Supplementary Information (SI)

Enhancing Sustainable Energy Production Through Co-Polyamide Membranes for Improved Pressure-Retarded Osmosis Performance and Environmental Impact: Synthesis and Life Cycle Analysis

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S1. Laboratory Bench-Scale PRO Setup to Measure Power Density

A laboratory bench-scale Forward Osmosis (FO)/Pressure Retarded Osmosis (PRO) system was meticulously designed and assembled to facilitate systematic evaluation of membrane performance in both FO and PRO processes. This apparatus allows the application of elevated pressure on the membranes, while concurrently evaluating their transport characteristics and quantifying the extractable power density. The experimental apparatus is illustrated in **Figure 2**, incorporating a detailed schematic flow diagram. In the experimental setup, the feed solution's circulation flow and the draw solution's constant operating pressure are maintained by a high-pressure positive displacement pump, controlled through bypass valves. To reduce pulsation and ensure consistent discharge flow and pressure, pulsation dampeners are strategically placed subsequent to the pumps. Given the high corrosivity associated with saltwater, tubing within the system is constructed from food-grade stainless steel 316, with a diameter of 1/8 inches, to ensure durability. On the retentate line of the draw side, a needle valve is installed to regulate solution velocity. For monitoring purposes, the system is equipped with three pressure gauges and two flow meters: the pressure gauges display the inlet pressures on the draw and feed sides, with an additional gauge positioned immediately after the membrane on the draw side to measure trans-membrane pressure, while the flow meters track the flow rates in different lines. In this experimental configuration, both the feed and draw solutions are contained in 6-liter reservoirs. The permeate flow rate is accurately measured using a digital balance with a precision of 0.01 g, through the timed collection of the draw solution. To enable precise inline monitoring, temperature and conductivity sensors are installed on both the draw and feed circuits, providing real-time measurements of temperature and electrical conductivity within the pipes. To regulate the conductivity of the draw solution during operation, a metering pump adds a salt stock solution of

a specified concentration. Additionally, the draw solution's temperature is controlled via a circulating water bath, which includes a coiled stainless-steel tube and a temperature control unit. The system employs a poly (methyl methacrylate) membrane cell, designed for crossflow operation, with an active membrane area of 20.6×10^{-4} m^2.
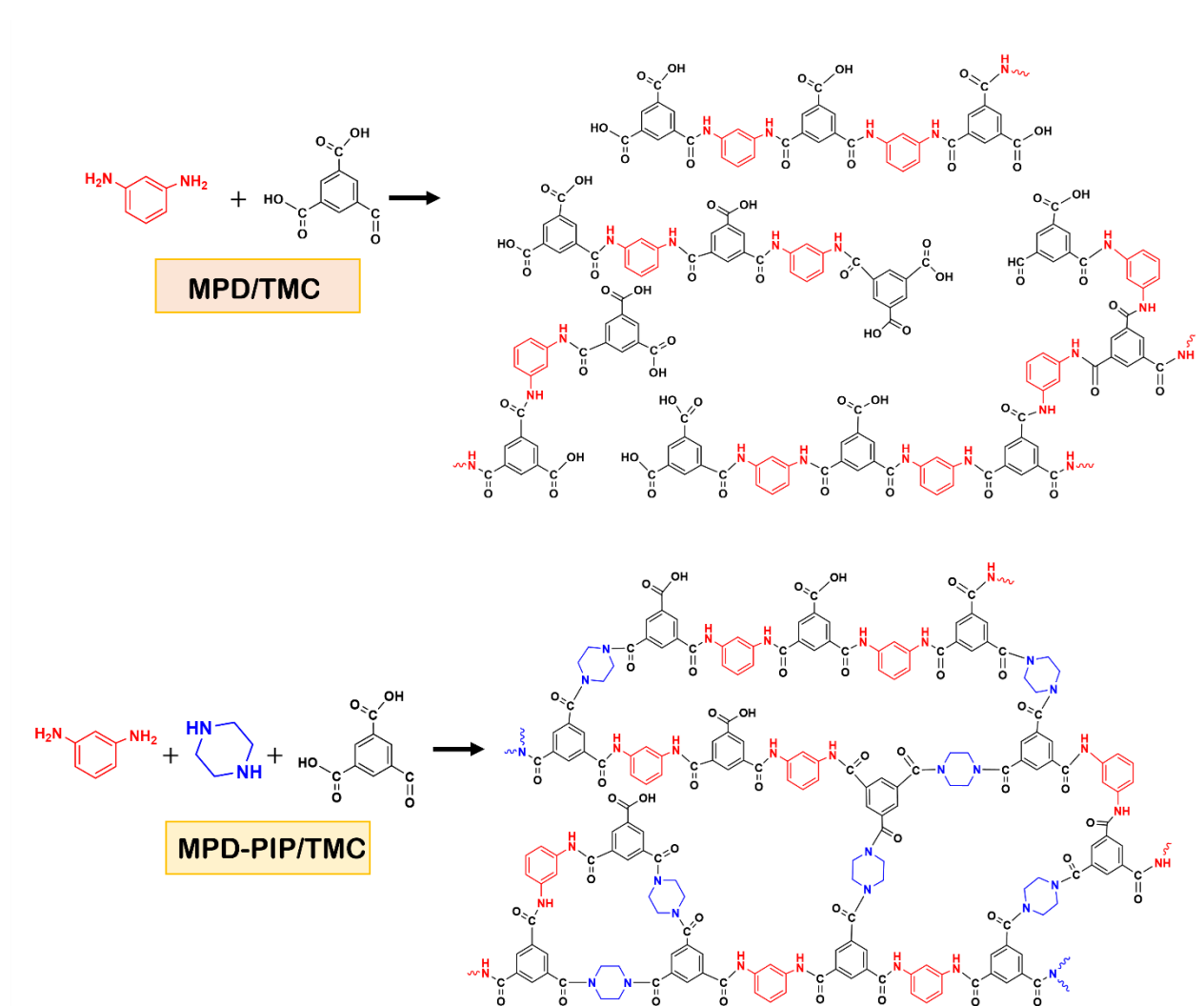

Figure 1S. Possible structure of the resultant mixed polyamide network utilizing a mixture of m-Phenylenediamine (MPD) and piperazine (PIP) as monomers.

The atomic concentrations of elements at the membrane surface are used to quantify the degree of cross-linking in the selective layer, as outlined in Equation (1).[1-3] In this context, m and

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n denote the cross-linked and linear segments of the selective layer, respectively.

$$\text{Degree of Crosslinking} = \frac{m}{m+n} \times 100 \qquad \text{Equation (1)}$$

The values of *m* and *n* can be determined from the experimental O/N ratio obtained through XPS analysis, utilizing Equations (2) and (3).

$$m + n = 1 \qquad \text{Equation (2)}$$

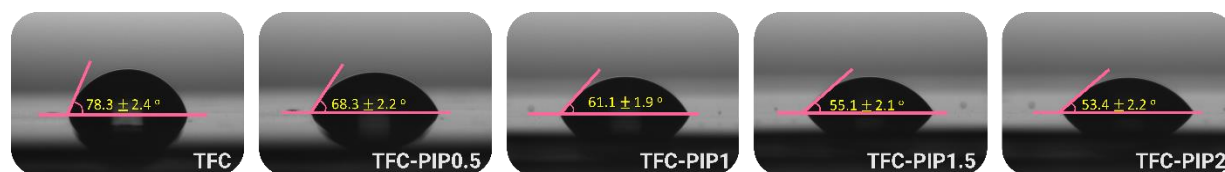$$\frac{O}{N} = \frac{3m+4n}{3m+2n} \qquad \text{Equation (3)}$$

Figure 2S. Water contact angle measurements indicate improved hydrophilicity for TFC-PIP membranes. The presence of the higher loading of PIP into the polyamide membrane further promotes a much rougher surface with roughness up to 328 nm along with a greater presence of unreacted amino groups, resulting in a more hydrophilic membrane surface. The increased surface wettability facilitates higher water flux and better performance in the PRO process.[4]

S2. Life Cycle Assessment Supporting Information

S2.1. The Definition of Environmental Impact Categories

The amount of resource utilization, chemical emissions, and the stressor's predicted potency are taken into account by the underlying techniques in TRACI.[5] The most accurate models and data are used for each impact category to evaluate potency. There is an international agreement on the relative potency of the chemicals listed for some impact categories, such as potential ozone depletion and global warming effects. The relative potency for other impact categories may be based on chemical and physical principles and/or experimental data models. Details on specific effect categories are provided below, along with descriptions that go into further detail regarding the modeling that underpins each category. In some impact categories, the location of the emission

or resource use affects how potent the stressor is, so it is urged to keep track of the location with each stressor. In these situations, each stressor has a different potency factor at each location rather than just one potency factor overall. To determine the overall impact of the study, the calculations should be completed at each location and then added. For instance, if a destiny factor (F) and potency factor (P) are present for an impact category (i), the site-specific analysis might be computed as follows:

$$I^i = \sum_s \sum_x \sum_m F_{xms}{}^i P_{xms}{}^i M_{xms}{}^i$$

(1)

I^i represents all compounds' potential impacts (x) for a particular impact category of concern (i). $F_{xms}{}^i$ shows the fate of chemical (x) emitted to media (m) at the site (s) for impact category (i). $P_{xms}{}^i$ is the potency of chemical (x) emitted to media (m) at the site (s) for impact category (i), and $M_{xms}{}^i$ is the mass of chemical (x) emitted to media (m) at the site (s). It frequently happens that the site-specific location is not being used. For some specific effect categories, for instance, location has little to no impact on fate, transit, and potency; as a result, just one characterization factor is offered for usage globally (e.g., global warming, ozone depletion). In other cases, the precise locations of the emissions for given research are unknown, and as all impact categories permit non-site-specific characterization, the more generic characterization parameters for the site may be applied. The generalized equation in these circumstances, concerning location, would be:

$$I^i = \sum_{xm} CF_{xm}{}^i \times M_{xm}$$

(2)

Here $CF_{xm}{}^i$ is the characterization factor of chemical (x) emitted to media (m) for impact category (i) and M_{xm} is the mass of chemical (x) emitted to media (m).

Ozone depletion, global warming, human health criteria, smog formation, acidification, and eutrophication are examples of conventional pollution categories incorporated into TRACI due to numerous EPA programs and laws and the need to mitigate their effects.[6] To better represent the focus of EPA rules and enable technique development consistent with US regulations, handbooks, and guidelines, the human health category was further separated into cancer, non-cancer, and the six criteria air pollutants (with an initial focus on particulate matter). In the US, smog production is acknowledged as a severe environmental problem, and specific legislation deals with its prevention.[6] Because the environmental effects connected to smog production would have been lost in the process of aggregation, the effects of smog generation were maintained separate and were not further aggregated with other human health impacts. Particulate pollutants in TRACI include different types and sizes of particulate matter (such as PM 2.5 and PM 10), as well as pollutants that have respiratory effects related to particulates (e.g., sulfur oxides and nitrogen oxides). They were kept as a separate human health impact category so that modeling approaches may benefit from the vast epidemiological data linked to these thoroughly researched effects. The US recognizes the importance of the resource depletion categories, particularly for fossil fuels, land use, and water use. Below is the thorough definition of each impact category.

Acidification. Acidification is the process of a local environment becoming more and more saturated with hydrogen ions (H^+). This may be brought on by the introduction of acids (such as nitric and sulfuric acid) or other substances (such as ammonia) that increase the acidity of the environment as a result of various chemical reactions and/or biological activity, or it may result from unavoidable events like a change in soil concentrations brought on by the expansion of local plant species.

Eutrophication. Enriching the aquatic habitat with nutrients (nitrates, phosphates), which

increase biological productivity (growth of algae and weeds) and lead to an unwelcome accumulation of algal biomass, is known as eutrophication. Although nitrogen and phosphorus are essential for fertilizing agricultural lands and other vegetation, their excessive discharges could have unfavorable impacts on the streams they pass through and the areas where they end up. According to the US EPA, nitrogen frequently harms coastal habitats more than phosphorus does to freshwater lakes and streams.

Global Warming. Global warming is the average rise in temperature of the atmosphere close to the surface of the Earth and in the troposphere, which can affect changes in the patterns of the planet's climate. There are many causes of global warming, both natural and caused by humans. In everyday speech, "global warming" frequently refers to the warming that may develop due to elevated greenhouse gas emissions from the human. The term "climate change" is increasingly being used in place of "global warming" to refer to potential changes other than temperature change.

Ozone Depletion. Radiation can increase the incidence of skin cancer and cataracts in humans and is shielded from them by ozone in the stratosphere. Ozone has also been shown to impact human-made objects, marine life, and other plants and animals. Chlorofluorocarbons, used as solvents, foam blowing agents, and fire extinguishing agents, as well as halons, which are utilized as fire extinguishing agents, are substances that have been reported and connected to lowering the stratospheric ozone level. The United States joined the Montreal Protocol more than 20 years ago to reduce chlorofluorocarbons production and adopted even stricter reductions that resulted in the cessation of chlorofluorocarbons production (1996) and halons (1994). Since 1998, global inorganic chlorine levels have been dropping, and the ozone layer is predicted to recover in around 50 years.

Respiratory Effects. Despite the name " Respiratory Effects," this category only includes particulate matter and precursors to particles as its subject matter. Particulate matter is a group of microscopic airborne particles that can have a detrimental impact on human health and even result in death from respiratory diseases. Numerous epidemiology studies demonstrate that elevated levels of ambient particulate matter are associated with increased mortality risk. Particulate matter can be released as particles or produced due to airborne chemical processes (secondary particulates). Nitrogen oxides (NO_x) and sulfur dioxide (SO_2) are the most prevalent precursors to secondary particles. Primary and secondary particulates are frequently produced by burning fossil fuels, wood, and dust from fields and highways. Inhalable coarse particles, which are classified as being between 2.5 micrometers and 10 micrometers in diameter and include road dust, and fine particles, which are classified as being less than or equal to 2.5 micrometers in diameter and frequently the byproducts of combustion, are the two main groups of concern for particulate matter. Persons with asthma, older adults, and other sensitive groups are more likely to have adverse effects. Even stricter standards were imposed in 2006, despite the fact that national US standards have been in existence since 1971.

Carcinogenic, non-carcinogenic, and Ecotoxicity. Any substance, radionuclide, or radiation that encourages carcinogenesis—the development of cancer—is a carcinogen. The Comparative Toxic Unit for Humans (CTUh) measures the estimated increase in morbidity in the entire population of humans per unit mass of a chemical released, serving as an impact indicator for carcinogenicity. According to the definition, ecotoxicity refers to the effects of toxic substances on the urban air, nonurban air, freshwater, seawater, natural soil, and agricultural soil. The Comparative Toxic Unit for ecosystem (CTUe) is the unit indicator for ecotoxicity.

Smog. Smog, often known as smoke fog, is a severe kind of air pollution. The chemical

reactions between sunlight's volatile organic compounds and nitrogen oxides produce ground-level ozone. Respiratory problems caused by human health difficulties can include worsening bronchitis, asthma, and emphysema symptoms. Prolonged ozone exposure may cause permanent lung damage. Crop destruction and harm to diverse ecosystems are examples of ecological repercussions. The primary sources of ozone precursors are electric generating plants, industrial establishments, and motor vehicles.

Ozone and Fossil Fuel Depletion. Fossil fuel depletion refers to the decline in the future availability of fossil fuels caused by their extensive exploitation for fuel, energy production, and the manufacturing of various industrial inputs. Ozone depletion is the slow disappearance of ozone in the upper atmosphere as a result of industry and other human activities that emit chemical compounds with gaseous chlorine or bromine.

S2.2. Detailed LCA Data: Calculations, Inputs, and Assumptions:

Substrate Membrane Fabrication							
Inputs		**Units**	**Product**	**Units**	**Waste**		**Units**
DMF	16.6	g	PES Substrate	1.6	m^2	DMF (in bath)	20	g
PES	3.2	g				DI Water	1980	g
PVP	0.2	g						
DI Water	1980	g						
DMF (in bath)	20	g						
Power (stirrer)	0.15	kwh						

The PES substrate was created via the phase separation technique. This involved casting a solution of PES (16 wt%) and PVP (1 wt%) in DMF (83 wt%), followed by immersing the cast into the coagulation bath containing water and DMF (1 wt%). We consider 5 g of casting solution gives 0.04 m^2 substrate. 3 hours of stirring is considered for casting solution.

TFC - PA Membrane Production							
Inputs		Units	Product		Units	Waste		Units
PES Substrate	1272.4	cm^2	PA membrane	769.6	cm^2	SDS	0.002	g
SDS	0.2	g				MPD	0.005	g
MPD	0.5	g				DI Water	0.983	g
DI Water	98.3	g				TEA	0.01	g
TEA	1	g				TMC	0.004	g
TMC	0.4	g				Hexane	0.996	g
Hexane	99.6	g						
Power (stirrer)	0.025	kwh						
Power (oven)	0.053	kwh						

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution using a roller, the TMC solution comprising 0.4 wt./v% TMC was poured on the membrane surface and kept for 60 sec to allow the polymerization reaction to complete (referred to as pristine TFC membranes without PIP). It is considered that each 5 g of aqueous solution gives one disc 7 cm diameter PA membrane. 5 g of organic solution is considered for each disc of 7 cm diameter. 20 discs are considered for this production. 15 minutes of stirring is considered for each solution. 1 wt% of chemical is considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

	TFC-PIP0.5 – Co-PA Membrane Production					
Inputs		Units	Product	Units	Waste	Units
PES Substrate	1272.4	cm^2	PA membrane	769.6	cm^2	SDS	0.002	g
SDS	0.2	g			MPD	0.005	g
MPD	0.5	g			DI Water	0.978	g
DI Water	97.8	g			TEA	0.01	g
TEA	1	g			TMC	0.004	g
TMC	0.4	g			Hexane	0.996	g
Hexane	99.6	g			PIP	0.005	g
Power (stirrer)	0.025	kwh					
Power (oven)	0.053	kwh					
PIP	0.5	g					

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 0.5 wt% PIP, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution using a roller, the TMC solution comprising 0.4 wt./v% TMC was poured on the membrane surface and kept for 60 sec to allow the polymerization reaction to complete (referred to as TFC-PIP0.5 membranes). It is considered that each 5 g of aqueous solution gives one disc 7 cm diameter PA membrane. 5 g of organic solution is considered for each disc of 7 cm diameter. 20 discs is considered for this production. 15 minutes of stirring is considered for each solution. 1 wt% of chemical are considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

TFC-PIP1.0 – Co-PA Membrane Production							
Inputs		Units	Product	Units	Waste		Units
PES Substrate	1272.4	cm^2	PA membrane	769.6	cm^2	SDS	0.002	g
SDS	0.2	g				MPD	0.005	g
MPD	0.5	g				DI Water	0.973	g
DI Water	97.3	g				TEA	0.01	g
TEA	1	g				TMC	0.004	g
TMC	0.4	g				Hexane	0.996	g
Hexane	99.6	g				PIP	0.01	g
Power (stirrer)	0.025	kwh						
Power (oven)	0.053	kwh						
PIP	1	g						

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 1 wt% PIP, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution using a roller, the TMC solution comprising 0.4 wt./v% TMC was poured on the membrane surface and kept for 60 sec to allow the polymerization reaction to complete (referred to as TFC-PIP1.0 membranes). It is considered that each 5 g of aqueous solution gives one disc 7 cm diameter PA membrane. 5 g of organic solution is considered for each disc of 7 cm diameter. 20 discs is considered for this production. 15 minutes of stirring is considered for each solution. 1 wt% of chemical are considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

TFC-PIP1.5 – Co-PA Membrane Production							
Inputs		Units	Product		Units	Waste		Units
PES Substrate	1272.4	cm^2	PA membrane	769.6	cm^2	SDS	0.002	g
SDS	0.2	g				MPD	0.005	g
MPD	0.5	g				DI Water	0.968	g
DI Water	96.8	g				TEA	0.01	g
TEA	1	g				TMC	0.004	g
TMC	0.4	g				Hexane	0.996	g
Hexane	99.6	g				PIP	0.015	g
Power (stirrer)	0.025	kwh						
Power (oven)	0.053	kwh						
PIP	1.5	g						

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 1.5 wt% PIP, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution using a roller, the TMC solution comprising 0.4 wt./v% TMC was poured on the membrane surface and kept for 60 sec to allow the polymerization reaction to complete (referred to as TFC-PIP1.5 membranes). It is considered that each 5 g of aqueous solution gives one disc 7 cm diameter PA membrane. 5 g of organic solution is considered for each disc of 7 cm diameter. 20 discs is considered for this production. 15 minutes of stirring is considered for each solution. 1 wt% of chemical are considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

TFC-PIP2.0 – Co-PA Membrane Production								
Inputs		**Units**	**Product**		**Units**	**Waste**		**Units**
PES Substrate	1272.4	cm^2	PA membrane	769.6	cm^2	SDS	0.002	g
SDS	0.2	g				MPD	0.005	g
MPD	0.5	g				DI Water	0.963	g
DI Water	96.3	g				TEA	0.01	g
TEA	1	g				TMC	0.004	g
TMC	0.4	g				Hexane	0.996	g
Hexane	99.6	g				PIP	0.02	g
Power (stirrer)	0.025	kwh						
Power (oven)	0.053	kwh						
PIP	2	g						

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 2.0 wt% PIP, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution using a roller, the TMC solution comprising 0.4 wt./v% TMC was poured on the membrane surface and kept for 60 sec to allow the polymerization reaction to complete (referred to as TFC-PIP2.0 membranes). It is considered that each 5 g of aqueous solution gives one disc 7 cm diameter PA membrane. 5 g of organic solution is considered for each disc of 7 cm diameter. 20 discs are considered for this production. 15 minutes of stirring is considered for each solution. 1 wt% of chemical is considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

Substrate Membrane Fabrication (Scaled up)							
Inputs		Units	Product		Units	Waste	Units
DMF	1307.25	g	PES Substrate	30	m^2	DMF (in bath)	0.01	kg
PES	252	g	DMF (in bath)	0.99	kg	DI Water	0.99	kg
PVP	15.75	g	DI Water	98.01	kg		
DI Water	99	g					
DMF (in bath)	1	g					
Power (stirrer)	0.016	kwh					
Power (roller)	0.022	kwh					

The PES substrate was created via the phase separation technique. This involved casting a solution of PES (16 wt%) and PVP (1 wt%) in DMF (83 wt%), followed by immersing the cast into the coagulation bath containing water and DMF (1 wt%). We consider 1575 g of casting solution gives 30 m^2 substrate. 3 hours of stirring is considered for casting solution. The power of the roll-to-roll setup is 65 w. The water bath is designated for reuse, with 1 wt% being accounted for as waste.

TFC-PIP1.5 Scaled up – Co-PA Membrane Production							
Inputs		Units	Product	Units	Waste		Units
PES Substrate	30	m^2	PA membrane	30	m^2	SDS	0.12	g
SDS	12	g				MPD	0.3	g
MPD	30	g				DI Water	59.76	g
DI Water	5976	g				TEA	0.6	g
TEA	60	g				TMC	0.15888	g
TMC	15.888	g				Hexane	39.56112	g
Hexane	3956.112	g				PIP	0.9	g
Power (stirrer)	0.033	kwh						
Power (roller)	0.044	kwh						
PIP	90	g						
Power (air blower)	0.0047	kwh						

For the thin film membrane fabrication, the PES support was first submerged in the MPD solution containing 0.5 wt% MPD, 1.5 wt% PIP, 0.2 wt% SDS, and 1 wt% TEA for 3 min. After removing the excess MPD solution, the membrane surface was soaked with the TMC solution comprising 0.4 wt/v% and kept for 60 s to allow the polymerization reaction to complete (referred to as TFC-PIP1.5 Scaled up membranes). 6 liters of the mixture of 0.4 wt% TMC and 99.6 wt% n-Hexane would weigh approximately 3972 g. 15 minutes of stirring is considered for each solution. 1 wt% of chemical is considered waste. All membranes are subjected to a 4-minute heat curing process in the oven.

Membrane Area Calculations for Energy Generation:

We calculated the energy requirements for a population of 2,000 people over a period of 10 days. Assuming an average daily energy consumption of 25 kWh per person, the total energy needed amounts to 500,000 kWh. To calculate the membrane area required to produce 500,000 kWh of energy over a 10-day period using various membrane types, the following steps are applied: First, convert the target energy production from kilowatt-hours (kWh) to watt-hours (Wh) by multiplying by 1,000 (as 1 kWh equals 1,000 Wh). Next, calculate the total energy required in watts (W) by dividing the watt-hours by the total number of hours over 10 days. Finally, for each membrane type, determine the required membrane area by dividing the total energy output (in W) by the power density (W/m²).

The following steps outline the calculation process:

- Convert the target energy production to watt-hours: 500,000 kWh * 1,000 = 500,000,000 Wh

- Calculate the total energy production needed in watts: 10 days * 24 hours/day = 240 hours

- Total energy production needed = 500,000,000 Wh / 240 hours = 2,083,333.33 W

Now, we determine the membrane area required for each type:

MPD: Area = 2,083,333.33 W / 5.333376 W/m² ≈ 390,697.49 m²

MPD-PIP0.5: Area = 2,083,333.33 W / 7.333392 W/m² ≈ 284,103.22 m²

MPD-PIP1: Area = 2,083,333.33 W / 8.444512 W/m² ≈ 246,681.74 m²

MPD-PIP1.5: Area = 2,083,333.33 W / 10.222304 W/m² ≈ 203,787.24 m²

MPD-PIP2: Area = 2,083,333.33 W / 10.00008 W/m² ≈ 208,333.30 m²

To generate 500,000 kWh of energy in 10 days, we would need approximately:

390,697.49 m² of MPD membrane

284,103.22 m² of MPD-PIP0.5 membrane

246,681.74 m² of MPD-PIP1 membrane

203,787.24 m² of MPD-PIP1.5 membrane

208,333.30 m² of MPD-PIP2 membrane

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