A SIMPLIFIED ANALYSIS OF REACTION AND MASS TRANSFER IN UASB AND EGSB REACTORS

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ABSTRACT

The paper is focused on the study of the hydrodynamics, reaction kinetics and mass transfer in lab-scale Upflow Anaerobic Sludge Blanket (UASB) and Expanded Granular Sludge Bed (EGSB) reactors treating wastewaters with low volatile fatty acids concentrations. The fluid pattern in the UASB approached a plug-flow reactor because of the low turbulence provided by the upflow velocity and gas production. The EGSB behaved as a continuous-stirred tank reactor due to the effects of the high recirculation rate. Liquid film mass transfer resistance seemed negligible in the EGSB but important in the UASB reactor. This should be the consequence of the different hydrodynamic conditions in the two reactors. The rate of acetate degradation was best fitted by an apparent half-order kinetics in both reactors. Internal mass transfer limitations, associated to an intrinsic zero-order kinetics, was detected in the degradation of low acetate concentrations by the granular biomass.

Keywords. Anaerobic digestion, EGSB and UASB reactors, low strength wastewaters, kinetics, mass transfer.

INTRODUCTION

The application of anaerobic processes for the treatment of effluents with COD values above 1500-2000 mg/L is increasing due to their economic feasibility. However, full-scale anaerobic systems for the treatment of wastewaters with lower COD values are not very common [1].

The Upflow Anaerobic Sludge Blanket (UASB) reactor is one of the anaerobic technologies that since its initial development recorded extensive full scale applications [2]. Recently, there are some attempts to use it for the treatment of low strength wastewaters. Moreover, for the treatment of such wastewaters there were also suggestions to implement the Expanded Granular Sludge Bed (EGSB) reactor [3,4]. The only difference between the EGSB reactor and the UASB reactor is that in the former the effluent is recirculated, providing much higher liquid velocity for the same residence time.

The design of full scale UASB reactors has been based in the experimental data obtained in treatability and performance studies. Models describing the apparent kinetics have been applied to steady-state degradation of non-inhibitory soluble organics [2, 4]. However, external and internal mass transfer resistance may originate substantial differences between apparent and intrinsic kinetics as indicated by Rodrigues et al., [5] and Arvin and Harremoes [6]. Moreover, the fluid mixing pattern within the reactor may also have an impact in reactor performance, depending on effluent type and removal rate kinetics [5, 7].

This research is focused on the feasibility of UASB and EGSB reactors for the treatment of low strength soluble wastewaters. In order to contribute for their development, the present study performs an evaluation of the flow pattern and of the apparent kinetics, as well as of the possible role of external and internal mass transfer resistance in acetate degradation.

MATERIALS AND METHODS

The present analysis of UASB and EGSB reactors was based on the application of models and techniques, as briefly is indicated. The flow pattern was assessed by residence time distribution experiments (RTD) with an inert tracer [7]. The kinetic of acetate removal was modelled by a power-law equation, as sugested by Rodrigues et al., [5] and Arvin and Harremoes [6]. These authors rely upon the analogy between biofilm and catalytic reactors to incorporate mass transfer resistance in the modelling of substrate degradation. This approach considers that some approximations concerning dominant features of the process should be made, in order to apply simple expressions useful for reactor design [6]. Major assumptions in the present case are: i) steady-state conditions,
ii) acetate as limiting substrate, iii) diffusion as the predominant mass transport mechanism and iv) the granules of biomass are considered homogeneous and spherical. The apparent rate law of acetate degradation was deduced from the fitting with first, zero and half-order equations. The presence of an external mass transfer resistance was assessed combining mass balances and a flow model, as described by Rodrigues et al., [5]. The role of diffusional limitations was evaluated in batch tests, by measuring the acetate removal rate obtained using different granule sizes (obtained by disintegration) with the subsequent determination of the best fit kinetics.

Reactors

Both reactors (UASB and EGSB) were made of acrylic glass, with an empty volume of 480 ml, an internal diameter of 41.8 mm and a height of 353 mm. The reactors had a thermo-regulated water jacket for temperature control and the feed was pumped by a Watson Marlow-101 peristaltic pump. Liquid recirculation in the EGSB was obtained with a Ihein centrifugal pump.

Biomass

The inoculum was an anaerobic culture structure in granules. The UASB inoculum was obtained from a full scale reactor operating at a wheat starch factory, in Latenstein, The Netherlands, with 74 kg.m⁻³ of volatile solids (VS). The granule diameter was measured with an image analyser Leica Quantimet 500. Most equivalent diameters were between 1.6-2.7 mm and an average diameter of 2 mm was considered for data analysis. The EGSB inoculum was provided by another UASB reactor from a recycled paper factory, in Roermond, The Netherlands, with 117 kg VS.m⁻³ and a granule average diameter of 1.6 mm. Porosity of the sludge bed was assessed measuring the difference in the volume of a bed of granules (filtered onto a membrane) and the same bed of granules filled with a known volume of water. Measured values range indistinctly between 0.36-0.43. Therefore, the value adopted for calculations was approximated to decimals, that is 0.4.

Substrate

The EGSB substrate was acetic acid and the UASB substrate was a mixture of acetic and propionic acids, at a proportion of 4.5 to 1. In this reactor, the acetate produced by the propionate degradation was considered as substrate feed in removal rate calculations, causing an increase in this rate near 10%. Both substrates were supplemented with a solution containing macro and micronutrients. The macronutrients solution, 2 ml, of substrate, was as follows, in mg.m⁻³: NH₄Cl: 164, KH₂PO₄: 28.3, (NH₄)₂SO₄: 28.3, MgCl₂: 25, KCl: 45, yeast extract: 3. The micronutrients solution, 0.03ml, in mg.m⁻³: FeCl₃·6H₂O: 2, H₃BO₃: 0.05, ZnCl₂: 0.05, CuCl₂·2H₂O: 0.038, MnCl₂·4H₂O: 0.5, (NH₄)₂Mo7O₂4·4H₂O: 0.05, AlCl₃: 0.09, CoCl₂·6H₂O: 2, NiCl₂·6H₂O: 0.092, Na₂SeO₃·5H₂O: 0.164, EDTA: 1. The substrate was previously neutralised with NaOH.

Analytical Techniques

Volatile fatty acids: Volatile fatty acids (VFA) determination was performed by HPLC, Jasco 880-PU, equipped with a detector Jasco 870-Vis and a Chrompack column, under a temperature of 40°C. The eluent was 0.01 N H₂SO₄, at a flow rate of 0.7 ml.min⁻¹. All samples were centrifuged (15 000 g, 5 min.).

Solids: Volatile solids concentration was determined according Standard Methods [8].

Lithium: Lithium, as Li⁺, was analysed by a flame photometer, Jenway PFP 7.

Residence time distribution

The RTD tests were performed with lithium chloride as a tracer. The tracer was injected as a pulse in the feed line, 4 cm below the reactor entrance and sampled in the effluent in a mixing cup. The injection volume in the EGSB was 1 ml of a solution of 3.2 g.1⁻³ Li⁺.

<table>
<thead>
<tr>
<th>Table 1. Operational parameters of the UASB and EGSB reactors.</th>
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<tbody>
<tr>
<td><strong>Temperature</strong></td>
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<td>-----------------</td>
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<tr>
<td></td>
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<tr>
<td><strong>pH</strong></td>
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<tr>
<td><strong>Hydraulic residence time</strong></td>
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<td><strong>Upflow fluid velocity</strong></td>
</tr>
<tr>
<td><strong>Reynolds number (reactor)</strong></td>
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<tr>
<td><strong>Reynolds number (particle)</strong></td>
</tr>
<tr>
<td><strong>Average granule diameter</strong></td>
</tr>
<tr>
<td><strong>Biomass concentration</strong></td>
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<tr>
<td><strong>Substrate</strong></td>
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</table>

(*) This biomass concentration is an average value: initially, the volume of granules was 90 ml; then, it was lowered gradually down to 70 ml due to partial wash-out.
The effluent lithium was sampled during at least 3 times the HRT of the reactors and the recovery was always near 95%. In the UASB the injection was 2 ml of 5 g l⁻¹ of a lithium chloride solution.

Experimental Conditions

Reactors:
The operating conditions of the UASB and EGSB reactors are summarised in Table 1.

The volumetric organic loading was increased by raising the substrate concentration in the feed. The maximum acetate concentration in the feed was 1.6 kg m⁻³ in the UASB and 1.2 kg m⁻³ in the EGSB reactor.

Batch degradation tests

Acetate degradation batch tests were performed with EGSB type of granules of different sizes with the purpose of detecting diffusional limitations within the granules. The granules range sizes were 1-2 mm (intact granules), 0.1-1.0 mm (obtained by crushing the granules in a mortar and passing them through a needle using a hypodermic syringe) and 0.01-0.1 mm (granules disintegrated by stirring with 2 mm glass beads). The tests were performed at 30°C, at 140 rpm and pH 7. The initial acetate concentration was inferior to 570-640 mg l⁻¹ and was measured until depletion in order to approach the conditions in the UASB and EGSB reactors. Biomass concentration ranged from 3 to 6 kg VS m⁻³. Granules were previously acclimatised during 5 days. The assessment of the order of reaction was performed by an integral method. A power rate law was considered, assuming first, 1/2 and zero-order reaction kinetics. The differential rate equation was integrated to give a relationship between acetate concentration and time. If the correct order has been chosen, the data should be well correlated by a straight line passing through the origin. The fitting was obtained by a least squares method.

RESULTS AND DISCUSSION

Fluid Mixing Pattern

EGSB and UASB acetate concentration profiles along the reactor are presented in Figure 1 under typical operating conditions. The normalised RTD in the EGSB is presented in Figure 2. The same type of function for the UASB reactor is plotted on Figure 3.

The flow patterns in the lab-scale EGSB and UASB reactors are very different. Figure 1 and 2 show that the EGSB mixing properties approach those of an ideal continuous stirred tank reactor (CSTR) [7]. In general, a high interstitial velocity tends to increase plug-flow characteristics, but the recirculation increases back mixing and this effect predominated in the EGSB. The determination coefficient of the fitting of a perfectly mixed CSTR model to the experimental data was 0.92 (dotted line in Figure 2). A model incorporating a hydraulic short circuit assumption did not lead to a significant improvement, with a small increase of the determination coefficient to 0.95. Therefore, for the sake of simplicity, the ideal mixing model was considered an appropriate representation of the flow conditions in the EGSB reactor. This approach has also been used in other studies with the EGSB [5, 6].

Unlike the EGSB reactor, the acetate profile in the UASB displays a gradient throughout the reactor, Figure 1. This fact is the consequence of its operation at a low upward velocity and of the weak gas production, both insufficient to produce the turbulence required for fluid backmixing. The RTD also displays a plug-flow trend, Figure 3. Considering the tank-in-series model [7], a determination coefficient of 0.97

![Figure 1. Acetate concentration profile along the EGSB and UASB reactors.](image)
using the RTD experimental data was obtained with 4 equivalent reactors, as shown by the dotted line on Figure 3. The observed tracer tail may be an indication of stagnant zones, with interchange between flowing and non-flowing zones [7]. Several other authors have published RTD essays in UASB systems, although very few were performed on full scale reactors as done by Heerjes et al. [9]. These authors proposed a model based on two mixers in series, represented by the sludge bed and the sludge blanket. Lettinga and Hulshoff Pol [2], mentioned indeed a general pattern of the UASB as a reactor with a substrate gradient in it. Probably, UASB systems display a certain range of mixing patterns as a function of the hydraulic load and gas production and also of the inlet feed system and sludge bed hold-up. In the present case, the lab-scale UASB was considered as a plug-flow reactor for the purpose of kinetic analysis.

Acetate Degradation Rate

Evaluation of the apparent kinetics:

Mass balances and kinetic equations in UASB and EGSB reactors were integrated in accordance with the mixing.
pattern indicated in the previous section, plug-flow for the UASB and completely mixed for the EGSB respectively. The acetate removal rate was fitted by testing first, 1/2 and zero-order reactions. The 1/2 order reaction produced the best fitting in both systems. Figures 4 and 5 present EGSB and UASB apparent (or "observed") removal rates per mass of volatile solids (r_c), plotted versus the bulk concentration in the reactors. With an apparent 1/2 order reaction, the average bulk concentration is represented in the UASB by

\[ \frac{Ac_{in} - Ac_{out}}{Ac_{in}^{1/2} - Ac_{out}^{1/2}} \] (plug-flow) and in the EGSB by \[ Ac_{out}^{1/2} \] (CSTR).

The fittings of half-order kinetics presented in Figure 4 and Figure 5 have a determination coefficient of 0.98 (EGSB) and 0.96 (UASB).

Observable zero-order kinetics can be discarded because the removal rates were lower than 1-2 kg COD.kgVS\(^{-1}\).day\(^{-1}\), which is commonly recognized as a minimum value for zero-order reactions.

Reported saturation coefficients for anaerobic granules are 10 mg VFA-COD\(^{-1}\) [6] and 70 mg l\(^{-1}\) [10]. Since the range of bulk acetate concentration in the EGSB (50-5 mg l\(^{-1}\)) was not significantly lower than those coefficients, first order kinetics are not probable to occur, as well. The same can be said about the UASB reactor, where the acetate concentrations were much higher (250-350 mg l\(^{-1}\), in general). It is here suggested that first order kinetics should be more appropriate at concentrations much lower than those measured in these reactors.

Values of the rate coefficients for anaerobic digestion processes are scarce, although there are a significant number of published data on 1/2 rate reactions in aerobic and anoxic processes, as reviewed by Arcangeli and Arvin [11]. Table 2 presents apparent 1/2 order rate coefficients for methanogenic reactors, including also an anoxic process, denitrification.

![Figure 4](image-url)  
**Figure 4.** EGSB acetate specific removal rate as a function of the bulk concentration with an apparent half-order reaction.

![Figure 5](image-url)  
**Figure 5.** UASB acetate specific removal rate as a function of the bulk concentration with an apparent half-order reaction.
Table 2. Apparent order 1/2 rate coefficients in anaerobic and anoxic conditions.

<table>
<thead>
<tr>
<th>System</th>
<th>Substrate</th>
<th>Temp. (°C)</th>
<th>( K^* ) reactor volume (kg_{s}^{1/2}m_{p}^{-3/2}s^{-1})</th>
<th>( K^* ) granule area (kg_{s}^{1/2}m_{p}^{-1}s^{-1})</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>UASB</td>
<td>acetate</td>
<td>30</td>
<td>0.254*10^3</td>
<td>0.5*10^4</td>
<td>This study</td>
</tr>
<tr>
<td>EGSB</td>
<td>acetate</td>
<td>24</td>
<td>0.256*10^3</td>
<td>0.6*10^4</td>
<td>This study</td>
</tr>
<tr>
<td>Upflow filter</td>
<td>molasses</td>
<td>35</td>
<td>0.9<em>10^{-3}-0.14</em>10^{-3}</td>
<td>0.3<em>10^4 - 4.8</em>10^{-4}</td>
<td>[12]</td>
</tr>
<tr>
<td>Downflow filter</td>
<td>acetate</td>
<td>35</td>
<td>-</td>
<td>1.2*10^4</td>
<td>[13]</td>
</tr>
</tbody>
</table>

In the present study, the apparent 1/2 order rate coefficients of acetate degradation by UASB and EGSB reactors are analogous, as displayed in Table 2. This result was expected since both reactors were fed with a similar substrate. It is worth noting that apparent rate coefficients were not affected by different temperatures (UASB at 30°C and EGSB at 24°C). Moreover, the presence of a higher bulk acetate concentration in the UASB reactor was not able to induce an increased performance. Some speculations are possible about such questions. First, the fact that the granular inoculum was not from the same source does not seem to be significant: activity measurements performed with the UASB type of granules showed only a slightly higher activity than the EGSB type [14]. Second, an increased affinity to substrate at low temperatures, as discussed recently [15], does not also appear to be important because of the small temperature difference. Therefore, the presence of an external mass transfer resistance is a possibility and will be discussed in the next point.

Role of external mass transfer

The role of external mass transfer limitations on the performance of the EGSB and UASB reactors was evaluated considering the analysis for homogeneous biofilms with intrinsic zero-order kinetics presented by Rodrigues et al., [5]. By means of an algebraic manipulation of CSTR and PFR mass balances and by relating inlet substrate concentrations to the removal efficiency (E), it is possible to conclude that the data should be fitted by a straight line intercepting the axes at the origin if mass transfer limitations are absent. The combination of intrinsic parameters (diffusivity and reaction rate coefficient) with the effectiveness factor makes this analysis independent of intrinsic parameters and particle geometry. In the case of CSTR, the inlet substrate concentration is plotted versus \((1-E)/E^2\) and in the case of a PFR, versus \(1/(1-(1-E)^{1/2})^2\). Figure 6 and Figure 7 present those results for the EGSB and UASB reactors.

As it can be observed in Figure 6, in the EGSB system there is a linear correlation intercepting the axes origin. Consequently, it can be considered that liquid film mass transfer resistance is not important in the EGSB reactor. Most probably, this is the result of the high liquid velocity in the reactor. On the contrary, in the UASB reactor, external mass transfer limitations may be present, (Figure 7), since the plot of \(1/(1-(1-E)^{1/2})^2\) vs. the substrate inlet concentration reveals a significant deviation from the axes origin. The occurrence of such phenomenon is related to weak liquid turbulence and should be the consequence of the very low velocity and low gas production in the lab-scale reactor. Some experiments with the EGSB reported by de Man et al., [3] are not conclusive about the effect of the external mass transfer in the observed removal rates. In vinasse wastewaters containing 200-450 mg COD.l^{-1} at 8°C, the authors refer to a small gain in the removal rate when the recirculation ratio is increased. However, the reported presence of 5-10% of colloidal particles and their possible adsorption on the granules could cause an increase in COD removal. In the other case mentioned by these authors, with volatile fatty acids and influent concentrations of 2600 mg COD.l^{-1}, the recycle ratio had no effect in the removal rate, which seems in accordance with an improbable substrate limitation at this concentration level. Kato et al., [4] with an influent feed concentration of 100-200 mg COD-VFA.l^{-1}, at a loading of 6.4 kg COD.m^{-3}.day^{-1}, reported an increase in the removal efficiency with the increase in upflow velocity up to 14 m.h^{-1}. That could represent a typical effect of liquid film resistance in a mass transfer-limited region. In other experiments, the higher removal efficiency was attained around 3 m.h^{-1}, an indication that this value represented the minimum upflow velocity necessary to neglects the external mass transfer resistance.

Role of internal mass transfer:

The possibility of internal mass transfer limitations in the acetate degradation process was studied by examining the relationship between the degree of fitting of an apparent kinetics and the granule size.

Power-law equations, namely first, 1/2 and zero-order reaction, were integrated and plotted as a function of time and the determination coefficient of the linear fitting was assessed. Table 3 presents the results for the three sets of particle size that were obtained through the disintegration of EGSB granules. Figure 8 shows an example of the application of the zero-order kinetics to the three different granule sizes.
Figure 6. EGSB reactor: \( (1-E)/E^2 \) vs. inlet acetate concentration.

Figure 7. UASB reactor: \( 1/(1-(1-E)^{-1/2})^2 \) vs. inlet acetate concentration.

Table 3. Determination coefficients of the fittings of power-law kinetics vs. time.

<table>
<thead>
<tr>
<th>Apparent reaction rate order</th>
<th>intact granules ( 1 &lt; d &lt; 2 ) (mm)</th>
<th>semi-disintegrated granules ( 0.1 &lt; d &lt; 1.0 ) (mm)</th>
<th>disintegrated granules ( 0.01 &lt; d &lt; 0.1 ) (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>first order</td>
<td>0.919</td>
<td>0.882</td>
<td>0.793</td>
</tr>
<tr>
<td>1/2 order</td>
<td>0.993</td>
<td>0.987</td>
<td>0.949</td>
</tr>
<tr>
<td>zero-order</td>
<td>0.947</td>
<td>0.965</td>
<td>0.982</td>
</tr>
</tbody>
</table>
The results presented on Table 2 and Figure 8 show that the fitting with zero-order kinetics improves as the granule size decreases. First and 1/2 order fitting had the opposite behaviour. A similar experiment was performed with intact and semi-disintegrated UASB granules and revealed the same trend at such acetate concentrations. This pattern suggests that the granules had a zero-order intrinsic kinetics under the tested conditions. Presumably, as granules become larger, substrate limitations will tend to shift the observable kinetics to half-order. This is equivalent to stating that intrinsic first-order reactions remain observable first order reactions under substrate limitations and that apparent 1/2 order may result from intrinsic zero order reactions when the biofilm is partly penetrated [6]. In agreement with such results, Alkemaar et al., [14] observed that the apparent saturation coefficient of acetate decreased with granules disintegration for the same types of inoculum as those of the present study. In their work, the maximal methanogenic activity remained constant with disintegration, suggesting it was not affected by internal mass transfer limitations. However, this conclusions may be too general: in fact, as the experiments were carried out with high initial acetate concentrations, it is possible that in such specific conditions the granules were completely penetrated by the substrate. The same phenomenon could be present in some of the experiments by Dolfin [16], although he reported significant acetate limitations in granules of large diameter and high specific activity. In granules with different biomass structures [17] or subject to other environmental situations, namely under inhibitory conditions [18], additional interactions may hide or prevent the occurrence of acetate limitations.

CONCLUSIONS

Residence time distribution tests in the lab-scale UASB and EGSB reactors indicate that UASB reactor could be considered as an ideal plug flow reactor and the EGSB as a stirred tank reactor, for the purpose of discussing substrate conversion efficiency. Possibly, stagnant zone or short-circuiting phenomena could be also present.

The anaerobic degradation of low acetate concentrations, lower than 1500 mg.L$^{-1}$, was properly fitted by an apparent 1/2 order kinetics expression. The 1/2 order rate coefficients, $k'$, were $0.254 \times 10^3$ and $0.256 \times 10^3$ in the EGSB and UASB reactors, respectively.

In terms of external mass transfer, the presence of a liquid film resistance was detected in the lab-scale UASB reactor. Such resistance is most probably related to operating conditions, namely a very low upflow velocity and a concomitant low gas production. Both can reduce the advantageous effects of a high bulk substrate concentration. Unlike the UASB, in the EGSB reactor external mass transfer limitations could be neglected. The much higher upflow velocity, as a result of the high recirculation ratio in the EGSB.
seemed to be responsible for the minimisation of the liquid-granule mass transfer resistance.

The apparent half-order kinetics at low acetate concentrations may have been a consequence of an intrinsic zero-order reaction under internal substrate limitations. This hypothesis was suggested by the experimental results obtained with three different granules sizes. In fact, by decreasing the granules size, the fitting of the zero-order model improves.

In general, the overall results point out to an intrinsic zero-order kinetics at relatively low bulk acetate concentrations and, consequently, reinforce UASB/EGSB prospects for the treatment of low strength soluble wastewaters. On such wastewaters, it seems advisable to use a fluid velocity high enough to reduce external mass transfer resistance and the occurrence of dead zones. At the same time, minimal liquid recirculation ratio should be sought, as the plug flow pattern could be advantageous for maintaining a higher substrate level, which favours the system performance.

Finally, this study also shows that elementary models of RTD, kinetics and mass transfer are a helpful approach for the performance characterisation of UASB/EGSB reactors. Models with few parameters and appropriate rate data may provide, in certain troubleshooting situations, the information needed for process engineering analysis. Such analysis could be also used to complement reactor design.

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NOMENCLATURE

Ac - Acetic acid concentration (M.L⁻¹)
COD - Chemical Oxygen Demand (M.L⁻¹)
E - Efficiency of acetate removal or fraction of acetate removed (dimensionless)
E(t) - Normalised residence time distribution function (dimensionless)
dp - Granules diameter (L)
k' - Apparent order 1/2 rate coefficient per reactor volume (M¹/².L⁻¹/².T⁻¹)
k'' - Apparent order 1/2 rate coefficient per superficial granules area (M¹/².L⁻¹/².T⁻¹)
Re(p), Re(d) - Reynolds number, based on particle or reactor diameter (dimensionless)
r_s - Acetate removal rate per mass of volatile solids (Mₚ.M_s⁻¹.T⁻¹)
t₀ - Time divided by mean residence time (dimensionless)
VFA - Volatile Fatty Acids
VS - volatile solids (Mₚ.L⁻³)

Subscripts
r - reactor
s - substrate
x - biomass
in, out - at inlet or outlet of the reactor

REFERENCES


