Life Cycle Assessment of Carbon Capture Membrane Separation Technologies

A Thesis

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Abstract

Carbon capture has been a significant topic due to elevating carbon dioxide (CO_2) levels in the atmosphere. CO_2 in the atmosphere is above 420 parts per million (ppm) as of 2022, 70 ppm higher than it was 50 years ago. Carbon capture technologies (CCUs) are being heavily researched to tackle this problem. Researchers have been focusing on developing processes that can capture carbon dioxide from existing sources and utilize it to produce commercial products such as methanol, ethanol, fuels, etc. However, not all these processes may be environmentally feasible, as it has been established that some of these processes end up doing more harm to the environment. Capture technologies such as solvent-based, adsorption-based, cryogenic distillation, and pressure-swing adsorption are currently being researched, but are associated with higher costs and have historically been unsustainable. Membrane-based carbon capture technologies are considered economical and environmentally friendly alternatives. Idaho National Laboratory (INL) researchers have been developing new types of membranes for the last 30 years, one of them being polyphosphazene polymer membranes, specifically Poly[bis((2-methoxyethoxy)ethoxy)phosphazene] (MEEP), which has demonstrated the highest selectivity for CO_2 over nitrogen (N_2). Life cycle analysis (LCA) has become a crucial step in evaluating carbon capture processes for their suitability to be utilized on a small and large scale. An extensive life cycle assessment (LCA) was performed to determine the environmental feasibility of the entire life cycle of the MEEP polymer material compared to other CO_2-selective membranes and separation processes. The MEEP-based membrane processes have been shown to produce at least 42% less equivalent CO_2 emissions than Pebax-based membrane processes. Similarly, MEEP-based membrane processes produce 34–72% less CO_2 than commercial separation processes. In all studied categories, MEEP-based membranes report lower emissions than Pebax-based membranes and commercial separation processes.

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I would like to sincerely thank my mentors at INL, Dr. Daniel Ginosar and Dr. Birendra Adhikari, for providing me the opportunity to work under them, and the knowledge and skills necessary to become a scientist and researcher, without which the completion of this project would have never been possible. I would also like to thank my thesis advisor, Dr. Haiyan Zhao, for her continuous support, guidance, and mentorship, without which I would not have been able to complete my master's degree and thesis.

Dedication

I would like to thank my parents for their unconditional love and support, without whom I would have never succeeded in any facet of life. I would like to thank my grandparents, for raising me like their own, and instilling in me the values that have made me the person I am today. I would also like to thank my brothers, for always believing in me and encouraging me to strive to be the best person I could possibly be each day.

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Statement of Contribution

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Chapter 1: Introduction

Nilkar, A.; Orme, C.; Klaehn, J.; Zhao, H.; Adhikari, B. Life Cycle Assessment of Innovative Carbon Dioxide Selective Membranes from Low Carbon Emission Sources: A Comparative Study. *Membranes* 2023, *13*, x. https://doi.org/10.3390/membranes13040410

Carbon dioxide (CO_2) is the primary contributor to global warming and climate change [1], which is why in recent years many researchers have turned towards carbon capture utilization and sequestration technologies (CCUS). Carbon capture utilization is the process of capturing CO_2 emissions from direct sources, such as oil or coal-fired power plants, for the sake of utilizing it later for other uses such as creating value added products like methanol, acetic acid, and many others. Carbon capture utilization (CCU) and carbon capture sequestration (CCS) are generally used in concepts related to CO_2 management and mitigation of climate change [2], which is why they have been gaining popularity due to high demand for decarbonizing energy production. The Paris Agreement, an international treaty on climate change signed by 194 countries part of the United Nations Framework Convention on Climate Change (UNFCCC), has set a goal of reducing the average global temperature to below 2˚C, and possibly 1.5˚C, as asked for by countries most vulnerable to climate change [3].

One of the ways the USA is trying to tackle this hurdle, is by enabling infrastructure and financial incentives for research and development of CCUS [4]. Carbon capture utilization technologies (CCUs) are becoming increasingly popular due to high demand for developing technologies that can help industrial processes become more environmentally friendly and hence, mitigate climate change. CCUs primarily capture carbon dioxide (CO_2) from processes such as combustion of natural gas and CO_2 removal from flue gas streams, which is either kept for storage (carbon capture storage or CCS) or used as a feedstock for production of value-added products such as chemicals and fuels [5]. However, issues have risen in CCU technologies, where the need to identify ‘sustainable’ options has become necessary to evaluate [6]. ‘Sustainable’ in this context refers to the capability of reducing CO_2 emissions while being economically feasible, which is a key indicator of sustainable development and engineering.

Life cycle assessment **(**LCA) is an approach that has been gaining popularity over the last 2 decades. It is used to analyze the environmental impact of a product at every stage in its “life”, from start to finish, often referred to as “cradle to grave” by researchers and individuals who study and implement LCA [7]. LCA calculates environmental releases for a product, starting with extraction of raw

materials, manufacturing, transport, and the use and disposal of the product. LCA is a very useful and powerful tool in determining the effects caused by a product and the production process of that product, and whether it does more harm to the environment than good, which is why it is being coupled with sustainability analysis in many cases. LCA has a history of being used as a tool to determine the sustainability of energy systems. This has led to a new type of LCA, known as life cycle sustainability analysis (LCSA) [8].

The objective of this study is to assess whether membrane technologies for carbon capture are feasible from an environmental standpoint, and how it compares against other types of carbon capture technologies. This study will focus on analysis of a list of commercial membranes and membranes developed at INL, known as poly[bis((2-methoxyethoxy)ethoxy) phosphazene (MEEP) membranes, and how they compare to each other in terms of environmental impact with respect to their material, equipment used to design the system, and the electricity used to operate the system.

Chapter 2: Literature Review

2.1 State of Art for Carbon Capture Utilization Technologies

In recent years, decarbonization has become a widely studied topic as a method of mitigating climate change, and application of CCUs for obtaining value added products has been one of the main approaches towards achieving this decarbonization [9]. There are many different products that can be obtained from CCUs and carbon capture and storage technologies (CCSs), depending on the type of process involved in the CCU and CCS technology, and depending on the sector that it is involved in. Certain value-added products like methanol produced from CCUs are being studied extensively due to its applications in multiple sectors and products, such as in chemicals, paints, fuels, electricity, etc. The methanol that is produced from these technologies is called green methanol, due to it being made from captured CO_2. However, although it is coming from a renewable source, it is important that technoeconomic assessment (TEA) and LCA studies be conducted to determine the economic and environmental feasibility of these processes.

Cordero-Lanzac et. al [10] conducted a TEA using Aspen Plus® and LCA using Gabi® Pro for production of green methanol with the help of catalysts to determine whether the process was economically and environmentally viable. For the TEA, they assumed annual production to be 275 kton/year of 99.5% pure methanol (MeOH), which falls in line with standard plants. When they evaluated the economic feasibility of the simulated plant, they found that the cost of the raw materials (H_2 and CO_2) were twice the sales of the produced green MeOH, hence making the process unfeasible. They found that 67.5% of the final MeOH price came only from H_2, and CO_2 contributed 16.5%. They summarized that the overall production process is not capital intensive but is highly characterized by raw materials costs which is affected by costs required for compression of H_2.

They also found that with the referenced plant design, the process would be feasible with H_2 prices lower than 1.5 USD per kg with a CO_2 price of 50 USD per ton. If the current prices of 3.5 USD per kg of H_2 were to be used, a carbon tax of minimum 300 USD per ton would be needed to obtain a profit. For the LCA study, the researchers considered two scenarios, 1 in which the methanol was produced from renewable sources and the other process which had non-renewable sources. They found that production of hydrogen was the biggest environmental impact factor for both cases, and that the global warming potential (GWP) for the conventional case was -1.5 kg CO_2/kg MeOH, whereas the second case was higher than maximum threshold of 1.75, hence making the process environmentally infeasible. Hence, this process for producing green methanol is both environmentally

and economically unviable, which they conclude falls in line with recent literature on TEA and LCA of similar work.

Rigamonti et. al [11] conducted a study to determine the feasibility of a CCU technology process which produces methanol from CO_2 and H_2 obtained from steel mill gases for use as fuel in transportation via ships. The researchers conducted 2 sensitivity analyses, one focused on a scenario where the system boundaries were severed at the production of methanol, and the second sensitivity analysis focused on methanol being produced from a specific electricity mix. From their results, they found that in the first sensitivity analysis, there was an improvement in 15 out of 17 environmental impact factors, and that it was abundantly clear that this green methanol could be used as a replacement for conventional marine fuel, and that there were many advantages to doing so. However, they did not conduct a TEA, hence it is unclear whether the process is economically feasible or not. The second sensitivity analysis found that the alternate electricity mix had a worsening effect on the environment compared to the conventional electricity mix, having a worse impact factor for climate change. They concluded that sensitivity analyses were important to make decisions with respect to the impact a new technology will have on the environment and the uncertainties involved, and that more research must be done to specifically focus on uncertainty analyses and how it affects certain parameters and how it would affect the results.

2.2 State of Art for Carbon Capture Utilization Technologies using Membrane Separation

Membrane separation has been gaining popularity in carbon capture applications in the past few years due to their cost advantages, low energy consumption, ease of operation and high selectivity. They are now being extensively researched upon in gas separation due to their ability to enrich or reduce concentrations of desired gases depending on the material of the membrane. Karaszova et. al [12] conducted a review on membranes that were currently being used for CO_2 separation from flue gases and see if they can compete with a Monoethanolamine (MEA) scrubbing technology, which is the standard technology used in most CCS systems [13]. The researchers found from various literatures that certain membranes such as Polaris membranes can compete with MEA scrubbing under mild conditions. However, the selectivity will have to be very high which will affect permeability and costs of the membranes severely. For the most part, there are not many membranes that can compete with MEA scrubbing technologies due to high pressure conditions that will severely affect the performance of the membranes in question. However, the researchers also found that when factors such as toxicity of solvents and overall costs of the conventional processes were considered, membrane separation was the better option due to their low environmental impact. The researchers concluded that membrane separation could be a key process for carbon capture technologies on a large scale in the

future, but more research needs to be conducted to create and optimize membranes to get a good selectivity without compromising
the permeability of the membrane.

Fang et. al [14] conducted a study where they combined a mixed electron and carbonate ion conductor (MECC) membrane with a solid oxide electrolysis cell (SOEC) to capture CO_2 and H_2 and convert them into syngas, all in the same reactor. They then conducted an LCA study on this system and compared it with a conventional MEA plant. The researchers found that the energy consumed by the mixed reactor system was half that of the conventional plant, and the overall energy efficiency of the mixed system was 82%. However, the cost of CO_2 captured was found to be \$119/ton CO_2 captured, which was more than the US-DOE rate. They contribute this to high raw material costs, which is majorly the case in most CCU technologies. The researchers found that the cost of electricity was higher for the mixed system due to the large surface area of the SOEC, which has a linear correlation with the electricity costs. This ended up causing a higher cost for syngas production. The researchers also conducted a sensitivity analysis on renewable electricity prices, and they found that this membrane system can be used to produce synthetic fuel that can compete with biosynthetic fuel systems only if renewable electricity is cheap enough, which was an expected conclusion as seen from various literatures citing the same statement.

Potential methods for capturing CO_2 have been identified; however, it is yet to be widely accepted and used on a commercial scale [15]. Chemical absorption into liquid solvents from gas streams is currently the most popular method for capturing CO_2 [16]. However, the cost, reusability, and life cycle impacts of solvent-based processes make them unpopular [17]. In addition, solvent-based processes often require significant processing equipment, where the cost and life cycle impacts of the capital equipment by itself are very high [18]. This has prompted researchers to lean towards more cost-effective and environmentally sustainable carbon capture alternatives. Technologies such as pressure swing adsorption [19] and cryogenic distillation [20] are also considered alternatives to solvent-based capture but have high economic and energy costs, hence making them unsustainable [21]. Membranes are often more sustainable than other technologies [22–26], are known to have lower environmental consequences, and since no solvents are being used in most membrane technologies, their operating cost is lower [27]. However, the capital cost is generally higher for membrane processes [28]. In comparison to other processes, product purity of membrane processes are generally lower, thus requiring further processing [29]. Membranes have been considered for carbon capture and utilization recently due to their benefits over other systems due to lower carbon emissions, process simplicity, low production cost, ease of operation, compactness, and scale-up

feasibility [30]. Membranes with high CO_2 permeance and CO_2/N_2 selectivity possess great potential to capture CO_2 from any feed gas, including air; however, most membrane technologies still need to be thoroughly investigated for this application [31].

2.3 LCA Applications

LCA is used in analyzing the environmental impacts for virtually any product in any industry, ranging from climate change to depletion of water resources, from the fossil fuel industry to small laboratory scale research efforts. This is because any activity involved in a product's life has environmental impacts, due to resource consumption, emissions of various matter into the environment, environmental exchanges, etc. LCA has become essential in analysis of emerging technologies and the United States-Department of Energy (US-DOE) is asking researchers in certain fields to conduct LCA along with TEA at very early stages of research [32].

Biomass is a renewable, non-conventional, and sustainable energy source that has been gaining popularity in recent years. There is a lot of research and development that is being poured into bioenergy production, whose raw material is biomass. The main reason for this is the increase in greenhouse gas emissions (GHG). It is estimated that the transportation sector accounts for more than 66% of fossil fuel consumption [33] and generates a third of GHG emissions. Hence, by introducing bioenergy and biofuel as an alternative, it will stimulate the economy introducing more jobs, and make the process and supply chain more ecological and hence, more sustainable. An important issue that needs to be addressed in bioenergy production is the bioenergy supply chain, which is the process in which biomass is converted to bioenergy. It is important that the supply chain be analyzed by sustainability metrics i.e social, economic, and ecological impacts. TEA and LCA are tools that are becoming commonly used by researchers and analysts due to their versatility in estimating economic and environmental feasibility respectively. The aim of this paper is to identify and assess the economic and environmental sustainability benefits of bio-oil production from forest biomass using a mixed supply chain, consisting of mixed-pathway transportation and mixed-mode bio-refineries. The authors did this by developing a framework that could evaluate the biomass-based energy supply chain (BESC) by utilizing TEA and LCA to conduct the economic and environmental analyses and hence, judge the feasibility based on the results.

A stochastic optimization model was used for the cost analysis, utilizing a support vector machine (SVM), a supervised machine learning method, which uses past data to predict future trends for parameters of interest. The biomass quality rate was one of the primary indicators for this study, which was based on non-combustibles (ash) content (% of dry weight) and moisture content (% of

wet weight). The other indicator was biomass accessibility rate, which was based on the available amount of biomass and distance of the biomass to the staging site. The ash content, moisture content, available biomass, and distance to staging site define the input information. Also, the biomass quality rate and accessibility rate define the output information for each collection site, of which there were 10 in total. The LCA conducted consisted of a goal and scope, a life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation. The LCI was extracted from SimaPro, an LCA software, and GREET. The authors used global warming potential (GWP) as their parameter, which is an indicator of GHG emissions and climate change. The LCIA was conducted using SimaPro. This 2-phase analysis framework, consisting of TEA and LCA, was applied to a case study which considered 20 collection sites, which were spread across Clatsop, Tillamook, Washington, and Columbia counties.

Parameters such as type of biomass, equipment like trucks, tankers, fixed and mobile bio-refineries, storage capacities, and staging sites, were all used as input variables for both the models, as they all have costs and environmental consequences associated with them. The results indicated that the total bio-oil produced would be around 2.2 million gallons in one year, and the total costs came up to approximately 2.4 million USD. The LCA results indicated that the mixed pathway had lower GHG emissions in all cases that were considered. The authors concluded that the proposed mixed supply chain pathway and BESC framework can improve the sustainability performance of bio-oil production by reducing the cost and environmental impacts but needs to be researched upon more to improve the pathways and make it more sustainable.

An important issue that needs to be addressed in bioenergy production is the bioenergy supply chain, which is the process in which biomass is converted to bioenergy. It is critical that each step in the supply chain be observed and analyzed with care and scrutiny. It is because of this, the researchers in this paper have proposed their own biomass-to-bioenergy supply chain and have conducted a sustainability analysis to assess the feasibility of their process. The researchers devised a supply chain that consists of transportable and fixed bio-refineries and mixed-pathway (truck-tanker and truck-truck) transportation. A transportable biorefinery is a trailer mounted unit that utilizes pyrolysis to produce value added products like bio-oil and biochar from biomass. Their greatest advantage is their ability to produce biochar near the source of the raw material, improving the economic and ecological aspects of the bioenergy process. Since their source is near the raw material, transportable biorefineries reduce the transportation, handling, and storage costs that you would get in a typical stationary biorefinery. This leads to a significant drop in GHG emissions, making the process much

more ecofriendly. Mirkouei et al. [34] conducted a case study where they compared a mixed supply chain with a reference supply chain.

The mixed supply consisted of 20 harvesting sites, five staging sites, two transportable bio-refineries, and one fixed bio-refinery with a storage facility. An LCIA was conducted on both cases using LCA software SimaPro, and it was observed that the mixed pathway reduced GHG emissions by 365,000 kg CO_2 eq, having about 2.3% lower GHG emissions than the conventional bioenergy supply chain. A sensitivity analysis was conducted to understand the effects of parameters such as location and distance of biorefineries, number of trips the trucks had to make, and the effect of biomass on the total impact on the environment. It was found that when the distance between the transportable and fixed bio-refinery, as well as the distance between the staging site and bio-refinery increased by 50%, the global warming potential (GWP) increased by 248,900 kg CO_2 eq and was found to be lower than the traditional pathway by 3.5% for the same increase in distance. When the number of trips the tanks had to take to the bio-refinery and staging areas increased by 100%, there was an increase in 497,800 kg of CO_2 eq, which is an increase of 4.6% compared to the reference case. When the amount and contents of biomass were increased, there was an observed increase of 1,105,000 kg of CO_2 eq in GWP as compared to the reference case. The authors concluded from their results that the location of biorefineries and staging sites with respect to their proximity within the source of raw materials play a vital role in reducing GHG emissions as well as the overall cost of the process, as it also affects the number of trips trucks take to the biorefinery, which will affect fuel and transportation costs and their associated emissions. Optimizing the process will also make the LCA results more attractive.

As observed, there are many different applications of LCA in CCUs, as well as other industries. It is mostly being used in the renewable energy sector to convince researchers and investors that these new technologies are capable of being sustainable, and hence, capable of becoming commercialized to produce clean energy on a large scale. This study particularly focuses on how LCA can be applied to display the benefits of membrane separation in CCU applications.

Chapter 3: Experimental Design and Methodology

3.1 LCA Approach

According to the International Organization for Standardization (ISO), LCA is defined as the "compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle". According to the SETAC (Society for Environmental Toxicology and Chemistry) 'Code of Practice', LCA is divided into 4 stages: scoping, compilation of quantitative data, or commonly known as life cycle inventory (LCI), life cycle impact assessment (LCIA), and life cycle interpretation. [35]. The scoping stage of LCA gives the outline or description of the product in terms of the boundaries and a functional unit. The functional unit is the basis that allows quantities to be analyzed and compared. This is a crucial part of LCA as many researchers and industries use different units to measure the same quantity. Hence, it is important to use a unit that can be understood and used universally.

The second stage is LCI, a procedure used for approximating resource consumption and the amount of waste flows and emissions caused by a product, directly or indirectly at any stage in its life cycle. This stage accounts for the all the material flows, energy flows, and environmental exchanges with respect to the functional unit, that are modelled to represent the inputs and outputs of the product system. The assumptions made during the modelling of the system with respect to the system boundaries and the processes that occur within these boundaries are vital to the results of the LCA study. LCIA evaluates a product's life cycle in terms of the functional unit, for various impact categories such as global warming, ozone depletion, water, and land use, etc. The evaluation is determined by indicators that are quantified with the help of functional units that are then used for analyzing the likely contributions of the resource withdrawal and emission production from the product system, which are used as a basis for potential environmental impacts.

The final stage, life cycle interpretation in practice occurs at every phase in an LCA. If two products are compared and one consumes more resources than the other, then a researcher or practitioner can make a judgement purely based on this criterion. However, it is in the practitioner's best interest to also consider impact categories and environmental consequences, and then make the decision on which of the two products are the better fit.

In this project, the 'cradle to gate' approach is used for the LCA study, which accounts for the greenhouse gas emissions from the source until the point of transportation of the raw materials to the seller/distributor. Since this process is still being researched and the product has not been sold to the customer for use, it is more appropriate to take this approach rather than the conventional "cradle to

grave" approach. The cradle to grave approach accounts for extracting the raw material from the source ("cradle") to the phase where it has been utilized and is of no further use ("grave"). The primary advantage that cradle to gate approach has is that it allows researchers to develop a starting point with respect to the product's lifecycle. The disadvantage that it has is that it does not account for the impact made by the product when it is being used by the customer and it's impact when being disposed, which can be significant, which the cradle to grave approach accounts for. Figure 1 consists of a flowchart that illustrates these 2 LCA approaches. The raw materials refer to the material of the membrane, which is polypropylene, and the gases such as air that will be used for the DAC process. The material processing refers to the capital equipment, which is stainless steel. The sequence within the red dotted lines refers to the cradle to gate sequence or cycle.

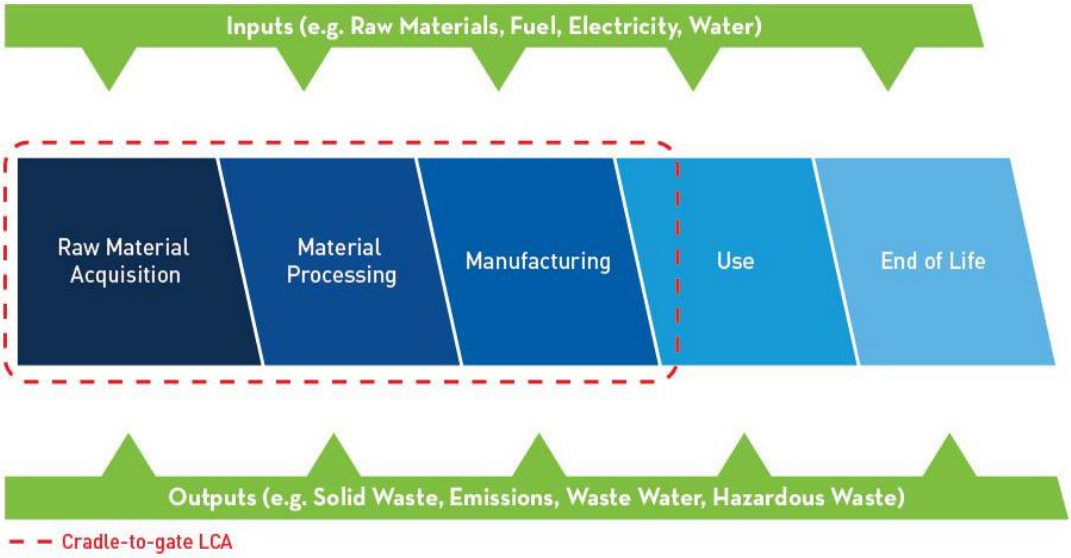


Figure 1. LCA flowchart explaining cradle to grave and cradle to gate approaches [36]

The database that was used for this project was obtained from GREET®, a fuel-cycle model that was developed by Argonne National Laboratory. The model calculates fuel cycle emissions of 5 pollutants: carbon monoxide, nitrogen oxides, sulfur oxides, volatile organic compounds and particulate matter with diameter of 10 microns or less [37], and three greenhouse gases: CO_2, methane and nitrous oxide. The model is also used to calculate the effects of different energy sources and emissions of different transportation technologies, and the different assumptions used to come to their respective conclusions. The values were directly extracted from the GREET® database and displayed

on an Excel spreadsheet, which was used for the subsequent LCIA. Table 1 is an example of the how the GREET database looks when entered in an Excel spreadsheet.

Table 1. LCI obtained from GREET database for 1 kWh of nuclear electricity generated.

Pollutants	Magnitude	Units
CO2 Total	5.584	g
CO2	5.584	g
CO2_Biogenic	0	kg
VOC	2.9147	mg
CO	11.2227	mg
NOx	11.1595	mg
PM10	0.7422	mg
PM2.5	0.5174	mg
SOx	2.6105	mg
CH4	15.674	mg
N2O	0.1084	mg

The LCIA was conducted based on the TEA data that was obtained from the INL team that were part of this project [38], based on the gas separation system that they designed. From the LCIA, the total impact of the system was calculated based on the impact categories that are standard in an LCA study. Impact categories are environmental parameters that are used to represent certain issues based on the data from the LCI or LCA database.

The impact categories that were used in this study are: global warming potential, acidification, respiratory effects, and fossil fuel depletion. These were the impact categories that could be determined from the data obtained from GREET. Global warming potential refers to potential of global warming caused by greenhouse gas emissions into the air by gases such as CO_2, methane, and nitrous oxide. Each has its own potency with respect to CO_2. For example, methane has been determined to be 25 times more potent than CO_2 with respect to its effect on climate change [39]. This impact category is measured in kg of CO_2 equivalent per functional unit. Respiratory effects is a measure of the amount of particulate matter smaller than 2.5 microns or less in width, emitted from the process that can cause respiratory problems. It is measured in kg of PM 2.5 equivalent per functional unit. Acidification potential is an evaluation of the amount of sulfur dioxide (SO_2) emissions caused from the process. It is measured in kg of SO_2 equivalent per functional unit. Fossil

fuel depletion is an indicator of the amount of fossil fuel the process consumes. It is measured in megajoule (MJ) per functional unit.

For each of these impact categories, different gases have different potencies with respect to the reference gases for those categories. To calculate the impact for a category having multiple pollutants, a characterization factor is assigned for each pollutant, which is then added to the amount of reference pollutant that is being emitted to the air. For example, GWP is usually measured in kg CO_2 equivalent, and the category accounts for multiple pollutants, such as methane and nitrous oxide. A characterization factor is assigned to both these pollutants based on the 2007 Intergovernmental Panel on Climate Change (IPCC) manual (25 for methane and 298 for nitrous oxide) The formula is given as follows:

$$\text{Impact (IMP)} = \sum_i M_i * CF_i$$

where, i is the inventory contributing to the impact, M_i is the amount of pollutant being emitted into atmosphere, and CF_i is the characterization factor for inventory i. In this study, inventory i is the input (electricity mixes, capital equipment and membrane material). The impact for each category was calculated by multiplying the percentage contribution of each electricity mix with the emission data for the respective pollutants, obtained from GREET.

The total impact for each category was obtained by adding the individual impacts obtained for each individual input. It can be calculated using the following formula:

$$\text{Total Impact (TIMP)} = \sum_i IMP_i$$

Table 2 summarizes the material and energy inputs to the membrane system. The life cycle inventory (LCI) is for the activities to produce a kg of CO_2 for every kg of CO_2 avoided by the system. The inputs are the same irrespective of how many membranes are used in the system. The distribution of the electricity mix in terms of types of electricity contributing to the mix has been broken down and represented in Figure 2. Approximately 40% of the electricity is produced from natural gas, 20.5% from nuclear energy, 20% from oil, less than 1 % from both biomass sources and coal, and the remaining 20% is produced from other renewable sources such as wind, solar, geothermal, etc.

Table 2. LCI data to produce 1 kWh of electricity from U.S mix, capital equipment, and membrane material.

Input parameters	**CO_2 (kg)**	**CH_4 (g)**	**N_2O (mg)**	**PM 2.5 (mg)**	**SO_2 (g)**	**Fossil fuel (MJ)**
U.S Mix	0.3902	0.854	7.7697	24.5123	0.247	5.463
Capital Equipment	0.7923	1.884	17.7302	563.1	0.9973	12
Membrane Material	1.5456	21.4185	33561.8	120.8	22.0667	73

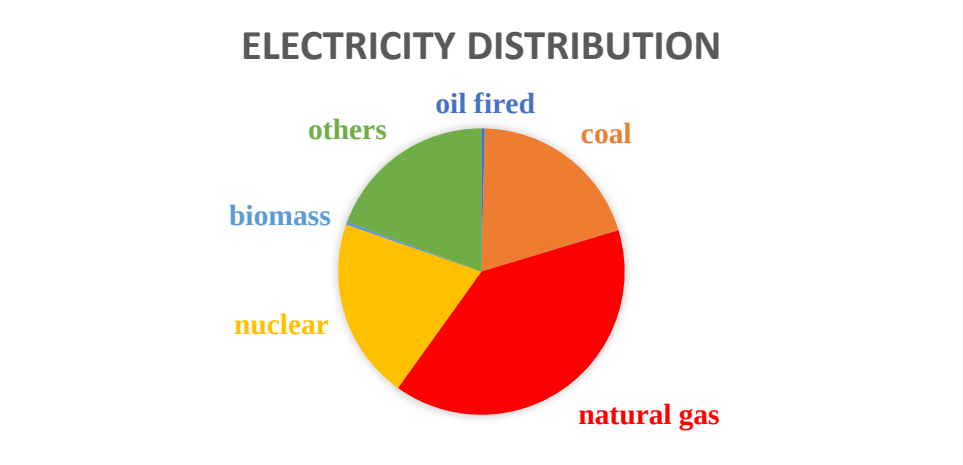


Figure 2. Distribution of energy sources contributing to US electricity mix

The LCI data for the input parameters are associated with the 3 major factors that affect the process: electricity, equipment material and membrane material. The inputs shown in table 2 signify the magnitude of pollutants emitted by the input parameters. For example, 1 kg of stainless steel (capital equipment) required to produce 1 kWh of electricity, emits 0.7923 kg of CO_2, and consumes 12 megajoules (MJ) of fossil fuel energy. Similarly, 1kWh of US electricity mix and 1 kg of membrane material (polypropylene) emit the magnitude of pollutants required to produce 1 kWh of electricity, as mentioned in Table 2. For each of these inputs, the impact parameters being considered are

components of the following impact categories: global warming potential (GWP), respiratory effects, acidification potential, and fossil fuel. The GWP is the most significant impact that will be analyzed, as it will indicate how much CO_2 the system is releasing into the environment for every kg of CO_2 it is capturing from the source.

3.2 CO_2 Separation System using Membrane Units

The gas separation system comprises of a maximum of four membrane units in series (*Figure 3*) [40]. The first membrane unit acts as a prefilter to remove dust and dirt particles that are usually found in air. A compressor is located before this membrane unit. The first gas separation membrane unit follows the prefilter, and a vacuum pump (denoted by VP1, VP2, and VP3) is installed on the permeate side of the membrane to increase mass transport. This is a technique commonly employed in large scale processes. The sizing of the pump (denoted by P1, P2, and P3), prefilter, the first gas separation unit, and the vacuum unit depends on the amount of feed gas that is being supplied and gas selectivity and permeability of the membrane. Flow meters (denoted by FM1, FM2, etc.) are placed at several locations throughout the whole process to monitor gas flow. The membrane modules following the first module are optional and chosen according to the process design. Both the compressor and vacuum pumps for each membrane unit are also optional after the first membrane unit. A second gas separation module follows the first unit sequentially where the permeate of the first unit becomes the feed to the second. In the model, a third unit is also considered and operated similarly. Three membrane units are used for the purpose of increasing the selectivity of the enriched CO_2 in N_2. The first membrane unit achieves 28% selectivity, the second unit achieves 94% selectivity, and the third unit achieves greater than 99% selectivity, hence making the process very efficient.

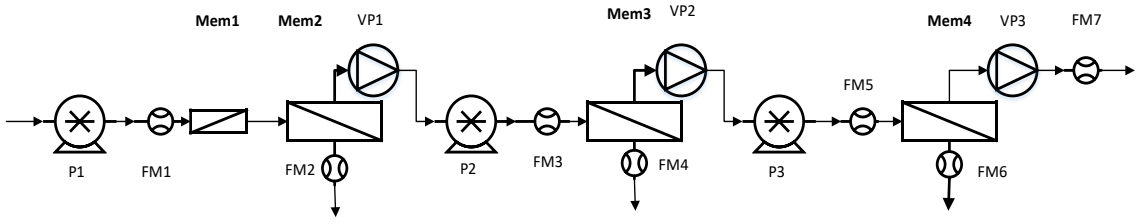


Figure 3. Flow Diagram of gas separation system

The cost of 17 different membranes were calculated in the TEA study, of which 2 were made at INL and the other 15 were made commercially available. The membranes' cost was calculated with respect to a pilot plant scale, and the life of the plant was estimated to be 30 years, as this is the standard timeline estimate. This data was then used for the LCIA. For CO_2/N_2 separation, Pebax elastomers from Arkema are used to develop state-of-art membranes with high CO_2/N_2 selectivity and CO_2 permeability. Emerging CO_2 selective membranes are always compared against pristine Pebax or Pebax derivatives. To quantify the environmental advantages of these membranes over other membrane and separation technologies, a detailed life cycle assessment (LCA) is necessary. A detailed life cycle assessment is performed and reported in this manuscript using the United States Department of Energy (US DOE) developed Greenhouse gases, Regulated Emissions and Energy in Transportation model and database, commonly known as GREET. Techno-economic analysis (TEA) results are reported separately. The basis of CO_2 separation is 1% CO_2 in nitrogen for MEEP membranes, while the basis of separation parameters for other membranes is reported in the literature and illustrated in table 3. Most membranes have used pure gas measurements, and some have used 50/50 CO_2/N_2 mix.

The emission factors obtained from GREET were then multiplied by the estimation factor, that was obtained by dividing the electricity use in kWh/year by the CO_2 avoided in kg/year. It is expected that the overall life cycle impact of the INL membranes, named MEEP membrane and MEEP/CN membrane respectively, will be lower compared to the commercial membranes, as indicated from the TEA data.

Different sources of electricity were used to compare the emissions from different sources of electricity, such as nuclear, geothermal, biomass, natural gas, coal fired, and oil fired. Each source had different impacts on the reference emission factors, known as impact categories in LCA terminology. The functional unit of each impact category is the kg of CO_2 avoided by the membrane system i.e., the net kg of CO_2 that was not released by the membrane system into the atmosphere. A better understanding can be obtained from observing the figure below (*Figure 4*). This figure is a representation of the percentage contribution of each source of electricity as the input of a single stage membrane process using a 100% stacked column bar chart.

Table 3. Details of membranes studied in this project.

Membrane	Membrane Thickness (μm)	Feed Gas (CO_2 volume%/N_2 volume%)	Separation Condition (Temperature ˚C/Pressure Bar)	Permeability (Barrier)	CO_2/N_2 Selectivity
Pebax LE	1	Pure gas	25/1	55.0	40.0
Pebax HE	1	Pure gas	25/1	100.0	70.0
Pebax/ZIF-8	105	Pure gas	25/1	105.0	34.8
Pebax/ZIF-8 (90 nm)	55	Pure gas	25/1	154.0	40.5
Pebax/ZIF-8-90(50)	75	Pure gas	35/--	217.5	54.1
Pebax/NH2-ZIF-8	-	Pure gas	25/1	163.8	62.0
Pebax/UiO-66	18	50/50	25/3	97.2	56.6
Pebax/NH2-MIL-53	75	Pure gas	35/10	120.0	55.5
Pebax/MoS2 nanosheet	28	Pure gas	30/1	52.3	90.6
Pebax/NaY	23	Pure gas	30/2	82.8	35.0
Pebax/NOTT300	38	Pure gas	25/10	395.2	61.2
Pebax/MCM-41	88	Pure gas	25/2	122.5	53.0
Pebax/GO	83	20/80	35/2	105.0	41.2
Pebax/aminosiliane-GO	83	20/80	35/2	166.3	45.2
Pebax/PEI-ZIF-8	1	50/50	25/1	13.0	49.0
MEEP	0.1	99/1	15/1	100.0	40.0
MEEP/CN	0.1	99/1	15/1	100.0	35.0

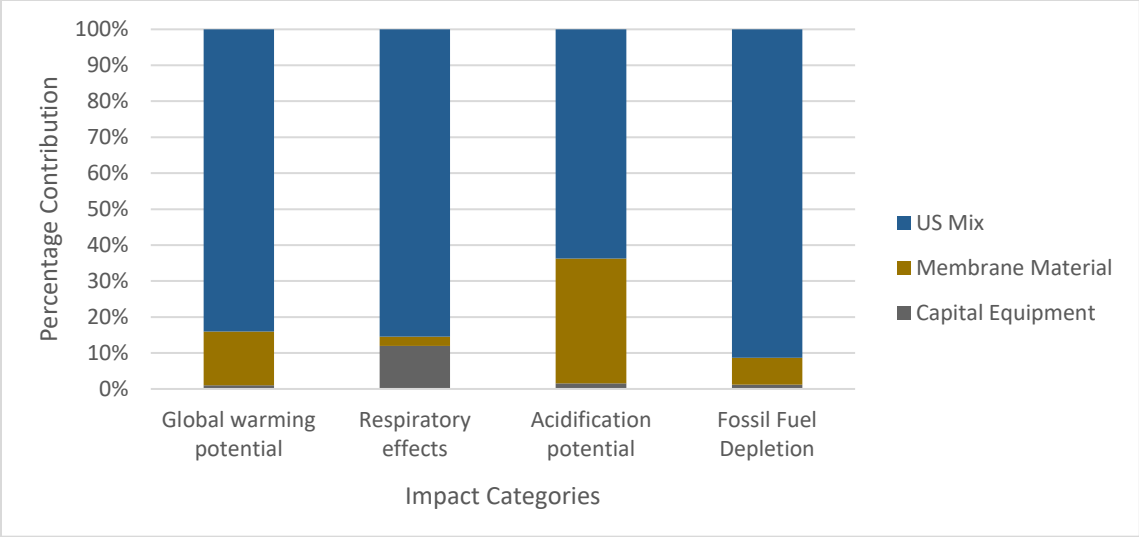


Figure 4. Contributional analysis for single stage membrane process for Pebax LE membrane

The contributional analysis was similarly conducted for MEEP membrane, MEEP/CN membrane Pebax HE membrane, and Pebax/ZIF-8 membrane. This was done to compare the environmental impacts of the MEEP membranes with the commercially used membranes. From the comparison, it was found that the MEEP membranes performed better than both the commercial membranes i.e., their environmental impacts were lower than the environmental impacts of the commercial membranes, except in the fossil fuel depletion category, where all the membranes performed the same. This was because this impact category was solely dependent on the sources of electricity and not the performance or material of the membrane. This analysis was done for single stage, double stage, and triple stage membrane systems. This will be highlighted in the results and discussion section.

Chapter 4: Results and Discussion

4.1 Life Cycle Assessment: Single-Stage Process

A single-stage membrane process is considered to enrich 1% CO_2 in N_2 to 28% CO_2 in N_2. This purity is achieved by recovering 90% of CO_2 present in the feed gas, which is in the permeate. The membrane area and number of membrane modules needed for the process were calculated based on CO_2/N_2 selectivity, CO_2 permeability, membrane thickness and operating parameters such as compressor pressure and vacuum pressure. Figure 5 shows the contribution of each component to the total environmental impact of a single-stage membrane process with MEEP, MEEP/CN, Pebax LE, Pebax HE and Pebax ZIF-8 membranes. The percentage contribution of each input is compared using a 100% stacked column bar chart. It is evident from the chart that capital equipment, membrane material, and electrical energy used in the membrane processes are significant in all the processes that have been studied. While comparing two MEEP-based INL membranes against three Pebax-based membranes in the four referenced environmental impact categories, it is found that for MEEP-based INL membranes, electricity has the highest impact in all the categories studied. Membrane materials have the highest impact on Pebax-based membranes in GWP, AP, and FFDP categories, while capital equipment has the highest impact in the RE category. This indicates that MEEP-based INL membranes use less membrane material and capital equipment, making electricity the highest contributor. Pebax-based membranes use a high amount of capital equipment and membrane material, and as a result, electricity becomes a less significant contributor. Pebax ZIF-8 membrane material has the highest membrane material contribution.

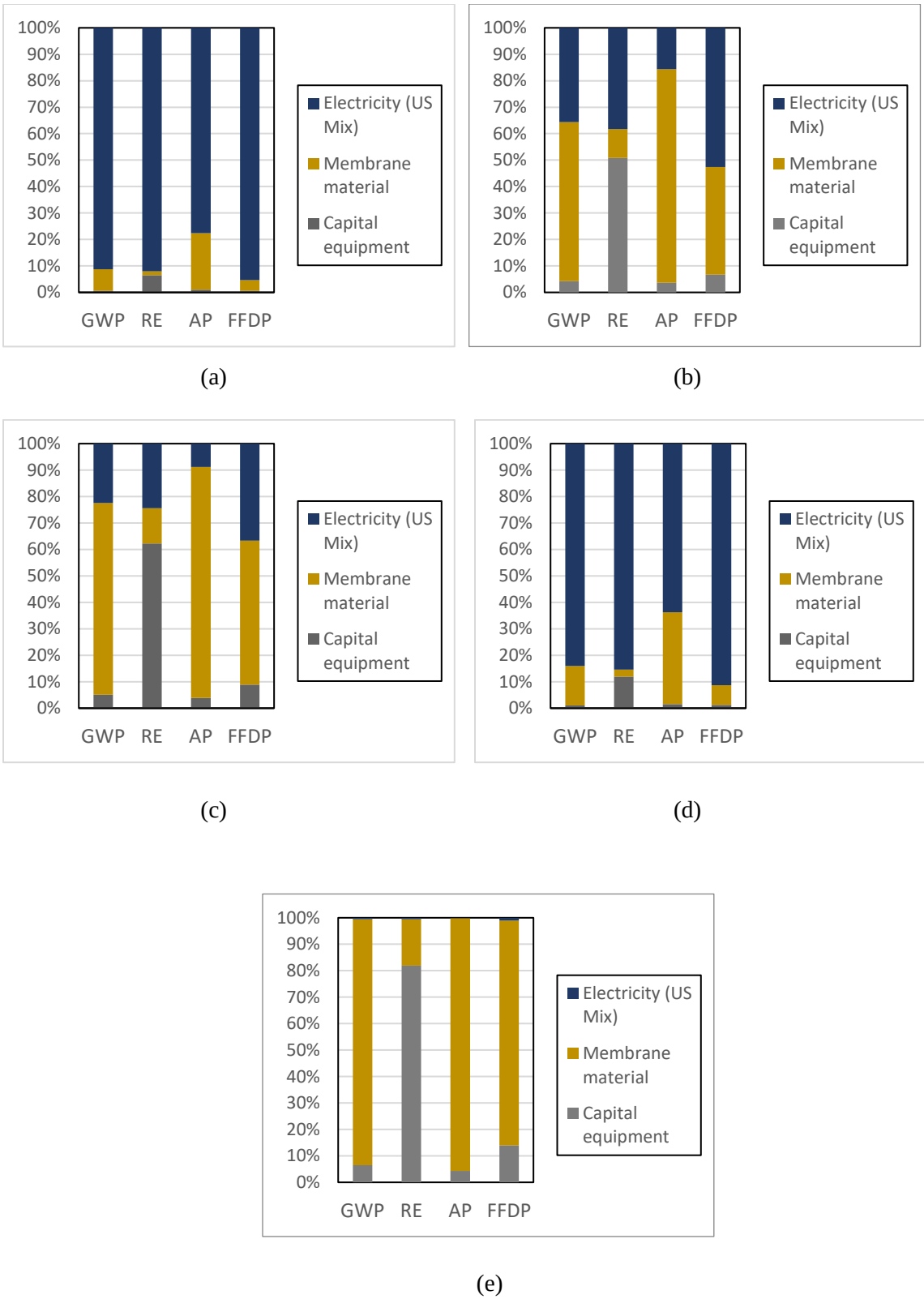


*Figure 5. Analysis of contributions of the single-stage CO_2 enrichment process with (**a**) MEEP, (**b**) MEEP/CN, (**c**) Pebax LE, (**d**) Pebax HE, and (**e**) Pebax ZIF-8 membranes.*

4.2 Life Cycle Assessment: Double-Stage Process

Figure 6 a to e show the percentage contribution of each of the five membranes in a double-stage process using a 100% stacked column bar chart. In this case, CO_2 is enriched from 1% in N_2 to 94% CO_2 in N_2 in two stages. Each stage has a 90% recovery of CO_2. All other parameters are the same as in the single-stage process.

Membrane material and capital equipment contribute the most significant environmental impact for the three Pebax-based membranes, but electricity still contributes the highest environmental impact for MEEP-based membranes. This trend is like that seen in the single-stage process, indicating that the MEEP-based INL membrane processes use far less membrane materials and capital equipment than the Pebax-based membrane processes.

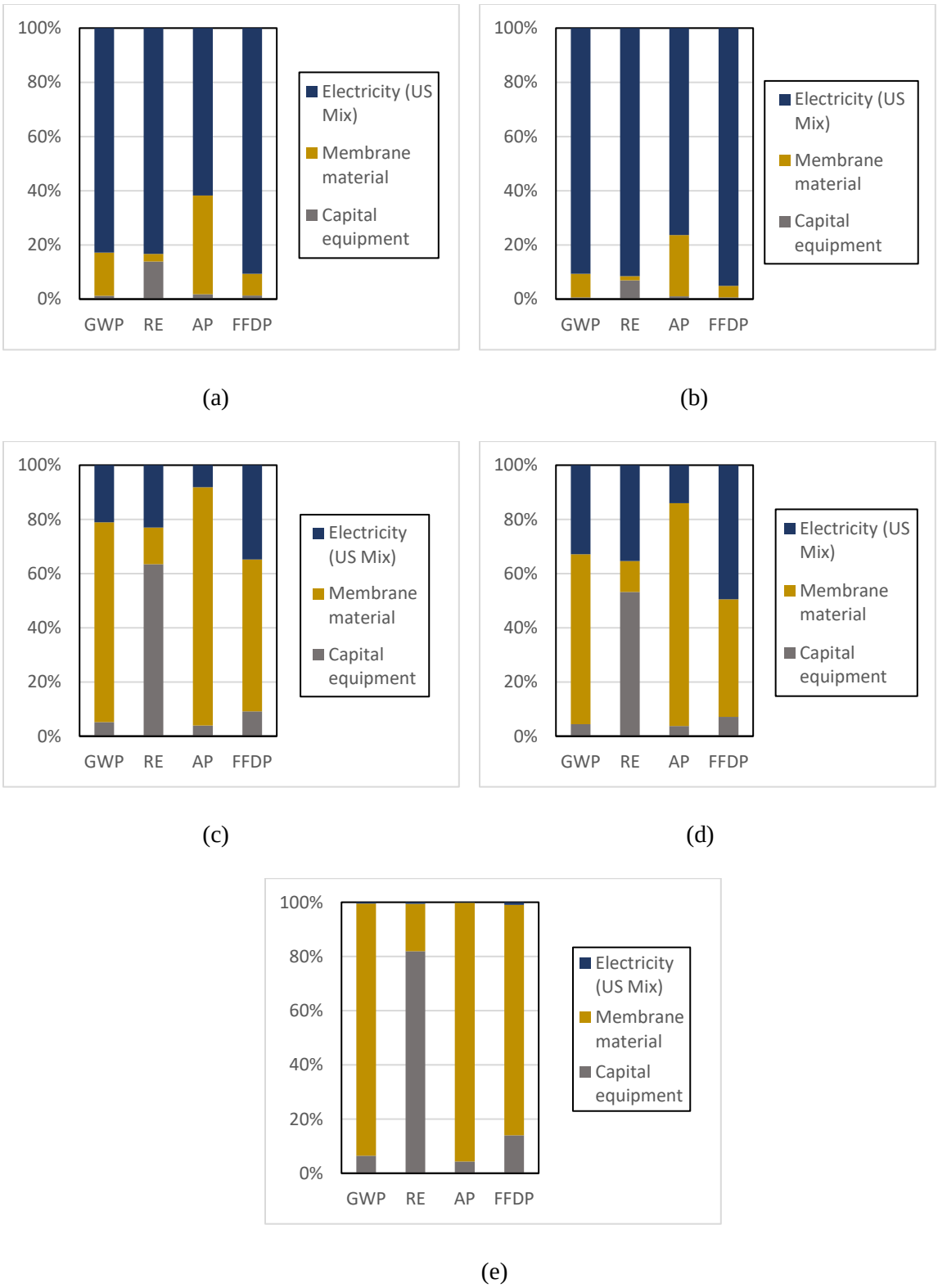
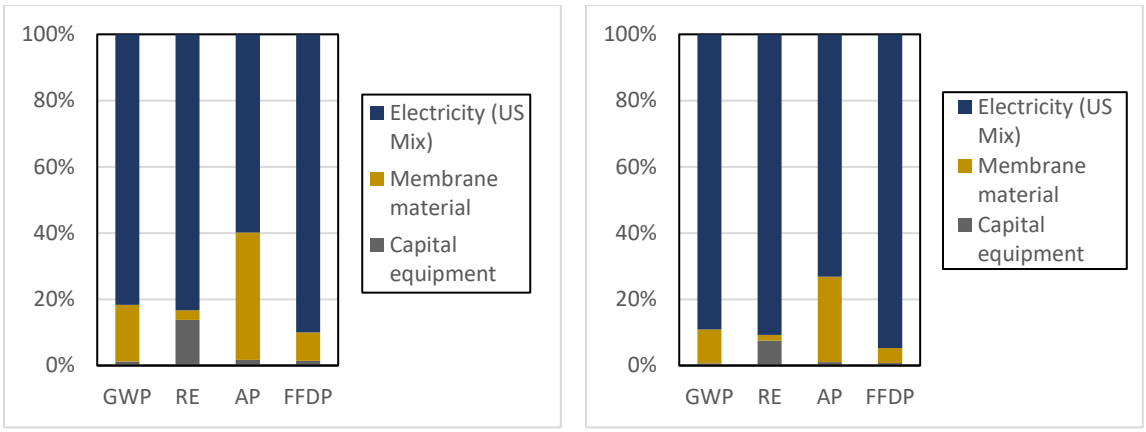


*Figure 6. Analysis of contributions of the two-stage CO_2 enrichment process with (**a**) MEEP, (**b**) MEEP/CN, (**c**) Pebax LE, (**d**) Pebax HE, and (**e**) Pebax ZIF-8 membranes.*

4.3 Life Cycle Assessment: Triple-Stage Process

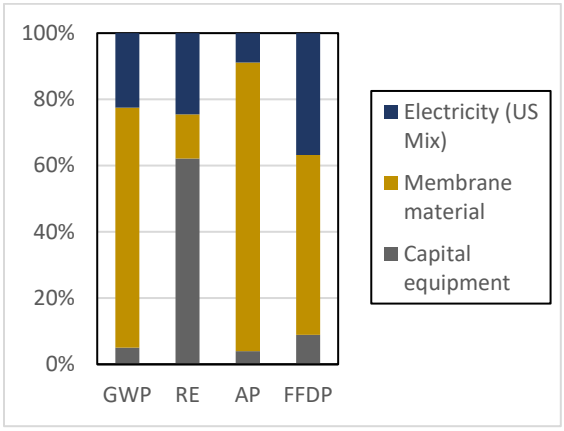
Figure 7 a to e show the percentage contribution of each of the five membranes in a triple-stage process using a 100% stacked column bar chart. CO_2 is enriched from 1% CO_2 in N_2 to greater than 99% CO_2 in N_2 using three stages and 90% CO_2 is recovered in each stage. All other parameters remain the same as in the single-stage and double-stage membrane processes.

Figure 8c–e indicate that membrane material and capital equipment contribute the greatest impact amongst the three Pebax-based membranes, compared to the two MEEP-based membranes (Figure 8a,b), where electricity contributes the greatest environmental impacts. This trend is like those in the single and double-stage membrane processes. This suggests that the MEEP-based INL membranes perform relatively consistently at varied purity levels. Capital equipment requirements drive membrane-based carbon capture processes, while MEEP-based capture processes are driven by electricity consumption from the viewpoint of life cycle impact.

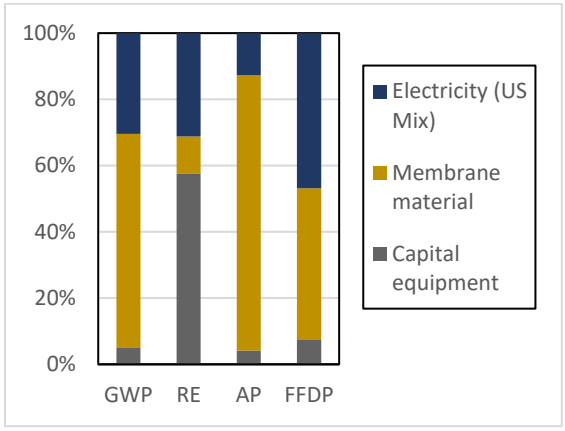


(a)

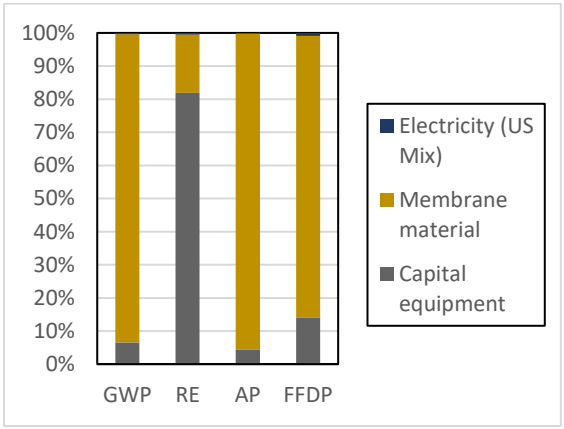
(b)



(c)



(d)



(e)

*Figure 7. Analysis of contributions of the three-stage CO_2 enrichment process with (**a**) MEEP, (**b**) MEEP/CN, (**c**) Pebax LE, (**d**) Pebax HE, and (**e**) Pebax ZIF-8 membranes.*

4.4 Comparison to Other Membrane Processes

MEEP-based INL membranes were compared against both Pebax-based membranes, as shown in Table 4. These results represent a single-stage system with CO_2 enriched from 1% CO_2 in N_2 to 28% CO_2 in N_2 in a single stage. The separation properties of these membranes are given in table 3. Four impact categories are computed for each membrane and compared against one another. Among all the analyzed membranes, MEEP and MEEP/CN membranes have the lowest impacts in all categories. Alone, the MEEP membrane has GWP emissions of 4.40×10^{-2} kg CO_2 eq/kg CO_2 avoided, and the MEEP/CN membrane has GWP emissions of 4.42×10^{-2} kg CO_2 eq/kg CO_2 avoided. State-of-the-art membranes Pebax LE and Pebax HE membranes have GWP emissions of 0.163 kg CO_2 eq/kg CO_2 avoided and 7.53×10^{-2} kg CO_2 eq/kg CO_2 avoided, respectively. This suggests that from GWP standpoint, MEEP membranes show a 73% performance improvement over the Pebax LE membrane and a 42% performance improvement over the Pebax HE membrane. In all other categories, MEEP and MEEP/CN membranes perform 50% better than Pebax LE and Pebax HE membranes and outperform all other membranes.

Table 4. Comparison of environmental impacts for different membranes.

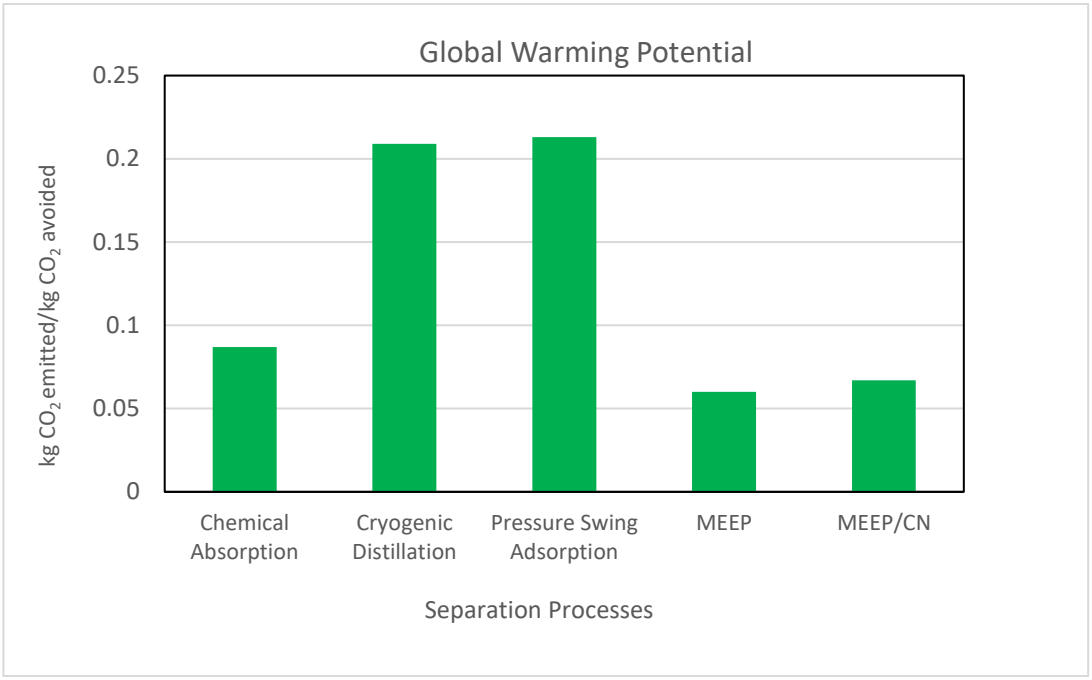
Impact Category	**Global Warming**	**Respiratory Effects**	**Acidification Potential**	**Fossil Fuel Depletion**
Unit	kg CO_2 eq/kg CO_2 avoided	kg PM2.5 eq/kg CO_2 avoided	kg SO_2 eq/kg CO_2 avoided	MJ surplus/kg CO_2 avoided
MEEP	4.40×10^{-2}	5.22×10^{-6}	5.70×10^{-5}	0.566
MEEP/CN	4.42×10^{-2}	5.48×10^{-6}	5.55×10^{-5}	0.592
Pebax LE	0.163	1.15×10^{-5}	2.69×10^{-4}	1.35
Pebax HE	7.53×10^{-2}	6.10×10^{-6}	1.19×10^{-4}	0.698
Pebax/ZIF-8	8.42	4.18×10^{-4}	1.5×10^{-2}	55.3
Pebax/ZIF-8 (90 nm)	2.49	1.35×10^{-4}	4.42×10^{-4}	1.66
Pebax/ZIF-9 90 (50)	1.98	1.07×10^{-4}	3.51×10^{-3}	13.2
Pebax/NH_2-ZIF-8	6.04×10^{-2}	5.45×10^{-6}	9.12×10^{-5}	0.613
Pebax/UiO-66	1.04	5.77×10^{-5}	1.85×10^{-3}	7.1
Pebax/NH_2-MIL-53	3.5	1.88×10^{-4}	6.23×10^{-3}	23.2
Pebax/MoS_2 nanosheet	2.26	1.21×10^{-4}	4.01×10^{-3}	15.0
Pebax/NaY	2.16	1.17×10^{-4}	3.83×10^{-3}	14.5
Pebax/NOTT300	0.531	3.04×10^{-5}	9.30×10^{-4}	3.71

Table 4 continued.

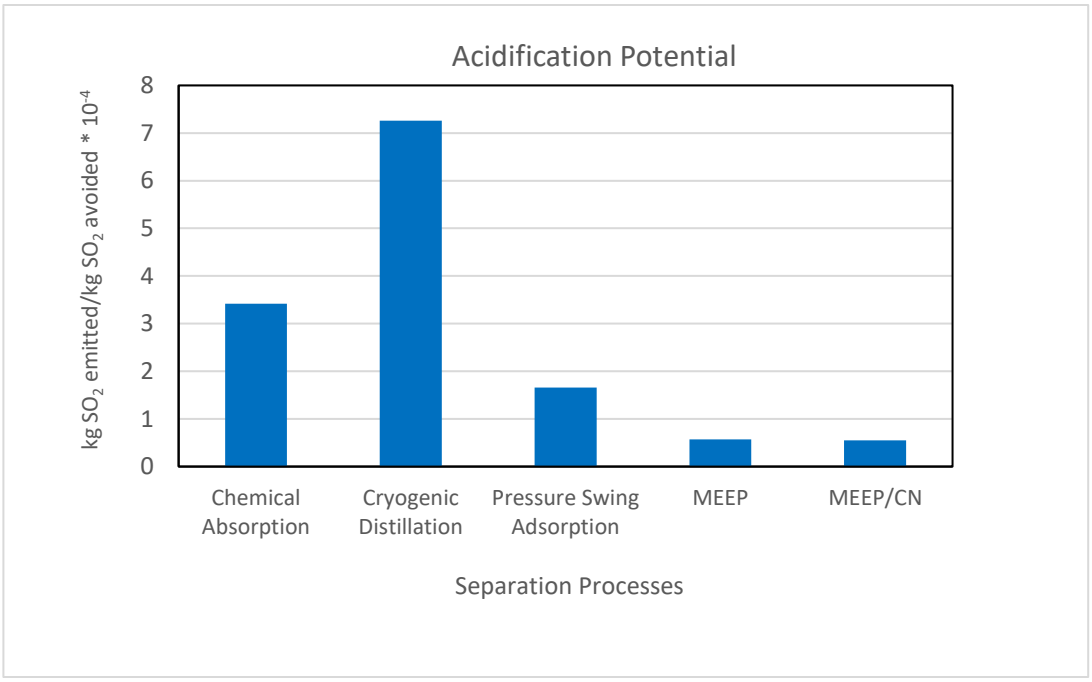
Pebax/MCM-41	4.14	2.21×10^{-4}	7.37×10^{-3}	37.5
Pebax/GO	5.4	2.89×10^{-4}	9.61×10^{-3}	35.8
Pebax/ aminosilane-GO	3.21	1.72×10^{-4}	5.71×10^{-3}	21.4
Pebax/PEI-ZIF-8	0.496	2.88×10^{-5}	8.66×10^{-4}	3.51

4.5 Comparison to Other Membrane Processes

In this section, other major technologies are compared against MEEP and MEEP/CN membrane processes. Given the complexity of each process, GWP and AP for energy use for each separation process were compared. It is important to understand that most of the separation processes are dominated by capital equipment costs while MEEP and MEEP/CN processes are driven by electrical energy demands. Khoo et. al. compared chemical absorption, generic membrane separation, cryogenic distillation, and pressure swing adsorption in terms of equivalent CO_2 emission per 950 kg of CO_2 captured with varying efficiencies [40]. Those results were compared with the MEEP-based membrane process by converting units to kg CO_2 emitted per kg of CO_2 avoided. Regarding GWP, chemical absorption, cryogenic distillation, and pressure swing adsorption processes emit 0.087, 0.209 and 0.213 kg CO_2 per kg of CO_2 avoided, respectively. This suggests that the MEEP-based membrane process emits 34%, 72%, and 72% less CO_2 than chemical absorption, cryogenic distillation, and pressure swing adsorption processes, respectively. For AP, cryogenic distillation, chemical absorption, and pressure swing adsorption emit 7.26×10^{-4}, 3.79×10^{-4} and 2.06×10^{-4} kg SO_2 per kg of CO_2 emitted, respectively. This implies that the MEEP-based separation process emits 95%, 91%, and 83% less SO_2 than cryogenic distillation, chemical absorption, and pressure swing adsorption separation processes, respectively. The following Figure 8 summarizes carbon capture and utilization (CCU) comparisons.



(**a**)



(**b**)

*Figure 8. (a) Comparison of GWP of CCUs. (**b**) Comparison of AP of CCUs.*

4.6 Sensitivity Analysis

A sensitivity analysis was conducted, and tornado charts were plotted based on a 10% change in the inputs to the system in terms of GWP [42]. This analysis helps to identify the most sensitive input for a specific impact category. The five membranes considered for the staging evaluations were again considered for sensitivity analysis. A single-stage membrane process is the basis of the sensitivity analysis.

For the MEEP membrane process, a 10% increase in electricity and membrane material demands resulted in an 8.42% and 1.47% increase in GWP, respectively. However, a 10% increase in capital equipment requirements resulted in an increase of 0.1% in GWP. For the MEEP/CN membrane single-stage process, a 10% increase in electricity demand resulted in a 9.14% increase in GWP. Similarly, a 10% increase in membrane material and capital equipment requirements resulted in only 0.81% and 0.05% increases in GWP, respectively. This indicates that the electricity required to drive the process is the most significant factor affecting the MEEP-based membrane process.

For the Pebax LE membrane process, a 10% increase in membrane material requirements resulted in a 7.22% increase in GWP, while a 10% increase in electricity consumption resulted in a 2.27% increase in GWP. A 10% increase in capital equipment requirements resulted in an increase in GWP of 0.51%. For the Pebax HE membrane process, a 10% increase in electricity and membrane material demands resulted in a 3.61% and 5.98% increase in GWP, respectively. However, a 10% increase in capital equipment requirements resulted in only a 0.42% increase in GWP. Similarly, for the Pebax ZIF-8 single stage process, a 10% increase in membrane material requirements resulted in a 9.30% increase; a 10% increase in capital equipment and electricity requirements resulted in only a 0.65% and 0.05% increase in GWP, respectively. This once again indicates that for MEEP-based membrane processes, electricity is the most dominant factor. In contrast, for Pebax-based membrane processes, the membrane material is the most dominant factor from the standpoint of GWP emissions. Figure 9 summarizes these results.

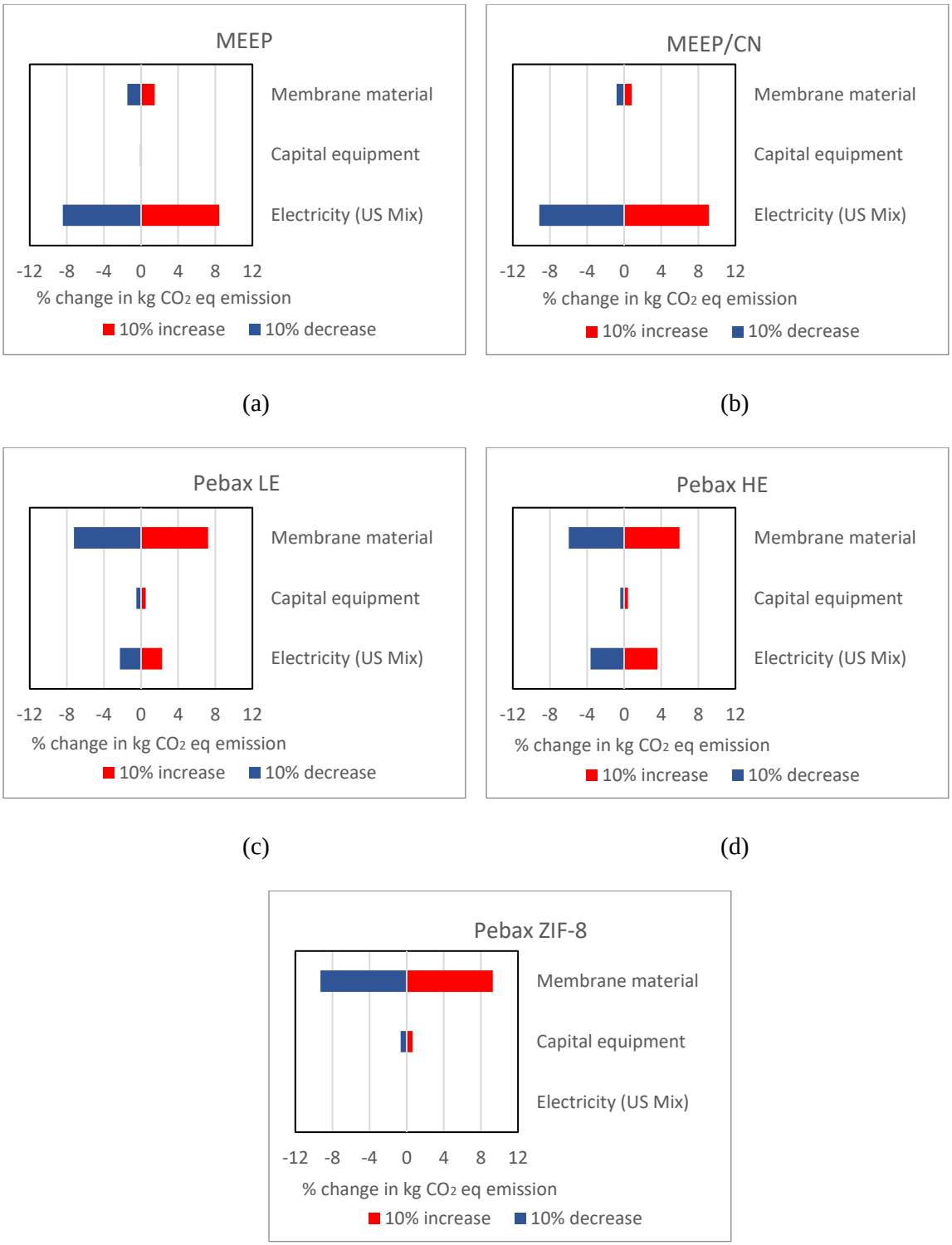


*Figure 9. Sensitivity analysis of GWP by increasing and decreasing by 10% membrane material, capital equipment, and electricity requirements of (**a**) MEEP, (**b**) MEEP/CN, (**c**) Pebax LE, (**d**) Pebax HE, and (**e**) Pebax ZIF-8 membrane processes*

Chapter 5: Conclusion

A comprehensive LCA of gas separation processes was performed using the GREET database by developing a Microsoft Excel-based model. Multiple scenarios of the CO_2/N_2 separation using Pebax-based membranes were compared with MEEP-based INL membranes in single-stage, two-stage, and three-stage processes. The results obtained in this study suggest that the MEEP-based INL membranes outperform Pebax-based membranes in all studied categories for a feed mixture of 1% CO_2/99% N_2 mixture. The most significant contributor to GWP and all other LCA metrics for the Pebax-based membrane processes is the membrane material utilized. For the MEEP-based INL membrane processes, electrical energy consumption is the most significant contributor to GWP. The MEEP-based membrane processes emit at least 42% less equivalent CO_2 than the best-performing Pebax-based membrane process. Similarly, MEEP-based membrane processes produce 34–72% less CO_2 than conventional state-of-the-art separation processes such as cryogenic distillation, pressure swing adsorption, and chemical absorption. The two MEEP-based membranes emit lower emissions in all major categories studied than all Pebax-based membranes and other state-of-the-art conventional CO_2 separation processes.

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