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Bioelectrochemical systems (BESs), which employ microbes as catalysts to convert chemical energy stored in organic matter into sustainable electricity and high-value chemicals, is an emerging and promising technology. BESs have broad applications including wastewater treatment, chemical production, resource recovery and waste remediation. Recently, new concepts of integrating BES with anaerobic digestion (AD) for process optimization have been proposed. The purpose of this work was to optimize the AD process using BES in two aspects: developing a new volatile fatty acid (VFA) monitoring system which can be used as the AD process indicator, and for improving biogas quality by removing CO_2. Firstly, a microbial desalination cell (MDC) was developed for measuring VFAs concentrations during AD process. The response time was approx. 5 h and the detection range was 1 to 200 mM. Secondly, in order to reduce the construction cost and response time, microbial electrolysis cell (MEC) was employed as VFA biosensor. The response of the biosensor was only 1 h due to the faster transfer of VFAs supported by the external voltage. The produced H_2 could potentially contribute to the energy needs for operating the biosensor and thereby to a self-sustaining system. Thirdly, to improve biogas quality, a microbial electrolytic capture, separation and regeneration cell (MESC) was developed. In MESC, acid and alkaline generation, CO_2 capture, biogas upgrading and wastewater treatment were simultaneously achieved.