

Short Communication

# Production of hydrogen and methane from organic solid wastes by phase-separation of anaerobic process

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## Abstract

Phase-separated two-stage anaerobic process was examined and evaluated using artificial organic solid waste in laboratory scale. Acidogenic process, which was combined with subsequent methanogenic process using packed-bed reactor, was operated emphasizing on either hydrogen production, or solubilizing efficiency of solid materials. In either effluent from hydrogenogenic, or solubilizing operation, maximum allowable OLR achieved at methanogenesis was higher than the single methanogenic process. Hydrogenogenic operation was more suitable to combine methanogenic process than solubilizing operation, since retention time of hydrogenogenic operation was much shorter than the solubilizing operation, obtaining almost the same levels of overall removal efficiency in both COD and VSS. The combination of hydrogenogenic operation in acidogenic process and methanogenic process produced approximately 442 mmol l-reactor<sup>1</sup> days<sup>-1</sup> of methane and 199 mmol l-reactor<sup>1</sup> days<sup>-1</sup> of hydrogen at 25 h of total retention time indicating 82% of COD removal with 96% of VSS decomposition.

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## 1. Introduction

Reuse and recycling of the organic wastes have been attracting social attentions and concerns, because effective utilization of limited resources and development of environmentally acceptable technologies are in great demand. Anaerobic process is one of effective technology to recover energetic materials from organic wastes as a simple and effective biotechnological means of reducing and stabilizing organic wastes (Archer and Kirsop, 1990).

The degradation of solid substances is the rate-limiting step for anaerobic digestion (Eastman and Ferguson, 1981). Thus, enhancement of solubilization is expected to improve the overall process performance. The phase-separated

anaerobic process including two-stage system basically comprises acidogenesis and methanogenesis process. In the first acidogenic process, organic polymers, carbohydrates, proteins, and lipids are degraded to VFAs, which are metabolized to methane in the subsequent methanogenic step. In such anaerobic digestion process, surplus electrons that can be recovered as hydrogen gas, are formed through fermentation metabolism, thus concurrent solubilization and hydrogen production may be practically possible in a single bioreactor. Although there has been considerable amount of the literature about two-stage process (Anderson and Björnson, 2002; Ghaly, 1996; Ghosh et al., 1975; Ng et al., 1985), most of them were combination of methanogenic process and acidogenic process, which is mainly focused on solubilization of solid materials. There have been a few reports on the phase-separation that is combined with hydrogen production.

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Hydrogen fermentation process of organic wastes has been studied and optimized at fundamental levels (Ueno et al., 2001; Lay, 2000). Continuous hydrogen production was observed from industrial wastewater (Ueno et al., 1996) and garbage slurry (Ueno et al., 2006) using natural population of microorganisms without sterilization.

The retention time of hydrogenogenic operation is relatively shorter than conventional methanogenic reactors (Ueno et al., 1996). This implies that great difference in the reactor size between two processes is made if methanogenic process, which has slow rate of digestion, is connected in series. It is, therefore, high efficient reactor would be suitable for such phase-separated process. Although up-flow anaerobic sludge blanket (UASB) process is known as one of high performance reactor, the UASB configuration is hardly applied to digestion of wastes containing high concentration of solid materials due to possible phase-separation between heavier solid particles and microbial sludge if high concentration of solid materials is contained in feedstock. Fixed-bed or packed-bed anaerobic reactors, on the other hand, are capable of efficiently converting organic wastes to biogas even if substantial amount of solid matters are contained in the wastes, because microorganisms are maintained on the surface of supporting media inside the reactor. A thermophilic down-flow packed-bed reactor (DPR) has been studied and reported as one of the reactor designs which enable digestion of solid organic wastes (Tatara et al., 2001).

In the present study, overall process performance of the two-stage biogas production process comprising acidogenic and methanogenic process was examined using artificial organic solid waste. Methane gas production by DPR from the effluent from both hydrogenogenic and solubilizing operations was evaluated in laboratory scale from the view points of recovery of total energy and retention time in comparison with the single stage system.

## 2. Methods

### 2.1. Feedstock and preparation of effluent from solubilization/hydrogen fermentation process

Artificial garbage slurry containing milled paper (AGSP) was used as a model waste material. The AGSP was prepared from 15 g l<sup>-1</sup> of a commercial dog food (Vitane, Nihon Pet Food Co Ltd., Japan) and 10 g l<sup>-1</sup> of milled paper. The milled paper was prepared from shredded waste paper by a vibrating sample mill (TI-200, CMT Co. Ltd., Tokyo, Japan). The average concentrations of COD<sub>cr</sub> and VSS are 37,100 and 23,700 mg l<sup>-1</sup>, respectively.

Hydrogenogenic microflora was prepared according to the previous report (Ueno et al., 1995). Fifty milliliter of the microflora was inoculated into 3 l of AGSP in a 5 l jar-fermenter. The cultivation was conducted at 60 °C and the pH was regulated at pre-set value by automatic titration of 10 N-NaOH. The supply of AGSP to the reactor was accompanied by the concomitant removal of an equal

amount of broth from the reactor. The effluent from the reactor was collected and stored at 4 °C in a sealed container until the methanogenic experiment.

### 2.2. Methane gas production from the effluent from hydrogenogenic/solubilizing operation

A thermophilic down-flow anaerobic packed-bed reactor (TDAPR) (Tatara et al., 2005) was used in the experiments. The TDAPR was an acrylic cylinder that was packed with supporting media of unwoven carbon-fiber textile in the direction of flow and had an effective volume of approximately 0.5 l. The reactor was filled with an anaerobic sludge collected from a commercial thermophilic methane fermentation reactor for garbage, and was acclimated using the AGSP before experiment. Contents in the reactor were moderately mixed by re-circulation of fermentation liquid from the bottom to the top using a peristaltic pump. Operation temperature was 55 °C.

### 2.3. Analyses

The amount of evolved gas and the composition was determined as described previously (Ueno et al., 1995). Analyses for the sampled broth for the determination of volatile suspended solids (VSS), chemical oxygen demand (COD), and volatile fatty acids (VFA) was carried out according to the previous report (Ueno et al., 1996).

## 3. Results and discussion

### 3.1. Acidogenic process for solubilization and hydrogen production

Culture conditions for acidogenic processes of AGSP were established according to our previous study (Ueno et al., 2006). The reactor performance on hydrogenogenic and solubilizing acidogenic process is summarized in Table 1. Hydrogenogenic operation was carried out at 0.5 days of HRT at pH 6.0. Stable hydrogen production

Table 1  
Process performance on acidogenic process

	Acidogenic process operation	
	Hydrogenogenic	Solubilization
<i>Culture condition</i>		
HRT (days)	0.5	4.0
pH	6.0	7.0
Hydrogen production (mmol l-reactor <sup>-1</sup> days <sup>-1</sup> )	199	0.1
<i>Removal efficiency (%)</i>		
Total COD	7.0	4.9
VSS	8.0	71.6
VFA formation (mg l <sup>-1</sup> )	3984	14,800

Data were average at steady state of operation. It was assumed that the steady state was established after the period at least three times as long as the new HRT had passed.

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