and selectivities. Mixed-matrix membranes (MMMs), composed of an organic matrix and an inorganic filler, have proven to be superior to polymeric and inorganic membranes in gas mixture separation [7, 8]. Also, the addition of ionic

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liquid (IL) has shown to enhance the mechanical strength and the CO_2 separation properties in MMMs [9].

Chitosan (CS) is a linear polysaccharide obtained by the deacetylation of chitin, an abundant natural polymer, which is cheap and obtained from renewable sources, i.e., the shell of crustaceans. CS has been proven to be a suitable organic framework to manufacture membranes for CO_2/N_2 membranes separation [10]. The only drawback occurs when the uncontrolled high hydrophilicity compromises the mechanical resistance. Mechanical strength of CS membranes has been improved incorporating IL as additive. The incorporation of microporous ETS-10 nanoparticles in CS/IL membranes has been also demonstrated by Casado-Coterillo et al. to increase tensile strength and CO_2/N_2 separation [11].

Metal-organic frameworks (MOFs) have been proposed as promising fillers for MMMs because of their organic nature and expected higher compatibility with polymer chains. Casado-Coterillo et al. have also been successful in synthesizing CS/IL with HKUST-1, which is a 3D porous MOF with high CO_2 sorption, and ZIF-8, being a zeolite imidazole framework subgroup of MOFs, as fillers in MMMs [12].

Biopolymers are proved to be a renewable and naturally occurring alternative to conventional petroleum-based polymers [13]. With the use of CS as membrane polymer matrix, it is expected that the environmental impacts of the manufacture of MMMs will be reduced in comparison with MMMs with polymers from fossil fuels. As a result, CO_2 capture by biopolymer-based membranes can avoid the depletion of fossil resources while contributing to the economy decarbonization. In light of the potential of these renewable materials for CO_2 capture, it is important to improve the understanding of their performance from an environmental perspective in order to avoid the risk of generating new environmental impacts. Consequently, in this work, the environmental impacts of the manufacture of CS MMMs are going to be compared with poly(1-trimethylsilyl-1-propyne) (PTMSP) MMMs, with Zeolite A as filler [14] since PTMSP is synthesized from fossil resources.

In this context, life cycle assessment (LCA) is a well-established and widely accepted tool for determining the environmental profile of a product or a process [15, 16]. There are not many studies about the environmental impacts of membrane synthesis by LCA, and less on biopolymer membranes [17]. In one study made by part of the authors of this work, the impacts of manufacturing a zeolite membrane on an industrial scale [18] were quantified by LCA. In a recent study, LCA was applied to quantify environmental impacts of Pd membranes synthesis, for H_2 separation [19]. In the case of CS membranes, the LCAs reported have been mostly limited to barrier films for packaging applications [20].

Only few articles exist comparing environmental impacts of polymeric membranes synthesis by LCA, analyzing different membrane base materials [21], different solvents for membrane synthesis [22], and quantifying the environmental impacts of polymeric membrane hollow-fiber manufacture [23]. In this last study, a comparison of the environmental impacts of membrane manufacture using polymers from fossils and renewable materials (cellulose) is made using the production of 1000 m^2 of permeation area as functional unit, concluding that biopolymers do not reduce environmental impacts. However, an effective LCA of membrane manufacturing should include as functional unit the permeation flux of the component to be separated.

To the best of our knowledge, this is the first study that compares the environmental feasibility of polymeric and CS biopolymeric membrane productions. The productions of four different MMMs for the same CO_2 permeation flux in CO_2/N_2 ideal separations have been analyzed from the LCA perspective. The main objective of this work is to quantify the environmental impacts of the manufacture of CS and PTMSP MMMs by LCA using as functional unit the same permeation flux of CO_2 in N_2/CO_2 ideal separations. Finally, permeation improvements of CS MMMs are suggested with the objective of making comparable the environmental impacts of CS MMMs with PTMS MMM.

2 Experimental

The LCA was carried out following the standards specified in ISO 14040 and ISO 14044 [24]. The study followed the four main LCA steps, which are: 1 – goal and scope definition, 2 – life cycle inventory analysis (LCI), 3 – life cycle impact assessment (LCIA) and, 4 – interpretation.

The lab-scale preparation of the MMMs studied in this work was described in the previous works cited above [25–27]. In a typical synthesis procedure, the membranes were prepared by solution-casting, dissolving the polymers in their respective solvents, and adding the allocated solutions to the dispersed fillers, then stirred to homogeneous mixture and cast on clean Petri dishes of 4.5 cm diameter that gave an effective membrane area of 15.55 cm^2. The membrane solvent was evaporated in a fume-hood at room temperature.

2.1 Goal and Scope

This work is focused on the impacts generated by the manufacture of four membranes: ETS-10/IL-CS, ZIF-8/IL-CS, HKUST-1/IL-CS, and Zeolite A/PTMSP. The purpose is to determine their impact from an environmental point of view, for the same CO_2 permeate flow rate in ideal CO_2/N_2 separation mixtures. In order to specify the inputs and outputs of the analysis, the study has been carried out considering the system from cradle to gate. The measure starts with the extraction of the raw materials and ends when the membrane is created, so the data on the materials and energy required throughout the process and also the emissions generated have been collected.

The LCA presented in this work follows the recommendations given by the European Platform on LCA [28] and the 16 environmental impact indicators (EIIs) used can be found in Tab. S1 in the Supporting Information (SI). The software GaBi® 9.5 Professional and its database [29] were used to conduct the LCA.

To determine the necessary area of each membrane to permeate the same CO_2 flow rate, the permeation in Barrer at a temperature of 25 °C and 1 bar of pressure was considered. Tab. 1 presents the properties of the membranes. Gas permeation tests were carried out with pure N_2 and CO_2 at room

temperature in a constant volume system. Membranes were placed in the permeation cell and tested for N_2 first and then CO_2. The steady-state permeability (1 Barrer = 10^{-10} cm^3 (STP)cm/cm^2s cmHg) was calculated from the steady-state flux.

Ideal selectivity $\alpha(CO_2/N_2)$ is defined as the ratio of the permeability values of the two gases, the faster gas permeability (CO_2) divided by the slower gas permeability (N_2). The ETS-10/IL-CS membrane was used as reference for the calculation, as it is the membrane with the lowest permeability, i.e., 300 Barrer [11]. Using the permeability equation (1), considering the thickness and experimental initial area, a permeate flow rate of 2.11×10^{-3} cm^3 (STP)/s CO_2 is determined. Thus, the functional unit of this study is the manufacture of a membrane capable of permeating a flow of 2.11×10^{-3} cm^3 (STP)/s CO_2. Despite permeation units usually being expressed in GPU (10^{-6} cm^3(STP)/cm^2s cmHg), the approach used in this work allows to compare the different experimental areas at the same CO_2 flux, cm^3 (STP)/s units. To determine the numerical factor to compare MMMs with ETS-10, first, the areas necessary to 2.11×10^{-3} cm^3(STP)/s of CO_2 for ZIF-8, HKUST-1 and Zeolite A MMMs are obtained (Tab. 1).

The areas for different membranes (A) are obtained with Eq. (1), permeation (P_i) and thickness (l) of each membrane, and pressure gradient of 760 mmHg (Δp_i) and 2.11×10^{-3} cm^3 (STP)/s of CO_2 (Q_i). Then, the numerical factors are obtained with Eq. (2) and considering selectivities CO_2/N_2, areas, and initial areas. The numerical factors will multiply the environmental impacts of each MMM manufacture in order to make them comparable in terms of obtaining the same flux of CO_2.

$$P_i = \frac{Q_i\, l}{A\, \Delta p_i} \tag{1}$$

$$\text{Numerical factor} = \left(\frac{\text{Area}}{\text{Initial area}}\right) \times \left(\frac{\text{Selectivity ETS } CO_2/N_2}{\text{Selectivity membrane } CO_2/N_2}\right) \tag{2}$$

The thickness of the Zeolite A/PTMSP membrane has been assumed to be 99 µm in this work. This assumption is based on the fact that the loading does not affect much in the thickness of the PTMSP membrane. The pure PTMSP membrane was 100 µm thick, while the membrane with a 20 wt % zeolite loading was 98.6 µm [14, 31].

Table 1. MMMs properties [26, 27, 30].

	Thickness [µm]	Initial area [cm^2]	Permeability [Barrer]	Area[a)] [cm^2]	Selectivity CO_2/N_2	Numerical factor
ETS-10/IL-CS	168	15.55	300	15.55	13	1
ZIF-8/IL-CS	90.4	14.45	2900	1.101	3	0.33
HKUST-1/IL-CS	91.8	14.45	2950	0.864	20	0.039
Zeolite A/PTMSP	99	14.05	13 000	0.196	10	0.018

[a)] Area for the permeation flow of 2.11×10^{-3} cm^3(STP) s^{-1} of CO_2.

2.2 Life Cycle Inventory

In this section, the main inputs for the manufacture of the four MMMs under study are compiled. The MMMs have been modeled following the method explained in literature [26, 27, 30]. The membranes consist of a polymer matrix and a dispersed phase. All membranes have a loading of 5 wt % with respect to the polymer concentration. The four analyzed membranes were divided into two groups. There are three with a biopolymeric CS base and the fourth with a PTMSP polymer base. The information for the synthesis of CS has been taken from literature [32].

Briefly, CS is produced from seafood wastes, mainly from crabs and shrimps [33], although in this work CS production has been considered coming entirely from shrimp shells. The route to prepare PTMSP, however, has been followed at the laboratory scale by the synthesis of the 1-trimethylsilyl-1-propyne (TMSP) monomer first and then its polymerization [34, 35]. The dispersed phase of the membranes is different in the four analyzed membranes. Zeolite A was used as inorganic filler for the PTMSP membrane [36], while the CS MMMs were loaded by small amounts of ETS-10 [25, 30], ZIF-8 [37], and HKUST-1 [38], respectively. These three latter membranes also contain a 5 wt % (referred to CS) of the ionic liquid (IL) [emim][Ac], with no reported toxicity [39] which provides the membranes with higher CO_2 solubility as well as better mechanical and thermal stability [40].

The inputs to the inventory of the membranes are given in Tab. 2 calculated with their respective areas. In the SI of this work the inventory for each membrane and for each component is given. It is worth noting that the impacts derived from shrimp shell were not considered as they are wastes and therefore, the burdens are assumed to be allocated to shrimps. In the PTMSP monomer process, the use of propene has been assumed instead of propyne, and dimethyldichlorosilane instead of trimethylchlorosilane.

The GaBi® Pro. database was used to estimate the datasets the background processes including chemicals, electricity, and other materials needed in the fabrication processes. Processes located in Spain were taken into consideration when it was possible, when not data from European Union were used. The environmental impacts were determined in the LCIA step by using the methods recommended by the European Platform on LCA [28], which include several midpoint categories according based on common mechanisms (e.g., climate change, acidification potential, abiotic resource depletion) (see SI). Because this is a first LCA approach intended to analyze hotspots of the manufacture performance, the transport step and the end-of-cycle are out of the scope of the present study.

3 Results

3.1 MMMs with the Same CO_2 Permeation Flux

Fig. 1 shows the comparison of the environmental impacts of the four

Table 2. Input material and energy values for each membrane to obtain their respective initial areas (ETS-10/IL-CS, 15.55 cm^2, ZIF-8/IL-CS, 14.55 cm^2, HKUST-1/IL-CS, 14.55 cm^2, Zeolite A/PTMSP, 14.05 cm^2).

	ETS-10/IL-CS	ZIF-8/IL-CS	HKUST-1/IL-CS	Zeolite A/PTMSP	Units
$Al(OH)_3$	–	–	–	3.5	[mg]
Ammonia	6.36	66.6	382.9	–	[mg]
Benzene	–	–	3.0	–	[mg]
Copper(II) nitrate	–	–	16.9	–	[mg]
Diesel	9240	3.5	3.5	–	[mg]
Dimethyldichlorosilane	–	–	–	217.8	[mg]
Electricity	2151.8	1779.0	2598.0	108.9	[J]
Ethanol	15.3	9.2	651.3	63.6	[mg]
Formaldehyde	29.4	307.8	1772.6	–	[mg]
Glyoxal	52.5	550.0	3160.6	–	[mg]
HAc	22.8	299.2	13.7	–	[mg]
HBr	27.1	16.3	16.3	113.2	[mg]
HCl	717	430.5	429.8	–	[mg]
Heat	7196.8	4798.8	6033.4	23 687.3	[J]
K_2O	6.7	–	–	–	[mg]
Lead	27.5	417.9	16.5	–	[mg]
Magnesium	–	–	–	40.6	[mg]
Methanol	–	114.4	–	9458.2	[mg]
Methylamine	11.3	118.4	678.9	–	[mg]
Na_2CO_3	–	–	–	1.9	[mg]
NaClO	65.8	863.6	39.6	–	[mg]
NaOH	1404	838.7	839.5	1.8	[mg]
Propene	–	–	–	88.1	[mg]
Shrimp shell	9240	5538.5	5551.9	–	[mg]
Silica flour	–	–	–	10.0	[mg]
Ta_5Cl	–	–	–	8.8	[mg]
Tetrahydrofuran	–	–	–	1103.5	[mg]
TiO_2	3.0	–	–	–	[mg]
Toluene	–	–	–	10 025.6	[mg]
Water	273.4	3223.4	921.6	219.3	[mg]
Waterglass	23.3	–	–	–	[mg]

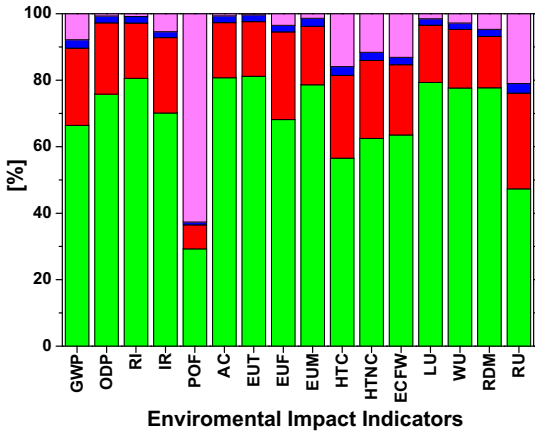

Figure 1. Contribution to the total environmental impact indicator value of the four MMMs with the same CO_2 permeation flow. Green, ETS-10/IL-CS; red, ZIF-8/IL-CS; blue, HKUST-1/IL-CS; magenta, Zeolite A/PTMSP).

membranes for the same CO_2 permeation flow (2.11×10^{-3} cm^3 (STP)/s CO_2). EIIs of different membranes have been multiplied by their numerical factor (Tab. 2) to consider different permeabilities and selectivities. Tab. S12 in the Supporting Information collects the numerical values of EIIs for the four membranes. Fig. 1 shows the relative contribution of each membrane to the sum of the indicator for the four membranes.

It is clear from Fig. 1 that the largest impact is generated by the ETS-10 membrane because it has the lowest permeability (300 Barrer). The ETS-10/IL-CS membrane has the highest impact on 15 of the 16 indicators. Due to the lower permeability of the ETS-10/IL-CS MMM, it needs a larger area for the same CO_2 permeation flux. It is worth noting that the ETS-10/IL-CS membrane has the EIIs multiplied by a factor of 3 compared with ZIF-8 membrane, by a factor of 25.6 compared with HKUST-1 membrane, and by a factor of 55.56 compared with the PTPMS membrane. So, these permeability differences are determinant when comparing the four MMMs EEIs.

However, in Fig. 1 it can be observed that the membrane with the lowest environmental impacts is the HKUST-1/IL-CS membrane, although it has not the highest permeability. These facts let suggest that the EIIs of the different membranes based on the same permeation area instead of the same CO_2 permeability could lead to different conclusions.

3.2 MMMs with the Same Area

In Fig. 2, the influence of the different permeabilities and selectivities has been removed (the numerical factor is not considered) and the EEI values are compared for the four MMMs with the same area (15 cm^2). Fig. 2 indicates that the contribution of each membrane to the total value of the different 16 EIIs (Tab. S13 gives EIIs numerical values). In this case, the Zeolite A/PTMSP MMM has the highest impacts for 12 of 16 indicators and ETS-10/IL-CS MMM has the highest impact for the rest of indicators. It is clear from these results that in order to improve the environmental impacts of CS-based membranes

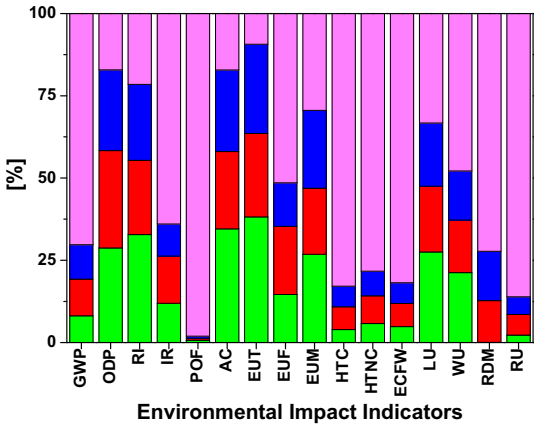

Figure 2. Contribution to the total environmental impact indicator value of the four MMMs with the same area. Green, ETS-10/IL-CS; red, ZIF-8/IL-CS; blue, HKUST-1/IL-CS; magenta, Zeolite A/PTMSP).

the permeabilities have to be increased. Another important conclusion is that the EII of CS-based membranes with the same permeation areas are almost the same independently of the type of filler used (ETS-10, ZIF-8, and HKUST-1).

3.3 Environmental Impacts of Each MMM

In this section, the environmental impacts of individual membrane production have been calculated as a function of the influence of each component on the total value. Fig. 3 illustrates the contribution of each membrane component to the total value of the different 16 EIIs for the 4 MMMs. Tabs. S14–17 present the EIIs numerical values for the four membranes with their respective component contribution.

As it can be seen in Fig. 3, the polymer is the main contributor for the total EIIs on the four MMMs studied. It is worth noting that although CS is obtained from a waste (shrimp shells), the impacts associated to the manufacture of CS from chitin continue to being high. The main cause responsible of these impacts is the high amount of sodium hydroxide and hydrochloric acid that are employed on chitosan deacetylation from the chitin in the shrimp shells (Tab. S5 in SI). Comparing the different CS-based membranes, the CS influence is higher for the ETS-10/IL-CS MMM than ZIF-8 and HKUST-1 MMMs where the fillers manufacture has a larger environmental influence. As to the Zeolite A/PTMSP MMM, the toluene used as PTMSP solvent during the synthesis has a high contribution in the final EIIs.

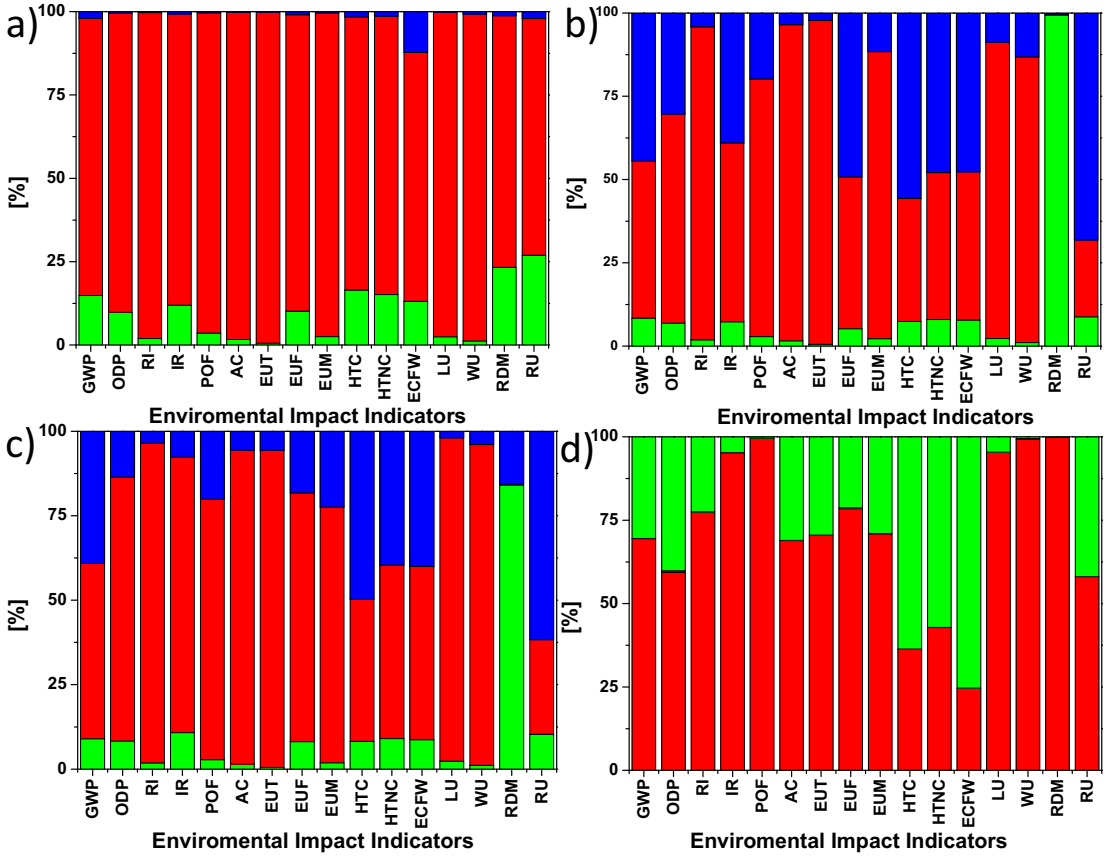

Figure 3. Contribution of each component to the total environmental impact of: (a) ETS-10/IL-CS; (b) ZIF-8/IL-CS; (c) HKUST-1/IL-CS; (d) Zeolite A/PTMSP. Red columns, membrane polymer: chitosan for (a), (b), and (c) and PTMSP for (d). Green columns, ionic liquid for (a), (b), and (c), and toluene for (d). Blue columns: fillers: (a) ETS-10, (b) ZIF-8, (c) HKUST-1, (d) Zeolite A.

3.4 Manufacture Improvement of MMMs

As it has been said before, the permeability is the main factor that determines the results when comparing EIIs of MMMs with the same CO_2 permeation flux. In this section, the permeabilities of the CS-based MMMs are going to be incremented to determine which value is sufficient to make CS-based MMMs environmental impacts comparable to Zeolite A/PTMSP MMMs to obtain the same CO_2 permeation flux. In Fig. 4, the evolution of the contribution of the ETS-10/IL-CS and Zeolite A/PTMSP MMMs to the total EEI values is represented when the permeabilities of the ETS-10/IL-CS MMM are improved by multiplying by a factor of: (b) 10, (c) 100, and (d) 1000. Fig. 4a denotes the initial situation. From Fig. 4 it can be concluded that only when the permeability of the ETS-10/IL-CS MMMs is multiplied by a numerical factor of 1000 (Fig. 4a), the 16 EIIs of this chitosan membrane are lower than those of the Zeolite A/PTMSP MMM.

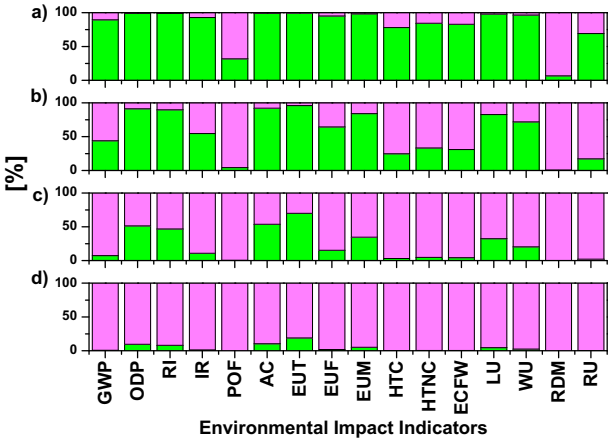

Figure 4. Contribution of ETS-10/IL-CS and Zeolite A/PTMSP to the total EIIs values for the same CO_2 permeation fluxes and different ETS-10/IL-CS permeabilities: (a) initial value of 300 Barrer; (b) initial permeability ×10; (c) initial permeability ×100; (d) initial permeability ×1000. Green columns: ETS-10/IL-CS; magenta columns: Zeolite A/PTMSP MMMs.

In Figs. 5 and 6 the same evolution of the contribution of CS-based MMMs and Zeolite A/PTMSP MMMs to the total value of the different EIIs is demonstrated. In Fig. 5, the ZIF-8/IL-CS permeabilities are multiplied by a numerical factor of (b) 10 and (c) 100, and in Fig. 6b those of HKUST-1/IL-CS are multiplied by a factor of 2. Fig. 5a and Fig. 6a show the initial experimental situations. It can be seen that in order to obtain EIIs lower than the PTMSP MMMs with the CS membranes, it is necessary to multiply the permeabilities of the ZIF-8/IL-CS MMM by a factor of 100 (Fig. 5). For the HKUST-1/IL-CS MMMs, the initial situation is more favorable and only a numerical factor of 2 on their permeability is necessary to reduce EIIs below the values of the PTMSP MMMs (Fig. 6).

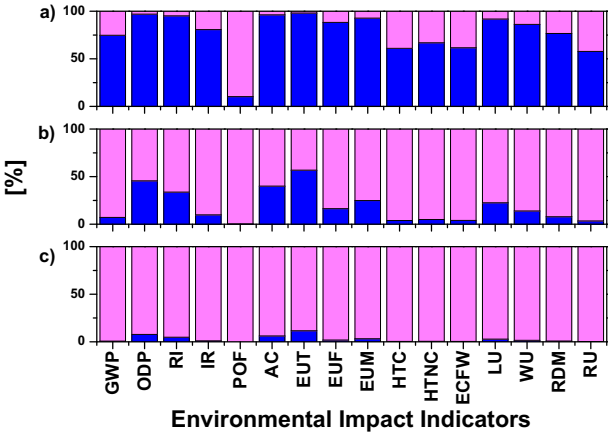

Figure 5. Contribution of ZIF-8/IL-CS and Zeolite A/PTMSP to the total EIIs values for the same CO_2 permeation fluxes and different ZIF-8/IL-CS permeabilities: (a) initial value of 2900 and 13 000 Barrer; (b) ZIF-8/IL-CS initial permeability ×10; (c) ×100. Blue columns: ZIF-8/IL-CS; magenta columns: Zeolite A/PTMSP MMMs.

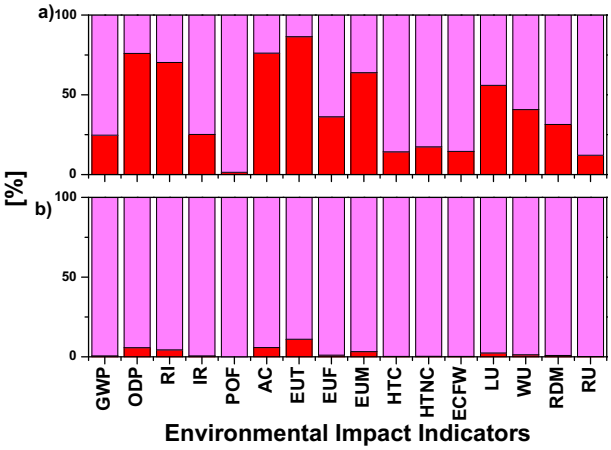

Figure 6. Contribution of HKUST-1/IL-CS and Zeolite A/PTMSP to the total EIIs values for the same CO_2 permeation fluxes and different HKUST-1/IL-CS permeabilities: (a) initial value of 950 and 13 000 Barrer; (b) HKUST-1/IL-CS initial permeability ×2. Red columns: HKUST-1/IL-CS; magenta columns: Zeolite A/PTMSP MMMs.

4 Conclusions

The environmental impacts of CS-based MMMs manufacture have been compared with PTMSP MMM by LCA. For an effective LCA study of membrane manufacture, the functional unit should be the permeation flux of the gas that needs to be separated. As a result, in this study the functional unit used is the CO_2 permeation flux 2.11×10^{-3} cm^3 (STP) GPU/s, i.e., the flow rate obtained with the ETS-10/IL-CS MMM. When comparing different environmental impact indicators, the ETS-10/IL-CS MMM is the one whose manufacture shows the highest impacts due to its lower permeability than the rest of the MMMs studied in this work.

The polymer manufacture (CS and PTMSP) is the main contributor to the EIIs values of the different MMMs. It is worth noting that although CS is obtained from shrimp shells, which is a waste and therefore with less environmental impacts, the synthesis process of CS from chitin significantly contributes to the total value. The EIIs of the manufacture of the ETS-10/IL-CS MMM are comparable to those of the Zeolite A/PTMSP MMM when the permeability of the former is multiplied by a factor of 1000, while this numerical factor is 100 for ZIF-8/IL-CS and 2 for HKUST-1/IL-CS MMMs. This proves the relationship between the membrane composition, permeability, and environmental impact on the development of new membrane materials.

Supporting Information

Supporting Information for this article can be found under DOI: https://doi.org/10.1002/ceat.202200397. This section includes additional references to primary literature relevant for this research [41–44].

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Abbreviations

CS	chitosan
EII	environmental impact indicator
IL	ionic liquid
LCA	life cycle assessment
LCIA	life cycle impact assessment
MMM	mixed-matrix membrane
PTMSP	poly(1-trimethylsilyl-1-propyne)

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