

Introduction

The urgent need to mitigate greenhouse gas (GHG) emissions has placed the energy sector at the center of global decarbonization efforts, as fossil fuel combustion accounts for a major share of anthropogenic CO₂ emissions. Post-combustion carbon capture represents a key strategy for reducing emissions from existing power plants, while enabling the valorization of captured CO₂ through Carbon Capture and Utilization (CCU) pathways. Among the available capture technologies, membrane gas separation has attracted increasing interest due to its compact design, low energy requirements, operational simplicity, and potential for modular deployment (Gkotsis et al., 2023).

This study focuses on the investigation of a membrane-based gas separation process for post-combustion CO₂ capture from flue gas streams. The proposed system aims to selectively separate CO₂ from nitrogen-rich exhaust gases of coal-fired power plants and produce a CO₂-enriched stream suitable for downstream utilization. In particular, the captured CO₂ is intended to be integrated with a biological methanation unit, where it can be converted into biomethane using renewable hydrogen, contributing to hybrid energy storage and sustainable fuel production.

Materials & Methods

CO₂/N₂ separation was investigated using polyimide (PI) hollow fiber membrane modules, characterized by a dense selective surface and a symmetrical internal morphology. The experimental study progressed from a laboratory-scale setup, utilizing back-pressure regulators and mass flow controllers to evaluate single-stage performance, to a sophisticated two-stage pilot-scale unit. This pilot configuration (Fig.1) consists of four modules arranged in a 3:1 stage sequence, integrated with a PLC control system and a dual-compression strategy (up to 7 barG). To simulate industrial post-combustion flue gas, synthetic mixtures (10-15 % CO₂) were analyzed using gas analyzers. The process incorporates a recycling loop for the 2nd stage retentate to maximize system efficiency, aiming to optimize operating parameters for achieving >90 % CO₂ purity with simultaneous high recovery rates.

Figure 1. Schematic Diagram of the membrane separation system.

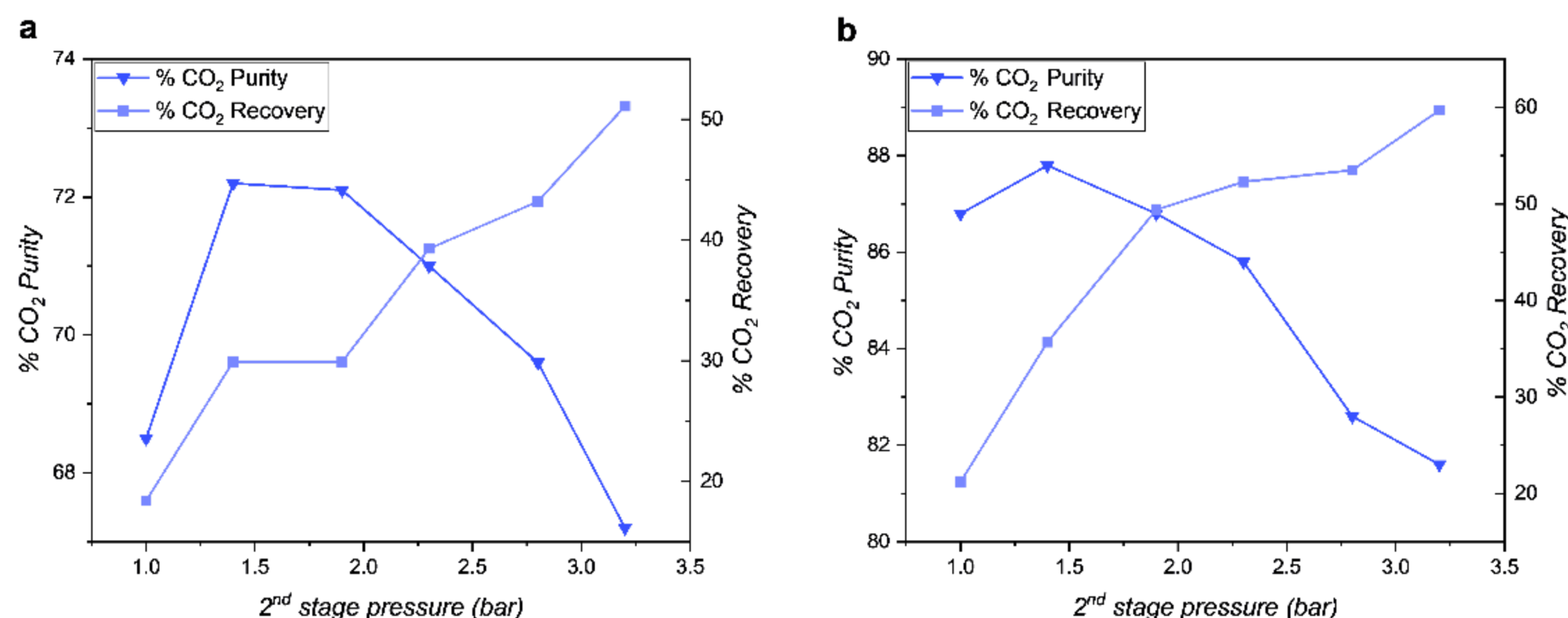
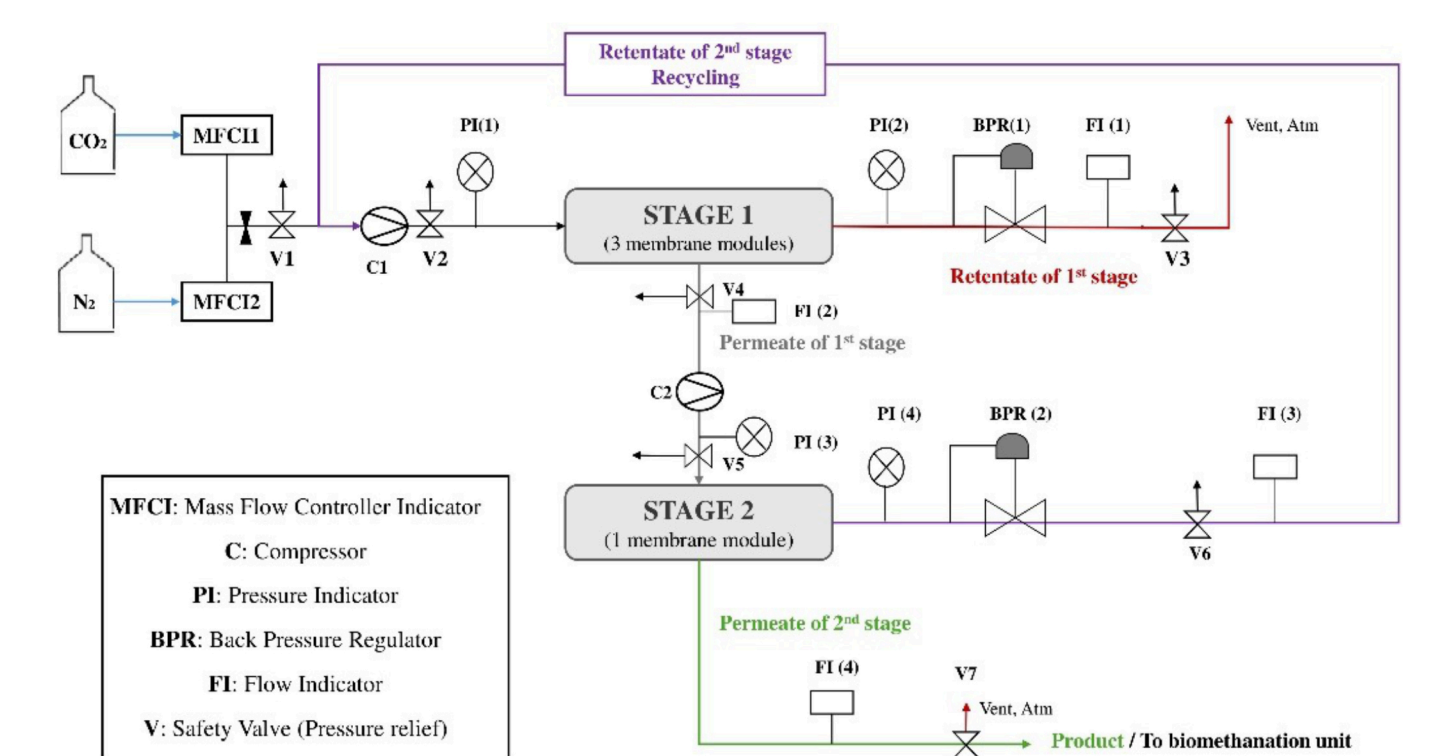


Figure 2. % CO₂ Purity and Recovery as a function of 2nd stage pressure for a feed composition of (a) 85% N₂ / 15% CO₂ and (b) 74% N₂ / 24% CO₂, with a feed flow rate of 10 L/min.

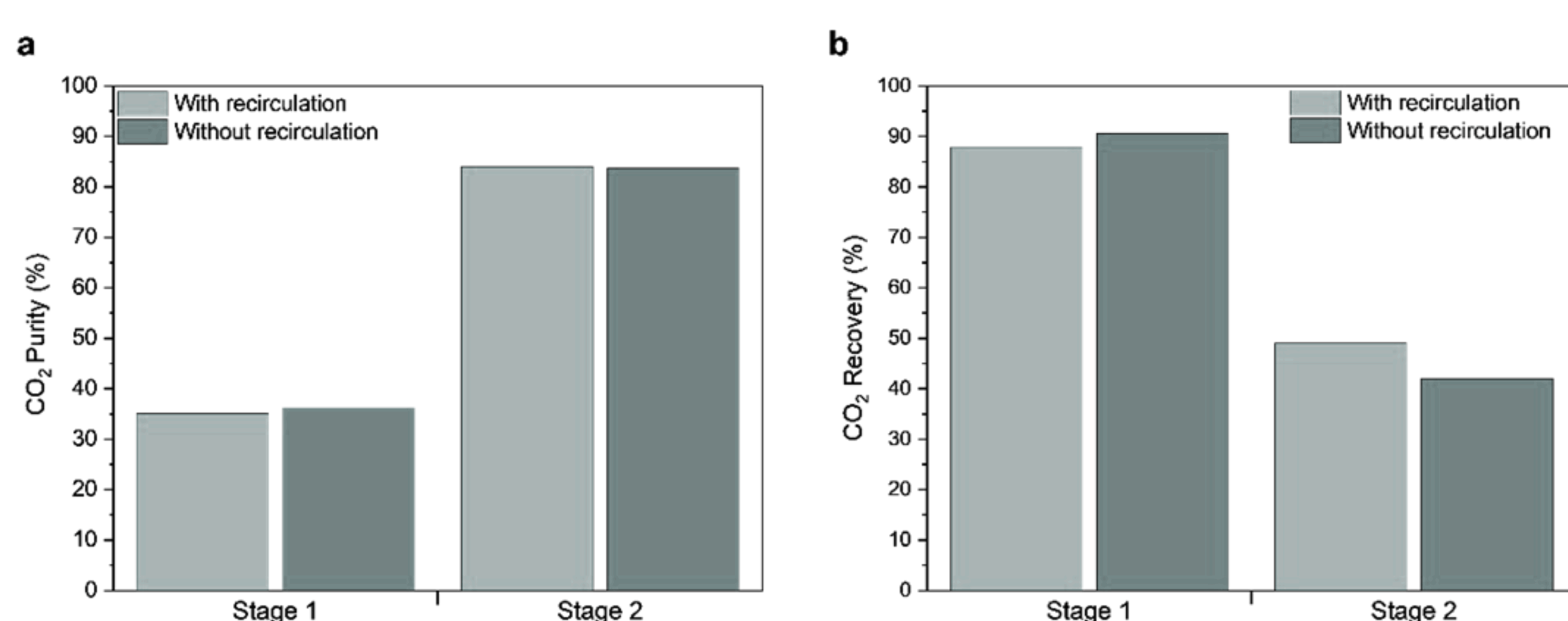


Figure 3. CO₂ purity and recovery in relation to system configuration.

References

P. Gkotsis, E. Peleka, and A. Zouboulis, "Membrane-Based Technologies for Post-Combustion CO₂ Capture from Flue Gases: Recent Progress in Commonly Employed Membrane Materials," *Membranes* (Basel), vol. 13, no. 12, 2023, doi: 10.3390/membranes13120898.

J. Xu, H. Wu, Z. Wang, Z. Qiao, S. Zhao, and J. Wang, "Recent advances on the membrane processes for CO₂ separation," *Chin J Chem Eng*, vol. 26, no. 11, pp. 2280–2291, 2018, doi: <https://doi.org/10.1016/j.cjche.2018.08.020>.

Results & Discussion

The pilot-scale experiments at the Agios Dimitrios Power Plant identified critical trade-offs between CO₂ purity and recovery. Increasing the 2nd stage separation pressure (up to 3.2 bar) significantly enhanced CO₂ recovery due to higher permeation rates, though it slightly compromised purity through dilution. Optimal stage pressures were established at 5.5 bar (1st stage) and 1.9 bar (2nd stage). Furthermore, the integration of a recycling loop for the 2nd stage retentate proved essential for system efficiency, improving overall CO₂ recovery by 7.1% while maintaining stable purity levels.

To meet the requirements for downstream applications like methanogenesis (>90 %), the effect of feed composition and flow rate was evaluated. It was demonstrated that lower feed flow rates (5.5 L/min) and higher inlet CO₂ concentrations significantly boost separation performance. The system achieved a maximum CO₂ purity of 94.3% and a recovery of 71% using a 27% CO₂ feed. These results confirm that the multi-stage membrane configuration is a viable, modular, and energy-efficient alternative to traditional capture technologies, capable of producing high-quality CO₂ streams for industrial utilization.

Conclusions

- The pilot-scale polyimide membrane unit demonstrated stable and continuous operation, successfully providing a high-purity CO₂ stream suitable for downstream utilization.
- The multi-stage system achieved a peak CO₂ purity of 94.3% and a recovery of 71% under optimized conditions (27 % CO₂ feed and 5.5 L/min flow rate).
- Performance is significantly enhanced when the initial CO₂ concentration exceeds 20%, indicating high suitability for industrial sectors such as cement manufacturing.

Acknowledgement:

The project is co-funded by LIFE, the EU's financial instrument supporting environmental nature conservation and climate action projects throughout the EU.