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AEROBIC COMPOSTING AND ANAEROBIC DIGESTION OF PULP AND PAPER MILL SLUDGES

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ABSTRACT

The feasibility of aerobic vessel composting and anaerobic digestion for the treatment of pulp and paper mill sludges were studied. The composting studies made use of primary and secondary sludge from a de-inking and paper mill. In six parallelly aerated 500 l vessels with various carbon : nitrogen (C:N) -ratios, the most optimal performance was obtained with C:N -ratios of c. 22-35, while higher and lower ratios delayed the temperature increase. With the optimal ratios, the thermophilic stage was reached within 36 h, and the stage lasted for about seven days. In the scale-up study (18 m^3 compost vessel), the thermophilic stage was reached within 24 h. An effective dehydratation of the mass was obtained as the total solids (TS) content of the compost increased from 31.3% to 63.8% within 21 days.

The anaerobic digestion of pulp and paper mill sludges was studied using two mesophilic 5 l digesters, their feed sludges consisting of a mixture of municipal sewage sludge and primary and secondary sludge from a pulp and paper mill. With this feed mixture and with a loading rate of about 1.0 kg volatile solids (VS)/m^3d a removal of about 27 to 40% VS and methane production of about 180 l/kgVS_{added} feed sludge were achieved during the 80 d study period.

The study showed that pulp and paper mill sludges are amenable to both aerobic composting and anaerobic digestion. © 1997 IAWQ. Published by Elsevier Science Ltd

KEYWORDS

Anaerobic digestion; composting; pulp and paper mill sludges; vessel composting.

INTRODUCTION

In the external treatment of pulp and paper industry wastewaters, mechanical treatment with primary clarifiers is usually applied to remove wastewater solids and the organic load thereof, while dissolved organic materials are commonly removed in biological aerobic treatment. The amounts and characteristics of the sludges generated in primary and secondary clarifiers (normally settling tanks) depend largely on both pulp and paper manufacturing processes and the wastewater treatment processes. For example, secondary

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sludge production has been 0.8-1.2 and 0.2-0.6 kg total solids (TS)/kg biological oxygen demand [BOD_{7rem}] for high (sludge load >0.65 kgBOD/kg mixed liquor volatile suspended solids [MLVSS]d) and low (sludge load <0.65 kgBOD/kgMLVSSd) loaded pulp and paper industry activated sludge treatment plants, respectively (Saunamäki, 1988). Primary sludge production varies from 5 to 60 kg per ton of pulp or paper, depending on the manufacturing process, and secondary sludge production e.g. 15 kg per ton of chemical pulp. Total daily sludge production varies from mill to mill, e.g. in Finnish pulp and paper mills from less than 10 to 55 t/d (about 30% TS) (Saunamäki, 1988).

In general, primary sludge consists mainly of cellulose, hemicellulose, lignin, and other components of wood furnish, and it may additionally contain process chemicals (such as filling chemicals) and bark. In primary sludge, fiber and ash may account for 40 to 95% and 5 to 60% of the solid matter, respectively (Kyllönen, 1986). Secondary sludge comprises biomass, cellulose fibers and wood-derived slowly biodegradable substances (such as lignin). Pulp mill activated sludges contain protein (22-52%), lignin (20-58%), carbohydrate (0-23%), lipid (2-10%), and cellulose (2-8%) (Kyllönen *et al.*, 1988). Primary sludge is readily dewatered (up to 50% TS), while secondary sludge is more difficult to dewater (to about 20% TS). Primary and secondary sludges are usually combined for dewatering, and about 30 to 40% TS is achieved with suitable polymer additions.

These sludges have been disposed of mainly through landfilling, incineration in pulp and paper mill power plants, earth filling, and recycling in the paper-making processes. However, the present policy aims to diminish landfilling of organic wastes (including sludges). Furthermore, because of the high water content in the sludge, its incineration is mostly not (without additional fuel i.e. bark or coal) economic.

Biological treatment of pulp and paper mill sludges could help reduce their volume, degrade potentially harmful compounds, and recover the energy and nutrients therein. Biologically treated sludge could then be used in farming, forest fertilisation, or e.g. even in manufacture of construction materials. Both composting and anaerobic digestion constitute common treatment of sludges from municipal wastewater treatment plants and, increasingly, of municipal solid waste. However, few studies have been reported on pulp and paper mill sludges (e.g. Campbell *et al.*, 1991; Puhakka *et al.*, 1992).

The objective of the present study was to evaluate the feasibility of aerobic composting and anaerobic digestion for the treatment of pulp and paper mill sludges.

MATERIALS AND METHODS

Composting

The composting studies were carried out in batch mode by using six 500 l and one 18 m^3 vessels. Both composting systems were equipped with forced aeration and a biological gas treatment unit. The compost and feed air temperatures in the larger vessel were measured continuously, and the flow rate of air in the larger vessel was adjusted to maintain the highest optimum composting temperature (assumed to be about 55-60°C) throughout the thermophilic composting period. The sludge, comprising primary (about 87% vol.) and secondary sludge (about 13% vol.), originated from a paper mill wastewater treatment plant, receiving about 60% of its flow from a de-inking and 40% from a paper-making process. The mixture of the sludges was mechanically dewatered to 29.4% TS (VS 72.7% of TS). The large amount of primary sludge apparently accounted for the high C:N-ratio (69.2) in the sludge. The sludge used in the larger vessel was stored outdoors for a week at about 25°C prior to composting, while the sludge for the composting vessels was used immediately after receipt.

Coarse pinewood bark was used in the small vessels as bulking agent (in 1:1 sludge to bark ratio), and a series of urea amendments were added (C:N-ratios: 69.2, 34.6, 29.6, 21.8, 19.9, and 18.1). In the larger vessel, coarse sprucewood bark was used as bulking agent (in 0.5:1 bark to sludge-ratio) with no amendments.

Anaerobic digestion

Anaerobic digestion was studied by using two 5 l completely stirred (magnetic stirrer) glass digester reactors (referred to as D1 and D2) with a liquid volume of 3.5-4.0 l, placed in a temperature controlled chamber at 37°C. The digesters were fed manually every workday (5 days a week), while an equal amount of the reactor content was removed before feed addition. Methane was measured using a wet test gas meter after passing the generated biogas through a 5 M sodium hydroxide solution to remove carbon dioxide.

The digesters were inoculated with 4 l (2% VS) of digested (37°C) sewage sludge from the wastewater treatment plant in Viinikanlahti, Tampere, Finland. The digester studies made use of primary (2.3-6.2% TS, 1.4-2.8% VS) and secondary (1.2-1.3% TS, 0.8-0.9% VS) sludges from a pulp and paper mill, manufacturing thermomechanical pulp and newspaper, and dewatered sewage sludge (13-16% TS, 8.5-9.5% VS) from a municipal wastewater treatment plant. D1 was fed with a mixture of primary, secondary and sewage sludge (ratio of 2:3:1, wet weight) and D2 with a mixture of secondary and sewage sludge (ratio of 3:1, wet weight). Feed TS was adjusted to about 5% with tap water. During the study, two batches of sludges were obtained from the mill and the municipality. The sludge characteristics in the different batches varied, resulting in different feed VS contents (Table 1).

Table 1. Percentage of different sludges of feed VS (total 100%) in the digester studies

Digester and experimental days	Primary sludge, %	Secondary sludge, %	Sewage sludge, %
D1, 1-49	39	14	46
D1, 50-80	19	17	64
D2, 1-49	-	23	77
D2, 50-80	-	21	79

Analyses

Chemical oxygen demand (COD, alkalinity, TS and VS were determined according to the *Standard Methods* (APHA, 1985). In composting studies, pH and electric conductivity were measured immediately after sampling with an Orion model 520 A and an Orion model 120, respectively. Ammonium and nitrate were measured with a Dionex DX 500 ionchromatograph. Filtered samples were obtained by filtering through Schleicher & Schüll 597 filters. The total concentrations of carbon (C), hydrogen (H) and nitrogen (N) were analysed according to method no: 820 L of the Finnish Geological Surveying Centre, which calls for readily dried and homogenised samples to be ignited in an oxygen stream. The concentrations were analysed with a Leco CHN-600 analyser: an IR- detector was used for the total concentration of carbon and hydrogen and a TC-detector for the total concentration of nitrogen.

In the anaerobic digestion studies, volatile acids (VA) and pH were analysed as described elsewhere (Rintala and Järvinen, 1996). The samples for soluble COD and VA were filtered using Whatman GF/A glass-fibre filters. The methane content of the biogas was analysed with a Perkin-Elmer gas-chromatograph.

Calculations

The C:N-ratio was calculated based on the results of the CHN-analysis. The loading rates, hydraulic retention times (HRTs), and methane productions in the anaerobic digester studies were calculated using weekly periods; e.g., the loading rate (gVS/ld) was calculated using the total amount of VS_{added} per week (in five days), divided by seven days.

RESULTS

Composting

The 500 l vessels were used to study the effect of the C:N -ratio on composting performance. The lag-period, which preceded the exponential temperature rise, lasted c. 36 hours, being shortest with a moderate urea addition (C:N-ratio 34.6 to 21.8) (Fig. 1), while a longer lag period was observed when no urea was added. All the composts reached the thermophilic stage within three days, except the two with the highest urea addition, which took 8 to 14 days to reach 50°C. The other composts demonstrated typical composting temperature performance with the thermophilic stage lasting an average 10 days. After the three week composting period, TS ranged between 27.3 and 32.3%, while the ash content increased from 16% to 19-20%.

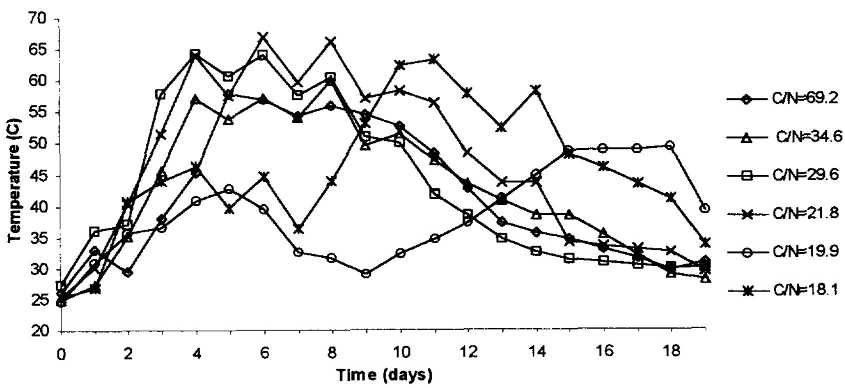

Figure 1. Temperature in the composting of pulp and paper mill sludgewith different initial C:N-ratios in the 500 l vessels.

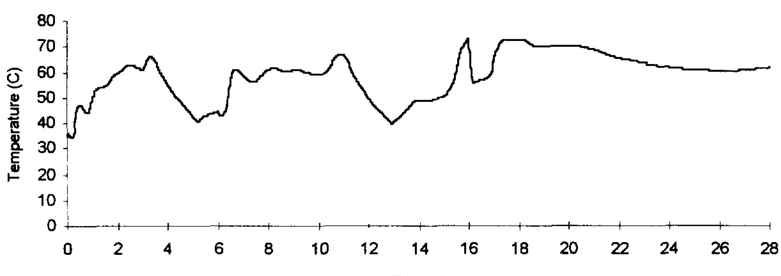

Figure 2. Temperature in the composting of pulp and paper mill sludge in the 18 m^3 vessel.

While the larger vessel was used to obtain design parameters for a full-scale composting facility, the results of the small vessels study showed the sludge compostable without urea amendment. Furthermore, in the larger vessel the large particle size of the sprucewood bark contributed to a lower bark/sludge-ratio. The initial C:N-ratio of the feed sludge mixture was 42.7, the lag-period about 12 h, and the process reached the thermophilic stage (50°C) within 24 h (Fig. 2). The temperature remained above 50°C, being on average 60°C for the rest of the 28 d study, except for two temporary coolings of the compost (Fig. 2). The highest temperature, reached on day 16, was about 73°C.

The most recognisable phenomenon during the composting was the fast dehydratation of the mass: within the 28 day period, TS increased from 31.1% to 66.0-74.0% (Table 2), while VS decreased (VS of TS) within 28 days by 8.2-9.4%. The C:N-ratio decreased from the initial 42.7 to the final 28.1. pH increased slightly, while conductivity decreased consistently with the reduction in the ammonium-nitrogen content already within seven days of the composting.

Table 2. Compost characteristics during the 28 day composting period (18 m^3 compost)

	Day				
	0	7	14	21	28
TS (%)	31	34	45	64	70
VS (% of TS)	77	76	73	72	71
pH	6.2	6.5	6.5	6.7	6.5
Conductivity (dS/m)	1.5	0.45	0.53	0.4	0.4
NH4-N (mgN/l)	895	175	136	143	85
Carbon, % of TS	37.6	nd	nd	37.6	36.6
Nitrogen, % of TS	0.88	1.1	1.2	1.2	1.3
Hydrogen, % of TS	5.4	nd	nd	4.7	4.7

nd not determined

Table 3. Performance of the anaerobic digesters treating pulp and paper mill sludges (days 63-80; number of samples 4-7; the means are shown; standard deviations were less than 5%)

	Reactor D1	Reactor D2
Loading rate[1], gTS/ld	1.5	1.7
Hydraulic retention time[1], d	c. 30	c. 30
Feed pH	7.0-7.5	7.1-7.5
Feed CODtot., mg/l	44600	58500
Feed CODsol., mg/l	3600	4700
Feed VA, mg/l	1200	1830
Feed alkalinity, mg/l	1730	2180
Feed TS, %	4.4	5.2
Feed VS, %	2.6	3.1
Digested sludge pH	7.3	7.4
Digested sludge CODtot., mg/l	30500	30200
Digested sludge CODsol., mg/l	2870	2750
Digested sludge VA, mg/l	260	270
Digested sludge alkalinity, mg/l	3480	4040
Digested sludge TS, %	3.6	3.7
Digested sludge VS, %	1.9	1.9
VS-removal, %	26.9	39.7
Methane content in the gas, %	57.5	58.7
Methane production[1], ml/gVS_{added}	185	189
Methane production[1], l/$gVS_{removed}$	688	476

[1] calculated for the whole period between days 63-80 (e.g. total methane production per total amount of VS added during the period)

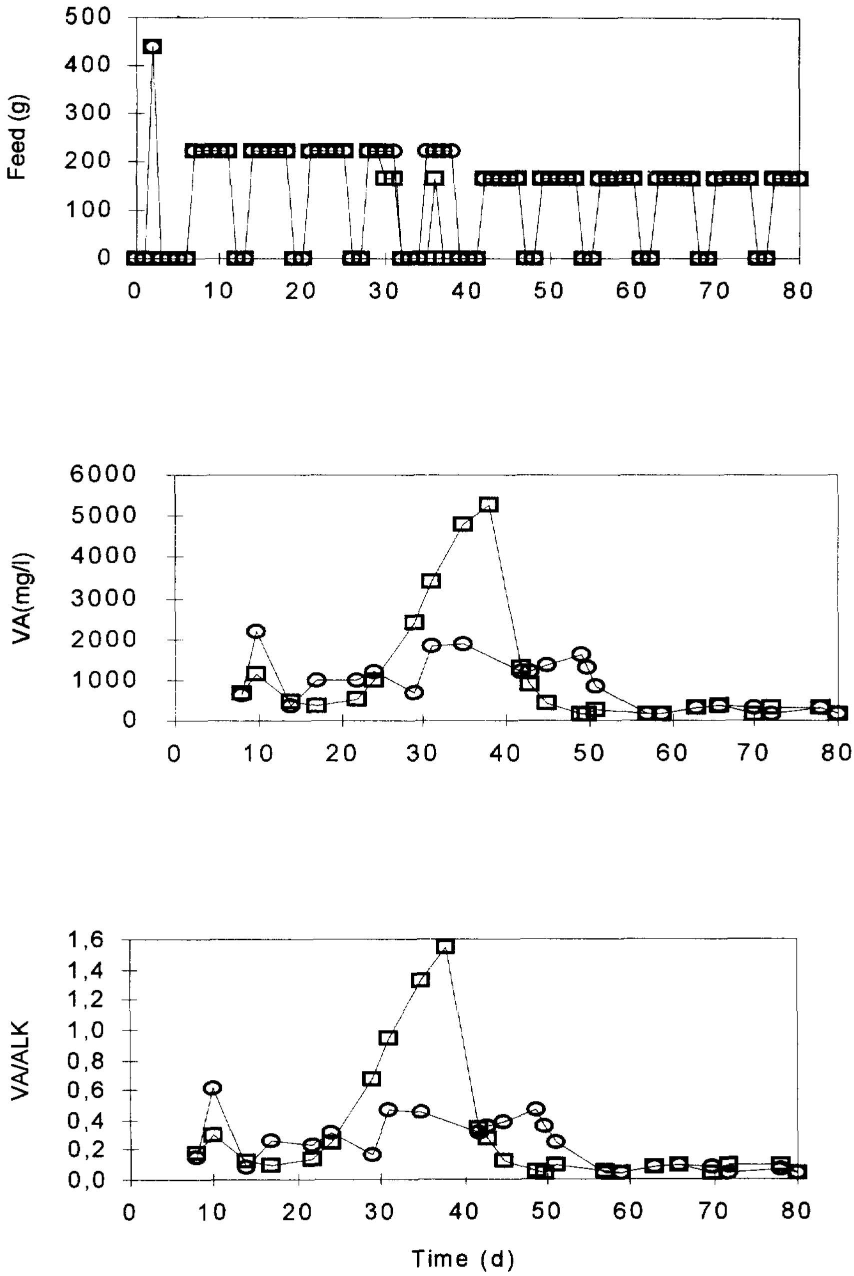

Figure 3. Performance (feed amounts, digested sludge VA and VA to alkalinity-ratio) of anaerobic digesters (❑, D1; o, D2) treating pulp and paper mill sludges during the runs.

Anaerobic digestion

Both anaerobic digesters were inoculated on day 1 and supplied with 440 g of feed on day 2. The methane content in the biogas reached c. 55% on day 7, whereupon a normal feeding procedure was started. The amount of feed in D1 and D2 was 220 g per each addition until day 29 and 42, respectively (Fig. 3).

On starting the runs the loading rate was c. 2 gTS/ld, but apparently because of overloading, the performance of both digesters, and more so of D1, deteriorated after about 20 days of operation, as indicated by the elevated VA and VA/alkalinity-ratios and decreased pH (not shown). The digesters recovered (VA and the VA/alkalinity-ratio decreased) after feeding was discontinued for a few days (D1 was unfed on days 32, 35, and 37-39, and D2 on days 32 and 39). Subsequent feeding was started at decreased loading rates (feed 165 g / addition), and for the rest of the runs the process parameters indicated undisturbed performance.

Table 3 summarises the digester performance of the end-of-the run 18 day period (days 63-80) with a constant feed pattern. The methane content and specific methane production (per g VS_{added}) were similar in both reactors, while removal of organic material (measured as VS and COD) was higher in D2 than in D1.

DISCUSSION

The results suggest that pulp and paper mill sludges are treatable both anaerobically and aerobically. The studied composts reached high temperature rapidly, indicating a prompt start of composting. A high proportion of the de-inking wastewater sludge was compostable. The results also indicate that the C:N-ratio affected the onset of composting, hence necessitating some urea addition to ensure optimal composting. Both too low and too high C:N-ratios resulted in delayed temperature increase with the low ratios further impeding transition to the thermophilic stage. Changes in pH and the ammonium-nitrogen content indicated a rapid ammonification of urea, which with the highest urea additions may have resulted in inhibitory ammonia concentrations.

In the larger compost vessel, the coarse spruce bark bulking agent made possible the lower bark/sludge-ratio. The larger vessel's shorter lag- period in temperature increase and in reaching the thermophilic stage was probably caused by the outdoor storage before composting, which apparently initiated hydrolysis of the sludge. The compost mass was thermally rather unstable due to a high and increasing volume of free air space in the mass during composting. The fast dehydratation was apparently due to an intensive degradation of organic matter, as also indicated by the decrease in VS (VS from TS) from 77.3% to 70.0-71.0% within 28 days, while TS increased from 31.3 to 66-77%. In the composting of combined (refiner mechanical pulp / chemimechanical pulp) pulp and paper mill sludge (C:N -ratio 20 to 29, TS 40 to 45%, VS 94 to 95% of TS) using a 1.1 m^3 static-pile process (without bulking agent or urea amendment) with forced-aeration (Campbell *et al.*, 1991), the maximum temperatures (60-67°C) were reached in approximately 72 h, dropping to 30-35°C after three weeks of composting. After six weeks, little change in mass or volume was observed, except for the change in the TS content from 45% to 67%, attesting to water loss.

In the anaerobic digestion study, the specific methane yields for primary, secondary, and sewage sludge were about 45, 85 and 220 $l/kgVS_{added}$, respectively, as determined in batch assays (data not shown) and estimated from digester studies. Methane production for the sewage sludge was typical of municipal sludge digesters in Finland (e.g. Rintala and Järvinen, 1996). For pulp and paper mill sludges, gas productions of 220-250 $l/kgVS_{added}$ have been reported (Boman and Bergström, 1985; Puhakka *et al.*, 1992), which with reported c. 40-45% VS removals and 60% methane content signify about 100 to 120 l $CH_4/kgVS_{added}$. The present low methane production of the primary sludge may be ascribed to the low VS to TS-ratio of the sludge used in the study (VS 45% of TS). The sludge contained high concentrations of inorganic material from paper machines, which may have limited the bioavailability of the biodegradable material for microbes. The VS removals in anaerobic digesters were lower than found for pulp and paper mill sludges. Up to 74% VS removal was obtained with kraft pulp mill primary sludge (Boman and Bergström, 1985) and 55-65% VS removal with papermill primary sludge (Gijsen *et al.*, 1988). About 40-50% VS removal has been obtained with secondary sludges and mixtures of primary and secondary sludges (Boman and Bergström, 1985; Puhakka *et al.*, 1988), and 20 to 50% with municipal sewage sludge.

In the present study, the loading potential of the anaerobic digestion process was limited apparently by the relatively short operation period and the semicontinuous feeding pattern of the digester. A 7 days a week feeding has been found to promote the anaerobic digestion process, compared to a 5 days a week loading procedure (Puhakka *et al.*, 1992).

Even though both composting and anaerobic digestion showed potential for the treatment of pulp and paper mill sludges, the high lignin and the high inorganic content of the sludges (especially in primary sludge) makes them less biodegradable by anaerobic and aerobic bacteria. In aerobic composts, fungus may degrade lignin compounds and thus enable higher VS removal than obtained in anaerobic digestion or in intensive vessel composting. Therefore vessel-composted and anaerobically digested material could be matured in static pile composts. On the other hand, anaerobic and aerobic degradability may be enhanced by pretreating the sludges e.g. thermally and chemically, which, on the other hand, may generate inhibitory compounds.

The actual feasibility of the studied processes for pulp and paper mill waste sludge management depends to a great extent on individual factors such as the generation of different wastes, existing facilities, and the end use of treated materials. In some countries the trend of centralised treatment plants for various municipal, industrial, and agricultural organic wastes means combining the various materials (sludges and wastes) for treatment. The combination should be worked out to optimise treatment performance. Some studies have shown the suitability of composted pulp and paper mill sludges for either agriculture or forest fertiliser. In case the market for compost is limited, incineration of low moisture compost could be considered.

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