# Intermittent Effluent Recirculation for the Efficient Treatment of Low Strength Wastewater by an EGSB Reactor

Yoochatchaval, W.<sup>1, 4\*</sup>, Ohashi, A.<sup>2</sup>, Harada, H.<sup>3</sup>, Yamaguchi, T.<sup>1</sup> and Syutsubo, K.<sup>4</sup>

<sup>1</sup>Nagaoka University of Technology, Niigata, Japan
<sup>2</sup>Hiroshima University, Higashihiroshima, Japan
<sup>3</sup>Tohoku University, Sendai, Japan
<sup>4</sup>National Institute for Environmental Studies, Ibaraki, Japan

Received 3 March 2008; Revised 15 April 2008; Accepted 2 May 2008

**ABSTRACT:** In order to establish the appropriate methane fermentation technology for low strength wastewater, a 2.0 L EGSB reactor was operated at 20°C with 0.3 - 0.4 g COD/L of sucrose-based synthetic wastewater for 500 days. At the start up period, the reactor was operated in EGSB mode with a 5 m/h up flow velocity by continuous effluent recirculation. However, the EGSB reactor exhibited insufficient COD removal (50-60%) at COD loading of 7.2 - 9.6 kg COD/m<sup>3</sup>·day due to the low COD concentration in the sludge bed. Therefore, we proposed the new operation mode by switching to UASB mode (without recirculation, 0.7 m/h up flow velocity) for 30 minutes and EGSB mode for 10 minutes in 40 minutes cycle. Moreover we added sodium sulfide to make the low ORP condition. In this operation, COD removal increased dramatically, from 65% to 91%. Additionally, physical properties of the retained sludge were well maintained in this operation mode. Furthermore, the retained sludge possessed appropriate levels of methanogenic activity (0.2-0.4 g COD/g VSS/ day) at 20°C.

Key words: Anaerobic treatment, EGSB, Low strength wastewater, Methane fermentation, Moderated temperature

## **INTRODUCTION**

Most of organic wastewater is discharged at a moderate temperature and with a low or medium organic strength (Rebec *et al.*, 1999; Angenent *et al.*, 2001). Application of methane fermentation process for such wastewater is important to reduce the energy consumption. However, treating this kind of wastewater by an anaerobic biofilm system (UASB fluidized bed and fixed bed) leads to a shortage of substrate and deterioration of the physical and biological characteristics of biofilm. Furthermore, low temperature caused of increase of water viscosity which retards the accessibility between substrate and biofilm (Lettinga *et al.*, 1983; Kato *et al.*, 1997; Driessen and Yspeert, 1999; Uemura and Harada, 2000; Price and Sower, 2004). To solve these problems, EGSB (Expanded Granular Sludge Bed) reactor was developed to improve the wastewater-biofilm contact via expanding the sludge bed and increase hydraulic mixing by effluent recirculation system (Kato et al., 1994, 1997; Rebec, 1998). In previous report, the EGSB reactor was possible to treat low-strength wastewater (< 1 g COD/L). The effluentrecirculation might be used to improve accessibility of wastewater to biofilm. The net effect of recirculation could be an increase in the efficiency of substrate removal. However, when we apply EGSB reactor with very low strength wastewater (0.3-0.4 g COD/L), we found some difficulty in terms of granulation of

<sup>\*</sup>Corresponding author:Email-wilasinee.y@nies.go.jp

the retained sludge and stable operation of the reactor. The difficulty appears to arise from a loss of substrate concentration that caused by effluent recirculation. Process kinetics, and in particular biological growth kinetics based on growth rate and substrate utilization rates, is key to the operation and development of anaerobic wastewater treatment systems. Limitation of substrate utilization during the operation of an anaerobic system always relate to the halfsaturation constant  $(K_{c})$  value based on Monod kinetics. In these cases, an extremely low substrate concentration retards the rates of growth and substrate utilization of bacteria, resulting in levels of sludge activity that are less than optimal.

In our previous work, we successfully operated an anaerobic EGSB reactor at 20°C with 0.6 - 0.8 g COD/L sucrose-based wastewater (Kawasaki et al., 2005). The EGSB reactor could maintain a sufficient sludge retention time (SRT) and showed excellent process performance. The genus Methanosaeta was confirmed as the dominant acetoclastic methanogen and has a comparatively high half-saturation constant for acetate. The reported Ks values for acetoclastic methanogenesis in granular sludge ranged from 0.01 to 0.4 g COD/L (Kato, 1994; Rebec, 1998; Dolfing, 1985; Pavlostatis and Giraldo-Gomez, 1991). Oxidation-reduction potential (ORP) is the measurement of the potential for a reaction to occur. It represents an electron concentration and activity level. During anaerobic degradation of organic matter, the methanogenic reaction proceeds well under low ORP conditions, approximate levels of less than -260 mV (Lettinga et al., 2000). Therefore, maintenance of low ORP conditions in the reactor had positive effects and possible to keep the good process performance for the anaerobic treatment of low strength wastewater. Building on this background, we sought to investigate how influent COD strength can limit efficient treatment of wastewater using an EGSB reactor. Furthermore, we investigate the conditions of effluent recirculation and influent ORP in order to improve the efficiency of COD removal.



Fig.1. Schematic diagram of an EGSB reactor

### **MATERIALS & METHODS**

A 2.0 L volume EGSB reactor (Fig. 1) was operated for more than 500 days with 0.3 - 0.4 g COD/L of synthetic wastewater (simulating food processing wastewater) composed of sucrose, acetate, propionate and yeast extract in a COD ratio of 4.5: 2.25: 2.25: 1. Basal mineral and trace element compositions were as described previously (Syutsubo *et al.*, 2001).

This reactor was inoculated with 20°C grown granular sludge (46.1 g VSS/reactor) (Kawasaki et al., 2005). The influent and EGSB reactor temperatures were maintained at 20°C. During the start-up period (phase "A" in Table 1), the reactor was operated in EGSB mode with an up flow velocity of 5 m/h by continuous recirculation of the effluent. The HRT was 1 h. and the organic loading rate was 7.2 - 9.6 kg COD/m<sup>3</sup>·day at 20°C (after 21 days; see Table 1). The influent ORP was maintained at a negative value (-140~ -170 mV) by adding 20 mg/L of Na<sub>2</sub>S·9H<sub>2</sub>O after 123 days.To investigate the influence of effluent recirculation conditions on process performance, the reactor was operated at 0.7 m/h of up flow velocity, (UASB mode, without recirculation) for 30 min and then switched to 5 m/h of up

Day	0-21	21-101	102-123	124-283	284-503	
Operational Phase		А	B1	B2	С	
Upflow velocity	5 m/h (EGSB)		0.7 m/h (UASB:30 min)+ 5 m/h (EGSB:10 min)			
(Operational mode)						
Influent ORP (mV)	-		+ 22~-70	-140~-170		
COD loading (kg	4.8-6.4		7.2-9.6			
COD/m <sup>3</sup> /day <sup>1</sup> )						
HRT(hour)	1.5			1.0		

Table 1. Operation conditions for the EGSB reactor during continuous flow experiment

flow velocity (EGSB mode, continuous recirculation) for 10 minutes in phase B and C. We found the loss of sludge during phase A and B; therefore we added external granular sludge as same level of start-up period at the end of phase B.

Regular sampling and analysis was conducted five days a week. The influent and effluent of the reactor were each sampled and assays were conducted to test pH, COD (COD<sub>...</sub>), volatile fatty acid (VFA), suspended solid (SS) and sulfate analyses. It took 40 minutes to collect effluent when the reactor was running in either EGSB or UASB mode. To measure effluent COD, a small amount of sulfuric acid was added to the collected sample and then purged with nitrogen gas to remove sulfide. The physical and microbial properties of the retained sludge were occasionally analyzed; these included sludge concentration (MLVSS), sludge volume index (SVI) and methanogenic activity (sampled from port No.2, 22.5 cm from the bottom). Batch feed experiments were conducted to determine the COD profile of the sludge bed after operation under different influent COD concentrations and different operational modes. To do this, the reactor was fed with a prepared substrate and operated in different modes (without recirculation or continuous recirculation). After substrate was provided for 3 - 4 times of HRT, the COD concentration of the samples which taken from the sludge bed were determined. The methanogenic activity of retained sludge samples was determined in duplicate for samples taken at day 0 (seed), 98, 259 and 500 as previously described (Syutsubo et al., 1997).

The test substrates were acetate, propionate and  $H_2/CO_2$  (80%:20%, V/V). All vials were incubated on a reciprocal shaker (120 rpm) at 20°C.

## **RESULTS & DISCUSSION**

Figure 2 shows the process performance of the EGSB reactor. The efficiency of COD removal was 63% during the first 21 days when an HRT of 1.5 hours were used.



Fig. 2. Process performance of the EGSB reactor





Fig.4. Changes of COD profiles in UASB and EGSB mode (day 187)

In order to keep the same COD loading with our previous study (7.2 - 9.6 kg COD/m<sup>3</sup>·day), the HRT was subsequently decreased to 1 hour. As a result, the COD removal efficiency decreased to 57%. In our previous experiment, the influent COD was set to two times of this experiment and the efficiency of COD removal was 70% at the same COD loading conditions. Thus, decreasing the influent COD caused deterioration in the COD removal efficiency using an EGSB reactor. Additionally, in phase A, acetate and propionate were detected in the effluent at concentrations of about 0.14 g COD/L and 0.10 g COD/L, respectively (Fig. 3).A low COD removal efficiency and deterioration of the physical properties of the retained sludge were observed during the first 101 days (phase A) of treatment.

These phenomena may result from a shortage of substrate in the sludge bed. To address this, we assayed the COD profile at different height of the reactor in the operation with recirculation (EGSB mode; up flow velocity 5 m/h) and without recirculation (UASB mode; up flow velocity 0.7 m/h) as shown in Fig. 4. The efficiency of COD removal (as measured by effluent COD quality) clearly improved in UASB mode. Moreover, in this mode, COD strength in the sludge bed kept at a high level as compared with the levels observed in EGSB mode (Fig. 4). Maintaining a high concentration of COD in the sludge bed may be effective in activation of anaerobic bacteria. In the batch feed experiment, we found the granular sludge floated up from the bottom that interferes with the operation of the reactor. Moreover, we had difficulty with bio-gas detachment from retained granular sludge in UASB mode due to the high viscosity of wastewater. The effluent recirculation may be able to improve the biogas detachment. Based on these assumptions, we next changed the operational mode to the combination of UASB and EGSB mode (intermittent effluent recirculation) after day 101. We noticed a change in average methane production during different phases of treatment and determined the rate of methane recovery, as a percentage of COD removal (Fig. 5).



Fig. 5. Changes in methane production, efficiency of COD removal and the rate of recovery of methane during different phases of treatment



Fig. 6. Physical properties of sludge retained in the EGSB reactor [MLVSS;(a), SVI;(b)]

Between the later part of phase A and phase B1 of treatment, the efficiency of COD removal increased from 57% to 61%. However, the methane recovery rate dropped from 43% to 39% during the same treatment period. By the end of phase B1, the methane production became 0.8 g COD/day for methane gas and 2.4 g COD/day for soluble methane. At this time, the influent ORP was 25±45 mV and the effluent ORP (measured at the gas-solid separator portion of the reactor) increased from -220±13 mV to -173±18 mV, presumably as a result of stopping effluent recirculation when switching to UASB mode. However, this value is not suitable for methane fermentation and thus, we next tested the effect of adding sodium sulfide to the influent (phase B2). The influent ORP was effectively lowered during phase B2 by addition of sodium sulfide. As a result, the influent-ORP and effluent-ORP reduced to -96±52 mV and -238±11 mV, respectively. In addition, the rates of recovery of methane gas and soluble methane clearly increased. Moreover, the efficiency of COD removal increased from 61% to 65% and the methane recovery rate increased from 39% to 52%. However, we also observed that

deterioration of sludge physical property continued during this phase of operation (phase "B", see Fig. 6 (b)). Thus, we added extra granular sludge in the end of phase B until the sludge concentration became 39.9 g VSS/L (the same level when reactor was started up). Then we re-started the reactor with the combination of UASB mode and EGSB mode (phase C). Within one month after re-starting the reactor, production of methane gas and soluble methane levels rose to 5.1 g COD/day (about 2 times of phase B2) and 3.5 g COD/day, respectively. The efficiency of COD removal and the rate of methane recovery clearly increased as well, reaching 91 % and 60 %, respectively. At the same time, the levels of effluent acetate and propionate were both less than 0.01 g COD/L (Figs. 2, 3 and 5). Moreover, we found that these excellent rates of methane production and recovery, in addition to the higher efficiency of COD removal, could be maintained for over 200 days. This result shows the effectiveness of our proposed operational mode in treatment of low strength wastewater. We also assayed the physical properties of retained sludge in continuous flow experiment (Fig. 6).

During phase A, sludge concentration (MLVSS) decreased from 39.9 g VSS/L to 27.4 g VSS/L, presumably due to wash-out of sludge and sampling loss. By the end of phase B, the sludge concentration had dropped to 17.6 g VSS/L.



Fig. 7. Methane producing activity of the retained sludge at 20°C (with H<sub>2</sub>/CO<sub>2</sub>, acetate and propionate as substrates)



Sludge settle-ability, as described by the sludge volume index (SVI), deteriorated during both phase A and phase B, reaching 33 mL/g SS at the end of phase B. After the addition of 28.5 g VSS of external granular sludge (phase C), the retained sludge concentration and sludge settle-ability kept at 33.4 g VSS/L and 25 mL/g SS (port 2) for more than 200 days. This maintenance of good physical properties of the retained sludge in phase C can be attributed to sufficient COD removal under the modified conditions; alternating operation in UASB and EGSB modes (intermittent effluent recirculation).

The methanogenic activity of retained sludge was determined at 20°C in duplicate (Fig. 7). When H2/CO2 was used as a substrate, we found a clearly increasing in activity, reaching 0.64 g COD/g VSS day at day 259, followed by a reduction to 0.22 g COD/g VSS day by day 500. The increase of hydrogen-fed activity in phase B may attribute to the loss of MLVSS concentration in the reactor (increase of COD-sludge loading). In contrast, acetate - utilizing methanogenic activity increased continuously, reaching 0.42 g COD/g VSS day by 500 days of operation. When a propionate substrate was assayed, we found that methanogenic activity was fairly stable throughout (0.095 g COD/g VSS day at day 500). Based on these results, it appeared that acetoclastic



Fig. 9. Effluent concentrations of acetate and propionate in UASB and EGSB mode

methanogenic bacteria became active and proliferated during the continuous flow the experiment, which we operated the reactor for a long period in the dual EGSB, UASB mode. Moreover, we confirmed the proliferation of genus Methanosaeta in the retained sludge by molecular biological analysis (data not shown).In order to understand how changing the influent COD concentration can affect the rate of COD consumption in the reactor, we conducted a batch feed experiment at day 487 (phase C). Moreover, we were able to determine the difference in the rates of COD consumption during UASB mode (at an up flow velocity of 0.7 m/h) and EGSB mode (at an up flow velocity of 5 m/h). During the test, the influent COD was changed from 0.1 to 0.4 g COD/L at HRT of 1 h. (Fig. 8).

At a COD influent of 0.25 to 0.4 g COD/L, COD consumption was clearly higher during operation in UASB mode than what was observed in EGSB mode. These results imply that the use of UASB mode may improve process performance when the influent COD concentration is in the range 0.25 - 0.4 g COD/L. Moreover, we also confirmed the effectiveness of use of UASB mode at inf. COD of 0.6 – 0.8 g/L in another granular sludge bed reactor (data not shown). However, for influent of less than 0.2 g COD/L, treatment in UASB versus EGSB mode have not much affect on the rate of consumption of COD. In addition, use of EGSB mode (effluent recirculation) internally had some good physical effects on the retained sludge, such as improvement of gas detachment from granular sludge and maintenance of dense structure of granular sludge. When we continued operation in UASB mode, floatation of the sludge bed caused deterioration of COD removal efficiency was observed (data not shown). In the batch feed experiment, we also assayed degradation of acetate and propionate at HRT of 1 h. Fig. 9 shows the effluent concentrations of acetate and propionate at different influent COD concentrations under operation in UASB and EGSB mode. Use of UASB mode has advantage in VFA removal as compared with EGSB mode. Effluent propionate concentration was clearly lower than effluent acetate

concentration in the same mode. The differences of effluent VFA concentrations between UASB mode and EGSB mode became small when influent COD strength lowered from 0.2 g COD/L. This suggests that propionate-degrading bacteria may have a higher affinity for the substrate than acetate-degrading bacteria. Thus, the results confirm that the half saturation constant  $K_s$  for acetate are higher than that for propionate (0.01-0.4 g COD/L for acetate and 0.007-0.014 g COD/L for propionate (Dolfing, 1985, Pavlostatis and Giraldo-Gomez, 1991 and Kato *et al.*, 1994, 1997).

### CONCLUSION

After operation in combined UASB and EGSB mode (named intermittent effluent recirculation), we observed that treatment proceeded well and was stable in the granular sludge bed reactor system. We suggest that this operation mode with low ORP of the influent may be useful for the successful anaerobic treatment of low strength wastewater (0.25-0.4 g COD/L) at moderate temperature.

### **ACKNOWLEDGEMENTS**

This study was supported by the New Energy and Industrial Technology Development

Organization (NEDO) and by the NIES special research program. We are grateful to Mr. Keita Nishiyama, Mr. Masashiro Okawara, Ms. Kaori Ooki and Ms. Yue Qin Fang for the technical assistance.

## REFERENCES

Angenent, L.T., Banik, G.C. and Sung, S. (2001). Anaerobic migrating blanket reactor treatment of low-strength wastewater at low temperature. Wat. Env. Res., **73**, (5), 567-574.

Dolfing, J. (1985). Kinetics of methane formation by granular sludge at low substrate concentrations. Appl. Microbiol. and Biotech., **22**, 77-81.

Driessen, W. and Yspeert, P. (1999). Anaerobic treatment of low, medium and high strength effluent in the agro-industry. Wat. Sci. Tech., **40**, (8), 221-228.

Kato, M.T., Field, J.A., Versteeg, P. and Lettinga, G. (1994). Feasibility of expanded granular sludge bed for the anaerobic treatment of low-strength soluble wastewaters. Biotech. and Bioeng., **44**, 469-479.

Kato, M.T., Field, J.A. and Lettinga, G. (1997). The Anaerobic treatment of low strength wastewater in UASB and EGSB reactors. Wat. Sci. Tech., **36**, (6-7), 375-382.

Kawasaki, T., Ohashi, A., Harada, H. and Syutsubo, K. (2005). High rate treatment of low strength wastewater by an EGSB reactor. Env. Eng. Res, **42**, 39-49(in Japanese).

Lettinga, G., Roersma, R. and Grin, P. (1983). Anaerobic treatment of raw domestic sewage at ambient temperatures using a granular bed UASB reactor. Biotech. and Bioeng., **25**, 1701-1723.

Lettinga, G, Hushoff Pol, L.W. and Zeeman, G (2000). Biological wastewater treatment: Part 1 Anaerobic wastewater treatment. Wageningen University, Wangeningen, The Netherlands, 5.10.

Pavlostatis, S.G. and Giraldo-Gomez, E. (1991). Kinetics of anaerobic treatment. Wat. Sci. Tech., **8**, 35-59.

Price, P.B and Sowers, T. (2004). Temperature dependence of metabolic rates for microbial growth, maintenance, and survival. Proc. Natl. Acad. Sci. USA, **101**, (13), 4631-4636.

Rebec, S. (1998). Psychrophilic anaerobic treatment of low strength wastewater. Ph.D. Thesis, Wageningen Univ., Wageningen, The Netherlands.

Rebec, S., van Lier, J.B., Lens, P., Stams, A.J.M., Dekkers, F., Swinkels, K.T.M. & Lettinga, G (1999).

Psychrophilic anaerobic treatment of low strength wastewaters. Wat. Sci. Tech., **39**, (5), 203-210.

Syutsubo, K., Sinthurat, N., Ohashi, A. and Harada, H. (2001). Population dynamics of anaerobic microbial consortia in thermophilic granular sludge in response to feed composition change. Wat. Sci. Tech., **43**, (1), 59-66.

Syutsubo, K., Harada H., Ohashi, A. and Suzuki, H. (1997). An effective start-up of thermophilic UASB reactor by seeding mesophilically-grown granular sludge. Wat. Sci. Tech., **36**, (6-7), 391-398.

Uemura, S. and Harada, H. (2000). Treatment of sewage by a UASB reactor under moderate to low temperature conditions. Biores. Tech., **72**, 275-282.