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ANAEROBIC TREATMENT OF LEACHATE WITH HIGH SODIUM CONCENTRATIONS

By

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Introduction

Leachate production is one of the effects of sanitary landfill operations which must be controlled. For many landfill sites, collection, treatment and disposal, of the leachate which drains from the fill, are used to insure ground-water protection. Leachate may be treated and dispersed into the environment in a harmless manner as other wastewaters are. Treatment could be achieved by a number of different methods. Despite some investigations (1, 2, 3) the design engineer is generally faced with limited information and data on the treatability of this strong, variable wastewater.

The research presented in this paper examines anaerobic decomposition as a leachate treatment process. In addition to presenting data for operating criteria, this research also investigates the effect of sodium, the cation present in highest concentration in leachate, on the efficiency of the anaerobic leachate treatment process.

Information presented in this paper is also applicable to an understanding of anaerobic decomposition and the inhibition of methane fermentation in landfills. This information would be of value in cases where methane is collected from the fill on a commercial basis.

Leachate Composition

Leachate is liquid drainage from a solid waste disposal area and contains high concentrations of dissolved organic and inorganic materials. Leachate composition varies considerably depending upon refuse composition, infiltration, age of fill, decomposition and chemical reactions. Steiner, et. al. (4), have reported leachate composition to vary as shown in Table 1. Table 2 contains the variation in leachate composition reported in another study (5). A more comprehensive sampling and analyses of leachates from fills in several areas of the country would undoubtedly reveal a wider range of concentration levels than depicted in Tables 1 and 2.

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In addition to analyses of individual leachate samples, some laboratory and field investigations have been conducted in an attempt to determine the total amounts of specific contaminants which might be leached from a given amount of fill. Quasim and Burchinal (6) constructed test columns filled with mixed refuse and simulated normal rainfall patterns by dosing the columns with water. The results indicated that 12.33 tons (11,186 kg) of BOD, 0.87 tons (789 kg) of chloride, 4.20 tons (3,810 kg) of hardness, and 19.6 tons (17,780 kg) of solids could be extracted from 10 acre-feet of refuse fill in a 4 to 6 month period. Investigations at the University of Southern California (7) determined that 1 acre-foot of refuse fill would produce a minimum of 1.5 tons (1,360 kg) of sodium plus potassium, 1.0 tons (910 kg) of calcium plus magnesium, 0.91 tons (826 kg) of chloride, 0.23 tons (209 kg) of sulfate, and 3.7 tons (3,530 kg) of bicarbonate by a continuous leaching process.

TABLE I

VARIATION IN LEACHATE COMPOSITION (4)

Constituent

Concentration Range

Iron	200-1700
Zinc	1-135
Phosphate	5-130
Sulfate	15-500
Chloride	100-2400
Sodium	100-3800
Nitrogen	20-500
Hardness (CaCO ₃)	200-5250
COD	100-51,000
Total Residue	1000-45,000
Nickel	0.01-0.8
Copper	0.10-9.0
pH	4.00-8.5

Note: All values in mg/l except pH

Anaerobic Treatment of Leachate

Anaerobic decomposition is one of the logical leachate treatment alternatives for several reasons. First, the leachate is a concentrated wastewater with high BOD and high alkalinity generated in relatively small amounts. Second, many of the organic materials present are intermediates of the nat-

ural anaerobic decomposition process which occurs in the landfill. Finally, the gas production from such treatment can be utilized expecially if it is combined with methane collected from the landfill which appears to be a likely future practice.

TABLE 2

LEACHATE CHARACTERISTICS (5)

Constituent	Min. Value	Ave. Value	Max. Value
pH	5.60	6.55	7.63
CaCO ₃	650	3633	8120
Alkalinity (CaCO ₃)	730	4629	9520
Calcium	. 115	1047	2570
Magnesium	64	181	410
Sodium	85	940	1860
Potassium	28	959	1860
Total Iron	6	110 -	305
Ferrous Iron	2	24	93
Chloride	96	1814	2350
Sulfate	39	248	730
Inorganic Phosphate	0.2	7	29
Organic Nitrogen	. 2	163	550
Ammonia Nitrogen	0.2	437	845
BOD	81	10850	33100

Note: All values in mg/l with the exception of pH.

Boyle and Ham (1) have conducted laboratory investigations on a wide variety of treatment processes and found anaerobic treatment to be one of the more successful methods of BOD reduction. The results of their investigation indicated that BOD_5 removals greater than 90% were achieved at detention times of 10 to 12 days and organic loading rates of 0.22 to 0.27 lb. of volatile solids per cubic foot per day. The average gas production rate in this case was 7.2 cubic feet per pound of COD reduction.

Cation Toxicity and Anaerobic Treatment

While anaerobic treatment of sanitary landfill leachate is a feasible process, the designer of such a system must compensate for the major cations, metals and other materials which occasionally reach toxic or inhibitory concentrations in the leachate. The inhibitory and toxic effects of the major cations on the anaerobic treatment process, as used for municipal wastewater sludges, have been well documented (8-12).

McCarty and McKinney (11) demonstrated cation toxicity in anaerobic treatment investigating the cations likely to occur in high concentration

during the anaerobic digestion of municipal wastewater sludges. They listed the cations investigated in order of increasing toxicity as calcium, magnesium, sodium, potassium and ammonium, and noted that divalent cations were less toxic than monovalent cations. McCarty (10) noted that anaerobic bacteria could be stimulated or inhibited by the common cations depending upon the concentration of the cation. Table 3 indicates the concentration ranges which McCarty (10) felt were required for stimulation or inhibition.

TABLE 3

STIMULATORY AND INHIBITORY CONCENTRATIONS OF ALKALI AND ALKALINE-EARTH CATIONS IN THE ANAEROBIC PROCESS (10)

Cation	Stimulatory	Moderately Inhibitory	Strongly Inhibitory
Sodium	100-200	3500-5500	8000
Potassium	200-400	2500-4500	12000
Calcium	100-200	2500-4500	8000
Magnesium	75-150	100-1500	3000

Note: Concentrations in mg/l

Sodium and Other Cations in Leachates

Sodium, potassium, calcium, ammonium, magnesium, iron and zinc are the cations present in substantial concentrations in most leachates. Magnesium, iron and zinc come from metal objects and metal coating in the refuse. Ammonium originates from the anaerobic decomposition of organic materials which contain nitrogen, sodium, potassium and calcium originate in salts, "solid fill" and paper products in the refuse. Table 4 lists values of the higher concentrations of each of the cations which have been reported. The values listed in Table 3 are not absolute maximums since analysis data on leachates is not extensive.

Sodium is the cation on which this investigation focused. This was due to the high sodium levels found in some of the leachate samples which were collected. Sodium is a fairly common element and sodium hydroxide is the most common base used in production of industrial products. Paper and paper product manufacturing utilize large amounts of sodium, calcium and potassium.

Kugelman and McCarty (9) confirmed the original work by McCarty and established optimum ionic concentrations for maximum methane fermentation efficiency, which were reported to be 230 mg/l for sodium; 390 mg/l potassium, 140 mg/l ammonium-nitrogen, 200 mg/l calcium, and 122 mg/l magnesium. The inhibitory effect caused by excessive concentrations of any one cation could be minimized by the addition of an optimum concentra-

tion of at least one of the other cations. The cations were listed in order of increasing inhibition as sodium, ammonium, potassium, calcium and magnesium.

Kugelman and McCarty (9) point out that prior data was in error due to the fact that low concentrations of cations could produce synergistic or antagonistic effects which were previously overlooked.

TABLE 4

MAXIMUM REPORTED CATION CONCENTRATIONS IN LEACHATES

Cations	Concentration mg/l	Reference
	• •	
Na ⁺	7600	7
K+	1860	5
Ca++	2570	5
Mg ⁺⁺	410	5
NH₄+*	1070	5
Fe ⁺⁺ ,Fe ⁺⁺⁺	1700	4
Zn ⁺⁺	135	4

Note: Based on total ammonia nitrogen.

Inhibition of the anaerobic process, caused by the excess of one cation can be antagonized by the addition of an optimum concentration of one of the other cations. Maximum antagonism can be obtained by the addition of several antagonistic cations. The addition of one cation to an inhibitory concentration of another cation can produce a synergistic effect, increasing the toxicity. The cations which form antagonistic and synergistic pairs are shown in Table 5. As illustrated by this table, sodium and potassium are excellent antagonists and potassium is the only antagonist for a sodium inhibited anaerobic environment.

Research by Chin (8) demonstrated the mutually antagonistic effects of sodium and potassium. The anaerobic process was demonstrated to have a tolerance level of approximately 2500 mg/l of sodium at a potassium concentration level of 5 mg/l. If the sodium level was increased beyond this point, the digester efficiency would be adversely affected. Shock loadings of up to 8000 mg/l sodium could be tolerated by acclimated methane forming bacteria by increasing the potassium level to 78 mg/l. Chin concluded that sodium could be antagonized by potassium and confirmed the threshold level for sodium inhibition of methane fermentation to be 3500 mg/l.

Kugelman and McCarty (9) present some data in terms of a 50 percent inhibition index. The 50 percent inhibition index is the concentration of

inhibitory cation which causes the process reaction rate to be one half the rate without any action inhibition. In laboratory investigations Kugelman and McCarty (9) found a 50 percent inhibition index for anaerobic digestion unit containing approximately 7400 mg/l of sodium. The control in this case was a unit with a concentration of 485 mg/l of sodium. An antagonism of the 50 percent inhibition was obtained by increasing the potassium concentration to 1340 mg/l.

TABLE 5

CATION PAIRS SHOWING ANTAGONISTIC AND SYNERGISTIC EFFECTS IN AN ACETATE FED SYSTEM

Effect Toxic Cation					
Antagonistic	Na+ K+	NH₄+ Na+	K+ Na+ NH₄+ Ca++ Mg++	Ca++ Na+ K+	Mg++ Na+ K+
Synergistic	NH+ Ca++ Mg++	K+ Ca++ Mg++		NH4 ⁺ Mg ⁺⁺	NH₄+ Ca++

Experimental Methods

The anaerobic leachate treatability experiments in this investigation were performed using laboratory anaerobic treatment units set up as shown in Figure 1. Each unit contained 7.2 liters of leachate solution. The units were mixed continuously and fed daily with leachate samples collected at a solid waste disposal site in Danvers, Massachusetts. Five separate leachate samples were collected from the fill, two from a recently deposited section and three from a 3 year old fill section. All leachate samples were stored at 4°C prior to analysis.

All analyses were conducted in accordance with procedures described in *Standard Methods* (13) with the exception of COD, TC, TOC, volatile acid and metal determinations. Chemical oxygen demand was measured using a short method procedure described in *Chemical Engineering News* (14) after comparison between this method and the standard COD test (Section 220, *Standard Methods*) on 15 leachate samples yielded values with less that 1% variation between methods. Total carbon and total organic carbon were measured on filtered samples using a Dohrman Envirotech Model DC-50 Total Organic Carbon Analyzer. Volatile acids concentrations were determined by the direct titration method outlined by Dilallo and Albertson

(15). The analysis for aluminum, cadmium, calcium, chromium, copper, iron lead, magnesium, potassium, sodium, and zinc were conducted using a Perkin-Elmer Model 303-Atomic Absorption Spectrophotometer. Ammonia analyses were conducted using an Orion membrane ammonia probe with an expanded scale pH meter. The carbon dioxide, methane and hydrogen sulfide gas analyses were performed using a Norelco, WG PYE Series 104 gas chromatograph.

FIGURE I EXPERIMENTAL APPARATUS



Experimental Results and Discussion

A fairly complete analysis of each leachate sample was conducted as soon as possible after sample collection. The results of the leachate analyses are presented in Table 6 and Table 7. Samples 1 and 2 were samples of a leachate produced from recently placed refuse which had been in the fill for a year or less. Samples 3 thru 5 were produced from a section of the fill completed 3 years before sampling. The first samples (1 and 2) contained concentrations of sodium which were high enough to prevent the methane fermentation phase essential to successful anaerobic digestion. The concentrations of sodium were higher than anticipated based on previous leachate analysis reports (4). Higher concentrations of volatile acids were observed in the first two samples. This was apparently due to the fact that the solid waste at the first sampling location was undergoing more active decomposition. It should be noted that values for total organic carbon reported in Table 6 are for soluble TOC since the leachate samples were filtered before testing.

TABLE 6

ANALYSIS OF LEACHATE SAMPLES

Sample Number

Constituent	1	2	3	4	5
Date Sampled	6/27/73	7/19/73	8/21/73	9/18/73	10/12/73
Total Solids	11411	12051	13629	10876	9500
Total Vol. Solids	5404	4761	1983	2136	2276
Total Fix. Solids	6007	7290	11380	8740	7224
Total Di. Solids	9872	8677	7924	4107	4750
Vol. Dis. Solids	5007	4199	2514	1196	1708
Fix. Dis. Solids	4865	4478	5410	2911	2942
Suspended Solids	1539	3374	5705	6769	4750
Vol. Sus. Solids	946	1293	1173	2088	1571
Fix. Sus. Solids	593	2081	4532	4681	3179
COD	16800	14900	7743	8100	6220
BOD ₅	8710	7810	4100	3870	3860
Soluble BOD ₅	6542	5400	2760	2150	2510
Total Organic					
Carbon	1560	4840	1260		
рH	5.60	5.65	6.00	6.05	6.00
Acidity (CaCO ₃)	1705	1960	1670		
Total Alkalinity					
(CaCO ₃)	3840	5470	3340	2700	3470
Bicarbonate					
Alkalinity	940	1150	1250	800	1340
Volatile Acids	4350	6470	3030	2970	3200

Note: All values with the exception of pH are in units of mg/l.

The first phase of the experiment was the acclimation and start up of the laboratory anaerobic units. All units were acclimated at a feeding rate which allowed for a 20 day hydraulic retention time. Units 1 and 2 were started using the first leachate sample before analyses of the metal cation concentrations were completed. Consequently, the start-up was inhibited by the sodium concentrations of the leachate sample. Figures 2 and 3 depict the operation of the first units. Unit 1 was started using leachate from Sample 1 only with a sodium concentration of 7600 mg/l. In addition to the sodium concentration, a low pH hindered the acclimation of this unit. Little methane fermentation occurred in this unit until the concentration of sodium began to decrease after 17 days when leachate sample 2 was first used as feed. Unit 2 was initially charged with a mixture of one part leachate.

TABLE 7

ANALYSES OF METAL IONS IN LEACHATE SAMPLES

Sample Number

			•		
Constituent	1	2	3	4	5
Date sampled	6/27/73	7/19/73	8/21/73	9/18/73	10/12/73
Total Iron (Fe)	629	474	109	170	
Calcium (Ca)	1420	987	330	290	
Magnesium (Mg)	140	110	92	71	
Cadmium (Cd)	*0.1	*0.1	*0.1		
Nickel (Ni)	0.33	1.0	0.30	0.22	
Chromium (Cr)	3.32	3.75	*0.1	*0.1	
Copper (Cu)	*0.1	*0.1	*0.1		<u> </u>
Total Carbon (C)	4140	5684	3700		
Zinc (Zn)	13.0	12.5	0.75	1.3	
Lead (Pb)	*0.1	*0.1	*0.1		
Ammonia (NH ₃)	153	102	360	72.0	35.0
Potassium(K)	275	225	149	127	169
Sodium (Na ⁺)	7600	6800	4200	3840	3570
Aluminum (Al-3)	0.5	0.75	0.62		

Note: (1)All values in units of mg/l

(2) *means the actual value is less than the stated value.

ate from Sample 1 and one part digested sludge from the Nut Island Wastewater Treatment Plant in Boston. Table 8 contains the results of a partial analysis of the digested sludge from the Nut Island Wastewater Treatment Plant. The concentration of sodium in Unit 2 began at approximately 3800 mg/l and increased as more leachate was fed to the unit. At this concentration range, the unit began to function as shown on Figure 3.

Figures 4 through 7 present the operational parameters during the acclimation and operation of units 3 through 6. Units 4, 5 and 6 were started and acclimated using leachate sample number 2. For unit 4, daily feeding was initiated immediately while units 5 and 6 were operated as batch units for three days and eight days respectively before beginning the daily feeding. Unit 3 was initially charged with a second sample of digested sludge from the Nut Island Treatment facility and leachate from sample 2 was gradually introduced by daily feeding. In addition to the monitoring parameters shown in Figures 2 through 7 for the anaerobic units gas composition and BOD₅, COD and Volatile Solids reduction data were also collected during the acclimation period.



FIGURE 2:. OPERATIONAL PARAMETERS; UNIT 1





TIME OF OPERATION DAYS

TABLE 8

CHARACTERISTICS OF NUT ISLAND ANAEROBIC SLUDGE

Sample No.	1	2
Date Sampled	7/13/73	7/25/73
Total Solids	25,420	26,530
Volatile Solids	14,820	14,600
Volatile Acids	520	495
Total Alkalinity	1750	950
Bicarbonate Alkalinity	1410	620
рН	7.15	7.0

Note: All values in mg/l except pH

After the acclimation period was completed, the experimental units were paired into three groups. One pair was adjusted to a 10 day hydraulic retention period. A second pair was adjusted to a 15 day hydraulic retention period. The third pair was allowed to remain on a 20 day hydraulic retention period. Once the units had adjusted to the change in hydraulic retention period the normal operational parameters were determined for each hydraulic retention period. During this adjustment period the leachate feed was changed to samples collected from the second location. This reduced the concentration of sodium in the units to the 4000 mg/l range. The normal operational parameters for anaerobic leachate treatment observed in this investigation for the 10, 15 and 20 day hydraulic retention times are shown on Figures 2 through 7. Some measures of degree of treatment and relative gas production are indicated on Tables 9 and 10.

TABLE 9

PERCENT REDUCTION FOR 10, 15 AND 20 DAY HYDRAULIC DETENTION PERIODS

Hydraulic Detention Period and Percent Reduction

	10 Days	15 Days	20 Days	Overall
COD	90.5%	91.1%	94.3%	· 92.0%
BOD ₅	91.8%	92.6%	94.8%	92.9%
Volatile Solids	40.2%	40.9%	40.6%	40.6%

TABLE 10

AMOUNT OF GAS PRODUCED PER POUND OF COD, BOD₅ OR VOLATILE SOLIDS DESTROYED

Hydraulic Detention Amount of Gas Per Pound Destroyed Period Volatile Solids COD BOD₅ 9.35 53.35 6.13 10 Days 7.14 13.22 56.34 15 Days 46.09 5.68 11.97 20 Days 51.92 Overall 6.36 11.27

FIGURE 4: OPERATIONAL PARAMETERS; UNIT 3



TIME OF OPERATION DAYS



FIGURE 5: OPERATIONAL PARAMETERS; UNIT 4





TIME OF OPERATION DAYS



FIGURE 7: OPERATIONAL PARAMETERS: UNIT 6

Table 9 indicates the percent reduction in COD, BOD_5 and volatile solids for the various hydraulic retention times. The percentage reduction of BOD_5 and COD was greater than 90% for all the units during the normal operating period. Units with greater hydraulic retention times exhibited only slightly higher percentages of BOD and COD reduction. Volatile solids reduction was around 40 percent, a figure which did not increase noticeably with increased hydraulic detention time. During the normal operating period the leachate feed changed from sample 2 to sample 3. The latter sample had approximately half of the organic concentration of the original which resulted in a lower gas production rate. However, none of the relative operating parameters exhibited a significant change.

Table 10 relates gas production to organic matter destruction. As indicated in the table, the units which were operated on a 15 day hydraulic retention time produced the greatest amount of gas per pound of COD, BOD_5 or volatile solids destroyed. For the units with a 15 day hydraulic retention time, 7.14 cubic feet of gas were produced per pound of COD destroyed. This is comparable to the value of 7.2 cubic feet per pound of COD destroyed for a unit with a 12.5 day hydraulic detention period reported in another study by Ham and Boyle (1). The leachate treatment units yielded 51.9 cubic feet of gas per pound of volatile solids destroyed. This is considerably more gas production per pound of volatile solids destruction than normally obtained from the digestion of municipal wastewater sludge. The difference in values could be due to differences in the nature of the volatile solids measured in both solutions and inherent error in the measurements. Gas composition measurements during this normal operating period indicated that the gas from the digestion units contained between 65 and 75 percent methane.

The last phase of this investigation was a series of experiments designed to document the effect of high sodium concentrations in the leachate on the anaerobic digestion process. In the first toxicity experiment, the sodium concentration in three units was increased to determine the concentration range in which methane fermentation of leachate could be severely inhibited or stopped. Toxic inhibition of this type is a function of the concentration, the change in concentration, and the rate of change of concentration. In this case the concentration of sodium in the units was increased from about 4100 mg/l by increments of 400 mg/l per day to 7600 mg/l. This rate of increase was a relatively sudden toxic slug for which the methane forming bacteria could not adapt. Units 1, 4 and 6 were the units in which the sodium concentration was increased while the other units in each pair remained as controls. Unit 1 had a hydraulic detention period of 20 days while Units 4 and 6 were operated at hydraulic detention periods of 15 and 10 days respectively. In this case all units began to fail on the seventh day of the toxicity experiment when the sodium concentration reached 6800 mg/l. Figure 8 indicates how the COD removal efficiency, based on the control unit, diminished with increasing sodium concentration.

A second toxicity experiment was conducted in order to determine the effects of a more gradual increase in sodium concentration. This experiment was begun after Unit 6 had recovered from the first toxicity experiment. Units 3 and 5 were used as control units while units 2 and 6 were used as test units. The hydraulic retention times for Units 2 and 6 were 15 and 10 days respectively. The initial concentration of sodium in the test units was first raised to 6500 mg/l. Next the sodium concentration was increased in increments of 500 mg/l every four days thereafter until the experiment was terminated 32 days later. Gas composition analyses indicated that methane fermentation was occurring at sodium concentrations of 9000 mg/l and 9500 mg/l. Gas production decreases were evident after the sodium concentrations reached 9000 mg/l. Figure 9 indicates how the COD removal efficiency diminished with increasing sodium concentration.

The difference in toxicity of rapid versus slow increase in sodium concentration is evident when Figures 8 and 9 are compared. Rapidly increasing sodium concentrations caused failure of the experimental units at lower sodium levels than when the sodium concentrations were increased slowly. The decline of the methane fermentation at sodium concentrations of 7200 mg/l was expected, based on results by Chin (8). In the second experiment the continued performance of methane fermentation at levels up to 9000 mg/l and 9500 mg/l of sodium was better than expected.

FIGURE 8 PERCENT EFFICIENCY AT VARIOUS SODIUM CATION CONCENTRATIONS; FIRST TOXICITY EXPERIMENT



FIGURE 9 PERCENT EFFICIENCY AT VARIOUS SODIUM CATION CONCENTRATIONS, SECOND TOXICITY EXPERIMENT



Often the presence of an antagonistic substance will allow operation at higher concentrations. Potassium is the only commonly occurring cation which is antagonistic to sodium toxicity in the methane fermentation process. During the toxicity experiments the potassium concentration ranged from 127 mg/l to 169 mg/l. According to the potassium concentration requirements determined by Chin (8) for sodium toxicity antagonism in methane fermentation, 169 mg/l of potassium would have only a minimal antagonistic effect when sodium concentrations were 6000 mg/l and greater.

Summary and Conclusions

Anaerobic decomposition appears to be an effective method of treatment for reducing the organic materials found in leachates generated by landfills. Anaerobic digestion units can be started without too much difficulty as the leachate naturally contains anaerobic organisms. The methane fermentation process commonly employed for the digestion of municipal wastewater sludges would result in better than 90 percent removal of BOD when applied to leachate. In this experiment COD reductions were equally high. The best performance was exhibited when anaerobic experimental units were operated at a 15 day hydraulic detention period.

A disadvantage of anaerobic digestion or any other biological treatment process applied to leachate treatment is the possible presence of toxic substances. In general, the anaerobic treatment process will resist changes in leachate composition but there are some substances, principally heavy metals and other cations, which can inhibit or kill the essential methane fermentation stage. In this investigation, the problem of high sodium ion concentrations was investigated. It was determined that an anaerobic leachate treatment unit could operate at increased sodium concentration levels greater than 6000 mg/l. If the sodium ion concentrations increased beyond the 6000 mg/l level the methane fermentation process efficiency would decrease. Relative decreases in efficiency were obtained for rapid and gradual sodium concentration increases. The amount of efficiency decrease in the process, due to sodium toxicity, was found to be a function of the increased sodium level and its rate of increase.

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