

Supporting Information

**Sustainable Utilization of Waste Expanded Polystyrene in Cellulose Acetate Polymer Blend
Membranes for CO₂/CH₄ Separation**

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To check the mechanical behavior of the prepared blend membranes, simple bending and folding tests were performed. Upon incorporation of waste expanded polystyrene (EPS) up to 10 wt% the membranes retained their structural integrity and remained sufficiently flexible. The macroscopic images clearly show that even when the membranes were twisted and folded no visible cracks, fractures, or signs of brittleness were observed. EPS is inherently more rigid and brittle than CA. Therefore, increasing EPS content generally tends to reduce the overall flexibility of polymer blends. However, up to 10 wt% loading the dispersed EPS phase appears to be well accommodated within the CA matrix while maintaining a continuous polymer network capable of withstanding moderate mechanical deformation. This suggests good compatibility and interfacial interaction between CA and EPS at lower blend ratios. At higher EPS loadings (more than 10%) increase in stiffness and reduction in flexibility was observed which indicates the due to the dominant influence of the rigid polystyrene segments brittleness increases. This behavior is consistent with polymer blending theory where excessive incorporation of a brittle polymer phase disrupts chain mobility and reduces deformability.

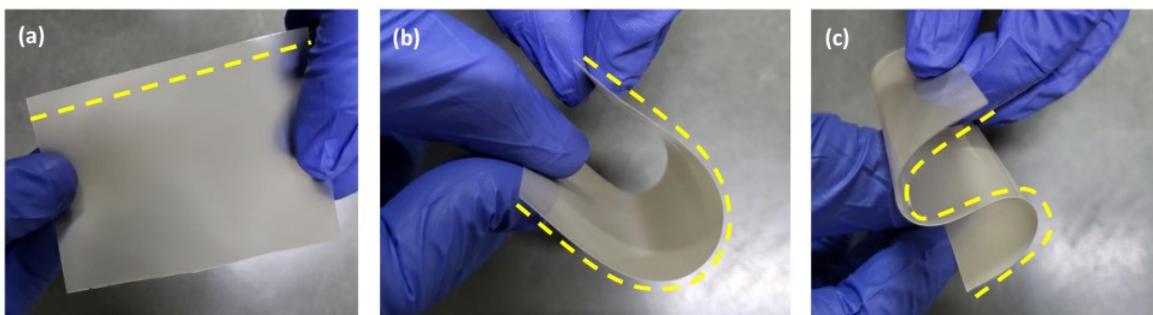


Fig. S1. Macroscopic image of 10% EPS/CA blend membrane

To check the thermal stability of the pure cellulose acetate (CA), waste expanded polystyrene (EPS), and the CA/EPS blend membranes containing 1, 5, and 10 wt% EPS Thermogravimetric analysis (TGA) was conducted. A multistage degradation pattern for all samples was observed in the TGA pattern (Fig. S2) with distinct improvements in thermal stability upon incorporation of EPS.

In the neat CA membrane's TGA curve an initial minor weight loss below ~ 120 °C was observed which is attributed to the evaporation of physically adsorbed moisture. The major decomposition

occurred between ~280-380 °C which corresponds to the breakdown of acetyl groups in the polymer backbone of CA.

Waste EPS showed a single dominant degradation step beginning near ~350 °C and progressing to ~470 °C which is typical thermal behavior of polystyrene. This indicates the depolymerization of polystyrene into styrene monomers and oligomers. EPS displayed comparatively higher thermal stability in the initial stage (<300 °C), but left negligible solid residue due to its non-char-forming nature.

The CA/EPS blend membranes demonstrated intermediate but enhanced thermal behavior. The initial decomposition temperature of the blends gradually shifted to higher values with increasing EPS content. The 1 wt% and 5 wt% EPS membranes showed a slight but noticeable increased initial decomposition compared to neat CA which indicated a stabilizing effect introduced by the thermally robust polystyrene component. This enhancement became more pronounced in the 10 wt% EPS blend, which exhibited the highest temperature among the membranes and a broader degradation profile, suggesting improved resistance to thermal scission. The presence of EPS appears to restrict chain mobility and delay the onset of thermal degradation, likely due to partial physical interactions and a dilution effect of the more stable polystyrene.

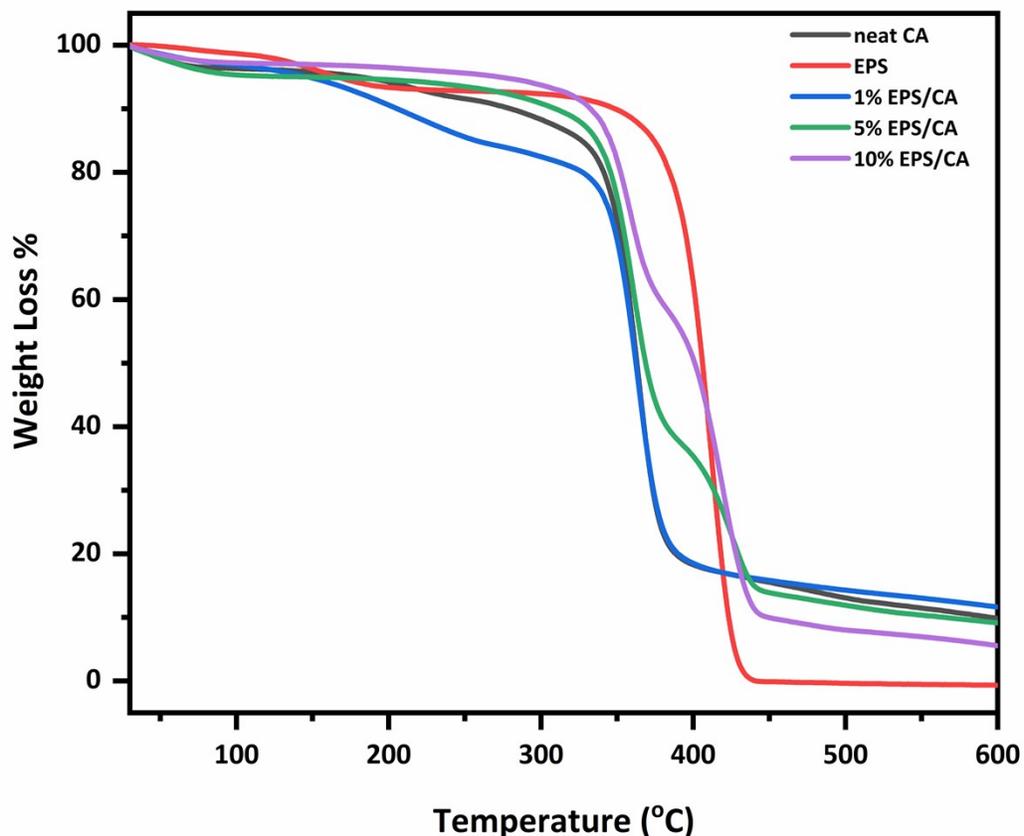


Fig. S2. Thermogravimetric analysis of neat CA membrane, EPS, 1%, 5%, and 10 EPS/CA blend membrane

Gas permeation experiments were conducted at 25°C to assess the transport behavior of CO₂ and CH₄ through neat CA and EPS/CA blend membranes at pressures of 1.5 and 3 bar. A model biogas mixture with a composition of CO₂ and CH₄ in a 40:60 ratio was used as the feed gas. The biogas contains trace amounts of nitrogen (N₂), oxygen (O₂), hydrogen (H₂), hydrogen sulfide (H₂S), and volatile organic compounds (VOCs). Therefore, model biogas was used as we could concentrate more on the membrane's basic separation behavior. Each membrane test sample was fixed in between a stainless-steel permeation cell with an effective area of 3.14 cm² and then sealed with two O-rings to prevent gas leakage. For sweep gas high purity nitrogen (N₂) gas was used. A back pressure regulator (BPR) had been used to ensure a consistent pressure throughout the experiment. To measure the gas flow rates on the feed and permeate sides a calibrated digital bubble flow meter was used. The composition of the outlet gas was determined with a gas chromatograph (GC, Shimadzu GC-2014, Japan) that consisted of a thermal conductivity

detector (TCD) and was operated using LabSolutions software. To ensure the accuracy and reproducibility of the results, a minimum of three different replicates of the same composition were tested under the same conditions in order to ensure the accuracy and reproducibility of the results. Using a digital micrometre, the membrane's average thickness was found to be between 200 to 220 μm . A systematic schematic (Fig. S3) of the gas permeation setup is shown in our earlier study ¹.

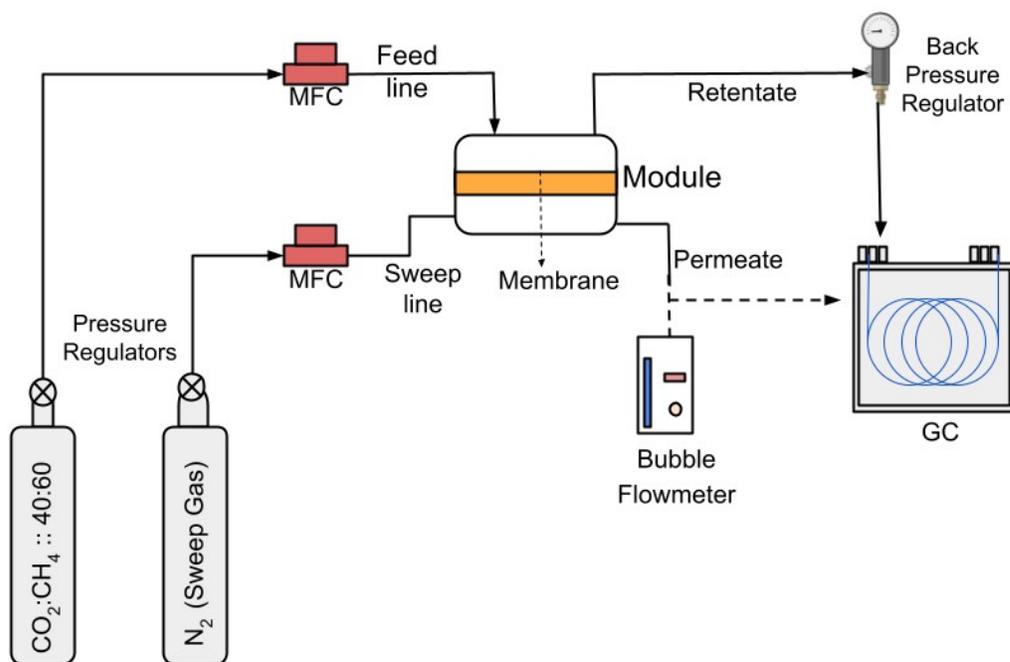


Fig. S3. Schematic of gas permeation setup

The position of the EPS/CA membranes on the Robeson 2008 upper bound plot (Fig. S4) further highlights their competitiveness among existing polymer blends ². The blend membranes were not able to cross the upper bound. The 5 and 10 wt% EPS/CA membranes are positioned close to the 2008 upper bound which shows a balanced improvement in permeability without a substantial loss in selectivity. This indicates that the incorporation of waste derived EPS not only enhances gas transport through microstructural modification but also enables sustainable upcycling of plastic waste into high performance gas separation membranes. In general, the EPS/CA blend membranes presented in this work can be utilized as a new class of eco-friendly

and cost-effective blend membranes with performance comparable or exceeding to available state of the art blend systems for biogas upgradation.

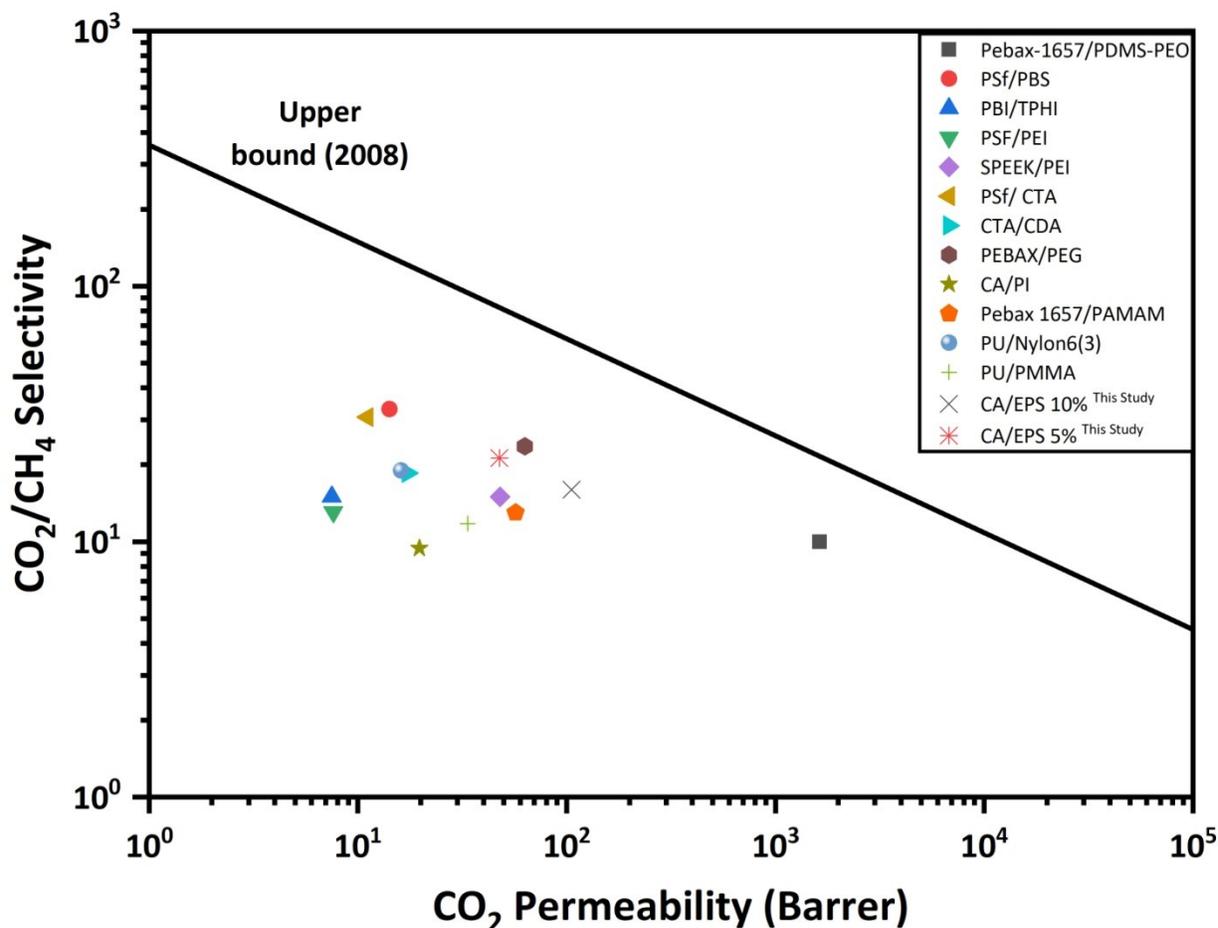


Fig. S4. Position of EPSC/CA blend membranes on the Robeson 2008 upper bound plot for CO₂/CH₄ separation. Data points correspond to measurements conducted at 25 °C using a mixed gas feed of CO₂/CH₄ (40:60) at pressures of 1.5 and 3 bar.

Table S1. Thickness of the membrane calculated using digital micrometer

Membranes	Thickness
neat CA	200±6.07
1% EPSC/CA	220.8±8.42
5% EPSC/CA	208.2±5.30
10% EPSC/CA	222.6±3.55

References

- 1 B. Nayak and B. V. R. Kuncharam, *Carbon N. Y.*, 2025, **236**, 120082.
- 2 L. M. Robeson, *J. Memb. Sci.*, 2008, **320**, 390–400.