CO-DIGESTION OF SEWAGE SLUDGE AND MATURE LANDFILL LEACHATE IN PRE-BIOAUGMENTED SYSTEM

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The study examined the effects of co-digestion of sewage sludge and mature landfill leachate at the volumetric ratio of 95:5% in primarily bioaugmented system. Bioaugmentation was carried out with the use of commercial product Arkea® in the volumetric dose of 5% and lasted three months prior to the co-digestion start-up. Co-digestion was undergone without bioaugmentation. The results indicated that in the first period (of three months) following bioaugmentation, co-digestion led to biogas/methane yields only 5-8% lower as compared to anaerobic digestion of sewage sludge, and the differences were not statistically significant. Moreover, a comparable value of volatile solids removal was obtained. However, the effects became worse over time, i.e. a lower organics removal efficiency of 16% as well as 9.5-13% decreases of biogas/ methane yields were achieved by applying co-digestion for a further period (of the same duration). Co-digestion of sewage sludge and mature landfill leachate could be recognized as quite efficient in the system that was primarily bioaugmented with the use of Arkea®. However, the beneficial impact of bioaugmentation remained for the limited period of three months after its completion. To sustain the favourable effects a periodical, repeatable bioaugmentation of the co-digestion system is required.

Keywords: anaerobic co-digestion; primary bioaugmentation; sewage sludge; mature landfill leachate; biogas/methane yields.

INTRODUCTION

Landfill has been employed worldwide for municipal solid waste disposal due to low operational cost and cheap exploitation. Leachate generated there as a result of waste degradation in combination with percolating rainwater, is a high-strength contaminated liquid that may contain large concentrations of organic matter, inorganic macro components (i.e. ammonia nitrogen), heavy metals and xenobiotic compounds (i.e. aromatic hydrocarbons, phenols, chlorinated aliphatics and pesticides). Leachate composition depends on many factors, such as morphology and the type of waste disposed, compaction of waste layers, landfill hydrology, seasonal weather variation, site operations and management and, particularly, on landfill age and the corresponding waste decomposition stage [Kurniawan et al. 2006, Renou et al.

2008]. According to the latter factor, the leachate classification comprises three categories: young, intermediate and mature (old). With an increase of landfill age, leachate characteristics significantly differ. The mature one (classified as stabilized) is typically represented by relatively low chemical oxygen demand (COD < $3000-4000 \text{ mg} \cdot \text{dm}^{-3}$), low biodegradability (BOD₅/COD < 0.1) and dominance of refractory, high molecular weight compounds such as humic and fulvic substances that prevail as components of leachate organic fraction. On the contrary, high concentration of ammonia nitrogen (NH₄-N> 400 mg \cdot L⁻¹) and dissolved salts (notably chlorides, carbonates and sulphates) could be expected [Pi et al. 2009, Foo and Hameed 2009]. Other constituents are as follows: slightly basic pH 7.5-8.5, total organic carbon TOC/COD ratio > 0.5 and low heavy metals concentration (below 2 mg·dm⁻³) [Li et al. 2010].

The composition of mature leachate indicates both biodegradability enhancement and ammonia nitrogen removal to be a critical issue of the treatment [Pi et al. 2009]. Due to the presence of biorefractory and toxic contaminants [Eggen et al. 2010] suppressing the biodegradation, unassisted biological systems involving conventional aerobic activated sludge (CAS), aerobic or anaerobic sequencing batch reactors (SBR) and up-flow anaerobic sludge blanket reactors (UASB) did not assure a requisite efficiency, even if a multi-stage configuration was employed. In order to achieve the levels of purification required to reduce the environmental adverse effect, various frequently combined biological and physicochemical treatment of leachate has been applied [Wiszniowski et al. 2006, Abbas et al. 2009, Li et al. 2009, Cortez et al. 2011]. Among the physicochemical methods incorporated as an additional treatment stage, techniques such as coagulation-flocculation, ammonia-stripping, chemical precipitation, activated carbon adsorption, advanced oxidation (AOP) and membrane separation (MBR) have been widely investigated in the last decade [Kurniawan et al. 2006, Bohdziewicz et al. 2008, Foo and Hameed 2009, Li et al. 2010, Umar et al. 2010, Ahmed and Lan 2012]. Interestingly, two main approaches have been considered. One of them concerned the physicochemical pretreatment, converting initially biorecalcitrant compounds to more readily biodegradable components degraded during the subsequent biological stage. Post-treatment (last purification) could also be recommended when the removal of recalcitrant organic micro pollutants and toxicity is required.

Among many technological systems, codigestion of sewage sludge and mature landfill leachate has also been studied [Lebiocka et al. 2010]. The findings indicated the method was practically insufficient due to the unacceptably low efficiency resulting probably from the presence of hardly biodegradable organic compounds as well as possible toxic influence of leachate on digester' biomass. However, in the last decade the new promising approach appeared to be incorporating a strategy that leads to improvement of several biological and chemical processes. This technique, bioaugmentation, consists in the application of a specific strain or a consortium of microorganisms to enhance a required biological activity in the system investigated. In anaerobic digestion it has been used more frequently to improve both process stability and biogas yields.

Interestingly, Schauer-Gimenez et al. [2010] applied this method for reducing the recovery time of digesters exposed to toxic events. In order to obtain favourable results investigating co-digestion of sewage sludge and mature leachate, application of the organisms from the archaeal domain seems to be promising because of their adaptations to extreme habitats, including environments of high salt, high temperature, low pH and acute anoxia [McLain, 2007]. However, to make the process most cost-effective and minimize the operational costs, primary bioaugmentation should be considered.

In the present study, the effects of co-digestion of sewage sludge and mature landfill leachate have been examined in applying an anaerobic digestion system that was primarily bioaugmented with the use of the commercial product Arkea[®] containing organisms from the archaeal domain.

MATERIALS AND METHODS

Sewage sludge (primary and waste sludge after thickening) was obtained from the Puławy municipal wastewater treatment plant (Poland). Thickened sludge was sampled once a week in the WWTP, then transported and mixed at a volume ratio of 60:40 (primary waste : sludge) under laboratory conditions. The sludge samples were homogenized, screened through a 3 mm screen and stored at 4 °C in a laboratory fridge for a week at the longest. Sludge prepared in this manner fed the digester as mixed sewage sludge (SS). The main characteristics of SS during experiments are presented in Table 1.

Leachate was sampled once as a collected sample achieved from a storage tank of mature leachate in the Rokitno municipal solid waste landfill (the age of landfill exceeded 20 years). Under laboratory conditions it was homogenized and partitioned, then frozen and stored at 25 °C in a laboratory freezer. The leachate samples were thawed daily for 6 hours at 20 °C in indoor air. The mature leachate (L) composition is presented in Table 2.

The commercial product Arkea[®] containing organisms from the archaeal domain was used for prior bioaugmentation and prepared as a solution in continuous mode according to the procedure given by ArchaeaSolutions Inc. The average TS and VS of the Arkea[®] liquor were 0.47 and 0.042 g·kg⁻¹, respectively, and pH was 7.16.

Parameter	Unit	Experiment 1	Experiment 2	
Chemical oxygen demand (COD)	kg m ⁻³	41.7 ± 4.6	44.9 ± 8.0	
Total solids (TS)	g kg⁻¹	33.2 ± 4.31	36.7 ± 6.02	
Volatile solids (VS)	g kg⁻¹	25.3 ± 3.41	26.3 ± 5.05	
рН	-	6.89 ± 0.96	6.55 ± 0.33	
Total nitrogen (TN)	g m-3	2228 ± 468	3324 ± 327	
Ammonia nitrogen (N-NH4+)	g m⁻³	64 ± 43	49 ± 29	
Total phosphorus (TP)	g m-3	464 ± 131	582 ± 221	
Phosphate phosphorus (P-PO ₄ ³⁻)	g m-³	122 ± 29	118 ± 26	

Table 1. Sewage sludge (SS) composition (the mean value and standard deviation are given)

Table 2. Leachate composition (the mean value and standard deviation are shown)

Parameter	Unit	Mean value	
Chemical oxygen demand (COD)	kg m⁻³	5.6 ± 0.2	
BOD₅/COD	-	0.05 ± 0.01	
Total solids (TS)	g kg⁻¹	25.4 ± 0.95	
Volatile solids (VS)	g kg⁻¹	14.3 ± 0.62	
Alkalinity	g m ⁻³	3950 ± 65	
рН	-	7.95 ± 0.06	
Volatile fatty acids (VFA)	g m-3	3960 ± 225	
Total nitrogen (TN)	g m ⁻³	7200 ± 123	
Ammonia nitrogen (N-NH ₄ +)	g m-3	6915 ± 114	
Total phosphorus (TP)	g m-3	71 ± 15.1	
Phosphate phosphorus (P-PO ₄ ³⁻)	g m-3	39 ± 8.4	

The study was carried out in reactors operating at the temperature of 35 °C in semi-flow mode. The laboratory installation consisted of two completely mixed anaerobic digesters (with an active volume of 40 dm³) working in parallel, equipped with a gaseous installation, an influent peristaltic pump and storage vessels. The gas system consisted of pipelines linked with a pressure equalization unit and a mass flow meter. The scheme of the installation is shown in Figure 1. An inoculum for the laboratory reactors was obtained from the Puławy wastewater treatment plant as a digest collected from an anaerobic digester operating at mesophilic temperature. The adaptation of the digester biomass was achieved after 30 days. Then, the primary phase started: the first reactor was fed regularly once a day with sewage sludge (SS of 2 dm³ volume) and operated without bioaugmentation, the second one was supplied with SS with Arkea[®] addition in the dose 0.1 dm³. This phase lasted three months.

Actual investigations following by the primary phase were devoted to co-digestion of sewage sludge and mature leachate and undergone without bioaugmentation. The study was divided into two experiments taking into consideration the duration after the bioaugmentation: Experiment 1 was carried out immediately after, Experiment 2 appeared consequently in a further period to evaluate the system efficiency over time. Each of them consisted of two runs lasting 90 days (30 days for adaptation and 60 days for measurements) and were carried out simultaneously to the control in two parallel systems. The digesters were supplied regularly once a day with an applied volume of the SS or mixture of SS and L. The feed composition and operational parameters are shown in Table 3.



Figure 1. Laboratory installation of anaerobic digestion of waste in a wet system

Run	Food composition	Feed volume	HRT*	OLR**				
	Feed composition	dm³	d	kg VS m⁻³d⁻¹				
Experiment 1								
R 1.1 (control)	SS (sewage sludge)	2.0	20	1.27				
R 1.2	SS + L 95:5 v/v	2.0 + 0.1	19.1	1.30				
Experiment 2								
R 2.1 (control)	I) SS		20	1.32				
R 2.2	2.2 SS + L 95:5 v/v		19.1	1.35				

Table 3. Feed composition and operational regime during experiments

* HRT – hydraulic retention time, ** OLR – organic loading rate

In the sewage sludge and its supernatant the following parameters were analyzed once a week: total chemical oxygen demand (COD), total solids (TS), volatile solids (VS), volatile fatty acids (VFA), alkalinity and pH level. Moreover, total nitrogen, ammonia nitrogen (N-NH₄⁺), nitrite and nitrate nitrogen (N-NO_x⁻), total phosphorus and ortho-phosphate phosphorus (P-PO₄³⁻) were also determined. The supernatant samples were obtained by centrifuging the sludge at 4000 r min⁻¹ for 30 min.

The leachate composition was analyzed once after providing it to the laboratory. The values of the parameters were examined using the sewage sludge schedule besides biochemical oxygen demand (BOD_s).

In the digest and its supernatant, the specified parameters were determined twice a week, in accordance with the timetable accepted. Most analyses were carried out in accordance with Polish Standard Methods. The analyses of N-NH₄⁺, N-NO_x and P-PO₄³⁻ were performed with FIASTAR 5000 using FOSS analytical methods. Ammonium was determined according to ISO 11732, nitrite and nitrate according to ISO 13395, and orthophosphate in accordance with ISO/FDIS 15681-1.

The anaerobic digestion efficiency was controlled by the daily evaluation of the biogas yield and its composition. Moreover, the volatile solids removal efficiency was evaluated according to Polish Standard Procedure. Biogas production was determined using digital mass flow meter Aalborg (USA). The composition of the biogas was measured using gas chromatograph Trace GC-Ultra coupled with a thermal conductivity detector (TCD) fitted with DVB packed columns. The Rt-Q-Bond column was used to determine CH₄ and CO₂ concentrations. The parameters used for the analysis were as follows - injector 50 °C and detector 100 °C. The carrier gas was helium with a flux rate of 1.5 cm³·min⁻¹. Peak areas were determined by the computer integration program (CHROM-CARD).

RESULTS AND DISSCUSSION

The characteristics of feed (reactors' influent) and digest (reactors' effluent) for the specified experiments are listed in Tables 4 and 5 (the average value \pm standard deviation are given). It should be noticed that feed conditions varied throughout the runs, which was attributed to the changes in sludge characteristics and the leachate addition.

The composition of feed supplemented by leachate (runs R 1.2 and R 2.2) did not essentially differ compared to SS (runs R 1.1 and R 2.1) for total solids, volatile solids and pH. TS and VS average values decreased to a little extent, respectively 1% and 2%, while pH was much the same. On the contrary, a clearly visible influence was found in relation to alkalinity and VFA (Table 5). These values were higher in the presence of leachate (as its composition indicated); the alkalinity increased by 17 and 23%, respectively for Experiment 1 and 2, and the increment of VFA concentration was of 27% (for R 1.2) and 16% (for R 2.2). The differences observed were statistically significant. Amazingly, the leachate's VFA concentration was untypically high regarding both the landfill age and low BOD₅/COD ratio.

The changes in feed volume (resulting from using leachate apart sewage sludge) led to small decreases in the hydraulic retention time (HRT) from 20 to 19.1 days, while organic loading rate (OLR) increased by about 2% with the highest value of 1.35 kg VS m⁻³ d for R 2.2 (Table 3). Anaerobic digestion proceeded in a stable way and pH remained in the range typical of methanogenesis 7.56–7.77 (with higher values corresponding with co-digestion runs). In Experiment 1, the digestion efficiency based on VS removal was comparable for both runs (Table 1), despite the shortened HRT. Contrarily, a decrease of η_{VS} appeared over time (Experiment 2), indicating the co-digestion response to pre-bioaugmentation being time-depen-

	Experiment 1				Experiment 2				
Deremeter	Runs								
Falameter	R 1.1		R 1.2		R 2.1		R 2.2		
	feed	digest	feed	digest	feed	digest	feed	digest	
TS	33.2 ± 4.3	21.4 ± 0.6	32.8 ± 4.1	22.0 ± 1.1	36.7 ± 6.0	27.2 ± 2.7	36.2 ± 5.7	28.0 ± 4.1	
η_{TS}	35.9 ± 9.8		34.1 ± 10.0		29.7 ± 6.7		29.6 ± 5.3		
VS	25.3 ± 3.4	14.5 ± 0.5	24.7 ± 3.2	14.3 ±	26.3 ± 5.1	17.5 ± 1.3	25.8 ± 4.8	17.4 ± 1.1	
η_{vs}	42.7 ± 9.2		43.0 ± 9.7		38.0 ± 5.7		32.9 ± 13.7		

Table 4. Total solids and volatile solids, as well as their removal efficiency η for runs (TS and VS expressed in g kg⁻¹, efficiency reported as a percentage value)

Table 5. Alkalinity, pH value and volatile fatty acids (VFA) for runs (concentrations reported in g m-3)

Parameter	Experiment 1				Experiment 2				
	Runs								
	R 1.1		R 1.2		R 2.1		R 2.2		
	feed	digest	feed	digest	feed	digest	feed	digest	
Alkalinity*	853 ± 328	2916 ± 313	1001 ± 313	3333 ± 215	668 ± 68	2650 ± 87	824 ± 65	3034 ± 128	
pН	6.89 ± 0.96	7.56 ± 0.13	6.91 ± 1.04	7.62 ± 0.16	6.55 ± 0.32	7.67 ± 0.16	6.56 ± 0.47	7.77 ± 0.20	
VFA	591 ± 190	136 ± 47	751 ± 181	180 ± 124	894 ± 217	202 ± 74	1040 ± 206	215 ± 83	

* Alkalinity given in g CaCO₃ m⁻³

dent (the longer the duration after bioaugmentation, the minor the co-digestion efficiency).

The average biogas yields for the experiments are shown in Figure 2. It is worth noticing a similar, time-dependent tendency for the results obtained. In the first period following bioaugmentation (Experiment 1), the daily biogas production was a little lower for the run R 1.2 with leachate addition $(18.1 \pm 2.4 \text{ dm}^3 \cdot \text{d}^{-1}; \text{ mean} \pm \text{ standard de-}$ viation is given) as compared to sewage sludge $(18.7 \pm 2.2 \text{ dm}^3 \cdot \text{d}^{-1})$. Similarly, biogas yields calculated per kg VS removed amounted to 0.87 \pm $0.31 \text{ m}^3 \cdot \text{kg}^{-1}$ and $0.93 \pm 0.33 \text{ m}^3 \cdot \text{kg}^{-1}$, respectively. Analogous decreases were achieved per kg VS added (values of 0.35 ± 0.05 and 0.37 ± 0.05 m³·kg⁻¹, respectively) as well as per kg TS added and removed (Figure 2a, c). In all the cases the difference was not statistically significant and did not exceed a few percent.

According to the author's previous investigations [Lebiocka et al., 2010], application of the same dose of same-sourced mature leachate as a co-substrate for sewage sludge anaerobic digestion (pre-bioaugmentation was not involved) led to much higher, statistically significant decreases in biogas yields of 40%. The results achieved in the present study seem to indicate a favorable impact of pre-bioaugmentation on the co-digestion process immediately following and lasting for three months. The Arkea[®] addition could improve the sludge digestion, probably due to enhanced activity of microorganisms involved in bioaugumenting systems and their resistance to toxic factors. This explanation is consistent with the research by Duran et al. [2006] regarding selected strains of the *Baccillus*, *Pseudomonas* and *Actinomycetes* species used for bioaugmentation.

Experiment 2, scheduled as a continuation of SS and L co-digestion in a further period (of the same duration), indicated the worsening of the results, despite the sustained operational conditions. A comparison of biogas yields, calculated both per kg VS and TS added and removed, showed much lower values in the presence of mature leachate as compared to SS (Figure 2b, d). Moreover, the yields achieved exceeded the levels reported for Experiment 1, although the differences still had no statistical significance. The daily biogas production unfavorably decreased from $23.2 \pm 3.1 \text{ dm}^3 \cdot \text{d}^{-1}$ for sewage sludge (R 2.1) to $22.0 \pm 2.3 \text{ dm}^3 \cdot \text{d}^{-1}$ for co-digestion process (R 2.2). The results were consistent with the study of Nielsen et al. [2007], which indicated bioaugmented anaerobic digestion of cattle manure to assure a high increase of methane yield in a two-stage thermophilic system, however for only a limited time after inoculation.

A similar average methane content was noted in biogas for runs applying leachate $(53.4\% \pm 0.74)$ and $53.0\% \pm 1.01$, respectively for R 1.2 and R 2.2), however it was a little lower compared to



Figure 2. The biogas yields obtained in specified experiments and expressed per kg: a), b) of organic compounds removed; c), d) of organic compounds added to the system (error bars represent confidence levels, $\alpha = 0.05$)



Figure 3. The methane yields obtained in specified experiments and expressed per kg: a), b) of organic compounds removed; c), d) of organic compounds added to the system (error bars represent confidence levels, $\alpha = 0.05$)

sewage sludge ($54.0\% \pm 1.37$ and 53.60 ± 1.01 , respectively for R 1.1 and R 2.1). Consistently, the average methane yields decreased when co-digestion occurred (Figure 3).

Higher drops appeared in Experiment 2 that was carried out three months after the completion of bioaugmentation. The results seem to confirm the usefulness of pre-bioaugmentation with the commercial product Arkea[®] for enhancing co-digestion efficiency of sewage sludge and mature leachate, however the beneficial impact should be regarded as time-dependent. It remained for the limited period of three months after completion of bioaugmentation and then the results appeared to be worse. This indicates the requirement of periodical, repeatable bioaugmentation of the co-digestion system as the lifetime of the microorganisms added is limited in the reactor's conditions.

CONCLUSIONS

The results indicated that co-digestion of sewage sludge and mature landfill leachate mixed at the volumetric ratio of 95:5% could be quite efficient in the system that was primarily bioaugmented with the use of commercial product Arkea[®]. However, the beneficial impact of bioaugmentation remained only for the limited period of three months after its completion. To sustain the favourable effects a periodical, repeatable bioaugmentation of the co-digestion system is required.

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