# Effect of substrate-seed mixing and leachate recirculation on solid state digestion of biowaste

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**Abstract** Lab-scale experiments were performed and a mechanistic model was developed to simulate the solid state digestion of biowaste in a batch reactor. Both experiments and model showed that the substrate-seed mixing degree and leachate recirculation rate have a strong effect on the reactor performance. This is due to mass transport limitations of volatile fatty acids (VFA) within the biowaste-seed bed. In that case two regions are developed in the digester, so-called acidogenic and methanogenic pockets. Limitations in mass transport will prevent irreversible acidification during start-up of the reactor because whereas high VFA concentration is met in the fresh waste pockets, the VFA concentration in the methanogenic pockets will remain low. However, accumulation of VFA in the acidogenic pockets will reduce the hydrolysis rate of biowaste due to inhibition by VFA. Moreover, experiments and simulations showed that the reactor performance can be improved by varying the leachate recirculation rate or applying sequential batch operation.

**Keywords** Biowaste; mass transport; modelling; solid state digestion

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## Introduction

The recycling of the organic fraction of municipal solid waste (MSW) is the preferred concept in waste management in Europe. After composting or digestion the product can be used as organic fertiliser or soil conditioner in agriculture. In the Netherlands, the organic fraction of MSW is source separated (so-called biowaste) because this strategy provides the best quality compost with respect to contamination with heavy metals and plastics. In contrast to landfilling and incineration, application of compost comprises the recycling of nutrients and organic matter and reflects the Dutch policy towards resource conservation and sustainability.

Conversion of organic wastes into compost can be realised by both anaerobic digestion and aerobic composting. Anaerobic digesters for solid organic waste streams can be operated in different modes which are reviewed by Cecchi *et al.* (1988). A rather simple concept is the BIOCEL process, a batch-wise solid state digestion, in which biowaste is pre-mixed with seeding material and subsequently digested at a total solids content of 35–40% (Ten Brummeler and Koster, 1990). The digestion process is promoted by recycling leachate over the biowaste bed. The system can handle biowaste without any pretreatment steps such as sieving and particle size reduction and together with the simple batch process results in low construction and operation costs compared to other anaerobic technologies. The main disadvantage of batch-wise solid state digestion (BSSD) is the high ratio of seeding material which is needed to prevent irreversible acidification at the start-up of the process (Ten Brummeler and Koster, 1990). Moreover, long solids retention times up to 40 days are met in batch solid dry digester, long compared to the high intrinsic hydrolysis rates of biowaste components. It was suggested by Veeken and Hamelers (1999) that low conversion rates are due to limited solids and hydraulic mixing in the biowaste bed.

This study presents a concept for interpretation of BSSD operation with leachate recirculation which was developed on basis of the theoretical model proposed by Kalyuzhnyi *et al.* (1999). It is proposed that, due to incomplete mixing in the biowaste-seed bed, two

regions are present in the digester: so-called acidogenic and methanogenic pockets (Chanakya *et al.*, 1997). The acidogenic pockets are composed of fresh biowaste which is degraded to VFA and hydrogen. The methanogenic pockets consist of the seeding material which converts VFA and hydrogen to methane and carbon dioxide. The transport of VFA from the acidogenic to the methanogenic pockets can take place only through the liquid phase, i.e. the leachate. The performance of lab-scale solid state digesters was studied for varying leachate recirculation rates. A mechanistic model is presented which includes both microbial kinetics and mass transport in the liquid phase of the biowaste bed. Simulations are described for varying substrate-seed mixing degrees and leachate recirculation rates.

## Materials and methods

### Biowaste and seeding material

Biowaste (screened at 80 mm) and seeding material (seed) were collected at the VALORGA full-scale plant (Tilburg, The Netherlands). Both biowaste and seed were homogeneous in compostion and particle size and were directly transfered in 5 L plastic bags and stored at –20ºC. The samples were defrosted in tap water one day before the experiments. The characteristics of biowaste and seed are given in Table 1. The digestion experiments and acetoclastic activity tests were performed at 30ºC.

**Table 1** Characteristics of biowaste and seeding material originating from the VALORGA full-scale plant

| | | biowaste | seed |
|---|---|---|---|
| TS | (% of total) | 40 | 35 |
| VS | (% of TS) | 60 | 40 |
| acetoclastic activity | (g COD.kg$^{-1}$ VS.d$^{-1}$) | <1 | 12 |

### Leachate recirculation experiments

An experimental set-up had to be designed to simulate BSSD with leachate recirculation. Moreover, it was the aim to study both the solid and gas phase of biowaste and seed compartments without influencing the course of the digestion process. For this, a novel experimental set-up was designed, composed of 5 reactors of 3 L each, which has to be considered as a single digester (Figure 1).

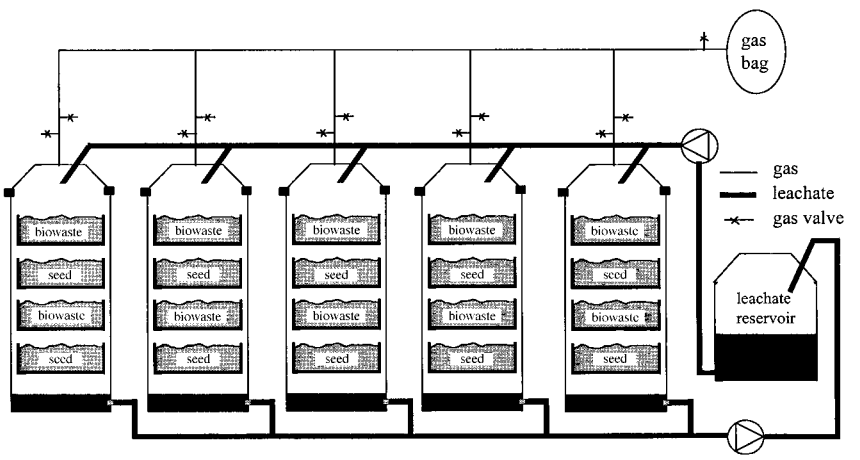


**Figure 1** Experimental set-up for simulation of leachate recirculation experiments

The biowaste and seed in each reactor were physically separated in 500 mL cups with perforated bottoms. The leachate from all reactors was collected in a 1 L magnetically stirred reservoir and recirculated back over the reactors. The produced biogas from the total system was collected in one gas bag. The gas phase could be sampled near to the gas bag, in the head space of the leachate reservoir and in each reactor. The leachate of the reservoir and the solid phases (comprised of solids and water) of biowaste and seed were sampled for VFA and pH. The solid phases could be sampled without disturbing the gas phase of the total system by disconnecting one reactor from the system and closing the total reactor system (using a special valve construction). After sampling of the solid phases, the sampled reactor was flushed with nitrogen and reconnected to the system. In this way, the gas phase of the system was diluted with approx 10% of nitrogen but this does not affect the digestion process. The solid phases of all five reactors were sampled consecutively. Two experiments were performed at a biowaste-seed ratio of 1:1 (w/w). For the first experiment no recirculation was applied and for the second a recirculation rate of 0.1 $m^3$ leachate per $m^3$ biowaste per day was applied. Additionally, the acetoclastic activity of biowaste and seed was measured for the second experiment.



#### Acetoclastic activity test

A standard acetoclastic activity test was adapted to monitor the activity in the biowaste and seed (Hwu *et al.*, 1996). Due to the low initial acetoclastic activity of biowaste and seed, no adaptation to VFA could be applied, and therefore the methane production was measured directly after the first addition of acetate. Taking into account biomass growth during the activity test, methane production can be described by $CH_4(t)=-[A(0)/\mu_{max}]\exp(\mu_{max}\ ?t)$, where $CH_4(t)$ is the cumulative methane production at time $t$ (in Nl, i.e. litres at 0ºC and 1 atm), $A(0)$ the initial acetoclastic activity (mg COD $?g^{-1}$ VS.$d^{-1}$), $\mu_{max}$ the maximum growth rate (in $d^{-1}$) and $t$ time (in d). The maximum growth rate was set at 0.2 $d^{-1}$ (Pavlostathis and Giraldo-Gomez, 1991) and as we are only interested in relative changes in activity, the exact value of the maximum growth rate is not relevant. The initial acetoclastic activity was estimated using non-linear least square curve fitting of the cumulative methane production.

#### Analyses

TS and VS were analysed according to standard methods (APHA, 1992). The concentration and composition of VFA were analysed with a Hewlett Packard 5890A gaschromatograph equipped with a glass column packed with Supelcoport and coated with 10% Fluorad FC 431 as described by Hwu *et al.* (1996). The gas (methane, carbon dioxide, oxygen, nitrogen and hydrogen) was analysed using a Hewlett Packard 5890 gaschromatograph equipped with a molecular sieve 5A as described by Hwu *et al.* (1996).

#### Model concept

The concept for BSSD of biowaste with leachate recirculation is given in Figure 2. Biowaste and seed are placed in layers which are assumed to be perfectly mixed. The number of layers and the order of biowaste and seed layers represents the mixing degree of the waste bed. Two types of water are distinguished in the solid phase, bound water and free water. It is assumed that anaerobic degradation of polymer substrate to methane takes place in the bound water of the solid phase. Degradation is described by a two step process: particulate organic matter (or polymers, $X_p$) is hydrolysed to VFA ($S$) and VFA is successively converted to methane. Both steps are inhibited by VFA which is represented by an inhibition constant $K_i$. Hydrolysis and methanogenesis can take place in both the biowaste and seed phase. Transport of VFA from biowaste to seed and vice versa can only take place through the free water, i.e. the leachate. Limitations in mass transfer are met for the trans-

port of VFA from the bound water ($S_b$) to the free water ($S_f$) for which mass transfer is described by an overall mass transfer coefficient ($k_{ov}$) whose magnitude depends on the particle size of the solid phase and the leachate distribition and rate over the waste bed. It is assumed that the liquid and gas phase are in equilibrium and the hydrogen partial pressure does not effect the degradation rate of VFA. The rate of hydrolysis ($r_h$) and methanogenesis ($r_g$) are given by the following equations:

$$r_h = k_h \frac{BO}{K_a + BO}\left(1 - \frac{S_b}{K_{i,h}}\right) X_p \quad (1) \qquad r_g = \mu_{max,g} \frac{S_b}{K_m + S_b}\left(1 - \frac{S_b}{K_{i,g}}\right) \frac{X_g}{Y_g} \quad (2)$$

where BO represent the occupation degree of polymer substrate by fermentative biomass, $BO = X_h/X_p$. The polymer concentration ($S_p$), VFA concentration in the solid phase ($S_m$) and leachate ($S_{mr}$) and methane production ($G$) are given by:

$$\frac{dX_p}{dt} = -r_h + bX_h + bX_g \quad (3) \qquad \frac{dS}{dt} = r_h(1-Y_h) - r_g - k_{ov}(S_b - S_f) \quad (4)$$

$$\frac{dS_r}{dt} = k_{ov}(S_b - S_f) + Q(S_r - S_f) \quad (5) \qquad \frac{dG}{dt} = (1-Y_g) r_g \quad (6)$$

The biomass concentrations of hydrolysing ($X_h$) and methanogenic ($X_g$) bacteria are described by:

$$X_h = Y_h r_h - b_h X_h \quad (7) \qquad X_g = Y_g r_g - b_g X_g \quad (8)$$

Table 2 gives the model parameters and their standard values.

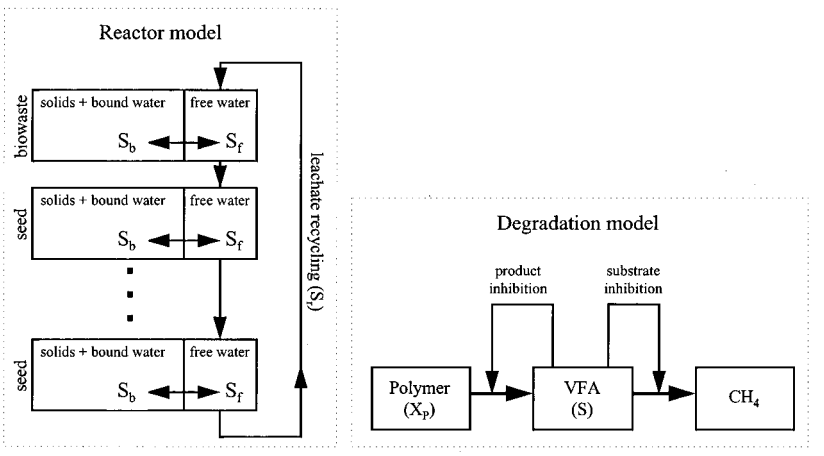


**Figure 2** Model concept for batch solid state digestion process

## Results and discussions

### Leachate recirculation experiments

From Figure 3 it can be seen that for the digestion without leachate recirculation only a small amount of methane is produced. This is due to the fact that VFA which is produced in the biowaste cannot be transported to the seed. VFA is accumulated in the biowaste bed but this stops at 40–50 g COD.$l^{-1}$ at which level hydrolysis is completely inhibited by VFA. Leachate recirculation is therefore necessary to keep the process going.

At a leachate recirculation rate of 0.1 $m^3$ ?$m^{-3}$ ?$d^{-1}$ methane production starts off after a lag phase of 5–10 days. Contrary to what might be expected for a completely mixed system

**Table 2** Parameter definition and values for model of biowaste solid state digestion with leachate recycling

| Parameter | Symbol | Value | Unity |
|---|---|---|---|
| degradable polymer concentration | $X_P$ | 600 (2)# | kg polymer.m$^{-3}$ |
| VFA in water bound to solid phase | $S_m$ | 9 (0)# | kg VFA.m$^{-3}$ |
| VFA in leachate phase | $S_{mr}$ | 1 | kg VFA.m$^{-3}$ |
| fermentative biomass | $X_h$ | 10 (10)# | kg biomass.m$^{-3}$ |
| methanogenic biomass | $X_g$ | 2 (20)# | kg biomass.m$^{-3}$ |
| overall mass transfer coefficient | $k_{ov}$ | 0.25 | d$^{-1}$ |
| yield for hydrolysis | $Y_h$ | 0.1 | kg biomass.kg$^{-1}$ substrate |
| yield for methanogenesis | $Y_g$ | 0.05 | kg biomass.kg$^{-1}$ substrate |
| decay rate of biomass | $b_h$, $b_g$ | 0.02 | d$^{-1}$ |
| inhibition coefficient for hydrolysis | $K_{i,h}$ | 30 | kg VFA.m$^{-3}$ |
| inhibition coefficient for methanogenesis | $K_{i,g}$ | 8 | kg VFA.m$^{-3}$ |
| affinity constant for hydrolysis | $K_a$ | 0.1 | kg biomass.kg$^{-1}$ polymer |
| affinity constant for methanogenesis | $K_m$ | 0.1 | kg VFA.m$^{-3}$ |
| hydrolysis rate of polymer | $k_h$ | 0.25 | d$^{-1}$ |
| maximum growth rate for methanogenesis | $\mu_{max,g}$ | 0.2 | d$^{-1}$ |
| leachate recirculation rate | $Q$ | 10 | m$^3$.m$^{-3}$.d$^{-1}$ |

# values for biowaste (and for seed between brackets)

with a VFA production rate of 20 g COD/kg biowaste/day and a methanogenic activity of 2 g COD/kg seed/day, no irreversible acidification of the batch process is met. Methanogenesis in the seed phase is not completely inhibited because a concentration gradient in VFA and pH is established (Figure 3). The gradient is due to mass transport limitations and thus has a positive effect on the start up of the batch process. The methane production (fitted from day 10) can be described by an apparent first order hydrolysis rate constant ($k_{h,app}$) of 0.02 d$^{-1}$, much lower then the intrinsic hydrolysis rate of biowaste components of 0.08–0.27 d$^{-1}$ as reported by Veeken and Hamelers (1999). The lower $k_{k,app}$ is due to the fact that VFA is accumulated in the biowaste and thus inhibits hydrolysis of biowaste. In this case, mass transport limitation of VFA has a negative effect on the process rate. These results suggest that the reactor performance can be improved by applying a variable leachate recirculation rate. At the start of the batch process the leachate rate is kept low to prevent irreversible acidification of the seed after which the leachate rate is increased to prevent inhibition of hydrolysis and stimulate the degradation rate of biowaste.

It was assumed that VFA production takes place in the biowaste from which VFA is transported to the seed where it is converted to methane. Figure 4 shows the acetoclastic activity of biowaste and seed during digestion at a recirculation rate of 0.1 m$^3$ ?m$^{-3}$ ?d$^{-1}$. Surprisingly, the activity remains constant in the seed but increases substantially in the biowaste bed after 20 days of digestion. Methane production can take place in biowaste when VFA drops below 10 g COD.l$^{-1}$ and pH increases above 7 (Figure 3). This implies that the batch process can be applied without inoculation when inhibitory fermentation products are fast removed from biowaste. This can be managed by leading the leachate from the biowaste reactor over a methanogenic reactor (reactor from which VFA are leached and methane production has started) before returning back to the top of the biowaste reactor. This strategy of sequential BSSD is described by Chynoweth *et al.* (1992).

#### Model simulations

Figure 5 shows the cumulative methane production for simulations at varying biowaste-seed mixing degrees (at $k_{ov}$ of 0.25 d$^{-1}$) and leachate recirculation rates (at Q of 10 m$^3$ ?m$^{-3}$ ?d$^{-1}$). Increasing the leachate recirculation rate from 1 to 100 m$^3$ ?m$^3$ ?d$^{-1}$ results in an

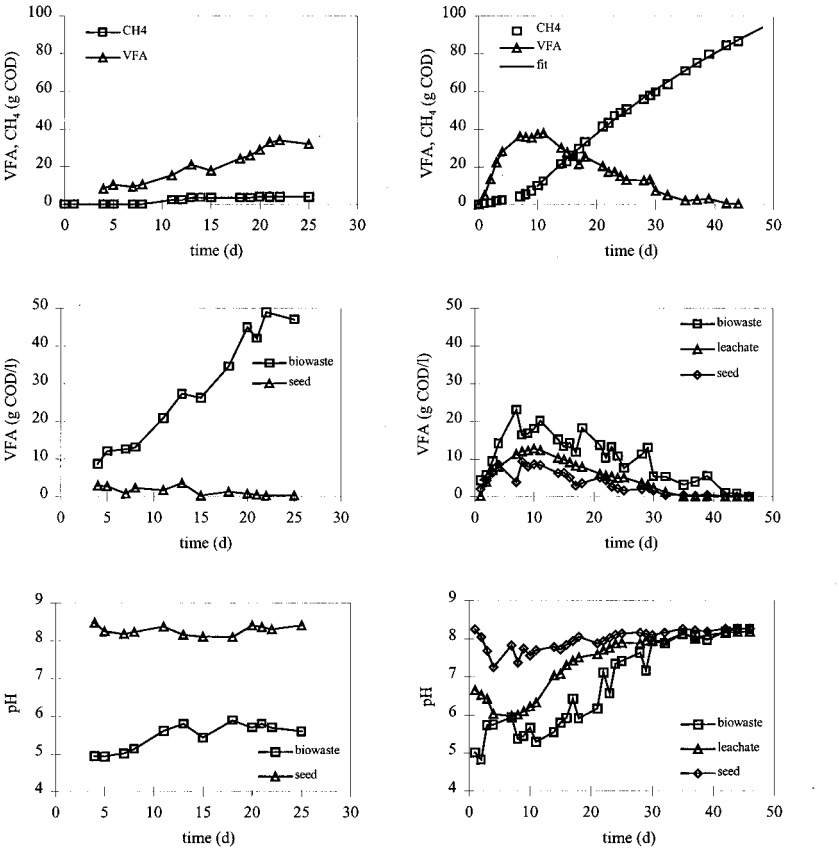


**Figure 3** Development of COD of total reactor system (top), VFA (middle) and pH (bottom) for biowaste digestion without leachate recirculation (left) and recirculation rate of 0.1 $m^3$ ?$m^{-3}$ ?$d^{-1}$ (right)

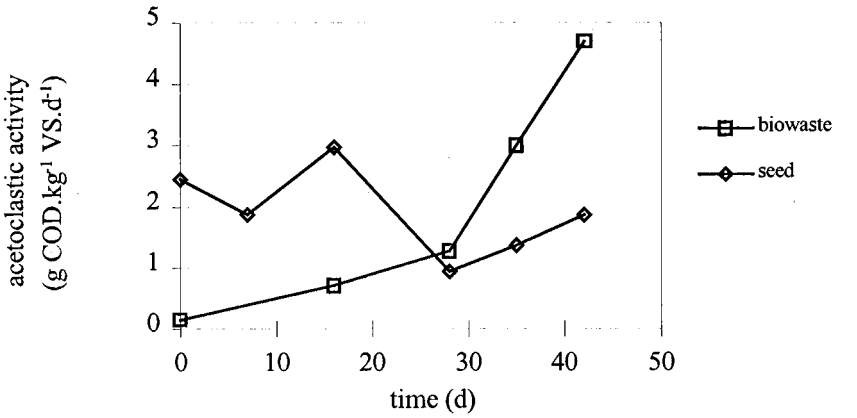


**Figure 4** Acetoclastic activity of biowaste and seed phases for batch digestion at leachate recirculation rate of 0.1 $m^3$ ?$m^{-3}$ ?$d^{-1}$

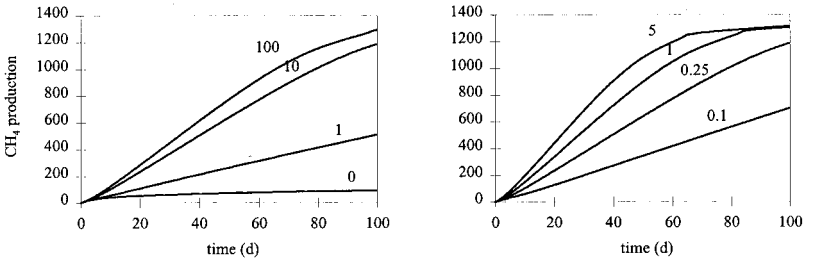


**Figure 5** Methane production for batch digestion of biowaste for simulations at varying leachate recirculation rates (left) and biowaste-seed mixing degrees (right)



increase in $k_{h,app}$ (fitted to the cumulative methane production) from 0.001 to 0.017 $d^{-1}$. Also an increase in the degree of biowaste-seed mixing results in an increase in methane production: an increase in $k_{ov}$ from 0.1 to 5 $d^{-1}$ results in an increase in $k_{h,app}$ from 0.006 to 0.032 $d^{-1}$. However, these conversion rates are still much lower than the intrinsic hydrolysis rates of biowaste (maximum hydrolysis rate under optimal conditions) which range between 0.08–0.27 $d^{-1}$ (Veeken and Hamelers, 1999). The lower conversion rate of biowaste in BSSD is due to limited mass transport of VFA within the waste bed from biowaste to seed. This results in accumulation of VFA in biowaste which inhibits the hydrolysis and results in a lower hydrolysis rate of polymer. The latter was also observed in the experiments (Figure 3). Figure 6 shows VFA in biowaste, seed and leachate for leachate recirculation rates of 1 and 100 $m^3 ? m^{-3} ? d^{-1}$ (at $k_{ov}$ of 0.25 $d^{-1}$). A leachate recirculation rate of 1 $m^3 ? m^{-3} ? d^{-1}$ results in accumulation of VFA in the biowaste which strongly inhibits hydrolysis of biowaste. The VFA concentration in the seed remains very low (<0.01 g COD.$l^{-1}$) because the assimilative methanogenic activity of the seed is greater than the VFA flux from biowaste to seed. Increasing the leachate recirculation rate results in a higher transport rate of VFA from biowaste to seed. This leads to less VFA accumulation in biowaste and thus higher hydrolysis rates. Therefore an increased leachate recirculation rate and improved mixing of biowaste and seed results in a higher biowaste conversion rate and consequently shorter solids retention times. However, the leachate recirculation rate and biowaste-seed mixing degree cannot be increased unlimitedly because a higher transport rate of VFA from biowaste to seed can result in irreversible acidification of the seed when the methanogenic activity of the seed is too low (results not shown here).

The BIOCEL process strongly resembles a landfill and is in fact derived from the landfill concept (Ten Brummeler and Koster, 1990). Therefore, the model can also account for the low methane production rates which are normally found in sanitary landfills (Borzacconi *et al.*, 1997). Rainwater infiltration rates of about 0.001 $m^3 ? m^{-3} ? d^{-1}$ are manifold lower then the leachate recirculations which are met in the BIOCEL process. These low infiltration rates together with the high heterogeneity and random distribution of the acidogenic and methanogenic pockets within the landfill will result in strong inhibition of hydroysis and very low apparent first order hydrolysis rates for MSW in landfills.

The model is a very valuable tool to get insight into the complex process and to develop new strategies for BSSD. At this stage, the model is of a qualitative degree and various model parameters still have to be validated after which the model can be calibrated for an extensive set of data. The model can also be applied to simulate leachate and biogas production in landfills.

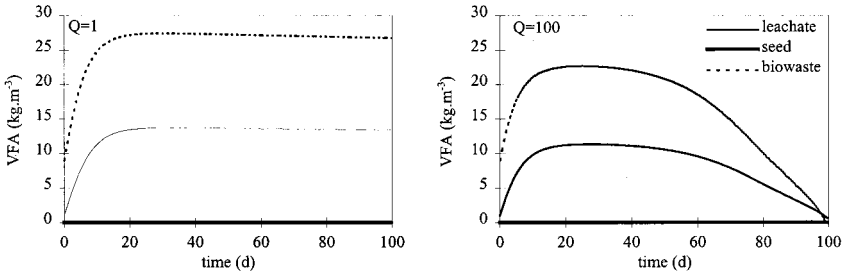


**Figure 6** VFA in biowaste, seed and leachate for batch digestion of biowaste for simulations at leachate recirculation rates of 1 and 100 $m^3$ ?$m^{-3}$ ?$d^{-1}$

## Conclusions

The experimental work and model simulations showed that the leachate recirculation rate and biowaste-seed mixing degree are very important parameters for the BSSD operation. Increasing the leachate recirculation rate and biowaste-seed mixing degree promotes mass transport of VFA from biowaste to seed regions. Optimum conditions for biowaste digestion are met when irreversible acidification of the seed and inhibition of biowaste hydrolysis are prevented. This can be reached by applying variable leachate recirculation rates: a low leachate rate at the start which can be increased gradually to higher leachate rates as the process progresses. Another strategy is the sequential BSSD operation as described by Chynoweth *et al.* (1992) without addition of seeding materal. In the first stage of the process, high leachate rates are applied to wash out VFA and prevent inhibition of hydrolysis. The leachate is fed over a reactor which is already in the second stage of the process, i.e. methanogenic activity which is established after VFA has been washed out of the reactor. Under optimal conditions with solids retention times of 3-4 weeks, the volumetric loading rate of the BIOCEL process can be increased to 20 tons/$m^3$reactor/d and a biogas production of 2 $m^3$/$m^3$reactor/d.

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