Supporting Information:
New strategies for economically feasible CO_2 electroreduction using a porous membrane in zero-gap configuration

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Simulation and technoeconomic calculation methods

- Unit production rate of CO:

$$\dot{n}_{CO} = \frac{CD \times FE_{CO}}{F \times z_{CO}} \quad [\text{mol s}^{-1}\text{cm}^{-2}]$$

where CD, FE_{CO}, F, and z_{CO}, indicate current density, Faraday efficiency of CO, Faraday constant, and electron number of the CO producing reaction.

- Unit production rate of H_2:

$$\dot{n}_{H_2} = \frac{CD \times FE_{H_2}}{F \times z_{H_2}} \quad [\text{mol s}^{-1}\text{cm}^{-2}]$$

where FE_{H_2} and z_{H_2} indicate Faraday efficiency of H_2 and electron number of the H_2 producing reaction.

- Required electrolyzer cell area A_{cell}:

$$A_{cell} = \frac{\dot{n}_{CO_2}^{in} \times X}{\dot{n}_{CO}} \quad [\text{m}^2]$$

where $\dot{n}_{CO_2}^{in}$ and X indicate molar flowrate of CO_2 into electrolyzer and one-pass CO_2 conversion.

- Base case for carbon capture process

The base carbon capture process that can captures 90% of inlet CO_2 was simulated via Aspen Plus. The CO_2 mass flowrate for the base case capture process was set to 12440 kg/h and the corresponding equipment cost was calculated as \$11.2 M. Cost calculation methods was referred from Seider et al.[1]

- Cost calculation for carbon capture

The equipment installation cost of carbon capture process was assumed to be obtained from the equation of CO_2 mass flowrate as below:

$$C_{cap} = C_{base} \times \left(\frac{\dot{m}_{CO_2}}{\dot{m}_{CO_2,\, base}} \right)^{0.6}$$

where C_{base}, $\dot{m}_{CO_2}$, and $\dot{m}_{CO_2,\, base}$ indicate the equipment cost for carbon capture process, the inlet CO_2 mass flowrate into capture process, and the inlet CO_2 mass flowrate into capture process for the base case.

The operating cost of carbon capture process was calculated using the amount of 50 psig steam required for regeneration of MEA.

$$C_{cap,\, op} = \dot{m}_{CO_2} \times G_{re} \times C_{steam,\, 50psig} \times T_{op} \text{ [\$ year}^{-1}\text{]}$$

where G_{re}, $C_{steam,50psig}$, and T_{op} indicate the unit MEA regeneration energy per ton CO_2, 50 psig steam purchase cost, and the process operating hours in a year (T_{op} = 8,000 hours).

- Capital cost calculation

Total bare-module investment (TBM)	= Total bare-module costs for equipment + costs for computers, and software ($20,000).
Total direct permanent investment (DPI)	= Cost of site preparation (10% of TBM) + TBM
Total depreciable capital (TDC)	= Cost of contingencies and contractor's fee (15% of DPI) + DPI
Total permanent investment (TPI)	= Cost of land (2% of TDC) + Cost of plant startup (2% of TDC) + TDC

* The bare-module costs for equipment were calculated via Guthrie's method[2] and the required data was obtained using Aspen Plus simulation

- Operating cost calculation

Cost Factor		Annual Cost ($)
Operations (labor-related) (Op)		
	Direct wages and benefits (DW&B)	$2,800,000
	Direct salaries and benefits	15% of DW&B
	Operating supplies and services	6% of DW&B
	Technical assistance to manufacturing	$200,000
	Control laboratory	$216,667
Maintenance (Ma)		
	Wages and benefits (MW&B)	3.5% of TDC
	Salaries and benefits	25% of MW&B
	Materials and services	100% of MW&B
	Maintenance overhead	5% of MW&B
Operating overhead		
	General plant overhead	7.1% of *Ma*+*Op*-SW&B
	Mechanical department services	2.4% of *Ma*+*Op* - SW&B
	Employee relations department	5.9% of *Ma*+*Op* - SW&B
	Business services	7.4% of *Ma*+*Op* - SW&B
Property taxes and insurance		2% of TDC
Depreciation		
	Direct plant	8% of (TDC-1.18alloc)
	Allocated plant	6% of 1.18alloc
General Expenses		
	Selling (or transfer) expense	3% (1%) of sales
	Direct research	4.8% of sales
	Allocated research	0.5% of sales
	Administrative expense	2.0% of sales
	Management incentive compensation	1.25% of sales

* The sales (CO sales) and utility costs (electricity, steam, and refrigerants) were calculated based on the results of Aspen Plus simulation

- Cash flow

Cash flow analysis was performed with 15 years plant life, 2 years plant construction period, 5 years of class life MACRS (Modified Accelerated Cost Recovery System) depreciation, 15% nominal interest rate, and 38.9% income tax rate to calculate net present value (NPV).

Table S1. Parameters for sensitivity analysis

Parameter	Unit	1st scenario		2nd scenario	
		PM	AEM	PM	AEM
Unit membrane cost	$ m^{-2}	316 ± 10%	3,167 ± 10%	316 ± 10%	3,167 ± 10%
CO_2 crossover ratio	-	0 ~ 10%	0	0 ~ 10%	0
Current density	mA cm^{-2}	500		100 ~ 2000	
CO Faraday Efficiency		0.9		0.5 ~ 0.99	
One-pass CO_2 conversion		0.1		0.01 ~ 0.5	
Cell voltage	V	3		1.3 ~ 3.5	
Unit electricity cost	$ kWh^{-1}	0.06		0.06 ~ 0.1	
Unit regeneration energy	GJ tonCO_2^{-1}	3 ~ 5			
Operating years	years	15			
Membrane replacement period	years	7			

Table S2. Parameters for technoeconomic analysis

Parameters	Unit	Value
Utility cost		
Steam, 450 psig[1]	\$ kg^{-1}	0.0145
Steam, 150 psig[1]	\$ kg^{-1}	0.0105
Steam, 50 psig[1]	\$ kg^{-1}	0.0066
Process water[1]	\$ m^{-3}	0.2
$KHCO_3$[3]	\$ kg^{-1}	1.38
Refrigeration, -150°F[1]	\$ GJ^{-1}	12.60
Refrigeration, -90°F[1]	\$ GJ^{-1}	10.30
Refrigeration, -30°F[1]	\$ GJ^{-1}	7.90
Refrigeration, 10°F[1]	\$ GJ^{-1}	5.50
Chilled water, 0°F[1]	\$ GJ^{-1}	4.00
Cooling water[1]	\$ m^{-3}	0.02
Direct wages and benefit (DW&B)[1]	\$ $operator^{-1}$ hr^{-1}	35
Number of workers	-	10
Tech assistance to manufacturing[1]	\$ shift $operator^{-1}$ yr^{-1}	60,000
Control laboratory[1]	\$ shift $operator^{-1}$ yr^{-1}	65,000
Operating hour	hr yr^{-1}	8,000
Catalyst prices		
Ag[4]	\$ kg^{-1}	490
Ir[4]	\$ kg^{-1}	46,940
Economic factors		

Plant life	year	15
Construction period	year	2
Income tax	%	38.9
Interest rate	%	15
MACRS	-	5-year class

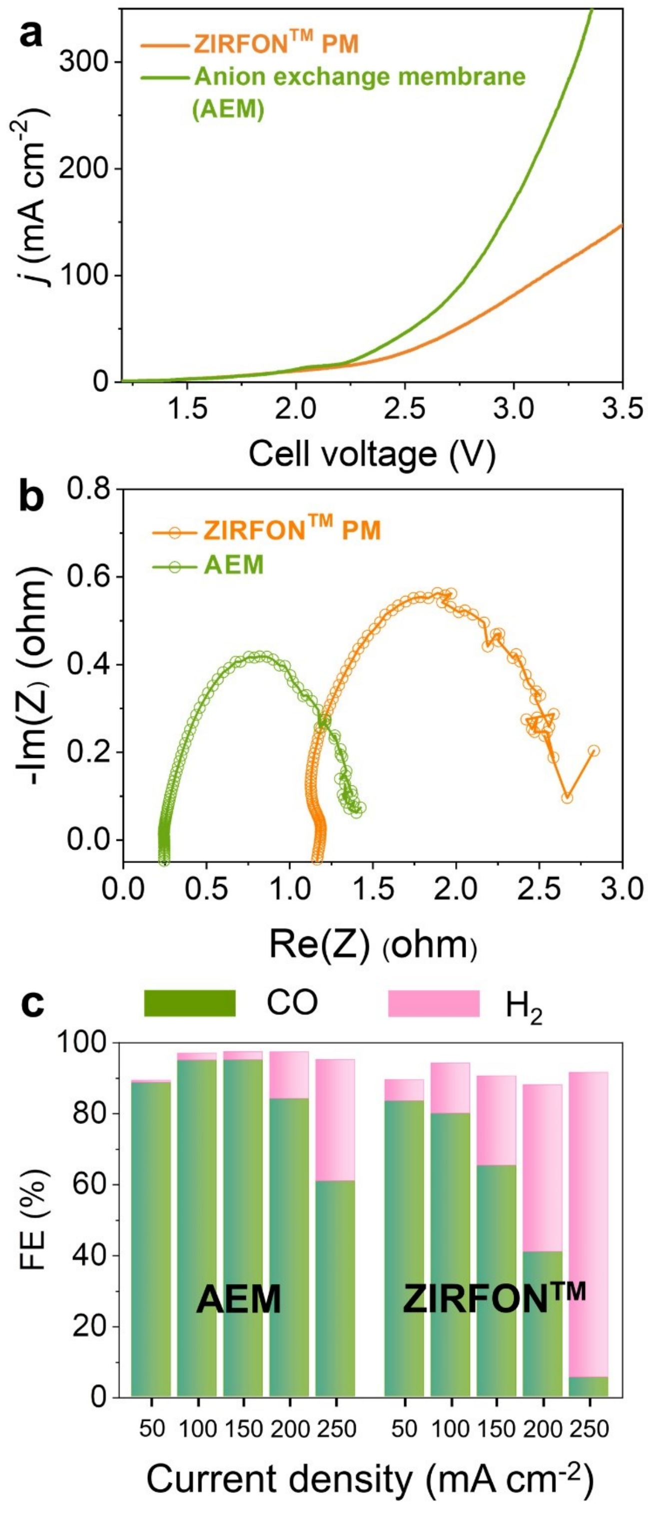

Figure S1. (a) Cell voltage and current density curve, (b) impedance and (c) Faraday efficiency (FE) of commercial ZERFON PERL^{TM} porous membrane applied zero-gap electrolyzer for CO_2RR. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. Anolyte: 0.5 M $KHCO_3$.

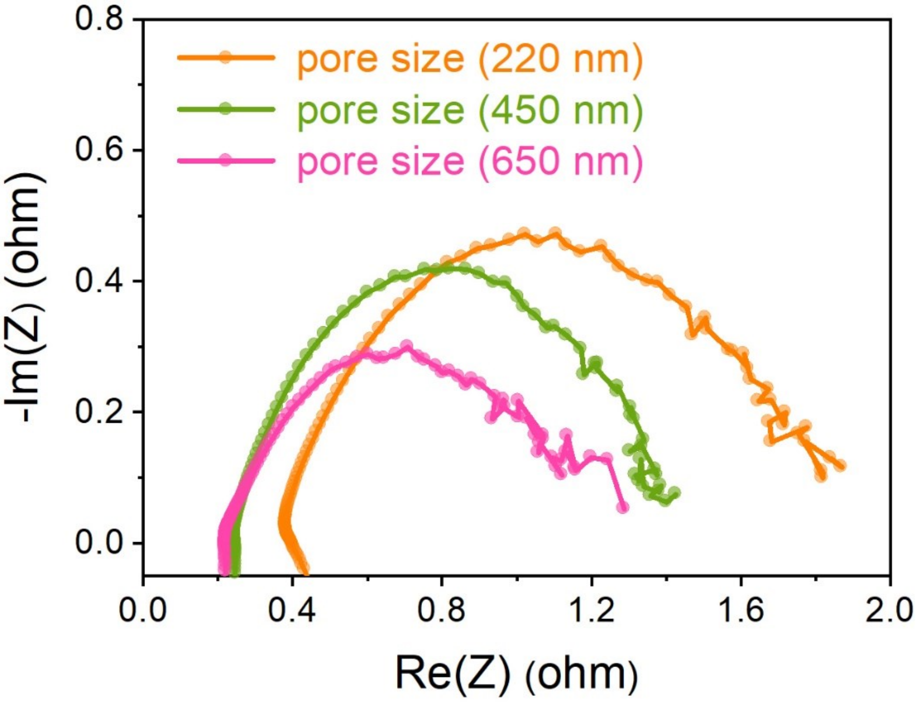

Figure S2. Impedance of porous membrane (PM) applied zero-gap CO_2RR device using Ag electrode with different pore size of PM. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. PM: polyvinylidene difluoride (PVDF) porous membrane. Anolyte: 0.5 M $KHCO_3$.

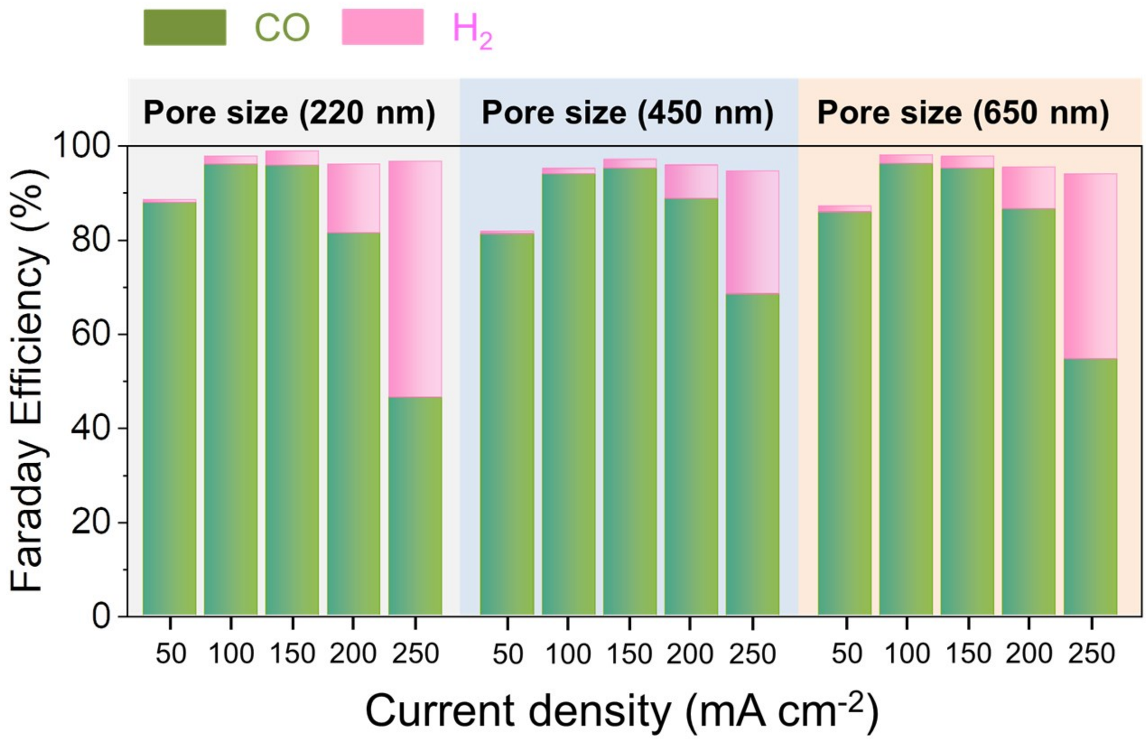

Figure S3. Faraday efficiency of zero-gap electrolyzer for CO_2RR using PM with different pore sizes. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. PM: polyvinylidene difluoride (PVDF) porous membrane. Anolyte: 0.5 M $KHCO_3$.

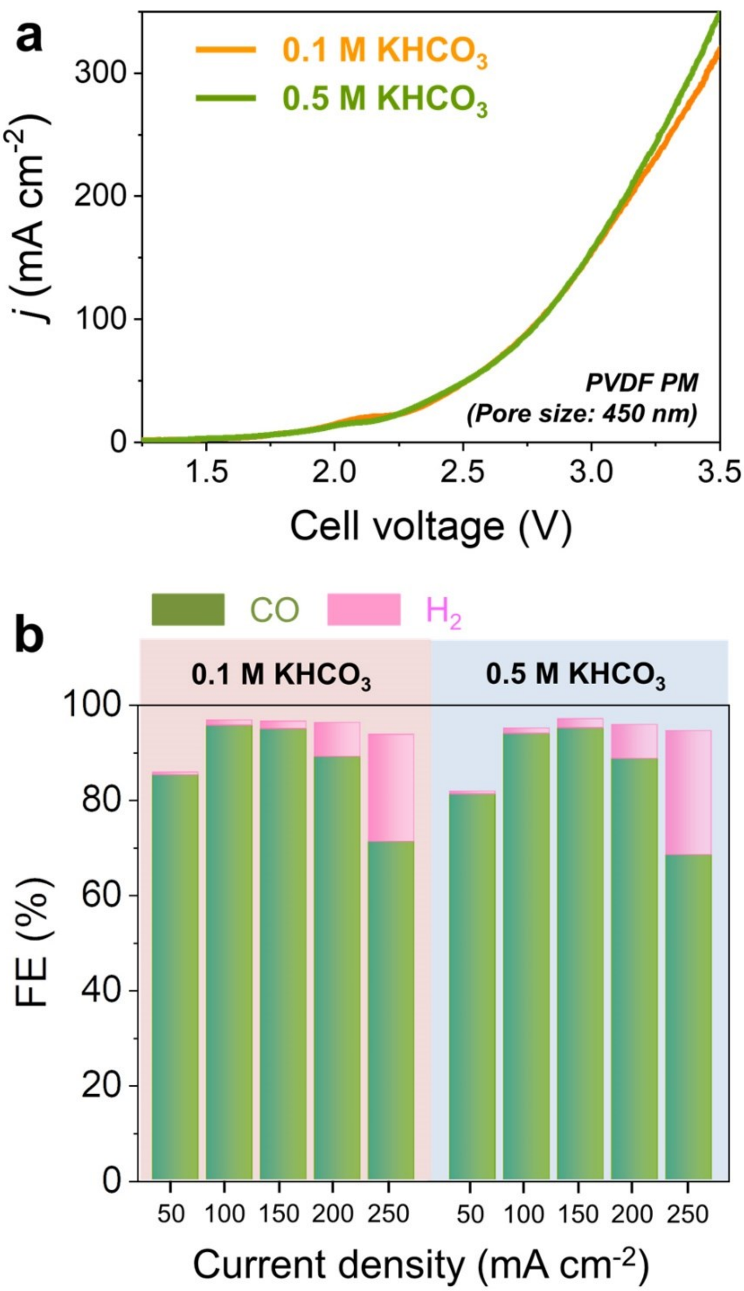

Figure S4. (a) Cell voltage and current density curve, and (b) Faraday efficiency of PM applied zero-gap electrolyzer for CO_2RR. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. PM: polyvinylidene difluoride (PVDF) porous membrane, 450 nm pore size, 125 μm thickness. Anolyte: 0.1 M and 0.5 M $KHCO_3$.

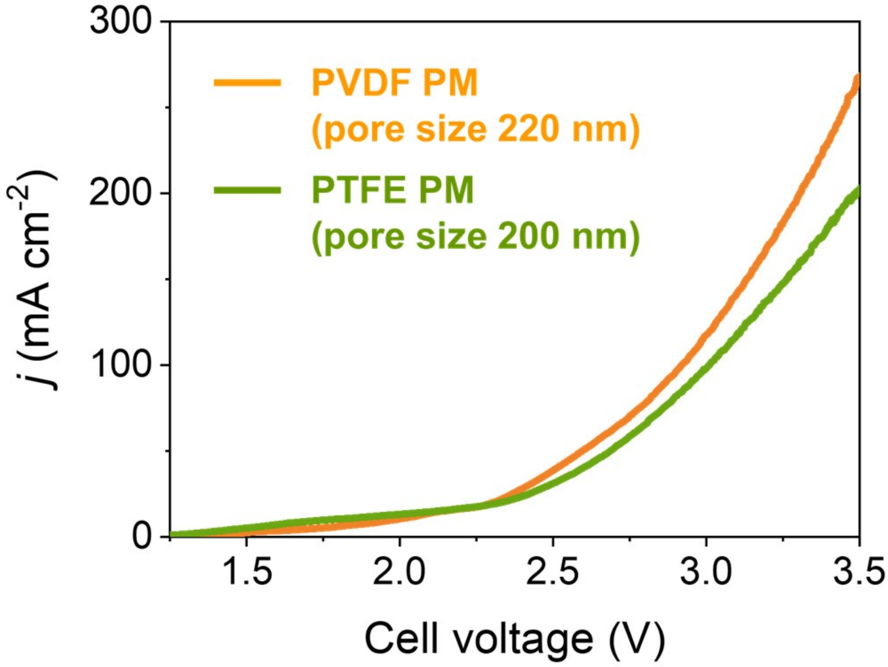

Figure S5. Cell voltage and current density curves of a zero-gap electrolyzer with hydrophilic and hydrophobic PM applied. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. PM: hydrophilic polyvinylidene difluoride (PVDF) porous membrane and hydrophobic polytetrafluoroethylene (PTFE) porous membrane. Anolyte: 0.5 M $KHCO_3$.

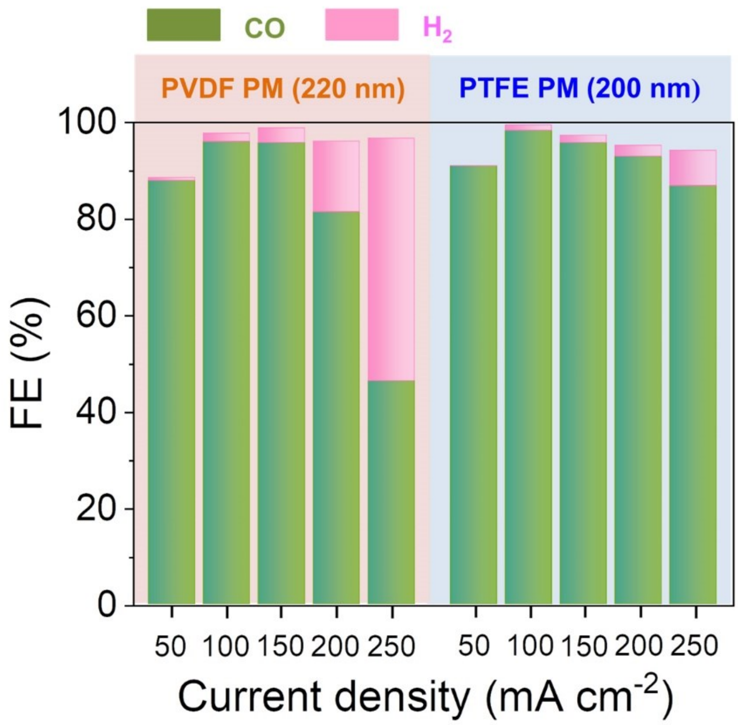

Figure S6. Faraday efficiency of a zero-gap electrolyzer with hydrophilic and hydrophobic PM applied. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Ag black on carbon paper. PM: hydrophilic polyvinylidene difluoride (PVDF) porous membrane and hydrophobic polytetrafluoroethylene (PTFE) porous membrane. Anolyte: 0.5 M $KHCO_3$.

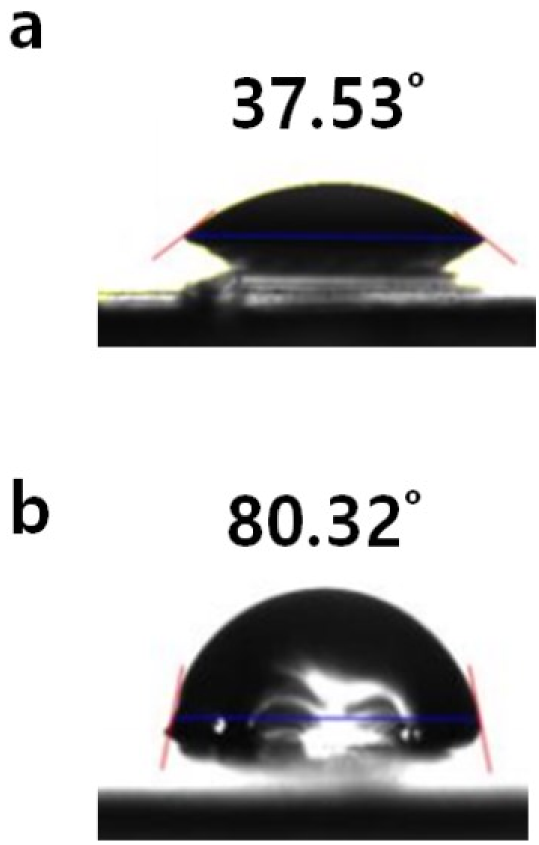

Figure S7. Water contact angle (WCA) photos of (a) porous PVDF membrane (pore size: 220 nm) and (b) porous PTFE membrane (pore size: 200 nm).

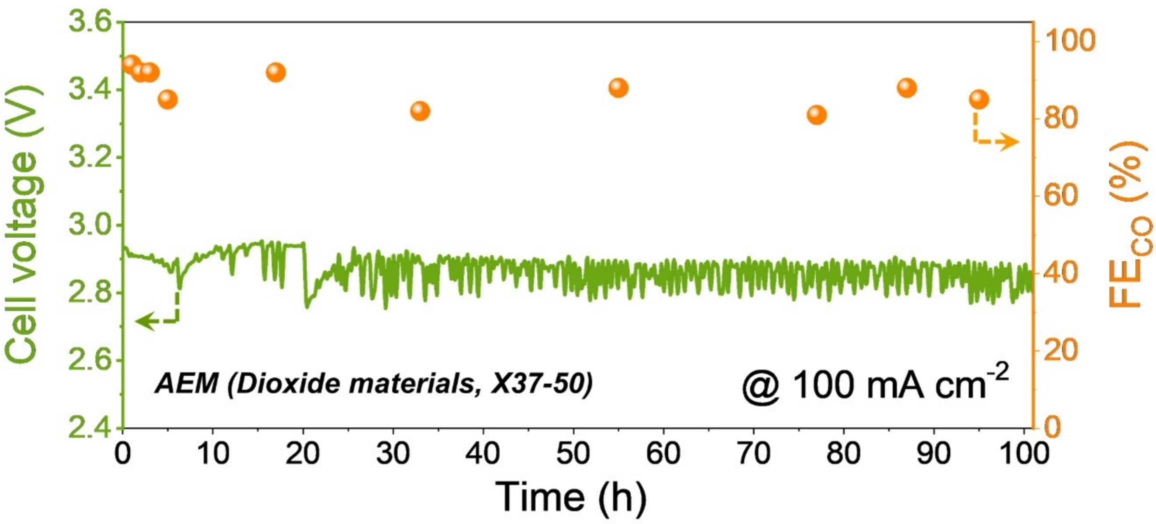

Figure S8. Durability test for the CO_2 electrolyzer employing AEM conducted for 100 h at a current density of 100 mA cm^{-2}. Separator: AEM (anion exchange membrane, Dioxide materials, X37-50). Anode electrode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode electrode: 0.5 mg cm^{-2} Ag black on carbon paper (Sigracet 39BB). Anolyte: 0.5 M $KHCO_3$.

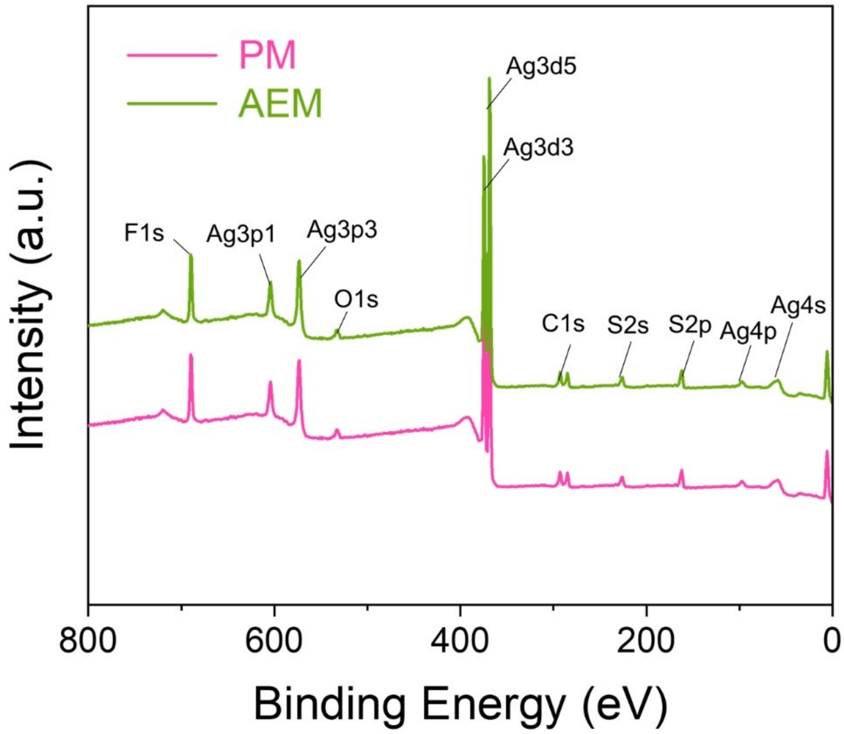

Figure S9. XPS survey spectra of Ag electrodes using PM- and AEM-applied CO_2 electrolyzer after stability test. PM (porous membrane): PVDF, 450 nm pore size, 125 μm thickness.

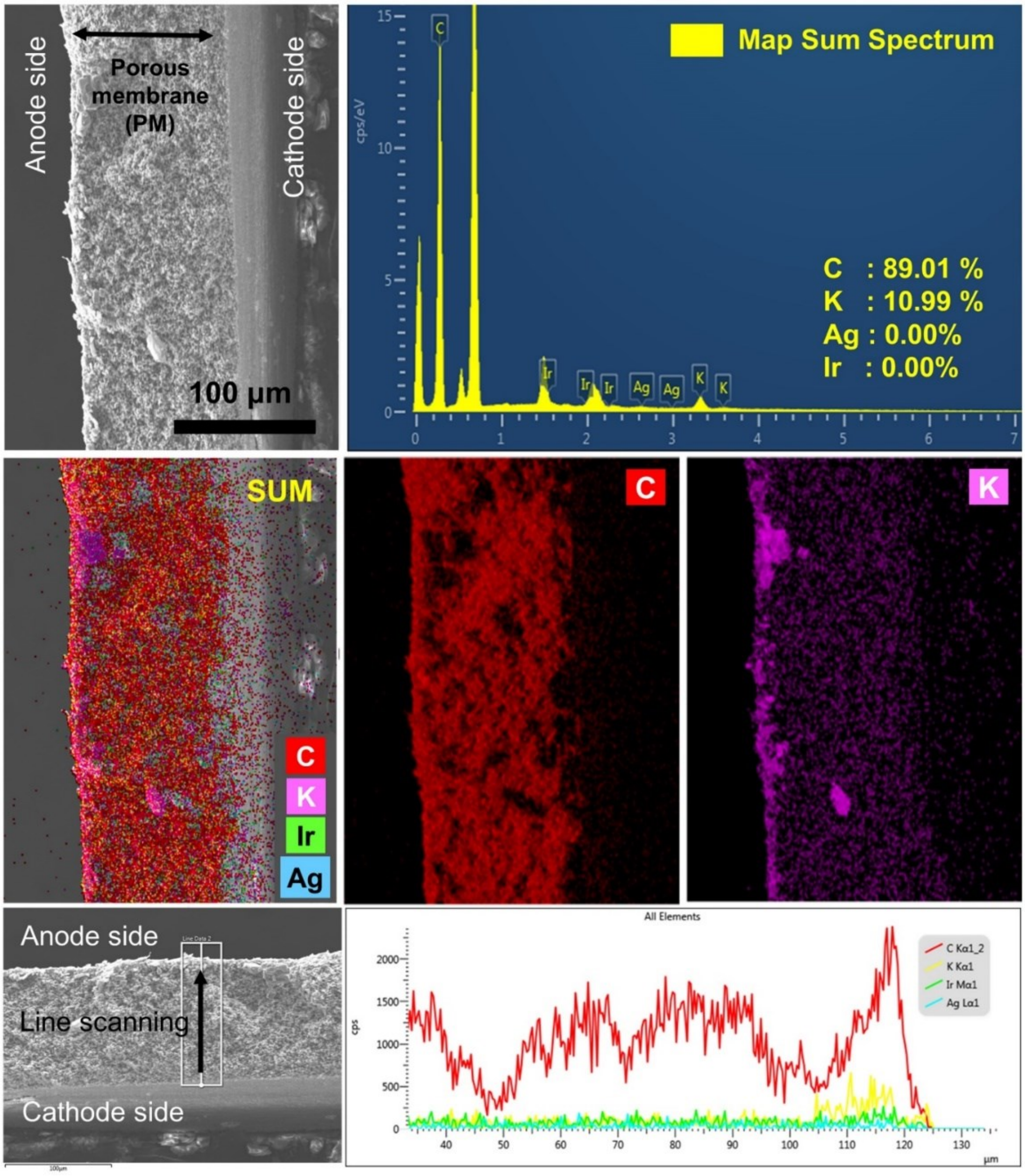

Figure S10. SEM, EDS elemental mapping, and cross section images of PM after CO_2RR stability test for 100 h. PM (porous membrane): PVDF, 450 nm pore size, 125 µm thickness.

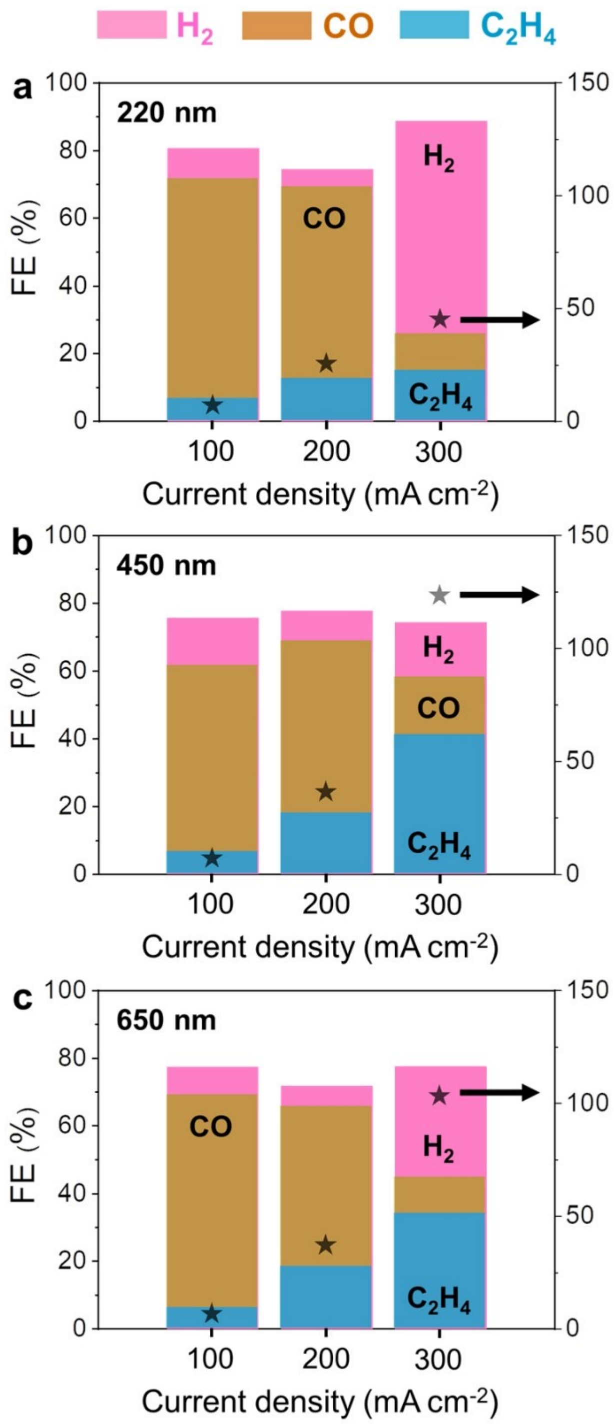

Figure S11. Faraday efficiency of zero-gap electrolyzer for CO_2RR using PM with different pore sizes. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Cu black on carbon paper. PM: polyvinylidene difluoride (PVDF) porous membrane. Anolyte: 0.5 M $KHCO_3$.

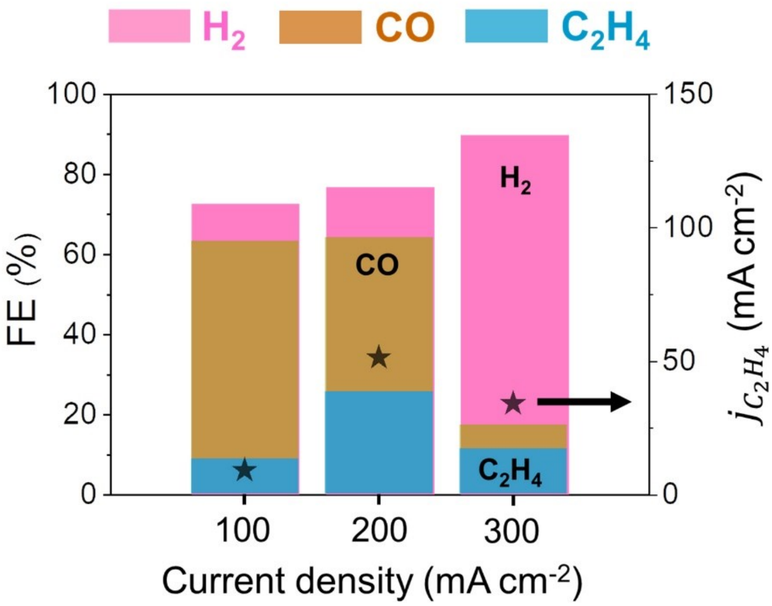

Figure S12. Faraday efficiency of zero-gap electrolyzer for CO_2RR using hydrophobic PM. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Cu black on carbon paper. PM: polytetrafluoroethylene (PTFE) porous membrane, 200 nm pore size, 125 µm thickness. Anolyte: 0.5 M $KHCO_3$.

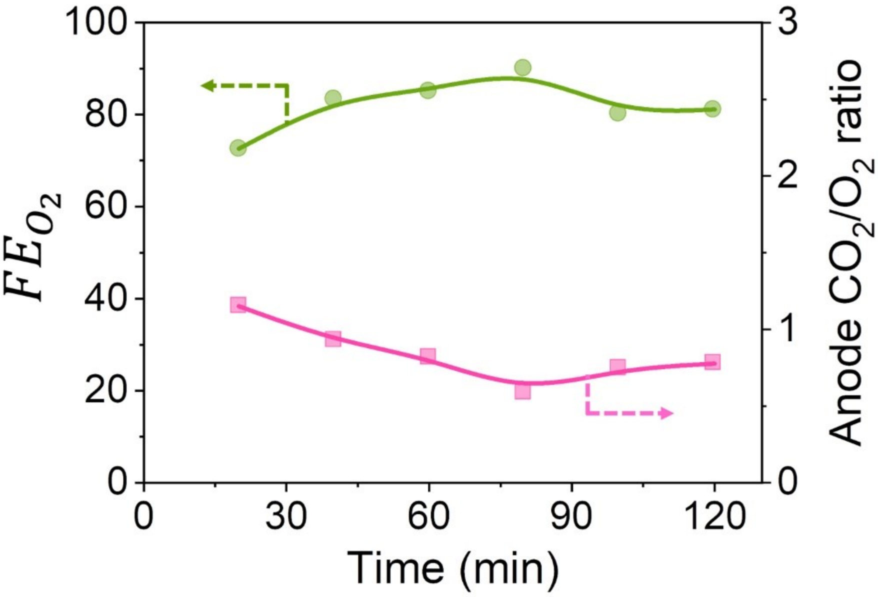

Figure S13. O_2 Faraday efficiency (FE) and CO_2/O_2 ratio at the anode outlet during CO_2 reduction reaction (CO_2RR) in PM-applied zero-gap electrolyzer using 1 M KCl anolyte. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. Cathode: 0.5 mg cm^{-2} Cu black on carbon paper. PM: polyvinylidene difluoride (PVDF) porous membrane, 450 nm pore size, 125 µm thickness.

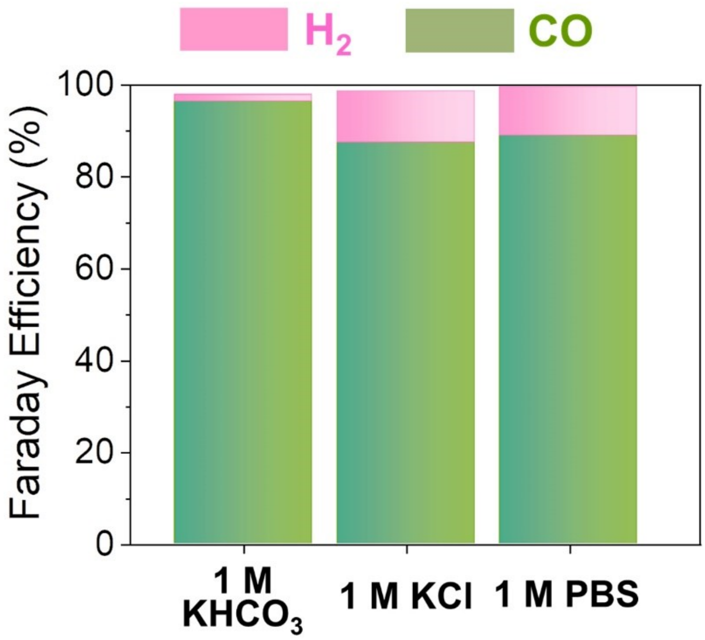

Figure S14. Faraday efficiency of PM-applied zero-gap electrolyzer for CO_2RR using Ag electrode with 1 M $KHCO_3$, 1 M KCl and 1 M PBS anolyte. Anode: 1 mg cm^{-2} IrO_2 on Pt-coated Ti mesh. PM: polyvinylidene difluoride (PVDF) porous membrane, 450 nm pore size, 125 μm thickness.

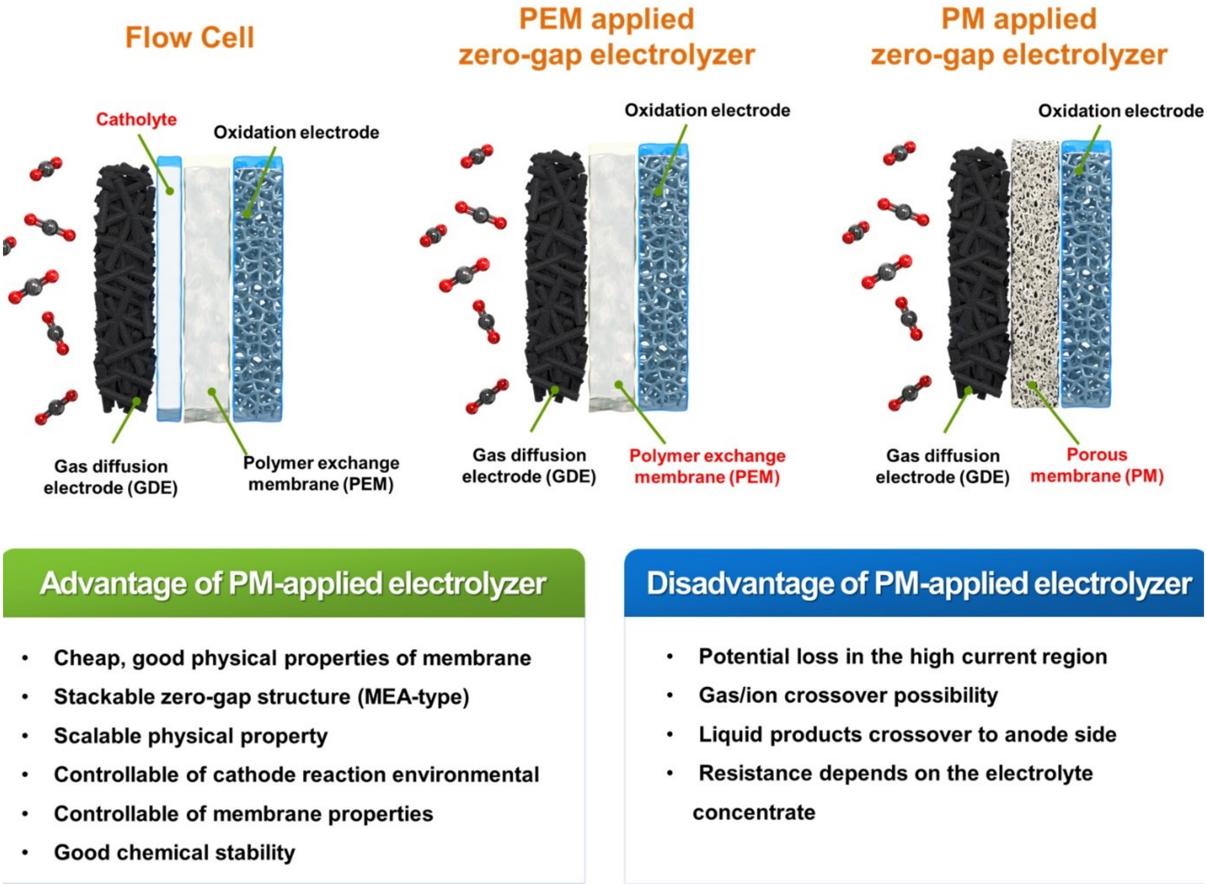

Figure S15. Advantages and disadvantages of zero-gap electrolyzer with PM applied for CO_2RR through comparison with flow cell and polymer exchange membrane (PEM) applied electrolyzers.

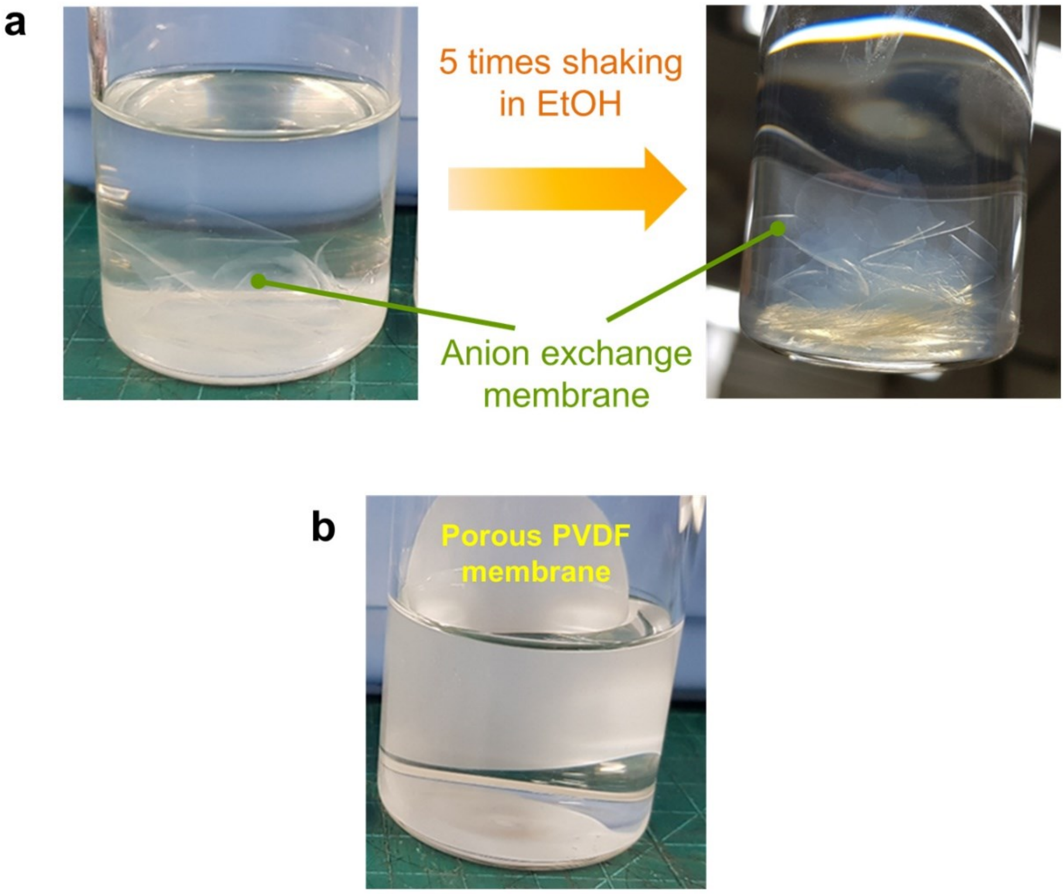

Figure S16. Mechanical strength of polymer exchange membrane (PEM) and porous membrane in ethanol. (a) Anion exchange membrane (AEM), (b) porous PVDF membrane (PM), 450 nm pore size, 125 μm thickness.

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Reference

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