Biohydrogen and Biomethane Production by Co-Fermentation of Food Waste and Paper Waste Using Two-Phase Anaerobic Digestion Process

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Human beings are facing two fatal crisis on the way of developing: the exhaustion of fossil fuels and the worsening global environment. Intergovernmental Panel on Climate Change (IPCC) has pointed out that it is urgently required to reduce the dependence on fossil fuels in the former energy structure and economic structure by promoting the occupation of renewable energy. Compared to fossil fuel energy, renewable energy releases no extra carbon dioxide to the atmosphere. Its property of renewability perfectly suits the goal of sustainable developing in the future of human beings.

Renewable energy has a wide extension, including but not limited to hydraulic energy, tidal energy, solar energy, wind energy, geothermal energy, nuclear energy and bioenergy. Most of these are directly converted into electricity to be transmitted to electric network. Bioenergy is the unique type that has real substance to be its carrier, which is biofuel, which means it could be stored before use. The two type of power with the highest specific energy, combustion engine and fuel cells, cannot work without fuels. The researches about biofuels has gained increasing attention in the fields of biodiesel, bioethanol and biogas.

It is considered that biogas has more advantages. First, unlike the other types of biofuel, being the gaseous characteristic renders the biogas free of the extraction or distillation from the liquid phase after the production. Second, it could be more easily compressed and then transported in the present gas network. Third, its virtual content, majorly referring to biomethane and biohydrogen, could serve as the raw materials for chemical engineering. Cheap catalysts have been found to synthesize
ethane or ethylene from methane. Also, biomethane has a promising role as the electron donor for denitrification in the environmental engineering. These path of biomethane utilization exhibits the ideas of material reusing in the 3Rs: reduce, reuse and recycle.

Anaerobic digestion is an ancient but advancing technology. It reduces organic matters in biomass. It recycles biogas with mild conditions, i.e. the temperature below 100 °C and the normal pressure as the atmosphere. It is the technology that converts biomass into biogas and realizes the 2Rs in the 3Rs. It is widely applied on the reduction of waste biomass, meanwhile easing the other problem mentioned above: the environment worsened by improper waste disposal.

Waste is produced wherever there is human activity. The urbanization even accelerates the waste production, especially for municipal solid waste (MSW). Approximately 1.3 billion tons of MSW was produced around the world in 2010 and the annual production was estimated to grow to 2.3 billion in 2025. MSW could be coarsely classified into these categories: organic waste, paper, plastics, glass, metal and others. They are generally separated manually at the source or by optical mechanics in the collecting center. Along with the trend of diverting the focus of human activity to the urban area, paper is becoming the second major solid waste, following the organic waste, in MSW. Factually, there is no legible boundary to distinguish paper from organic waste since paper products, such as tissue and paper bags, could be easily found in organic waste. So paper and organic waste are combined to be called the organic fraction of MSW (OFMSW). The anaerobic digestion of organic waste has been widely investigated and applied around the world. There comes some unknown questions about whether the complete separation of paper from OFMSW is necessary and how the paper fraction influences the anaerobic digestion of OFMSW. The effect of paper waste content on the anaerobic digestion will be targeted in this research.

Hydrogen energy is energy of the future. In the recent trend of researches, hythane (hydromethane or hydrogen enriched compressed natural gas, HCNG), the gas mixture of hydrogen and methane, is regarded as the important bridge fuel to connect the current type of energy consuming structure to the future type. It has been reported that hythane released less air-pollution gas and enhanced the combustion efficiency in the gas engine. Recent studies confirmed that the hythane that was suitable for the incineration turbine could be produced by two-phase anaerobic digestion.

Two-phase anaerobic digestion is a promising technique that can produce hydrogen and methane simultaneously. By controlling the operating parameters, such as temperature, hydraulic retention time and recirculation, acidogenic phase and methanogenic phase could be separated and distributed to the two reactors in the two-phase anaerobic digestion process. However, there is a void in the current knowledge of the effect of paper waste (PW) content on continuous two-phase process and the stable production of biohythane. Hence, following researches were systematically conducted to develop a two-phase process with stable biohythane production from the anaerobic co-digestion of food waste (FW) and PW.
This dissertation includes 6 chapters. Chapter 1 narrates a general background to emphasize the significance of waste-to-energy technology and lists the structure of the dissertation. Chapter 2 presents the fundamentals for studying anaerobic digestion and reviews the previous literatures on the anaerobic treatment of paper waste and the two-phase anaerobic digestion.

Chapter 3 describes the study on the effect of PW content on the two-phase anaerobic digestion for biomethane production. The OFMSW consisting of 0%, 20%, 40% and 50% PW (on total solids, TS) was prepared. Stable performance was observed in the long-term operation for PW ≤ 40%, with VS (volatile solids) removals around 80%. When fed with 50% PW, the VS removal rate decreased by 8% and the methanogenic reactor required extra NH$_4$HCO$_3$ addition. The ultimate concentrations under the steady state were predicted by simulation using the data from the transient state. Little difference was found between the simulated values and those obtained from experimental steady state. A comparison of the two alkalinity contributors and ammonia revealed that ammonia was the major contributor for pH-buffering alkalinity. The calculated microbial yield coefficients indicated that a higher C/N ratio in the feedstock stimulated the microbial yields and exacerbated the nitrogen deficiency. Energy estimations suggested that significant improvements on the determined amount of FW occurred only when PW content was higher than 40%. Finally, it was determined that the upper limit for PW was 40% and provided the optimal energy-economic value for two-phase anaerobic digestion of OFMSW.

Chapter 4 focused on upgrading the function of two-phase process from biomethane production to biohythane production by adjusting the recirculation ratio in two-phase process. The recirculated two-phase anaerobic digestion (R-TPAD) process was started up and operated with a recirculation ratio ($R$) of 1, and both stages produced methane. In order to acidify the first stage, recirculation was stopped until the production of hydrogen started. The $R$ was ultimately
adjusted to 0.4, and the process obtained stable hydrogen and methane production thereafter. The removal efficiencies of VS in the R-TPAD process were as high as 84.9%. In the R-TPAD process, the hydrogen production yield was 49.9 L/kg-VS_{fed} with an efficiency of 2.405 mol-H_{2}/mol-hexose_{reduced}. The hythane production rate was 1.623 L-hythane/L-reactor/d, with a hydrogen content of 10.5%. This study showed that transforming the function of the R-TPAD process was feasible by adjusting R with observations on hydrogen contents and pH. A pH lower than 5.0 was suitable for continuous hydrogen production.

Chapter 5 investigated the effect of PW content on the biohythane production in the recirculated two-phase process, by combining the feeding conditions applied in the first study and the recirculated two-phase system obtained from the second study. The feeding mixtures consisting of 0%, 20%, 40% and 50% (on TS) of PW and the rest of FW were put into R-TPAD. As a result, stable performance of biogas production was observed in the long-term operation. The process achieved the organic removals more than 78% of the VS and 89% of total carbohydrates. Hythane was harvested from the R-TPAD with the hydrogen content of 10~20%. The PW content elevated the butyrate-to-acetate ratios in the dark fermentation for hydrogen production. Adding PW into would lead to proportional increments on hythane production and bioenergy recovery.

Chapter 6 summarizes the conclusions of these researches and briefly portrays a future vision of resource-recycling society.