## Optimizing Biomethanation Efficiency: The Influence of Packing Materials in Trickle Bed Reactors

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The imperative for transitioning to sustainable energy has never been more critical. Traditional energy sources, predominantly fossil fuels, are finite and significantly contribute to global warming and environmental degradation. Climate change is widely recognized as a substantial challenge, with international efforts such as the Paris Agreement aiming to mitigate its impacts (United Nations, 2015). Achieving the objectives of the Paris Agreement necessitates a reduction in carbon dioxide (CO<sub>2</sub>) emissions and the adoption of sustainable energy solutions. Among these, renewable energy sources derived from organic waste are gaining considerable traction. These sources not only provide a sustainable means to reduce greenhouse gas emissions but also offer avenues for energy diversification and effective waste management. However, the low calorific value of biogas, due to its CO2 content, presents a significant barrier to its utilization as a sustainable energy alternative (Martin et al., 2013). To overcome this limitation, various biogas upgrading strategies have been developed. Although challenging, these processes are valuable as they enable the production of biomethane with a methane (CH<sub>4</sub>) content exceeding 95%, which can substitute natural gas across multiple sectors (Deng & Hägg, 2010). In Europe, there has been a notable increase in biogas plants incorporating upgrading technologies such as membrane separation, water scrubbing, chemical scrubbing, and pressure swing adsorption (EBA, 2021).

As the urgency to transition to environmentally friendly power generation methods intensifies, emerging biological technologies have garnered significant interest from researchers globally. Among these technologies, biogas upgrading via hydrogenotrophic methanogens presents a promising approach. This method utilizes specific microorganisms that employ induced hydrogen (H<sub>2</sub>) as an electron donor to facilitate the conversion of CO<sub>2</sub> into CH<sub>4</sub> (Holmes & Smith, 2016). The trickle bed reactor (TBR) is considered one of the most promising bioreactor designs for efficient biomethanation (Angelidaki et al., 2018). A TBR comprises a column filled with high-specific-surface-area packing material, which allows for effective immobilization of microorganisms.

This study investigates the impact of various packing materials on the performance and biomethanation efficiency of TBRs. Additionally, the study explores the process's flexibility to operate intermittently and perform "on-demand" biomethanation, simulating real-world scenarios where  $H_2$  availability is inconsistent.

The experiments used two custom-made stainless steel TBRs, each with a 1 L volume. Reactor 1 (R1) was filled with activated carbon pellets, and Reactor 2 (R2) with Raschig rings. Both operated at  $55 \pm 1^{\circ}$ C, maintained by thermal jackets. A synthetic gas mixture of 20% CO<sub>2</sub> and 80% H2 was supplied via peristaltic pumps. Nutrients came from recirculated digestate, refreshed twice weekly. Gas Retention Time (GRT) was tested from 12 hours to 45 minutes. The process was also validated under intermittent gas supply, with interruptions of 1 to 3 weeks to simulate real scenarios.

In the first experimental assay, GRT was gradually reduced, as shown in Figure 1. Both packing materials showed efficient biomethanation up to a GRT of 2 hours, with K1 Raschig Rings (R2) achieving CH<sub>4</sub> purity up to 99%. At a GRT of 1 hour, R1 experienced a significant drop in CH4 concentration but later recovered. R2 was only mildly affected and quickly stabilized. Further reducing the GRT to 45 minutes caused instability, with R1 showing low CH<sub>4</sub> concentration and R2 collapsing, as illustrated in Figure 1. In the second experimental assay, a GRT of 1 hour was used during gas mixture supply periods. Figure 2 shows that both reactors recovered after a 3-week starvation period. R1 established a more robust microbial community, unaffected by starvation, and quickly reached high methane purity upon reactivation. In contrast, R2 needed adjustment time after 1- and 2-week starvation periods but eventually also achieved high methane purity.

This study highlighted the significant impact of packing material on biomethanation efficiency. Both reactors tolerated GRT reductions to 1 hour, maintaining high efficiency. Raschig rings achieved up to 99%  $CH_4$  purity, outperforming carbon pellets. Both materials recovered from short and extended gas starvation without compromising methane production. However, the reactor with carbon pellets developed a more robust microbial community, showing an immediate and positive response upon resumption of feeding.

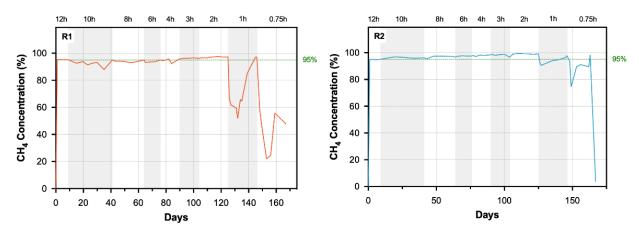


Figure 1. Concentration of methane at the output gas of R1 (A) and R2 (B) during the different Gas Retention Times.

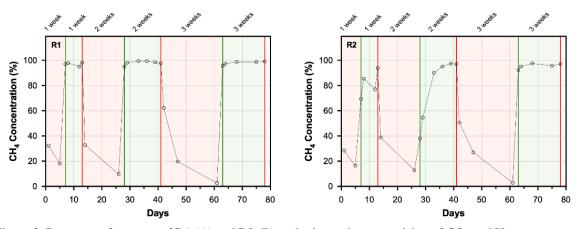


Figure 2. Process performance of R1 (A) and R2 (B) under intermittent provision of CO<sub>2</sub> and H<sub>2</sub>.

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