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Title: Assessment of microbial electrosynthesis technology for utilizing unpurified CO 2 from industrial sources.

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Abstract:

Carbon capture and utilization (CCU) technologies are highly desired to address the issues associated with industrial CO 2 release into the atmosphere and achieve a circular carbon use cycle. Although the direct use of unpurified industrial CO 2 at point sources minimizes the purification and transportation costs, its suitability for producing chemicals via different CCU technologies remains to be fully assessed. This thesis focused on testing unpurified industrial CO 2 for acetic acid production via microbial electrosynthesis (MES) technology by leveraging the inherent CO 2 fixation capabilities of anaerobic chemolithoautotrophic microorganisms. The selected industrial sources include breweries, biogas plants, steel processing units, sugar mills, and incineration plants. Considerable microbial growth and acetic acid production were observed only with brewery CO 2 and biogas in gas fermentation experiments conducted using H 2 and industrial CO 2 as the sole sources of energy and carbon, respectively. Both brewery CO 2 and biogas contained low impurities that showed no substantial detrimental effect on the growth of Acetobacterium-dominated enriched mixed and pure Clostridium ljungdahlii microbial cultures. In the proof-of-concept MES experiments with brewery CO 2 in bioelectrochemical reactors, the mixed microbial culture outperformed the pure culture in terms of acetic acid productivity due to its robust activity, synergistic microbial interactions, and tolerance to gaseous impurities. At an E cell of 2.9 V, up to 7.6 ± 0.65 g/L acetic acid was produced at a rate of 0.5 ± 0.03 g/L/d in the MES reactors operated with 0.7 L/d brewery CO 2 feed under catholyte recirculation conditions. It was achieved at $92\pm4\%$ and $34 \pm 2\%$ faradic and energy efficiencies. With raw biogas feed containing ~60% CH 4 and ~30% CO 2, MES produced methane-rich (>90%) off-gas and acetic acid in the same process. The follow- up process validation and scalability assessments conducted in liter-scale reactors suggested MES as a promising biogas upgradation technology. At 1 L/d raw biogas feed and a fixed current density of -0.5 mA/cm 2 in galvanostatically-controlled reactors, the methane content increased from 56 ± 2 to 86 ± 6% in upgraded biogas along with the production of 3.6 ± 0.6 g/L acetic acid at 82 % faradic efficiency. As per the electrochemical analysis, microbes lowered the hydrogen evolution overpotential and increased electrocatalysis at the cathode, thereby minimizing the overall energy input to the system. A techno-economic assessment revealed that the high cost of reactor materials, including electrodes and proton exchange membrane and a low acetic acid titer, are the major bottlenecks for realizing the near-future practical implementation of the MES technology. Overall, this research sheds light on the applicability of MES technology for utilizing unpurified CO 2 from industrial sources. MES is suitable for utilizing biogenic CO 2 derived from anaerobic digestion and fermentation processes, which are readily available yet underexplored carbon sources. Future research on MES technology development should consider alternative low-cost but efficient reactor materials without compromising the key bioproduction process indicators.

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