0273-2289/03/109/0227/$20.00

Electron Beam Pretreatment of Sewage Sludge Before Anaerobic Digestion

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Abstract

The pretreatment of waste-activated sludge (WAS) by electron beam irradiation was studied in order to improve anaerobic sludge digestion. The irradiation dose of the electron beam was varied from 0.5 to 10 kGy. Batch and continuous-flow stirred tank reactors (CFSTRs) were operated to evaluate the effect of the electron beam pretreatment on anaerobic sludge digestion. Approximately 30–52% of the total chemical oxygen demand (COD) content of the WAS was solubilized within 24 h after electron beam irradiation. A large quantity of soluble COD, protein, and carbohydrates leached out from cell ruptures caused by the electron beam irradiation. Volatile fatty acids production from the irradiated sludge was approx 90% higher than that of the unirradiated sludge. The degradation of irradiated sewage sludge was described by two distinct first-order decay rates (k_1 and k_2). Most initial decay reaction accelerated within 10 d, with an average k_1 of 0.06/d for sewage sludge irradiated at all dosages. The mean values for the long-term batch first-order decay coefficient (k_2) were 0.025/d for irradiated sewage sludge and 0.007/d for unirradiated sludge. Volatile solids removal efficiency of the control reactor fed with unirradiated sewage sludge at a hydraulic retention time (HRT) of 20 d was almost the same as that of the CFSTRs fed with irradiated sludge at an HRT of 10 d. Therefore, disintegration of sewage sludge cells using electron beam pretreatment could reduce the reactor solid retention time by half.

Index Entries: Sewage sludge; electron beam; sludge disintegration; biochemical acid potential test; first-order decay rate; anaerobic digestion.

Introduction

In Korea, a new regulation for the treatment of bio-solids will take effect in the year 2003. The discharge guideline prohibits the direct landfill of biowastes that contain more than 40% volatile solids (VS). Because of this

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measure, the processing and disposal of sewage sludge has become one of the major concerns in the field of biosolids treatment. Among options for the treatment of sewage sludge, anaerobic digestion is a possible alternative *(1)*. It has several advantages over other processes, such as less energy requirement, useful biogas production, higher VS reduction, and disinfection of pathogens. The digestion of waste-activated sludge (WAS), in which most organic substrate is entrapped within a cell membrane, requires the release of cell-bound substrate into the aqueous phase for viable anaerobic microbes. Therefore, the rate-limiting step of sludge digestion is generally believed to be the solubilization or hydrolysis of particulate organic matter into soluble forms *(2)*. Many novel pretreatment processes have been developed in order to disintegrate the sludge cell, leaching out the organic substance into the solution. The resulting soluble organics are obviously beneficial for improving the performance of the anaerobic process or to be used as a carbon source for the biological nutrient removal processes *(3)*. Several pretreatment alternatives for sewage sludge have been documented: mechanical disintegration by stirred ball mill, high-pressure homogenizer, and mechanical jet smash technique *(4–6)*; thermal hydrolysis *(7,8)*; chemical treatment with acid or alkali *(9,10)*; ultrasonication and ozonation *(11,12)*; and biologic hydrolysis with enzyme addition *(13)*.

In addition, a combination of simultaneous ultrasonic and alkaline treatments has recently been tried to improve the overall disintegration efficiency *(14)*. Electron beam irradiation of any system gives rise to the formation of highly reactive chemical species, such as free radicals, ions, and excited atoms and molecules. They are capable of accomplishing various radiolytic transformations of pollutants, such as redox reactions, decomposition of organic compounds, decolorization of dyes, and formation of precipitates. Thus, high-energy electron beam irradiation has been utilized in the field of groundwater remediation *(15)*; purification of drinking water *(16)*; and treatment of less biodegradable waste and wastewater, such as paper mill or dying wastewater *(17,18)*. The electron beam irradiation technique was also used to disinfect the pathogen in the field of sludge treatment *(19)* and to purify flue gas *(20)*.

In the present study, sewage sludge was irradiated with electron beam as one of the pretreatment alternatives in order to disintegrate sludge cells and consequently to enhance the organic removal efficiency in the subsequent anaerobic digestion. The specific objectives were to identify changes in physicochemical characteristics of sewage sludge on electron beam irradiation and to measure the degree of disintegration of sludge cells and to quantify the effect of electron beam irradiation on the organic removal efficiency of anaerobic sludge digestion.

Materials and Methods

Electron Beam Pretreatment and Examination of Sludge Disintegration

All experiments utilized two different kinds of sewage sludge: WAS and thickened sludge. The thickened sludge mainly consisted of primary

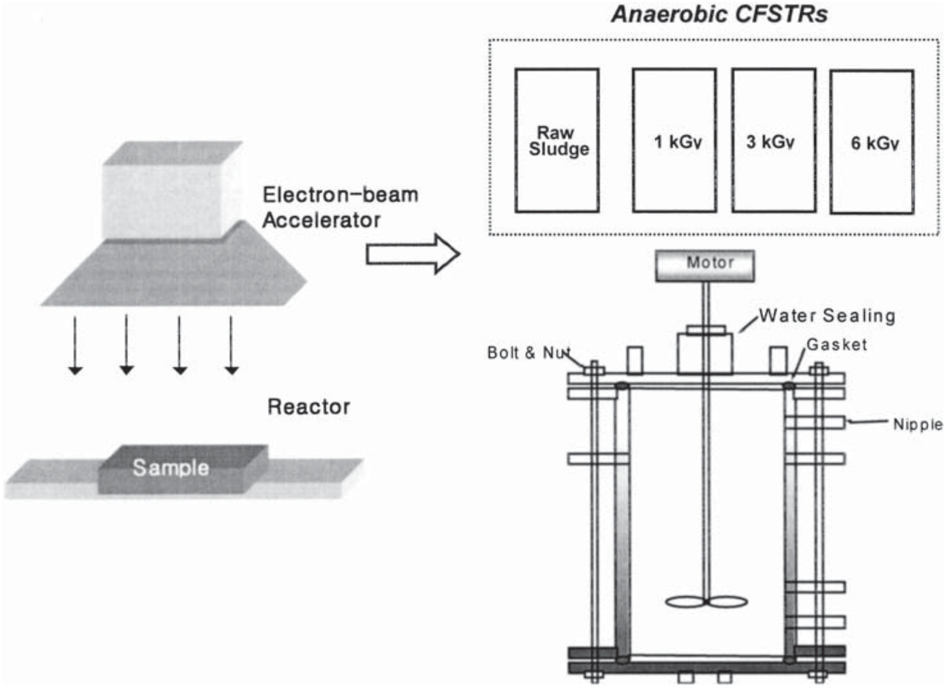

Fig. 1. Schematic diagram of electron accelerator and anaerobic digester.

sludge and WAS. The total solids (TS) content of the WAS was >1.5% and its VS was about 73% of the TS, whereas the TS content of thickened sludge ranged from 2.4 to 3.2%. As shown in Fig. 1, the sewage sludge was put in Pyrex trays (35 × 22.5 × 5.6 cm) and irradiated at different doses by a 1-MeV, ELV-4 Model electron accelerator. The degree of cell disruption and changes in physicochemical characteristics induced by electron beam irradiation was determined by measuring parameters such as pH, alkalinity, sludge volume index (SVI), specific resistance to filtration (SRF), soluble chemical oxygen demand (COD_{sol}), protein, and carbohydrate concentration in the aqueous phase before and after irradiation. The degree of disintegration was calculated as the ratio of increase in COD_{sol} by electron beam treatment and the increase in COD_{sol} by the addition of NaOH for 26 h.

Volatile Fatty Acids Production of Pretreated Sludge

The biochemical acid potential (BAP) test, a simplified version of the biochemical methane potential (BMP) test, was carried out to examine the volatile fatty acids (VFA) production from WAS irradiated at different doses. In this test, the irradiated WAS was mixed with acidifying bacteria that was derived from a food waste acid fermentor at a mixing ratio of 3:1 (v/v). The mixture was incubated in 3-L Erlernmeyer flasks in a temperature-controlled room maintained at 35 ± 1°C until the VFA production rate showed a decline.

Batch and Continuous-Flow Anaerobic Digestion

The batch tests were carried out in 5-L fermentation bottles at a constant temperature of 35°C for 50 d to compare the biodegradable substrate degradation rates of sewage sludge with and without electron beam pretreatment. The batch reactors were mixed by magnetic stirrers. Based on VS mass, the mixing ratio of substrate (irradiated WAS) to inoculum (anaerobic digester sludge) used in the batch test was 1:2.

Three continuous-flow stirred tank reactors (CFSTRs) and a control reactor were operated to evaluate the effect of electron beam treatment on the organic removal efficiency of anaerobic sewage sludge digestion. The reactors were fed with sewage sludge pretreated at an irradiation dose of 1–6 kGy. The anaerobic reactors were made of acryl and consisted of a cylindrical pipe tank whose effective volume was 18 L. All experiments were conducted in a temperature-controlled room maintained at 35 ± 1°C. The biogas was collected in a Teflon gas bag for later measurement.

Analytical Methods

The parameters of physicochemical characteristics such as soluble chemical oxygen demand (COD_{sol}), pH, alkalinity, TS, NH_3-N, total Kjeldahl nitrogen (TKN), and SRF were analyzed as proposed by standard methods *(21)*. The Anthrone carbohydrate method and the Lowry-Foline method were used to measure the carbohydrate and protein concentration in the aqueous phase of the samples, respectively. Oxidation-reduction potential (ORP) during the BAP test was monitored with a HORIBA D-25 ORP meter, and biogas volume produced was measured by a wet test gas meter. The biogas volume was then calibrated at the standard temperature and pressure (0°C, 1 atm) by compensating both the thermal expansion according to Charles' law and the volume occupied by water vapor. Normally, no pressure correction was required because biogas volumes were measured at about 1 atm. Biogas composition and VFA were analyzed using a Shimadzu Model GC-14A with a thermal conductivity detector and a flame ionization detector, respectively.

Results and Discussion

Pretreatment Results

The changes in the physicochemical characteristics of sewage sludge before and after electron beam irradiation at 0.5, 1, 3, 6, and 10 kGy are summarized in Table 1. After the disintegration of WAS and thickened sludge cells by irradiation, pH decreased slightly from 6.41 at 0 kGy to 6.17 at 10 kGy; therefore, pH was not influenced by electron beam irradiation. Note that COD_{sol} concentration was strongly related to the dose of irradiation. An increase in irradiation dose resulted in an increase in COD_{sol} concentration. As compared with unirradiated sludge (i.e., control), COD_{sol} production rates of the irradiated WAS and thickened sludge were much

Table 1
Physicochemical Characteristics of Sewage Sludge Before and After Electron Beam Pretreatment[a]

	WAS (kGy)						Thickened sludge (kGy)					
Parameter	0 (control)	0.5	1	3	6	10	0 (control)	0.5	1	3	6	10
pH	6.41	6.41	6.33	6.28	6.19	6.17	6.28	6.19	6.26	6.20	6.11	6.06
Alkalinity (mg/L as $CaCO_3$)	435	472	465	475	453	443	833	773	770	728	723	690
TS (%)	1.49	1.49	1.48	1.45	1.47	1.47	2.84	2.85	2.88	2.80	2.83	2.83
VS (%)	1.06	1.05	1.05	1.03	1.04	1.04	1.85	1.90	1.91	1.85	1.87	1.85
COD_{sol} (mg/L)	52	390	735	828	1072	1254	442	1259	1377	1560	1913	1970
Soluble protein (mg/L)	14.4	121.8	240.1	306	379.1	397.3	62.4	230.7	235.4	383.8	469.2	559.9
Carbohydrate (mg/L)	5.9	92.2	108.4	110.3	119.7	116.8	17.1	152.0	158.3	197.0	243.2	262.7
SRF ($\times 10^{16}$ m/kg)	0.45	1.31	1.70	1.72	2.04	2.24	0.37	2.08	2.29	2.52	2.06	2.27

[a]Average for 10 samples; SDs were less than ±10% over average value.

faster at a lower range of irradiation dose: 1 kGy for WAS and 0.5 kGy for thickened sludge, respectively. COD_{sol} production continuously increased but its rate leveled off when the irradiation dose exceeded 1 kGy. COD_{sol} concentrations of WAS and thickened sludge at an irradiation dose of 1 kGy were 14 and 3 times higher than those of the control, respectively. Changes in soluble protein and carbohydrate contents of WAS and thickened sludge after electron beam irradiation were more pronounced: 16.7 and 18.4 times greater than those of the control.

The significant increase in COD_{sol}, protein, and carbohydrates in the aqueous phase was attributed to the breakup of microbial cells leading to the release of intracellular substances. These highly solubilized organics could be a good substrate for the subsequent anaerobic digestion process.

The settleability of irradiated sludge was very good compared with the nontreated sludge. SVI decreased from 110–160 to 60–70 mL/g. However, the supernatant of pretreated sludge was very turbid because much of the soluble organic substances leached out into the aqueous phase owing to sludge cell disruption. The dewaterability evaluated by SRF became relatively worse after irradiation—from 0.37×10^{16} to 2.27×10^{16} m/kg.

Disintegration of Sludge After Irradiation

To quantify the degree of sludge disintegration (DR) by electron beam irradiation, Eq. 1 was described:

$$DR_{\text{based SCOD}}\ (\%) = (COD_{\text{EB treated}} - COD_{\text{untreated}})/(COD_{\max} - COD_{\text{untreated}}) \times 100 \qquad (1)$$

in which COD_{max} is the maximum particulate COD to be solubilized, which was determined by the alkali fusion process with NaOH in this study. As shown in Fig. 2A, no further soluble substances were produced, and saturation of COD_{max} was finally reached after 24 h of solubilization testing. Thus, the COD_{max} of WAS approached 11,600 mg/L out of the initial total COD (COD_{tot}) concentration of 15,300 mg/L. This means that only 76% of the initial COD_{tot} can be converted to COD_{sol} form during hydrolysis, whereas 24% cannot be used as a substrate in the biodegradation process. Figure 2B illustrates the degree of disintegration of WAS immediately after irradiation and 24 h after irradiation. It is obvious that the degree of disintegration was directly related to the irradiation dose. WAS that lasted for 24 h at room temperature after irradiation produced more soluble products than that of WAS immediately after irradiation.

Accordingly, the degree of disintegration was 20–35% for WAS immediately after irradiation, whereas it was 30–52% for WAS 24 h after irradiation. This was probably owing to the uncontrolled enzymatic activity as a result of enzyme release on cell disintegration.

BAP Test for Irradiated Sludge

Various kinds of VFA (C_2–C_8) of WAS pretreated at different irradiation doses were examined through BAP tests. VFA were summed up and

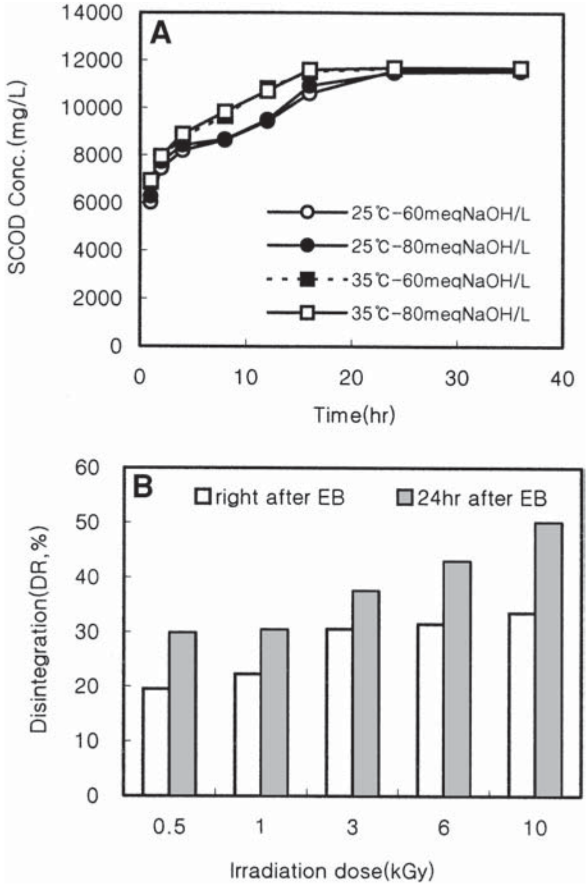

Fig. 2. Determination of COD_{max} of WAS by alkali fusion treatment **(A)** and degree of sludge disintegration after electron beam irradiation **(B)**. SCOD = COD_{sol}.

expressed as acetic acids concentration. Figure 3 shows the profile of COD_{sol}, VFA, and ORP during operation of the acid fermentors. Unirradiated WAS produced a very small amount of VFA only for the initial 16 h, whereas WAS irradiated at less than 1 kGy steadily produced VFA for up to 48 h of acidification. As shown in Fig. 3D,E, a further increase in the irradiation dose of 3 and 6 kGy prolonged VFA production for 72 h of experimentation. This phenomenon might be explained by the large amount of soluble substances leaching from the sludge cell owing to higher irradiation doses continuously being converted into short chain fatty acids during the longer period of the experiment.

Interestingly, it was observed that ORP was conversely related to COD_{sol}. As shown in Fig. 3, the ORP curves decreased with a significant increase in COD_{sol} for the initial 16 h. Afterward, the ORP curves increased whereas the COD_{sol} curves leveled off. This observation coincided well with the results of Chiu et al. in their alkaline hydrolysis experiments.

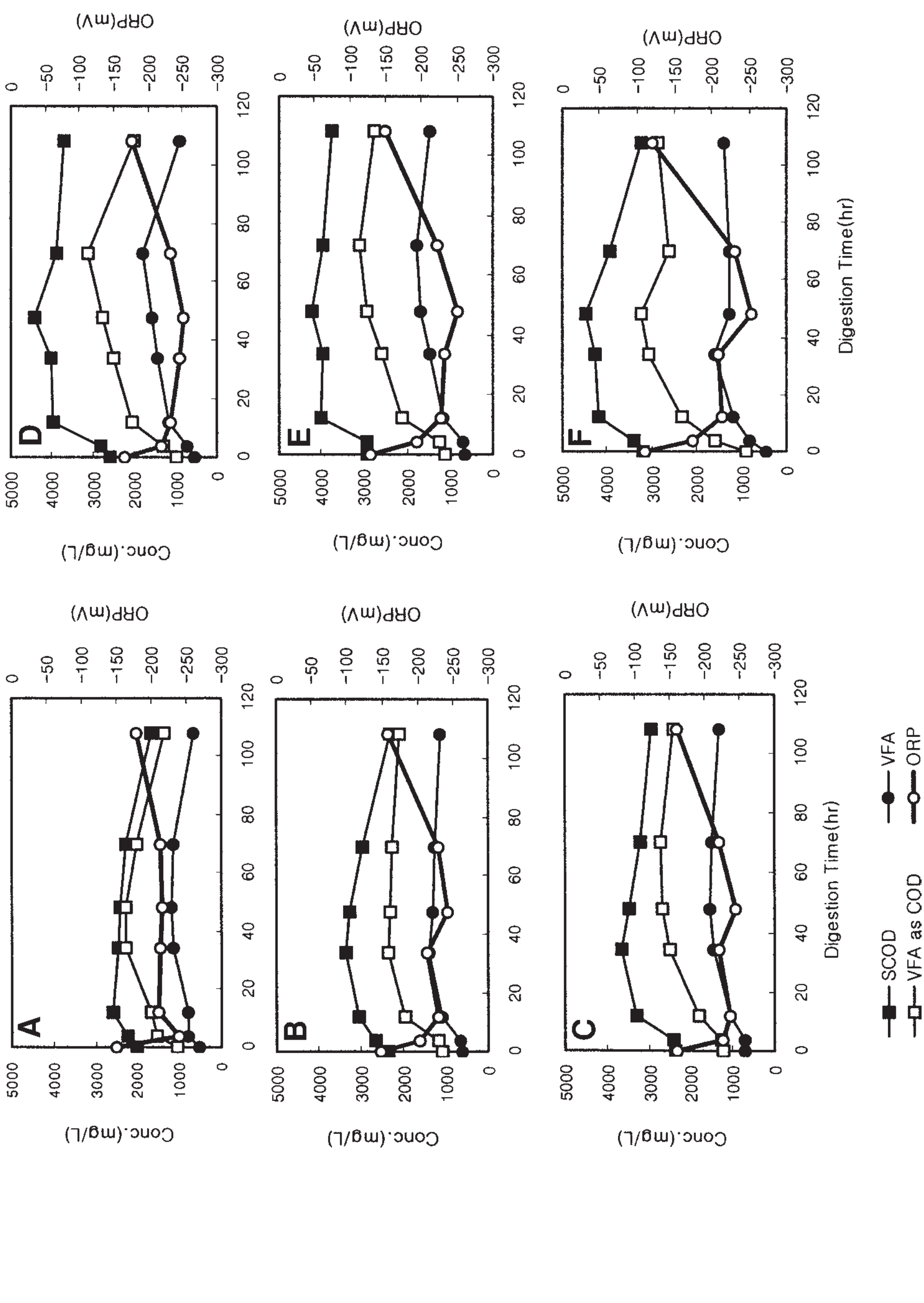

Fig. 3. Variation in VFA, COD_{sol} (SCOD) and ORP in BAP test: **(A)** untreated sludge; **(B)** 0.5 kGy; **(C)** 1 kGy; **(D)** 3 kGy; **(E)** 6 kGy; **(F)** 10 kGy.

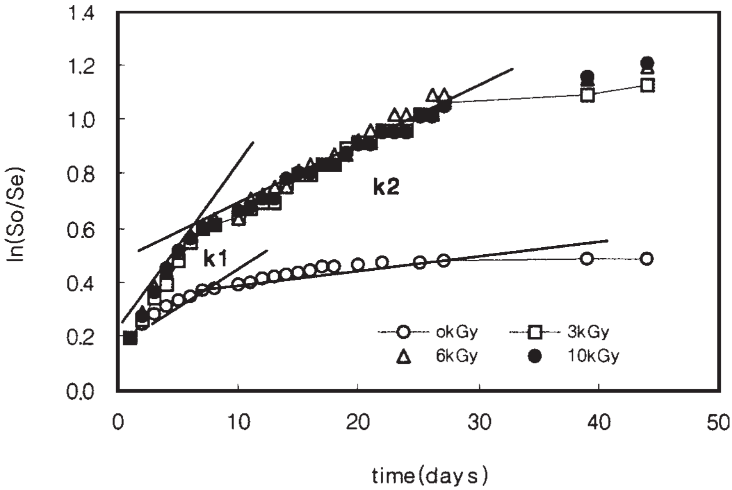

Fig. 4. Graphic illustration of first-order decay rate coefficients (k_1 and k_2) in mesophillic sewage sludge batch reactor at different irradiation dose.

However, in our study, the ORP might be used as one of the rapid tools to monitor acidification as well as hydrolysis of biosolids.

First-Order Decay Rate of Pretreated Sludge

To evaluate the effect of electron beam irradiation on the first-order decay rate of biodegradable sewage sludge, anaerobic mesophilic batch tests were carried out on substrate-to-inoculum ratios (S/I) of 0.5.

The integral form of the basic equation for the first-order removal of a biodegradable substrate in an anaerobic batch system is expressed in Eq. 2:

$$\ln(So/Se) = kt \tag{2}$$

in which, *So* is the initial substrate BVS mass (g); *Se* is the BVS mass (g) at time, *t*; and *k* is the first-order decay rate coefficient (d^{-1}).

The slope of the linear plot of $\ln(So/Se)$ vs *t* yields the first-order decay rate coefficient, *k*, that represents the change in biodegradable volatile solids (BVS) concentration with time. The first-order reaction yields a straight line, indicating a single decay rate coefficient. However, most batch reactions observed in our study were not continuously linear, as shown in Fig. 4. The reaction curves usually consisted of the linear form with a steep slope (k_1) during the initial period of operation, followed by a gradual nonlinear transition, and then a second linear line with a more gradual slope (k_2). It was easy to separate these two reaction rate coefficients (k_1 and k_2) by simple visual fitting. As shown in Fig. 4, at the S/I ratio of 0.5, most initial decay reactions accelerated within 10 d with an average k_1 of 0.06/d for sewage sludge irradiated at all doses. The mean values for the long-term batch first-order decay coefficient (k_2) were 0.025/d for irradiated sewage sludge and 0.007/d for unirradiated sewage sludge. These results indicate that disintegration of sewage sludge using an electron beam could accelerate the organic degradation rates and result in operating anaerobic digesters at a much shorter solid retention time (SRT).

Operation of CFSTRs

CFSTRs were operated to verify their reactor performance with and without electron beam pretreatment of sewage sludge. A stepwise decrease in hydraulic retention times (HRTs) and an increase in organic loading rates (OLRs) was used, reducing the HRT from 20 to 10 d. The results of operating CFSTRs fed with sludge irradiated at different doses are summarized in Table 2. OLRs based on VS at each reactor HRT were relatively constant, but OLRs based on COD_{sol} varied according to the different amount of soluble substance produced as a result of different irradiation doses applied to sewage sludge.

During the operation of the CFSTRs, the pH level of all four reactors remained neutral at all HRTs. The reactor alkalinity stayed above 2000 mg/L as $CaCO_3$ at an HRT of 20 and 15 d, respectively. However, alkalinity levels at an HRT of 10 d were slightly lower owing to the higher organic load.

VS removal efficiencies were compared among CFSTRs at different reactor HRTs and at different irradiation doses, as illustrated in Table 2. It became clear that the VS removal efficiencies of CFSTRs gradually increased as a function of irradiation dose but decreased as the HRT became shorter. At an HRT of 20 d, a conventional SRT for anaerobic digesters, the unirradiated control reactor measured 37% VS removal efficiency, whereas CFSTRs fed with irradiated sludge at 1, 3, and 6 kGy measured 51, 57, and 60%, respectively. Even at the much shorter HRT of 10 d, the CFSTR fed with irradiated sludge at 6 kGy resulted in a better VS removal efficiency (38.2%) than that of the unirradiated control reactor at the longer HRT of 20 d. Thus, it is likely that electron beam pretreatment of WAS could reduce its reactor HRT by half.

It was often observed that the biogas methane content decreased as the reactor OLR increased owing to the large quantity of CO_2 produced in the hydrolytic reaction. The biogas methane contents of CFSTRs operated at all operation conditions remained above the range of 60–72% for the entire experiment. The volume of biogas produced from each CFSTR was expressed as the biogas productivity, in liters of biogas produced per volume of reactor per day (L/[m^3·d]). The average biogas production rates of CFSTRs fed with sludge irradiated at 1, 3, and 6 kGy were 180, 260, and 290 (L/m^3·d), respectively, at an HRT of 15 d, while that of the unirradiated control was 95 (L/m^3·d). Accordingly, CFSTRs fed with sludge irradiated at 1–6 kGy yielded 189–287% more than that of the control reactor. These results are similar to those of Lin et al. *(10)*, who worked with WAS pretreated with 20–40 meq of NaOH/L. Such a remarkable increase in biogas production at a relatively short HRT was owing to the large quantity of soluble organic materials leached out by the electron beam irradiation.

Table 2
Results of Anaerobic Digesters Fed with Unirradiated and Irradiated Sludge[a]

	Reactor A (unirradiated sludge)			Reactor B (1 kGy)			Reactor C (3 kGy)			Reactor D (6 kGy)		
Parameter	20 d	15 d	10 d	20 d	15 d	10 d	20 d	15 d	10 d	20 d	15 d	10 d
OLR (g VS/[L·d])	1.06	1.33	1.84	0.94	1.31	1.92	0.94	1.25	1.90	0.96	1.25	1.87
OLR (g COD_{sol}/[L·d])	50.3	62.0	87.0	87.9	110.9	143.6	102.5	149.8	198.5	114.5	163.7	224.1
Reactor pH	7.1	7.3	7.1	7.0	7.1	6.8	7.0	7.1	6.9	7.0	7.1	6.9
Reactor alkalinity (mg/L as $CaCO_3$)	2050	2410	1980	2220	2350	2100	2380	2500	2150	2300	2460	1940
Influent VS (%)	1.92	2.15	1.81	1.91	1.96	1.83	1.88	1.91	1.85	1.90	1.89	1.85
Influent COD_{sol} (mg/L)	890	910	870	1675	1690	1550	2040	2270	2105	2290	2480	2350
VS removal (%)	36.7	32.5	22.3	51.4	42.0	30.2	56.7	48.1	32.3	60.3	50.4	38.2
Biogas (L/[m^3·d])	82	95	65	155	180	175	230	260	235	236	290	231
Methane contents (%)	69	65	59	68	66	60	72	69	61	71	64	62
VFA (mg/L as C_2)	97	102	100	123	109	121	129	135	154	152	156	142
SRF ($\times 10^{12}$ m/kg)	32.0	39.7	70.1	28.7	34.4	72.1	26.8	36.2	80.4	29.5	44.4	94.5

[a]SDs were less than ±10% over average value.

Conclusion

As one of the pretreatment alternatives for sewage sludge, electron beam irradiation was examined to identify changes in physicochemical characteristics of sewage sludge treated with electron beam irradiation and to measure the degree of disintegration of sludge cell and to quantify the effect of electron beam pretreatment on the destruction efficiency of anaerobic sludge digestion. Of the COD_{tot} content of the WAS 30–52% was solubilized within 24 h after electron beam irradiation at 0.5–10 kGy. The significant increase in COD_{sol}, protein, and carbohydrates was attributed to the cell rupture caused by irradiation. The solubilization of sewage sludge and its conversion to short chain fatty acids were strongly related to the degree of irradiation applied.

The degradation of irradiated sewage sludge was described by two distinct first-order decay rates (k_1 and k_2). Most of the initial decay reaction accelerated within 10 d with an average k_1 of 0.06/d for sewage sludge irradiated at all dosages. The mean values for the long-term batch first-order decay coefficient (k_2) were 0.025/d for irradiated sewage sludge and 0.007/d for unirradiated sludge. VS removal efficiency of the control reactor fed with unirradiated sewage sludge at an HRT of 20 d was almost the same as that of CFSTRs fed with irradiated sludge at an HRT of 10 d.

In conclusion, disintegration of sewage sludge cells using electron beam treatment could reduce the reactor SRT by half.

Acknowledgment

This research was supported by the Korea Institute of S&T Evaluation and Planning (KISTEP contract no. M2-0106-00-0056-01-B05-00-013-0-0, Korea).

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