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# Effect of mass transfer in dry anaerobic digestion systems

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## Abstract

The effect of the total solid content on anaerobic digestion performance was experimentally investigated (TS ranging from 10 to 35%). An inhibition of methanogenic step was observed for high solid content (>30% TS). Anaerobic Digestion Model No. 1 (ADM1) was then used to describe the experimental data and the effects of hydrolysis and liquid/gas mass transfer (*k<sub>la</sub>*) were more particularly investigated. From the model, it was identified that a limited overall mass transfer can clearly affect the methane production and can result in a considerably lower cumulative methane production. Based on this observation, an additional experiment was conducted in order to confirm the key role of liquid/gas mass transfer on the inhibition of the methanogenesis step for high-solid contents.

## Keywords

Dry anaerobic digestion, limitation, mass transfer, *k<sub>la</sub>*

## INTRODUCTION

Dry digestion technologies (Total Solid content TS > 20%), also called “high-solids” or solid-state processes are attractive because of the reduction of water addition and, consequently, the minimisation of the digester size and transport costs. However, difficulties in operating dry anaerobic digestion at both laboratory and industrial scales are related to the high concentration of total solids. For example, Forster-Carneiro *et al.* (2008) showed better performance of anaerobic reactors operated at 20% TS compared to 25% and 30% TS. It was suggested that a high TS content could reduce substrate degradation and, therefore, biogas production (Fernandez *et al.*, 2008). In addition, a high TS content also affects the physical properties of the digested solid waste. For dry AD systems, the high solid content strongly affects the rheological behavior of the digestates (Garcia-Bernet *et al.*, 2011). Digested media are visco-elastic materials characterized by high yield stress levels, and yield stress increases with TS content according to an exponential law. Under unmixed conditions, transport is likely governed by diffusion processes, which are strongly related to the porosity of the media and, thus, to the water content. Therefore, the diffusive transport resistance of soluble compounds (substrate or by-products) may have a strong influence on anaerobic digestion performance in dry systems.

The aim of this study is to assess the impact of mass transfer of soluble and solubilized compounds on anaerobic digestion. First the effect of the TS content on anaerobic digestion performance was experimentally investigated (TS ranging from 10 to 35%). Anaerobic Digestion Model No. 1 (ADM1) was then used to describe the experimental data and the effects of hydrolysis step and liquid/gas mass transfer were more particularly investigated. Based on this observation, an additional experiment was conducted in order to clarify the role of the liquid/gas mass transfer on the inhibition of the methanogenesis step for high TS contents.

## MATERIAL AND METHODS

### Batch preparation and operating conditions

A compact cardboard with a density of 1.42 kg m<sup>-3</sup> was used as a substrate since paper and cardboard represents usually one of the largest proportion of organic compounds in municipal solid waste. The cardboard was shredded and sieved at 2 mm. Basic experiments were carried out in 600

mL batch flasks with a working volume of 100 mL. A mixture of cardboard, water, inoculum and oligo-elements was prepared to reach six TS contents from “wet” to “dry” anaerobic conditions: TS = 10, 15, 20, 25, 30, 35 %. A substrate to biomass ratio  $S/X$  of 20 (w/w) was used to limit the influence of the inoculum composition on the degradation,  $S$  and  $X$  representing the initial TS contents of the substrate and the biomass, respectively. The inoculum corresponded to a leachate of pressed MSW digestate sampled in an industrial plant treating MSW. One mL of an oligo-element solution was added to the mixture. The experiments were run over 298 days under mesophilic conditions (35°C) without mixing. Each TS condition was tested in four replicates. Biogas production was measured by the water displacement method and its composition was determined using a gas chromatograph (Varian  $\mu$ GC-CP4900). Volatile fatty acids (VFA) and pH were analyzed at the end of the batch test.

### **ADM1 model**

The Anaerobic Digestion Model No. 1 (Batstone et al., 2002) was used to describe the experimental data. The characterization of the shredded cardboard with respect to ADM1 state variables, in particular carbohydrates and particulate inerts, was based on the results of the Van Soest fractionation and the degradable fraction of fibers (hemicellulose and cellulose). The model was calibrated to the experimental data of the batch experiments at 10% TS using a trial and error approach. Compared to the standard parameters for the anaerobic digestion of sludge, two main parameters were modified in order to account for the degradation of the specific substrate (i. e., cardboard) at high total solids contents: the first-order hydrolysis rate constant for carbohydrates ( $k_{\text{hyd},X_{\text{ch}}}$ ) was, thus, reduced from its default value to  $0.01 \text{ d}^{-1}$  (Qu et al., 2009); the volumetric liquid/gas mass transfer coefficient was reduced to  $0.5 \text{ d}^{-1}$  to account for a reduced gas transfer due to the pasty texture of the substrate-biomass mixture. Additional information for both the experimental conditions and the model structure can be found in Abbassi-Guendouz et al. (2012).

## **RESULTS AND DISCUSSION**

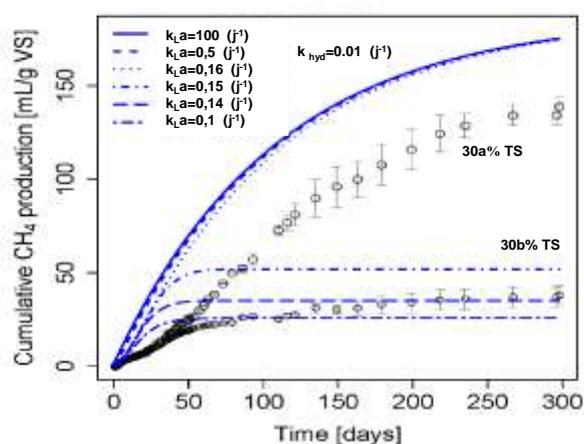
### **AD performance vs. TS content**

The cumulated methane production monitored throughout the experiment and the VFA concentration measured at day 298. The methanogenesis performances were closely related to the TS concentrations. Higher water content significantly enhanced methane production. The average cumulated methane production decreased from 183 to 168  $\text{mL.gVS}^{-1}$  when TS increased from 10% to 25%. Two distinct behaviour patterns were observed at 30% TS. Two replicates (group 30a%) showed performances similar to the 25% TS tests, with a methane yield of 142  $\text{mL.gVS}^{-1}$ . The two remaining replicates (group 30b%) were similar to the 35% TS tests with a lower methane yield of 28  $\text{mL.gTS1}$ , whereas at 35% TS the methane yield was 24  $\text{mL.gVS}^{-1}$ . For all the 30b and 35% replicates, high VFA accumulation (between 29  $\text{g.L}^{-1}$  and 36  $\text{g.L}^{-1}$  of water in the digestate) was observed. Acetate was the main dominant VFA, followed by propionate. Batch tests between 10% to 30a% TS presented effective methanogenic activity and their pH remained neutral ( $\text{pH } 7.4 \pm 0.3$ ). When AD was rapidly inhibited (30b and 35% TS), pH values were more acid due to VFA accumulation ( $\text{pH } 5.9 \pm 0.1$ ).

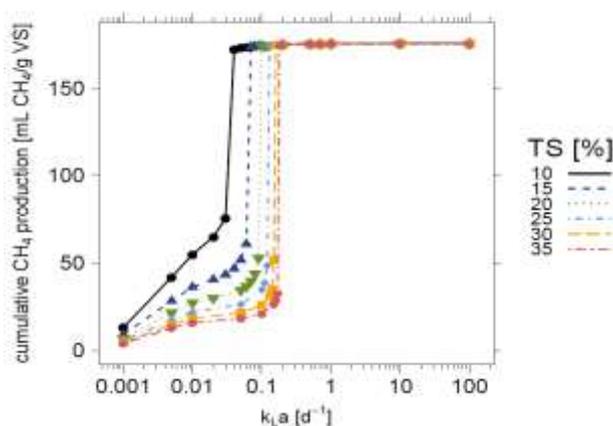
The difference in the behavior between the four replicates at 30% TS suggested that 30% TS could be considered as a threshold concentration for an inhibitory effect in high solids anaerobic digestion. This threshold could correspond to an inhibition of anaerobic digestion at high solids content due to a higher accumulation of metabolic by-products, such as volatile fatty acids. Further investigations were thus realized to better understand the origin of the limitation of anaerobic degradation at high solid contents.

## ADM1 model

For each TS concentration, the first-order hydrolysis rate constant for carbohydrates was calibrated for batch experiments that were not inhibited. The hydrolysis rate constants obtained for TS concentrations of 10% and 15% were similar ( $0.01 \text{ d}^{-1}$ ). Then, the hydrolysis rate constants linearly decreased with increasing TS concentration for  $15\% \leq \text{TS} \leq 30\%$ . This finding is in agreement with results presented by Pommier *et al.* (2007), who observed a strong impact of the moisture content on kinetic rates and maximum methane production in solid waste anaerobic digestion. Extrapolation of the hydrolysis rate constant for a TS concentration of 35% gave a value of  $0.00275 \text{ d}^{-1}$ . With this hydrolysis rate constant, the ADM1 predicted a total cumulative methane production of  $107 \text{ mL.gVS}^{-1}$  and no VFA accumulation for  $\text{TS} = 35\%$ . The total cumulative methane production predicted by the model was 4.5 times higher than that observed in the experiment ( $23.5 \text{ mL.gVS}^{-1}$ ). Thus, a further decrease in the hydrolysis rate constant could not explain why methane production stalled at a total solids concentration of 35%. As a consequence, decreasing hydrolysis rate constant cannot explain the considerable VFA accumulation, reactor acidification, and limited methane production observed in two batch experiments at 30% TS and all batch experiments at 35% TS.



**Figure 1.** Effect of  $k_{La}$  of the production of  $\text{CH}_4$  at 30% TS.



**Figure 2.** Influence of  $k_{La}$  on AD performance for different TS content.

The first-order hydrolysis rate constant for carbohydrates was kept constant at  $0.01 \text{ d}^{-1}$  for all simulations to focus on the effect of  $k_{La}$ . The overall mass transfer coefficient  $k_{La}$  was, therefore, decreased to match measured and simulated total cumulative methane production for the two inhibited batch experiments at 30% TS (Figure 1). Reducing the value of  $k_{La}$  from  $100 \text{ d}^{-1}$  to  $0.10 \text{ d}^{-1}$  resulted in an adequate representation of the final cumulative methane production for the two inhibited experiments. In simulations, methane production at 30% TS content dropped from  $174.5 \text{ mL.gVS}^{-1}$  to  $51.9 \text{ mL.gVS}^{-1}$  when the mass transfer coefficient was only reduced from  $0.16 \text{ d}^{-1}$  to  $0.15 \text{ d}^{-1}$ . A small variation of  $k_{La}$  in this range of magnitude can thus explain the shift observed experimentally at 30% TS. The different behaviors in the batch experiments at 30% TS content might be related to minor inaccuracies in the preparation of the bottles.

The influence of the overall mass transfer coefficient  $k_{La}$  was evaluated by varying its value between  $0.001 \text{ d}^{-1}$  and  $100 \text{ d}^{-1}$  for each studied TS concentration (Figure 2). A sudden and step decrease in the cumulative methane production can be observed for  $k_{La}$  values between  $0.04 \text{ d}^{-1}$  and  $0.18 \text{ d}^{-1}$  depending on the TS content. Above a critical  $k_{La}$  value, methane production was not limited by overall mass transfer and the cumulative methane production remained almost constant independently of the  $k_{La}$  value. Below the critical  $k_{La}$  value, a limited overall mass transfer clearly affected methane production and resulted in considerably lower cumulative methane production. Methane production was more sensitive to overall mass transfer limitations at high TS

concentrations as the methane production dropped already at higher *kla* values (at 0.18 d<sup>-1</sup> for 35% TS compared to 0.04 d<sup>-1</sup> for 10% TS). According to model results, *kla* limitation resulted in an accumulation of inorganic carbon (CO<sub>2</sub>), dissolved methane, and dissolved hydrogen, and can have an inhibitory effect on methanogenesis.

### **The overall mass transfer coefficient *kla*: a key parameter?**

In order to verify experimentally the importance of *kla* coefficient on the limitation of anaerobic digestion, a second set of experiment were performed. *kla* coefficient is composed of two terms: *kl*, the mass transfer coefficient, and *a*, the specific surface area of the interface. In a cylindrical flask, *a* corresponds to the ratio of the surface of the flask over the volume of the digestion medium. Two different flasks of 0.6 and 5 L were selected and were filled with 0.4 L of medium to be digested at 25% TS (triplicates). These conditions correspond to a specific surface area *a* of 12.6 and 50.3 m<sup>2</sup>.m<sup>-3</sup>, respectively. After 171 days of experiment, the methane yield was 71.4 mL.gvs<sup>-1</sup> for the reactor having a high surface *a* of 50.3 m<sup>2</sup>.m<sup>-3</sup>, whereas it was 6 times lower (12.4 mL.gvs<sup>-1</sup>) for the low surface *a* reactor. In the last case, methanogenic step was rapidly inhibited (50 days) and an accumulation of VFA was identified. As a consequence, specific surface area *a*, and thus the overall mass transfer coefficient *kla* of gas solubilized compounds must have be a key parameter for operating AD at high solid content.

## **CONCLUSION**

Based on both experimental observations and ADM1 simulations, it was shown that physical limitation related to liquid/gas mass transfer can affect the global anaerobic digestion performance for high solid content. *kla* limitation can result in an accumulation of dissolved carbon dioxide (and thus on pH) and hydrogen and can have an inhibitory effect on methanogenesis. Further investigations need to be done to understand these mechanisms of inhibition.

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