Distributed Model of Solid Waste Anaerobic Digestion

Effects of Leachate Recirculation and pH Adjustment

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Abstract: A distributed model of solid waste digestion in a 1-D bioreactor with leachate recirculation and pH adjustment was developed to analyze the balance between the rates of polymer hydrolysis/acidogenesis and methanogenesis during the anaerobic digestion of municipal solid waste (MSW). The model was calibrated on previously published experimental data generated in 2-L reactors filled with shredded refuse and operated with leachate recirculation and neutralization. Based on model simulations, both waste degradation and methane production were stimulated when inhibition was prevented rapidly from the start, throughout the reactor volume, by leachate recirculation and neutralization. An optimal strategy to reduce the time needed for solid waste digestion is discussed. © 2002 Wiley Periodicals, Inc. Biotechnol Bioeng 81: 66-73, 2003.

Keywords: solid waste; distributed model; hydrolysis; methanogenesis; leachate recirculation; pH adjustment

INTRODUCTION

In the anaerobic digestion of the organic fraction of municipal solid waste (OFMSW), under imperfect mixing conditions, methanogenic bacteria require sites where they will be protected from rapid acidogenesis. Volatile fatty acids (VFAs), which are transferred from the acidogenic to the methanogenic areas, serve as the precursor for methane production. High VFA concentrations inhibit both methanogenesis and hydrolysis/acidogenesis. Thus, mass transfer processes and adequate microbial seeding are important for anaerobic digestion of OFMSW (Martin, 2001).

Mixing as well as a reduction in the initial solid waste concentration may be a solution to avoid inhibition by high VFA levels, and several full-scale continuous-flow mixed

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bioreactors have been operated for over a decade. However, the cost of these systems is relatively high (Westegard and Teir, 1999). In "wet" complete mixed systems, the organic solid waste is diluted with water to less than 15% total solids (TS), while in "dry" systems, the waste mass within the reactor is kept at a solids content in the range of 20–40% TS. Because batch digesters are technically simple, the capital cost is significantly lower than for continuously fed digesters, though some technical problems still exist (ten Brummeler, 2000).

A landfill bioreactor approach has been developed to optimize landfills as biological treatment systems (Reinhart and Townsend, 1998). One of the most critical parameters affecting OFMSW biodegradation is the moisture content, which can be controlled via leachate recirculation. The idea of enhancing refuse decomposition by the addition of supplemental water and/or recirculating leachate was first proposed over twenty-five years ago (Pohland, 1975). Leachate recirculation is stimulatory because liquid movement distributes the inoculum, minimizes local shortages of nutrients, and dilutes potential toxins (Novella et al., 1997). However, in the absence of active acetogenic and methanogenic populations, recirculated leachate may cause an accumulation of VFAs. A combination of leachate recirculation and pH adjustment can minimize the inhibitory effects of acid accumulation and accelerate the rate of waste degradation (Barlaz et al., 1990; Komilis et al., 1999). In addition to moisture content, pH, waste particle size and density, and temperature may impact refuse biodegradation. A discussion of the effects of multiple parameters on waste decomposition has been presented (Barlaz et al., 1990).

To assist in comparison of landfill design alternatives on the basis of water balance and hydrological performance, the Hydrological Evaluation of Landfill Performance (HELP) model was developed (Schroeder et al., 1994). The HELP model is a quasi-two-dimensional, deterministic, wa-

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ter-routing model that accepts meteorological, soil, and design data, including various combinations of vegetation, cover soils, lateral drainage layers, low permeability barrier soils, etc. However, although the HELP model describes water content and moisture movement into the landfill body, it does not simulate the biochemical/microbiological processes occurring in the landfill. There have been many attempts to apply simple first-order or zero-order equations to describe the rate of solids degradation and methane production in landfills but the fit is typically poor (McBean et al., 1995). Structural modeling of anaerobic digestion, an approach that takes into account different stages of this complex process, is more promising (Batstone et al., 2002). Polymer hydrolysis, acidogenesis, acetogenesis, and methanogenesis are traditionally included in structured models as separate steps. Vavilin et al. (1999) developed a structured model of anaerobic digestion to simulate a batch laboratory reactor treating MSW, but a well-mixed system was assumed. The heterogeneous nature of the initial waste and biomass distributions were taken into account in a "twoparticle" model (Kalyuzhnyi et al., 2000; Veeken and Hamelers, 2000). However, this model as well as many other models (Haarstrick et al., 2001; Mata-Alvarez, 2000) developed earlier and based on ordinary differential equations could not describe non-synchronized processes of waste conversion into biogas in the total digester volume.

A distributed model of solid waste digestion in a 1-D bioreactor with leachate recirculation and pH adjustment that takes into account the initial waste and biomass distributions as well as the biochemical/microbiological processes, and based on partial differential equations, was developed and is described here. This distributed model was used to analyze the balance between the rates of polymer hydrolysis/acidogenesis and methanogenesis during the anaerobic digestion of MSW in batch laboratory reactors. The model was calibrated based on published experimental data (Barlaz et al., 1989) and builds on a previously developed model, where the processes of VFA diffusion and advection were considered (Vavilin et al., 2002a).

METHODS

Experimental Setup

Refuse decomposition was monitored in laboratory-scale simulated landfills. Thirty-seven, 2-liter (20 cm height) containers were filled with about 700 g of shredded refuse at the beginning of the experiment. The cellulose plus hemicellulose fraction of refuse accounts for ~63% of refuse weight. Leachate recirculation and neutralization were used in all containers. Deionized water was added to the refuse to adjust its moisture content to ~73% (wt/wt) and to ensure the availability of ample free liquid for leachate recirculation. The pH of the fresh refuse was 7.5. All leachate collected was neutralized and recycled through the top of the container on a daily basis. A container was dismantled approximately weekly for microbiological and chemical analysis.



Figure 1. Simplified kinetic scheme of anaerobic digestion of solid waste.

Complete microbial and chemical analyses required that the entire contents of a container be used. Thus, once sampled, a container could no longer be monitored. The most probable number (MPN) method was used to monitor bacterial population development. It should be noted that the standard deviation of the MPN data might be up to 1 order of magnitude. A detailed description of the experimental methods was published earlier (Barlaz et al., 1989).

Model

A simplified kinetic scheme (Fig. 1) was used in the distributed 1-D batch reactor model with waste initially loaded and leachate recirculated continuously. Polymer hydrolysis/ acidogenesis and acetogenesis/methanogenesis were included in the model as the two possible rate-limiting steps of the overall anaerobic digestion process. Inhibition of both of these steps by an intermediate product (VFAs) was taken into account. For simplicity, all transformation processes involved in the conversion of VFAs to methane (acetogenesis and both hydrogenotrophic and aceticlastic methanogenesis) were lumped together as a single step in the model. The mean of the acetate and H₂/CO₂-utilizing MPN data was used as the methanogenic biomass data in the model. A cell mass of 8.3×10^{-11} g/cell was used to convert cell number to cell mass as required by the model [Eq. (1)].

The following system of five parabolic partial differential equations in which Z is the vertical coordinate of the 1-D reactor of height L = 20 cm ($0 \le Z \le L$) was considered:

$$\begin{cases} \frac{\partial W}{\partial T} = -k \ W f(S), \\ \frac{\partial S}{\partial T} = D_{\rm S} \ \frac{\partial^2 S}{\partial Z^2} - q \ \frac{\partial S}{\partial Z} + \chi \ k \ W f(S) - \rho_{\rm m} g(S) \ \frac{SB}{K_{\rm S} + S} \\ \frac{\partial B}{\partial T} = D_{\rm B} \ \frac{\partial^2 B}{\partial Z^2} - q \ \alpha \ \frac{\partial B}{\partial Z} + Y \ \rho_{\rm m} g(S) \ \frac{SB}{K_{\rm S} + S} - k_{\rm d} B, \end{cases}$$
(1)
$$\frac{\partial P}{\partial T} = A(1 - Y)\rho_{\rm m} g(S) \ \frac{SB}{K_{\rm S} + S} \\ \frac{\partial N}{\partial T} = D_{\rm N} \ \frac{\partial^2 N}{\partial Z^2} - q \ \frac{\partial N}{\partial Z}. \end{cases}$$

$$W(Z,0) = \sigma(Z); S(Z,0) = \varphi(Z); B(Z,0) = \psi(Z); N(Z,0) = \xi(Z)$$
(2)

Boundary conditions

$$\frac{\partial S(0,T)}{\partial Z} = \frac{q}{D_{\rm S}} \left(S(0,T) - S(L,T) \right); \quad \frac{\partial S(L,T)}{\partial Z} = 0 \tag{3}$$

$$\frac{\partial B(0,T)}{\partial Z} = \frac{q}{D_{\rm B}} \alpha (B(0,T) - B(L,T)); \quad \frac{\partial B(L,T)}{\partial Z} = 0$$
(4)

$$N(0,T) = \frac{23}{60} \frac{S(L,T)}{1 + [H^+]/K_a}; \quad \frac{\partial N(L,T)}{\partial Z} = 0$$
(5)

where $W \equiv W(Z,T) \ge 0$; $S \equiv S(Z,T) \ge 0$; $B \equiv B(Z,T) \ge 0$; $N \equiv N(Z,T) \ge 0$ are the solid waste, total VFA, methanogenic biomass, and sodium concentrations, respectively; dP/ $dP \equiv dP/dt(Z,T) \ge 0$ is the methane production rate; $0 \le T$ $< +\infty$ is time; k is the first-order hydrolysis rate constant; $\rho_{\rm m}$ is the maximum specific rate of VFA utilization; k_{d} is the specific biomass decay coefficient; χ is a stoichiometric coefficient; A = 16/60 is the mass fraction of methane in biogas; $K_{\rm s}$ is the half-saturation constant for VFA utilization; Y is the biomass yield coefficient; $D_{\rm S}$, $D_{\rm B}$, and $D_{\rm N}$ are the diffusion coefficients for VFA, biomass, and sodium, respectively; q is the volumetric liquid flow rate per unit surface area (specific liquid flow rate); α is the fraction of biomass transferred by liquid flow; [H⁺] is the proton ion concentration; and K_a is the VFA (acetic acid) dissociation constant.

In Eq. (5) it is assumed that the VFA concentration is the sum of non-ionized and ionized VFA concentration $S = [VFAH] + [VFA^-]$. For the dissociation constant we have

$$K_{\rm a} = \frac{[\rm VFA^{-}][\rm H^{+}]}{[\rm VFAH]} \approx 2 \times 10^{-5},$$

and from the ionic balance we have $[OH^-] + [VFA^-] \equiv [Na^+] + [H^+]$ or for circumneutral conditions, $[VFA^-] \approx [Na^+] \equiv N$. Taking into account the molecular weights of sodium and VFA (acetic acid), the boundary condition, Eq. (5), for N(0,T) was obtained. A high sodium concentration may inhibit anaerobic digestion (Parkin and Owen, 1986). In this model, the sodium concentration was used for simulation purposes only to describe the pH influence on a process rate indirectly.

Darcy's law, as applied to one-dimensional (1-D) downward flow, can be used to describe leachate movement in the unsaturated landfill (Mc Bean et al., 1995):

$$q = K(\theta) - D(\theta) \frac{\partial \theta}{\partial Z}$$
(6)

where q is the volumetric liquid flow rate per unit surface area, K is the hydraulic conductivity in the porous medium, D is the liquid dispersion coefficient, θ is the moisture content, and Z is the vertical coordinate. In the case with large pore and particle sizes,

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$$\left| D(\theta) \frac{\partial \theta}{\partial Z} \right| << K(\theta) \tag{7}$$

In the system of partial differential equations [Eq. (1)], a constant value of volumetric liquid flow rate per unit surface area q is assumed. Using condition (7), one obtains from Eq. (6)

$$q \approx K(\theta) \tag{8}$$

Thus, for a fixed θ value throughout the Z coordinate, the specific liquid flow rate q is constant. It should be emphasized that q is not the only variable dependent on θ (and vice versa). The distribution of hydraulic conductivities within an actual landfill is dependent upon the character of the waste, waste disposal operations, and the age of the landfill (McCreanor and Reinhart, 2000). During leachate recirculation a channeled flow is a major leachate movement mechanism.

The dimensionless functions f(S) and g(S) describe the non-ionized VFA inhibition of hydrolysis/acidogenesis and methanogenesis, respectively. These functions can be written in the following explicit form:

$$f(S) = \frac{1}{1 + \left(\frac{I}{K_{\rm f}}\right)^{m_{\rm f}}}; \quad g(S) = \frac{1}{1 + \left(\frac{I}{K_{\rm g}}\right)^{m_{\rm g}}}, \tag{9}$$

where I = S - (60/23)N is the inhibiting concentration of non-ionized VFA in g/l units, $K_f > 0$, $K_g > 0$ are the inhibition constants, $m_f \ge 1$ and $m_g \ge 1$ are the corresponding inhibition degree indexes. Using the functions from Eq. (9), it is possible to describe sharp or smooth inhibition of hydrolysis/acidogenesis and methanogenesis by non-ionized VFA.

The initial waste and biomass distributions along the reactor height Z are simulated using the following functions:

$$\sigma(Z) = \gamma_1 \left(1 - \exp\left[-0.5 \left(\frac{Z - a_1}{\gamma_{21}} \right)^2 \right] \right) \\ \times \ldots \times \left(1 - \exp\left[-0.5 \left(\frac{Z - a_n}{\gamma_{2n}} \right)^2 \right] \right), \quad (10)$$

$$\psi(Z) = \gamma_3 \left(1 + \gamma_{41} \exp\left[-0.5 \left(\frac{Z - b_1}{\gamma_{51}} \right)^2 \right] \right)$$
$$\times \ldots \times \left(1 + \gamma_{4m} \exp\left[-0.5 \left(\frac{Z - b_m}{\gamma_{5m}} \right)^2 \right] \right), \quad (11)$$

where $\gamma_1, \gamma_{21}, \gamma_{22}, \ldots, \gamma_{2n}, \gamma_3, \gamma_{41}, \ldots, \gamma_{4m}, \gamma_{51}, \ldots, \gamma_{5m}$ are the distribution coefficients; *n* and *m* are the total number of depression/peak zones of waste and biomass, respectively. Eqs. (10) and (11) describe the case with a multidepression ($\sigma(Z)$) and multi-peak ($\psi(Z)$) distributions along the reactor height *Z* for waste (VFA) and biomass, which have the minimum and maximum at $Z = a_i$ and $Z = b_j$, correspondingly. A function that describes the distribution of VFA coincides with $\sigma(Z)$. For sodium concentrations, it was assumed that $\eta(Z) = 0$. The initial methane production rate was assumed to be zero. Numerical simulation based on the model described above was performed using vectororiented software (MATLAB, version 6.0).

After visual model calibration using previously published experimental data (Barlaz et al., 1989), the following parameter values were obtained: $k = 0.011 \text{ d}^{-1}$, $\chi = 0.48$, $\rho_{\rm m} = 0.32 \text{ d}^{-1}$, $k_{\rm d} = 0.001 \text{ d}^{-1}$, $K_{\rm S} = 1.2 \text{ g} \text{ l}^{-1}$, Y = 0.12, $m_{\rm f} = m_{\rm g} = 3$, $K_{\rm f} = 16.0 \text{ g} \text{ l}^{-1}$, $K_{\rm g} = 10.0 \text{ g} \text{ l}^{-1}$, $D_{\rm S} = 1.0 \times 10^{-3} \text{ L}^2 \text{ d}^{-1} \approx 4.7 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, $D_{\rm B} = 1.0 \times 10^{-4} \text{ L}^2$ $\text{d}^{-1} \approx 4.7 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$, $D_{\rm N} = 1.0 \times 10^{-3} \text{ L}^2 \text{ d}^{-1} \approx 4.7 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, $q = 1 \text{ L} \text{ d}^{-1}$, and $\alpha = 0.1$. It should be noted that the set of coefficients obtained as a result of model calibration is not unique. According to the parameter values calibrated, the initial waste, VFA, and biomass concentrations were localized in separate zones: maximum biomass and minimum waste and VFA concentrations placed at the same points (n = m = 6 was used in Eqs. (10) and (11)):

OFMSW distribution

 $\gamma_1 = 259$ g l⁻¹, $\gamma_{21} = \gamma_{22} = \ldots = \gamma_{26} = 0.01$ L, $a_1 = 0.15$ L, $a_2 = 0.3$ L, $a_3 = 0.45$ L, $a_4 = 0.6$ L, $a_5 = 0.75$ L, $a_6 = 0.9$ L

VFA distribution

 $\gamma 1 = 0.05$ g l⁻¹, $\gamma_{21} = \gamma_{22} = \ldots = \gamma_{26} = 0.01$ L, $a_1 = 0.15$ L, $a_2 = 0.3$ L, $a_3 = 0.45$ L, $a_4 = 0.6$ L, $a_5 = 0.75$ L, $a_6 = 0.9$ L

Biomass distribution

 $\begin{aligned} &\gamma 3 = 0.15 \text{ g} \text{ I}^{-1}, \gamma_{41} = \gamma_{42} = \ldots = \gamma_{46} = 10, \gamma_{51} = \gamma_{52} \\ &= \ldots = \gamma_{56} = 0.01 \text{ L}, b_1 = 0.15 \text{ L}, b_2 = 0.3 \text{ L}, b_3 = 0.45 \\ &\text{L}, b_4 = 0.6 \text{ L}, b_5 = 0.75 \text{ L}, b_6 = 0.9 \text{ L} \end{aligned}$

Assuming perfect mixing conditions along the *X* and *Y* axes, volume units for all concentration variables were used despite the 1-D character of the model.

RESULTS AND DISCUSSION

The model dynamics are shown in Figure 2. It can be concluded that the model describes the experimental data com-



Figure 2. Time profiles of waste (cellulose plus hemicellulose), VFA, and biomass averaged over the total reactor volume and methane production rate. Symbols refer to the experimental data; lines refer to the model predictions with pH adjusted to 7.0. Solid waste remaining is the ratio of the current weight of cellulose plus hemicellulose divided by the initial weight of cellulose plus hemicellulose. The VFA mass was divided by the initial solids mass.

paratively well except for the biomass data, apparently because of high experimental errors inherent to such measurements (Barlaz et al., 1989).

Methanogenesis and Hydrolysis as Rate-Limiting Steps

Figure 3 shows the concentration profiles along the coordinate Z at different times. Initially, the acidogenic and methanogenic areas were separated in space. During the first 55 days, the VFA concentration increased because of the relatively low initial concentration of methanogenic biomass. Evidently, during the initial time interval methanogenesis was the rate-limiting step. Additional simulations (results not shown) revealed that because of diffusion and advection, VFA and biomass concentration profiles became close to uniform around the 4th day of incubation. VFA concentrations above 16 g/L accumulated by the 20th day and inhibited not only methanogenesis but also polymer hydrolysis and acidogenesis. It should be noted that some authors (e.g., Veeken et al., 2000) consider that the hydrolysis rate constant is pH dependent. However, inhibition of hydrolysis at pH values around 7.0 during batch slaughterhouse solid waste degradation has been reported (Salminen et al., 2000; Vavilin et al., 2000). Nevertheless, Wang et al. (1997)



Figure 3. Sequence of distributions of waste, VFA, biomass, and sodium concentrations, and methane production rate throughout the coordinate *Z* at 0 (0), 24 (1), 48 (2), 72 (3), 96 (4), and 120 (5) days of incubation (standard set of model parameter values). VFA and sodium concentrations are about zero at 0 (0), 96 (4) and 120 (5) days. Waste means cellulose plus hemicellulose.

showed that the refuse ecosystem may tolerate a relatively high concentration of non-ionized carboxylic acids.

According to experimental data by Barlaz et al. (1989), an initial drop in pH from 7.5 to 5.7 corresponded to VFA accumulation. The pH level increased significantly when methanogenesis began. In Figures 2 and 3 it can be seen that active methanogenesis occurred between 55 and 95 days of incubation. The rate of waste degradation was approximately the same throughout the reactor depth due to the uniform VFA and sodium concentrations in the reactor during leachate recirculation and neutralization. After the 95th day, VFAs were depleted throughout the reactor depth. It can be concluded that after this time, methane production was controlled by the hydrolysis rate, i.e., hydrolysis became the rate-limiting step.

Effect of Liquid Flow Rate

The higher the specific liquid flow rate q, the more rapidly methanogenesis takes place (Fig. 4). Methane production did not occur without leachate recirculation. At a higher specific liquid flow rate q, a uniform sodium concentration is attained more rapidly, preventing inhibition of methanogenesis and hydrolysis/acidogenesis in any part of the reactor. Thus, waste degradation and methane production were stimulated when inhibition was prevented from the start, throughout the reactor volume, by leachate recirculation and neutralization as a consequence of a relatively fast specific liquid flow rate. Figure 4 shows that there was no difference practically in solids reduction and methane production at the rather high specific liquid flow rate of 1 and 0.1 L d⁻¹. Uniform concentrations of VFA, sodium, and biomass formed rapidly in both cases throughout the reactor depth.

The concentrations of VFAs, sodium, and biomass as well as the methane production rate became non-uniform



Figure 4. Simulated time profiles of waste, VFA and biomass concentrations, and methane production rate averaged over the total reactor volume with pH adjusted to 7.0 at different specific liquid flow rates of 1 (1), 0.1 (2), and 0.01 L/d (3).

over the reactor depth at the low specific liquid flow rate of $0.01 \text{ L} \text{ d}^{-1}$ (Fig. 5). A high rate of solids reduction and methane production was obtained near the top of the reactor, where inhibition of methanogenesis was minimal. Evidently, in the absence of inhibition of methanogenesis and hydrolysis/acidogenesis, pH adjustment will not increase the process rate. Such an effect, where pH adjustment had no effect on the waste degradation rate, was shown recently with laboratory lysimeters by Jokela et al. (2001). Rhew and Barlaz (1995) showed that the lime incorporated into the cover landfill soil allowed for acidic leachate to be neutralized as it percolated through the cover. This resulted in the refuse near the top of the reactor being neutral and supporting increased rates of methane production while the refuse at the bottom of the reactor was still acidic.

Effects of Initial Biomass Spatial Distribution and pH Adjustment

Six peaks/depressions in the initial distribution of biomass and waste were used for the simulation presented in Figure 2. Additional simulations (results not shown) revealed that at the rather high specific liquid flow rate $q = 1 \text{ L d}^{-1}$, there was not much difference in solids reduction and methane production if the averaged uniform initial biomass distribution was used instead of six-peak distribution with pH ad-



Figure 5. Sequence of distributions of waste, VFA, biomass, and sodium concentrations, and methane production rate throughout the coordinate *Z* at 0 (0), 32 (1), 64 (2), 96 (3), 128 (4), 160 (5) days of incubation (low specific liquid flow rate q = 0.01 L/d). Waste means cellulose plus hemicellulose.

justed to 7.0. At the low specific liquid flow rate ($q = 0.01 L d^{-1}$) for the rather high biomass concentration initially located around the top of the reactor (one-peak distribution), methanogenesis was enhanced substantially earlier (Fig. 6, profile 1) relative to the case where an initial uniform biomass distribution was used (Fig. 6, profile 2). Evidently, the high sodium concentration at the top of the reactor decreased the non-ionized VFA concentration and promoted more rapid methanogenesis.

Without leachate neutralization or with pH adjusted to 5.0 (Fig. 7, time profiles 2), a strong inhibition of methanogenesis takes place and the high VFA concentration causes inhibition of hydrolysis/acidogenesis.

Mass Transfer-Based Acceleration of Methane Production

Vavilin et al. (2001a,b) have formalized the conditions of a mass transfer-based acceleration of methane production, when the intensity of VFA utilization in a methanogenic area (Θ) is sufficient for the complete conversion of the incoming VFA flow. According to this model, the following condition must hold for all *T*:

$$J_{\Theta} + \int_{\Theta} R_{\mathrm{H}}(X,T) dX < \int_{\Theta} R_{\mathrm{M}}(X,T) dX, \qquad (12)$$

where $R_{\rm H} = \chi kWf(S)$ is the rate of VFA production by hydrolysis/acidogenesis; $R_{\rm M} = \rho_{\rm m}g(S)[SB/(K_{\rm S} + S)]$ is the rate of VFA consumption; and J_{Θ} is the incoming VFA flow through the boundaries of Θ area due to diffusion and advection. In the opposite case, the acidogenic area expands because of inhibition of methanogenesis by the high VFA concentration. Based on model simulations and comparison



Figure 6. Simulated time profiles of waste, VFA and biomass concentrations, and methane production rate averaged over the total reactor volume at the low specific liquid flow rate q = 0.01 L/d with one-peak (1) and uniform (2) initial biomass distribution. (For the one-peak distribution b = 0.1 L and $\gamma_4 = 60$; the initial averaged biomass concentrations were the same for both cases.)



Figure 7. Simulated time profiles of waste, VFA and biomass concentrations, and methane production rate averaged over the total reactor volume with pH adjusted to 7.0 (1) and 5.0 (2) (standard set of model parameter values).

with experimental data of Lagerkvist and Chen (1992), it was shown (Vavilin et al., 2001a,b) that optimum OFMSW digestion can be achieved by setting a low rate of mass transfer (non-mixing conditions) initially with a subsequent increase in the mixing intensity, once a significant increase in the methanogenic population has taken place. The benefits of delayed mixing have also been reported for MSW anaerobic digesters by Stroot et al. (2001).

Based on the results of model simulations presented in this paper, diffusion coefficients comparable to the specific liquid flow rate do not have much influence on the rate of methanogenesis. The model showed that the beneficial effect of the specific liquid flow rate is attributed to the distribution of sodium ions, which prevents inhibition of methanogenesis and hydrolysis. As shown in Figure 5, uniform concentrations of VFAs, sodium, and biomass formed rapidly throughout the reactor depth at the high specific liquid flow rate of 1 L d⁻¹. However, at the low specific liquid flow rate of 0.01 L d⁻¹ the methanogenic and acidogenic areas were separated in space, and with increased digestion time, expansion of the methanogenic area took place.

According to the model simulations, to reduce the time needed for significant solid waste destruction and methane production, a balance between the rates of hydrolysis/ acidogenesis and methanogenesis should be achieved. Optimal conditions for methanogenesis are more important at the initial stage, but hydrolysis/acidogenesis was important at the final stage. The key parameters for methanogenesis are the maximum specific rate of VFA utilization, the initial methanogenic biomass concentration, and the VFA mass transfer processes (diffusion and advection). Additional simulations showed that the model was very sensitive to changes in the maximum specific rate of VFA consumption ρ_m as well as in the hydrolysis rate coefficient *k*. An in-

crease in the value of the hydrolysis rate coefficient (e.g., smaller solids particle size achieved by shredding) leads to a higher level of VFA concentration in the acidogenic area. In turn, such condition stimulates the VFA flow into the methanogenic area because of diffusion and advection. If inhibition of methanogenesis by a high VFA concentration is avoided by leachate recirculation and neutralization, then methane production will be enhanced and consequently the rate of waste degradation will increase. The higher the initial biomass concentration, the higher the rate of methanogenic biomass expansion. In accordance with experimental observations (Reinhart and Townsend, 1998), the model showed that it is important to introduce leachate recirculation slowly at the initial digestion stage before the onset of methanogenesis. Pumping leachate from an older landfill cell to a new one may be useful by introducing an established microbial population to accelerate waste degradation (Chynoweth et al., 1991; Doedens and Cord-Landwehr, 1989). Evidently, leachate pumping from an older to a newer landfill cell may be equivalent to buffer addition during leachate recirculation. As shown above, the model adequately described the stimulation of waste degradation and methane production as a result of leachate neutralization and recirculation. However, it should be emphasized that this model, as well as any other simulation models, is a simplification of the actual processes occurring in anaerobic digestion systems.

CONCLUSIONS

Based on the model simulations, waste degradation and methane production are stimulated when inhibition of methanogenesis and hydrolysis/acidogenesis are prevented from the start, throughout the reactor volume, by leachate recirculation and neutralization which is achieved by increasing the liquid flow rate. Uniform concentrations of VFAs, sodium, and biomass formed rapidly throughout the reactor depth at the high specific liquid flow rate of 1 L d^{-1} . There was not much difference in solids reduction and methane production if a mean, uniform initial biomass distribution was used instead of the six-peak distribution with pH adjusted to 7.0. However, at the low specific liquid flow rate of 0.01 L d⁻¹ the methanogenic and acidogenic areas were separated in space, and with increased digestion time, expansion of the methanogenic area took place. In the lowflow case, for the one-peak distribution of relatively high biomass concentration located around the top of the reactor, methanogenesis was enhanced substantially earlier than in the case where a uniform initial biomass distribution was used. At the low specific liquid flow rate, the high sodium concentration at the top of the reactor decreased the unionized VFA concentration and promoted more rapid methanogenesis in this area. According to model simulations, the rates of hydrolysis/acidogenesis and methanogenesis should be balanced to reduce the time needed for significant solid waste destruction and methane production. Optimal conditions for methanogenesis are more important

in the initial stage, but hydrolysis/acidogenesis is more important in the final stage of digestion, once accumulated VFAs are depleted.

NOMENCLATURE

Α	mass fraction of methane in biogas
a_i	heights at which waste (VFA) concentration have the mini-
	mum values
В	methanogenic biomass concentration
b_i	heights at which methanogenic biomass concentration have the
	maximum values
D	liquid dispersion coefficient
$D_{\rm S}$	diffusion coefficients for VFA
$D_{\rm B}$	diffusion coefficients for biomass
$D_{\rm N}$	diffusion coefficients for sodium
dP/dT	methane production rate
f(S)	non-ionized VFA inhibition function of hydrolysis
g(S)	non-ionized VFA inhibition function of methanogenesis
$[H^+]$	proton ion concentration
Ι	inhibiting concentration of non-ionized VFA
Κ	hydraulic conductivity
$K_{\rm a}$	VFA dissociation constant
$K_{\rm f}$	inhibition constant of hydrolysis
$K_{\rm g}$	inhibition constant of methanogenesis
K _s	half-saturation constant for VFA utilization
k	first-order hydrolysis rate constant
k _d	specific biomass decay coefficient
L	reactor height
т	total number of peak zones of methanogenic biomass
$m_{\rm f}$	hydrolysis inhibition degree index
$m_{\rm g}$	methanogenesis inhibition degree index
Ν	sodium concentration
n	total number of depression zones of waste
q	volumetric liquid flow rate per unit surface area
S	total volatile fatty acids (VFA) concentration
Т	time
W	solid waste concentration
Y	methanogenic biomass yield coefficient
Ζ	vertical coordinate of the 1-D reactor
α	fraction of biomass transferred by liquid flow
$\gamma_1 – \gamma_{5m}$	distribution coefficients
θ	moisture content
$\xi(Z)$	initial sodium concentration distribution along the reactor
	height
$\rho_{\rm m}$	maximum specific rate of VFA utilization
$\sigma(Z)$	initial waste concentration distribution along the reactor height
$\varphi(Z)$	initial methanogenic biomass concentration distribution along
	the reactor height
χ	stoichiometric coefficient
$\psi(Z)$	initial VFA concentration distribution along the reactor height

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