Integration of anaerobic digestion of pre-treated sewage sludge with a membrane-based biogas upgrading system: Evaluation of process configuration through pilot-scale operation

G.-C. Mitraka¹, C. Koutsiantzi², M. Gaspari¹, K.N. Kontogiannopoulos¹, A.I. Zouboulis³, P.G. Kougias¹

¹ Soil and Water Resources Institute, Hellenic Agricultural Organization – Dimitra, Thermi, 57001, Greece ² Department of Chemical Engineering, Aristotle University of Thessaloniki, Thessaloniki, 54124, Greece ³ Laboratory of Chemical & Environmental Technology, Department of Chemistry, Aristotle University of

Thessaloniki, Thessaloniki, 54124, Greece

Keywords: anaerobic digestion, sewage sludge, biogas upgrade, membranes, biomethane Presenting author e-mail: <u>m.gaspari@swri.gr</u>

Sustainable waste management is one of the most vital strategies that need to be established to overcome suffering points associated with climate change, energy safety and resource depletion. In this framework, the methane (CH₄) content in the biogas produced by the anaerobic digestion (AD) of biomass is of increasing interest, considering that it can be directly utilized as a renewable energy resource for heat and electricity (Nguyen et al., 2021).

Sewage sludge is among the wastes that pose a significant environmental threat. However, its proper management and treatment through AD offer a solution to this problem, while it contributes to the production of renewable energy in the form of biogas. For this reason, further research in the anaerobic bioconversion of this substrate has been promoted, and various pre-treatment technologies have been developed, aiming to simplify sewage sludge's complex structure and composition to facilitate AD's efficiency (Mitraka et al., 2022). In this context, avenues in the development of biogas upgrading techniques have also been paved, in order to broaden the AD-delivered biogas' potential as an energy carrier in transportation or as a substitute for natural gas. To meet the specifications for this kind of utilization purposes, the upgrading technologies focus on removing carbon dioxide (CO₂) from biogas so that the CH₄ content to be upgraded at least to 95% (Fu et al., 2021). One of the "best practice" technologies for biogas upgrade is the membrane separation process. This method's mechanism of action is based on the selective permeability properties of the membranes. Therefore, considering the different permeability rates of the biogas components, this method aids in the selective retention of CH_4 in the inlet compartment, as the smallest molecule, i.e., CO₂, passes through the membrane pores (Khoshnevisan et al., 2022). Among the membranes commonly used for CO₂ removal from biogas and thus, for the enhancement of CH₄ purity, are the polymeric and inorganic. Nonetheless, the formers are preferred, mainly because of their lower cost compared to the inorganic ones (Garcia Gomes et al., 2019).

On accounts of the above, the main objective of the current study concerns the integration of an anaerobic reactor with a membrane-based unit for the upgrade of the biogas produced during the anaerobic bioconversion of sewage sludge and the implementation of this system at pilot-scale conditions. The development of this process configuration aimed at validating the scientific results obtained from preliminary laboratory scale trials. These trials were conducted to systematically determine the most effective method for sewage sludge pre-treatment and identify between two hollow fiber (HF) polymeric membranes the one revealing the optimum performance in terms of CH₄ purity and recovery in the retentate stream. These two HF membranes were composed of polysulfone (PSF) and polyimide (PI), respectively.

More specifically, the method chosen for the pre-treatment of the sludge used as the pilot-scale anaerobic reactor's feedstock involves its heating at 45° C for 48 h and then at 55° C for additional 48 h. This selection was made considering the results of preliminary Biochemical Methane Potential (BMP) batch experiments, which revealed this method's outperformance in enhancing methane yield compared to the other pre-treatment methods examined. Specifically, the increment achieved by using this method, when compared to the methane yield obtained from the untreated sludge, was statistically significant and reached a percentage of 24%. However, before seriously considering the implementation of this method at pilot conditions, another experimental set was also conducted to optimize and assess its performance and reliability through lab-scale continuous reactor operation. The set-up consisted of two replicate continuous stirred reactors (CSTR) that operated under mesophilic (37° C) conditions, with a hydraulic retention time (HRT) of 25 days and exclusive feeding with sewage sludge at an organic loading rate (OLR) of 1.05 g VS L⁻¹ d⁻¹. For comparison purposes, the first reactor (R1) was fed with raw sludge and the other (R2) with sludge treated at 45° C for 2 days and then at 55° C for additional 2 days.

Accordingly, a laboratory-scale single-stage membrane unit was designed and used to evaluate and compare the performance of the two polymeric membranes (i.e., the PSF and PI ones) regarding the CO₂ removal from biogas, aiming for the production of an enriched CH₄ gaseous stream of at least 95% purity. The gases utilized

as the feed gas mixture for the experiments of gas separation were CO_2 and CH_4 of 99.99% purity. The feed gas compositions used to evaluate both membranes' performance, varied between 55:45 and 70:30 % v/v (CH₄:CO₂). The results revealed that when the applied feed pressure ranged above 1 bar for PSF membrane and higher than 6 bar for PI, the CH₄ purity increased, exceeding the target value of 95%. Additionally, the PI membrane led to higher CH₄ recovery values in the retentate stream, rendering it the most promising for biogas upgrading and concomitant high biomethane production.

For this reason, the PI membrane was selected for further evaluation before considering its implementation at the integrated pilot-scale set-up. Indeed, another experimental trial was conducted to assess the separation performance of the PI membrane using feed gas compositions that simulated those of the real biogas produced during the operation of the lab-scale CSTR (R2) (74-84 vol% CH₄). The obtained results showed that as long as the feed pressure was above 6 bars, the CH₄ purity at the retentate side exceeded 95% for all the different feed compositions tested.

The abovementioned results enhanced the design and construction of the pilot-scale set-up, which consists of an anaerobic reactor coupled with a two-stage membrane unit for the upgrading of the biogas produced during the anaerobic bioconversion of the pre-treated sewage sludge. The reactor is made of stainless steel and was designed to be approx. 800 L with a working volume of approx. 600 L. Additionally, the reactor is equipped with a thermal jacket to maintain the operating temperature at 37° C $\pm 2^{\circ}$ C. The pressure inside the anaerobic reactor is monitored and logged through a pressure meter. The reactor feeding is performed periodically using a peristaltic pump at an OLR of 1.05 g VS L⁻¹ d⁻¹. The process configuration also consists of an influent tank containing the treated sewage sludge to be used as feedstock and an automated gas metering system for the measurement of the biogas produced.

The gas separation device comprises an oil-injected rotary screw gas compressor, sized for continuous operation at 11.3 bars. This serves to increase the feed pressure to the necessary level for the biogas to flow through the two-stage PI membrane system. In the first stage, once the pressure reaches the desired value, the biogas enters the first membrane, and biogas separation occurs. Thus, two gaseous streams are produced, one of high CO_2 content (permeate 1) and one of high CH_4 content (retentate 1). Following, the first retentate stream enters the second membrane, aiming to enhance the overall gas separation performance so that a final gas stream of high CH_4 purity and recovery values can be obtained in the retentate side (retentate 2). The second permeate stream is recycled and led to the compressor's suction. The pressure of each inlet and outlet gas stream is monitored and logged with the aid of pressure meters.

Throughout the pilot-scale operation, samples for the monitoring and recording of the reactor's biochemical process parameters will be collected, and the factors influencing the performance of the biogas upgrading unit (e.g. pressure, temperature, gas flow rate through the membranes) will be inspected. It is expected that process control will assist in optimizing the integrated system's performance while ensuring the reliability of its operation. Additionally, the techno-economic and environmental assessment of the integrated system will be performed based on the operational input and output data collected throughout the entire pilot-scale experimental trial. The complete set of results will be available upon the time of the conference.

Acknowledgements

This research has been co-financed by the European Union and Greek national funds through the Operational Program Competitiveness, Entrepreneurship and Innovation, under the call RESEARCH – CREATE – INNOVATE (project code: T2EDK-01293, BiogasUp).

References

Fu, S., Angelidaki, I., & Zhang, Y. (2021). In situ biogas upgrading by CO₂-to-CH₄ bioconversion. Trends in Biotechnology, 39(4), 336-347.

Garcia Gomes, M., de Morais, L. C., & Pasquini, D. (2019). Use of membranas for biogas purification: Review. Holos Environment, 19, 466-501.

Nguyen, V. K., Chaudhary, D. K., Dahal, R. H., Trinh, N. H., Kim, J., Chang, S. W., ... & Nguyen, D. D. (2021). Review on pretreatment techniques to improve anaerobic digestion of sewage sludge. Fuel, 285, 119105.

Khoshnevisan, B., He, L., Xu, M., Valverde-Pérez, B., Sillman, J., Mitraka, G. C., ... & Angelidaki, I. (2022). From renewable energy to sustainable protein sources: Advancement, challenges, and future roadmaps. Renewable and Sustainable Energy Reviews, 157, 112041.

Mitraka, G. C., Kontogiannopoulos, K. N., Batsioula, M., Banias, G. F., Zouboulis, A. I., & Kougias, P. G. (2022). A Comprehensive Review on Pretreatment Methods for Enhanced Biogas Production from Sewage Sludge. Energies, 15(18), 6536