MODELLING FREE OXYGEN EFFECTS IN ANAEROBIC DIGESTION

Deshai Botheju, Bernt Lie and Rune Bakke
Telemark University College, Norway

Corresponding Author: Deshai Botheju, Faculty of Technology, Telemark University College
Kjølnes Ring 56, 3901, P.O.Box 203, Porsgrunn, Norway; Deshai.Botheju@hit.no

Abstract: Interactions of free oxygen in bio-gasification is a sparsely studied area, apart from the common argument of oxygen being toxic and inhibitory for anaerobic micro-cultures. Recent research reveal, however, increased solubilisation of organic matter in the presence of some free oxygen in anaerobic digestion. This study analyses these counterbalancing phenomena with a mathematical modelling approach using the widely accepted biochemical model ADM 1. Aerobic oxidation of soluble carbon and inhibition of obligatory anaerobic organisms are modelled using standard saturation type kinetics. Biomass dependent first order hydrolysis kinetics is used to relate the increased hydrolysis rate with oxygen induced increase in biomass growth. The amended model, ADM 1-Ox (oxygen), has 25 components and 22 biochemical processes, presented in matrix form. Computer aided simulation tool AQUASIM 2.1 is used to simulate the developed model. Simulations are in accordance with common process observations. Low oxygen loading conditions, such as by oxygenated influent streams do not really induce significant effects in anaerobic digesters. The simulations indicate that this is primarily due to the rapid oxygen consuming ability of facultative acidogenic organisms. Free oxygen level is thereby maintained too low to cause inhibition of methanogenesis. Further model improvements and verifications are suggested to make the ADM 1-Ox a more precise tool to analyse overall oxygen effects in anaerobic digestion.
Keywords: anaerobic digestion, modelling, oxygen, simulation

1 Introduction

Conventionally the anaerobic digestion (AD) process should occur in a strict anaerobic environment with no free oxygen available. It is however not realistic to avoid all supply of free oxygen into anaerobic digester systems, and hence they can be exposed to considerable free oxygen loads. Such aerobic invasions can deteriorate the performance of digestion systems, but experience shows that many digesters are capable of maintaining high performance with significant aerobic loads [2], [4], [5], [9], [10], [11], [13]. Improved performances under mild aerobic conditions in AD are also observed [8], [12]. This is a first attempt to explain the dynamics of free oxygen in AD with a comprehensive mathematical modeling approach. The aim is to develop a sound model basis for analyzing these aerobic-anaerobic interactions by exploiting the latest knowledge base on biochemical reactions related to aerobic and anaerobic wastewater treatment.

2 Oxygen Effects – Theoretical and Experimental Basis

Anaerobic digestion is the totality of the collective interaction of at least three main microbial groups which are known as acidogens, acetogens and methanogens. Acidogenic organisms are the commonly known fermentative organisms which can ferment simple organic substrates in the absence of oxygen. The vast majority of this group of organisms are, however, facultative organisms, implying that they can also use oxygen as electron acceptor. They tend to prefer oxygen and do aerobic respiration whenever oxygen is available, as it is energetically more favourable. When oxygen level is sufficiently low they may switch back to fermentation for their energy needs. The consumption of oxygen and readily available carbon sources, growth rates and conversion into carbon dioxide and other products are well studied and standard kinetic and stoichiometric parameters are available [6],[7].

Macromolecular complex organic matter such as carbohydrates, proteins and fatty acids (the three main categories commonly encounter in AD) must be broken down into smaller soluble molecules to be consumed by acidogenic organisms. This process is commonly known as hydrolysis and is carried out by acidogenic micro-organisms using their extra cellular enzymes. Hydrolysis can occur under both aerobic and anaerobic conditions. Hydrolysis rates are observed to be significantly higher under aerobic conditions, probably due to higher production of enzymes or higher diversity in enzymes produced [1].

Acetogenic and methanogenic organisms are responsible for the final conversion of acidogenesis products into methane and carbon dioxide. These two groups of organisms are obligatory anaerobes and hence free oxygen can inhibit their functioning and can even lead to rapid cell lysis.

An experimental series testing oxygen effects in AD [8] found that oxygen can enhance the hydrolysis stage, while higher oxygen levels cause more of the available soluble carbon to be oxidized into carbon dioxide

(catalyzed by facultative acidogens), reducing the methane potential of the system. This may suggest that an optimized level of oxygen can enhance the digester performance with minimal detrimental effects.

The model development described below is based on the above mentioned main facts and findings regarding oxygen effects in anaerobic digestion.

3 Model development

The model is developed using the generally accepted anaerobic digestion model ADM 1 structure [1] which is developed by the Mathematical Modelling Task Group of the International Water Association (IWA). The ADM 1 contains 19 processes and 12 soluble components and 12 particulate components. The ADM 1-Ox extension proposed includes the incorporation of oxygen as one extra soluble component and 3 additional aerobic uptake processes.

3.1 Stoichiometric matrix and rate equations

The stoichiometric matrix for soluble components of the proposed ADM1-Ox model is shown in Table 1. The new processes j8, j9 and j10 represent the aerobic uptake of monosaccharides, amino acids and long chain fatty acids (LCFA), respectively. Note that ADM1-Ox does not introduce any new microbial groups in addition to the 7 groups already present in ADM 1. All three aerobic uptake processes are associated with the existing three acidogenic groups namely monosaccharide degraders (X_{su}), amino acid degraders (X_{aa}) and LCFA degraders (X_{fa}). It has been found that small aeration effects induce negligible impact on the phylogenetic diversity of anaerobic digesters [15]. Dissolved oxygen concentration S_{O2} is introduced as the 13th soluble component. The selected unit for oxygen in the model is kg O_2/m^3. The aerobic uptake rates are described using saturation type (Monod) kinetic equations shown in Eq. 1, 2 and 3. Other biochemical rate expressions in ADM 1-Ox also utilizes Monod type Kinetics.

$$r_{aer\_su} = k_{m,su} \frac{S_{su}}{K_{s,su,aer} + S_{su}} \frac{S_{o2}}{K_{o2} + S_{o2}} X_{su} I_1 \qquad (1)$$

$$r_{aer\_aa} = k_{m,aa} \frac{S_{aa}}{K_{s,aa,aer} + S_{aa}} \frac{S_{o2}}{K_{o2} + S_{o2}} X_{aa} I_1 \qquad (2)$$

$$r_{aer\_fa} = k_{m,fa} \frac{S_{fa}}{K_{s,fa,aer} + S_{fa}} \frac{S_{o2}}{K_{o2} + S_{o2}} X_{fa} I_2 \qquad (3)$$

Integrated inhibition term I_1 includes 2 inhibition type terms used in ADM 1 for describing microbial inhibition due to extreme pH conditions and limitation of soluble inorganic nitrogen. Inhibition term I_2 in Eq. 3 is the resultant of multiplying I_1 with one more inhibition term representing the hydrogen inhibition of LCFA degrading organisms. An additional inhibition term, I_{O2}, is introduced in ADM 1-Ox to account for oxygen inhibition effects on the strictly anaerobic acetogens and methanogens. A generally accepted non-competitive type inhibition function was used as a gradual oxygen switch (Eq. 4). The same oxygen inhibition function was also used to account for negative effects of oxygen on fermentation rate / acidogenesis.

$$I_{O2} = \frac{K_{O2}}{K_{O2} + S} \qquad (4)$$

3.2 Oxygen Stoichiometry

Stoichiometric coefficients for oxygen under aerobic uptake processes were approximated using representative chemical formula for 3 basic substrates (carbohydrates – $C_{10}H_{18}O_9$; lipids – $C_8H_6O_2$; protein – $C_{14}H_{12}O_7N_2$) [7]. Three new yield coefficients were introduced to represent the additional biomass growth under oxygen respiration. Then the total yields of three acidogenic biomass groups are the additions of anaerobic and aerobic yields (stoichiometric matrix for the particulate components is not shown here). Aerobic growth of 3 acidogenic groups lead to additional inorganic nitrogen assimilation and is taken into account by the products of their aerobic yields and biomass nitrogen content. Different to the fermentation processes which can produce multiple products, aerobic uptake results in oxidation of substrates into the single product of carbon dioxide. Hence the averaged carbon content values (kmol C/kg COD) of the three substrate groups (C_1 = 0.03125 for sugars, C_2= 0.030 for amino acids and C_3 = 0.0217 for LCFAs) can directly be used together with respective aerobic yield coefficients as the stoichiometric coefficients under inorganic carbon (IC) balance (component i10, Table 1).

Table 1: Stoichiometric matrix of the **ADM1-Ox** Model for soluble components. The newly included modifications are shown highlighted.

Components , i	i1	i2	i3	i4	i5	i6	i7	i8	i9	i10	i11	i12	i13	Rate (ρ_j) [kg cod/m^3.d]
Processes, j	S_{su}	S_{aa}	S_{fa}	S_{va}	S_{bu}	S_{pro}	S_{ac}	S_{h2}	S_{ch4}	S_{IC}	S_{IN}	S_{O2}	S_I	
j1.Disintegration													$F_{sI,xc}$	$k_{dis}X_c$
j2. Hydrolysis of Carbohydrates	1													$k_{hyd,ch}X_{ch}$
j3. Hydro. proteins		1												$k_{hyd,pr}X_{pr}$
j4. Hydro. lipids	1- $F_{fa,li}$		$F_{fa,li}$											$k_{hyd,li}X_{li}$
j5. Uptake of sugars	-1				(1-Y_{su})$F_{bu,su}$	(1-Y_{su})$F_{pro,su}$	(1-Y_{su})$F_{fa,su}$	(1-Y_{su})$F_{h2,su}$		-$\Sigma c_i v_{i,5}$ i=1-9, 11-25	-$Y_{su}N_{bac}$			$k_{m,su}\frac{S_{su}}{K_{s,su}+S_{su}}X_{su}I_1I_{O2}$
j6. Uptake of amino acids		-1		(1-Y_{aa})$F_{va,aa}$	(1-Y_{aa})$F_{bu,aa}$	(1-Y_{aa})$F_{pro,aa}$	(1-Y_{aa})$F_{ac,aa}$	(1-Y_{aa})$F_{h2,aa}$		-$\Sigma c_i v_{i,6}$ i=1-9, 11-25	N_{aa}-$Y_{aa}N_{bac}$			$k_{m,aa}\frac{S_{aa}}{K_{s,aa}+S_{aa}}X_{aa}I_1I_{O2}$
j7. Uptake of LCFA			-1				(1-Y_{fa})0.7	(1-Y_{fa})0.3		?	-$Y_{fa}N_{bac}$			$k_{m,fa}\frac{S_{fa}}{K_{s,fa}+S_{fa}}X_{fa}I_2I_{O2}$
j8. Aerobic uptake of sugar	-1									C_1(1-Y_{su_aer})	-$Y_{su,aer}N_{bac}$	-1.1		$k_{m,su}\frac{S_{su}}{K_{s,su,aer}+S_{su}}\frac{S_{o2}}{K_{o2}+S_{o2}}X_{su}I_1$
j9. Aerobic uptake of aminoacids		-1								C_2(1-Y_{aa_aer})	N_{aa}-$Y_{aa,aer}N_{bac}$	-1.2		$k_{m,aa}\frac{S_{aa}}{K_{s,aa,aer}+S_{aa}}\frac{S_{o2}}{K_{o2}+S_{o2}}X_{aa}I_1$
j10. Aerobic uptake of LCFA.			-1							C_3(1-Y_{fa_aer})	-$Y_{fa,aer}N_{bac}$	-2.03		$k_{m,fa}\frac{S_{fa}}{K_{s,fa,aer}+S_{fa}}\frac{S_{o2}}{K_{o2}+S_{o2}}X_{fa}I_2$
j11.Uptake of valerate				-1		(1-Y_{c4})0.54	(1-Y_{c4})0.31	(1-Y_{c4})0.15			-$Y_{c4}N_{bac}$			$k_{m,c4}\frac{S_{va}}{K_{s,c4}+S_{va}}X_{c4}\frac{1}{1+S_{bu}/S_{va}}I_2I_{O2}$
j12.Uptake of butyrate					-1		(1-Y_{c4})0.8	(1-Y_{c4})0.2			-$Y_{c4}N_{bac}$			$k_{m,c4}\frac{S_{bu}}{K_{s,bu}+S_{bu}}X_{c4}\frac{1}{1+S_{va}/S_{bu}}I_2I_{O2}$
j13.Uptake of propionate						-1	(1-Y_{pro})0.57	(1-Y_{pro})0.43		-$\Sigma c_i v_{i,13}$ i=1-9, 11-25	-$Y_{pro}N_{bac}$			$k_{m,pro}\frac{S_{pro}}{K_{s,pro}+S_{pro}}X_{pro}I_2I_{O2}$
j14.Uptake of acetate							-1		1-Y_{ac}	-$\Sigma c_i v_{i,14}$ i=1-9, 11-25	-$Y_{ac}N_{bac}$			$k_{m,ac}\frac{S_{ac}}{K_{s,ac}+S_{ac}}X_{ac}I_3I_{O2}$
j15.Uptake of hydrogen								-1	1-Y_{h2}	-$\Sigma c_i v_{i,15}$ i=1-9, 11-25	-$Y_{h2}N_{bac}$			$k_{m,h2}\frac{S_{h2}}{K_{s,h2}+S_{h2}}X_{h2}I_1I_{O2}$
j16 – j22 Biomass decay processes														$k_{dis}X_i$ i=18-24

3.3 Hydrolysis rates

Standard ADM 1 uses first order rate expressions of the form in Eq. 5 to represent hydrolysis, not taking the effect of acidogenic biomass concentration on hydrolysis into account. Oxygen is, however, expected to enhance hydrolysis through aerobic growth and higher biomass concentration, as observed in some experimental studies [8]. This effect but not this mechanism can be included by just increasing the hydrolysis rate constant (K_{hyd}). A more mechanistic hydrolysis model is chosen in ADM 1- Ox by using the modified first order rate expressions which include biomass concentration terms. The kinetic expressions for hydrolysis of carbohydrates (*ch*), proteins (*pr*) and lipids (*li*), in processes j2, j3 and j4, are modified accordingly (Eq. 6, 7 and 8). Note that the hydrolysis of *ch*, *pr* and *li* are catalysed by respective acidogenic biomass groups X_{su}, X_{aa} and X_{fa} which utilize the relevant hydrolysed products.

$$r_{hyd,x} = k_{hyd,x} X_x \tag{5}$$

$$r_{hyd,ch} = k_{hyd,ch} X_{ch} X_{su} \tag{6}$$

$$r_{hyd,pr} = k_{hyd,pr} X_{pr} X_{aa} \tag{7}$$

$$r_{hyd,li} = k_{hyd,li} X_{li} X_{fa} \tag{8}$$

3.4 Kinetic and stoichiometric parameters

The simulations carried out with ADM1-Ox uses a typical set of kinetic parameters suggested in standard ADM 1 model [1]. The few new oxygen related kinetic constants in ADM1-Ox are estimated or chosen by comparing mainly three sources: ADM 1 kinetics parameter list [1], ASM 2 kinetic parameter list [6] and Henze et al.[7]. Accordingly the three aerobic yield coefficients are given the values of $Y_{su\_aer} = 0.5$; $Y_{fa\_aer} = 0.3$; $Y_{aa\_aer} = 0.4$ (in kg COD biomass per kg COD substrate).

Saturation coefficients (K_s) under aerobic condition are set to be one fifth of the values used under anaerobic conditions in ADM 1. This is in agreement with the used K_s values in ASM 2 model under aerobic and anaerobic conditions. Therefore the used values are $K_{s\_aa\_aer} = 0.06$; $K_{s\_fa\_aer} = 0.08$; $K_{s\_su\_aer} = 0.1$ (in the units of kg COD/m^3).

A coarse calculation based on the formula, $K_m = \mu^{max}/ Y$ shows that the ratio of K_m values under aerobic and anaerobic (fermentative) conditions come closer to 0.9. Then by considering also the uncertainty of these parameters it is decided to use the same values under both conditions.

The oxygen inhibition parameter K_{O2} in Eq. 4 is given the same value as the half saturation constant for oxygen in aerobic uptake processes, as in the ASM 2 model [6].

The all three hydrolysis rate constants (K_{hyd}) are adjusted by one order of magnitude in order to compensate for the modification done to the hydrolysis rate equations.

4 Simulations and Discussion

The developed model ADM 1-Ox was implemented and simulated using the AQUASIM 2.1 software package [14]. The initial simulation results are satisfactory in the sense that simulations are similar to the behaviour observed and expected in anaerobic digesters where no or some free oxygen is introduced. Comparison of simulation results with and without oxygen interactions gives insight into the possible free oxygen effects in an anaerobic digester. It is expected that these initial model simulations would also give valuable directions in developing the model further and also planning validation experiments intended.

4.1 Oxygen Impacts

When comparing the cases of no oxygen in influent and 1 mg/L oxygen in influent, no difference at all is observed for most of the simulated parameters (including biomass concentrations, gas flow, methane concentration etc.). The free oxygen concentration in the reactor is kept below a very low value of 3.5 $x10^{-6}$ mg/L at steady state. The same fact is true for oxygen concentration of 7 mg/ L in the influent (nearly oxygen saturated). In this case the free oxygen concentration in the reactor at steady state is 2.5 x 10^{-5} mg/L , still a very low value. The simulation graphs for this scenario are shown in Figure 1(a)-(j). This influent oxygen is not sufficient to make significant changes in the reactor behaviour and the low free oxygen level cause insignificant inhibition. This explains the common field and laboratory observations ([2], [5], [9], [10], [11], [13]) of unhampered anaerobic digester performance regardless of oxygen loading with feed stream. The simulations

show that aerobic activity of facultative acidogens consuming oxygen lead to very low free oxygen concentrations, hence minimising any detrimental oxygen effects.

In order to introduce a higher oxygen load into the digester, an unrealistic oxygen input concentration of 500 mg/L is tested in simulations (this kind of an oxygen load can only be achieved through some other aeration means, like air injection or membrane diffusion directly inside the reactor). Some degree of oxygen inhibition of methanogenic and acetogenic organisms is exhibited in this case, but yet very low (around 0.99, where 1 means no inhibition). Free oxygen concentration in the reactor rose to the value of around 0.002 mg/L (simulation graphs not shown). Methane concentration is also slightly reduced.

Though it is logical to expect that oxygen in anaerobic digestion may lead to increased alkalinity through higher production of CO_2, the simulations performed here did not show any difference in alkalinity after the introduction of oxygen. This may be because increased CO_2 generation by aerobic uptake processes are compensated to a certain extent by the reduced acidogenic fermentation inhibited by oxygen (hence less CO_2 from acidogenic fermentation). Volatile fatty acids (VFA) and methane oxidation may also have to be included in the model to better simulate alkalinity. The current model only considers the oxidation of hydrolysis products.

4.2 Significance of Facultative Organisms

When the simulation for 1 mg /L influent oxygen concentration is repeated but after removing the three aerobic uptake processes of facultative organisms defined under ADM 1- Ox model (Figure 2 (a)-(j)), it is immediately clear that a high inhibition of acetogenic and methenogenic organisms occur (inhibition factor of approx. 0.2, where zero implies full inhibition). Growth rates of these organisms also reduced significantly and as a result, washout of biomass is observed. Free oxygen concentration in the reactor finally reaches the 1 mg/L influent value. Methane concentration starts decreasing with time. Also about three times higher VFA accumulation is observed compared to the case shown in Figure 1. Higher soluble concentrations of sugar and amino acids are noticed. Increased carbon dioxide content and decreased methane content with time can be seen. Total gas flow is also reduced. These observations are similar to the experimental observations made by Johansen and Bakke [8] at high oxygen loading conditions (with intermittent aeration of the digesters – 500 ml/d aeration in 500 ml batch reactors). This shows that even relatively low oxygen levels can cause dramatic detrimental changes in an anaerobic digester, but this situation will not occur under normal operating conditions due to the protective oxygen consuming activity of facultative organisms present in the reactor.

4.3 Further Improvements in the model

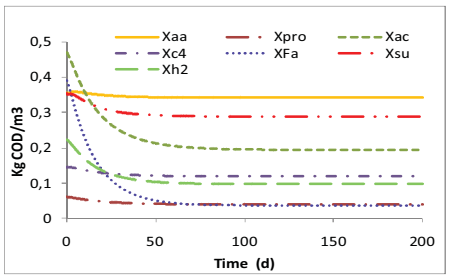
The current model can be improved in several ways to predict and analyse oxygen effects in anaerobic digestion in a more precise manner. Planned ADM 1-Ox improvements include testing different hydrolysis kinetics such as surface growth (Contois type) kinetics. It is expected that hydrolysis process is a surface phenomena rather than being volumetric [3]. Splitting hydrolysis in two categories, as aerobic and anaerobic, may also be required. This will facilitate using separate (and notably different) known kinetic parameters for aerobic and anaerobic hydrolysis.

In addition to the oxidation of hydrolysis products, the oxidation of fermentation products and acetogenesis/methanogenesis products (volatile fatty acids, acetates and also methane) in the presence of oxygen are also likely to occur, but may not play a significant role. These phenomena are not yet included because it would make the model complicated with a considerably higher number of processes.

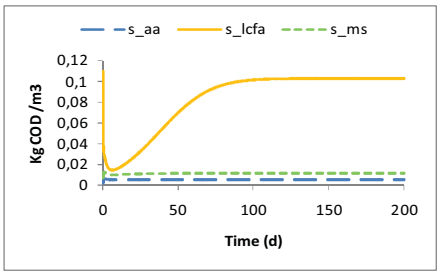
Acclimatization/adaptation of microbes to inhibiting toxic compounds is a known phenomena and the same is true for oxygen inhibition. One way of incorporating this effect in the model is to use a time dependent oxygen inhibition function. However such time dependant functions can be awkward to use in simulations. The alternative approach would be to use a separate differential equation to describe the inhibition constant K (e.g. K_{O2} in Eq. 4)

Estimating or finding good enough values for kinetic constants is a challenge in modelling/simulating biochemical reactions. Most of the time they have to be estimated using model associated parameter estimation tools using experimentally determined data. Further validation of the model based on experimental data should have to be done.

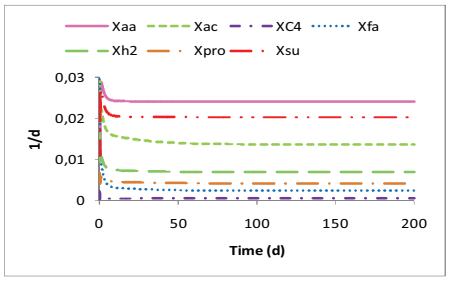
Improved model may be used to investigate the possible existence of an optimal oxygen load giving enhanced digestion with minimal detrimental effects.



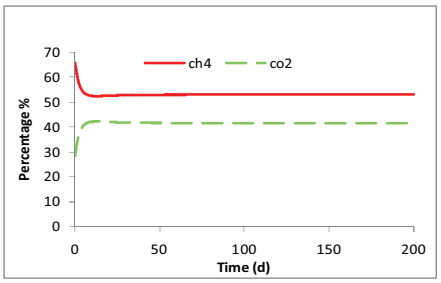
(a) Reactor biomass



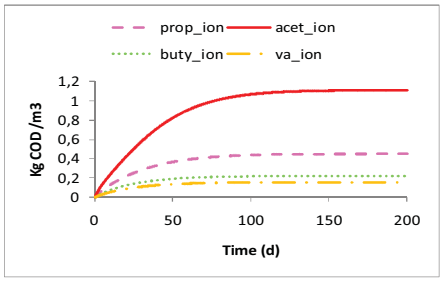
(f) Solubles



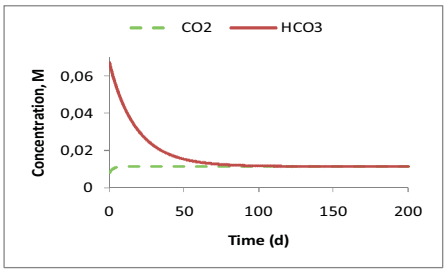
(b) Growth rates



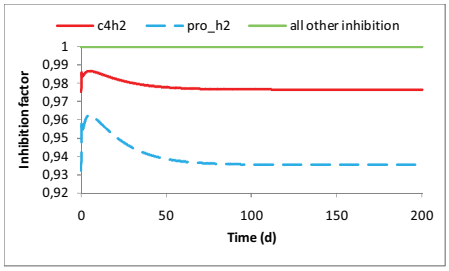
(g) Gas composition



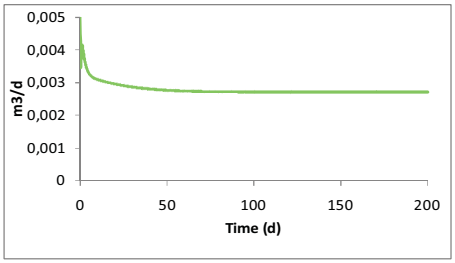
(c) Volatile fatty acids



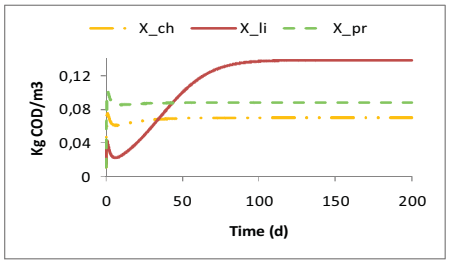
(h) Inorganic carbon



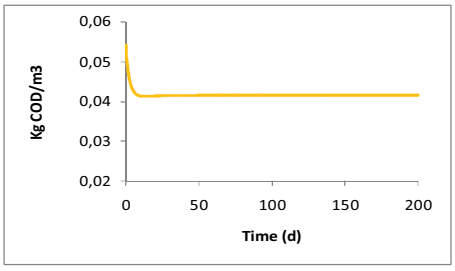
(d) Inhibition factors



(i) Total gas flow

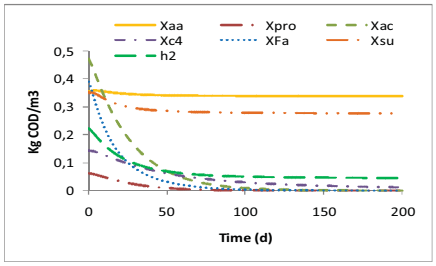


(e) Particulates

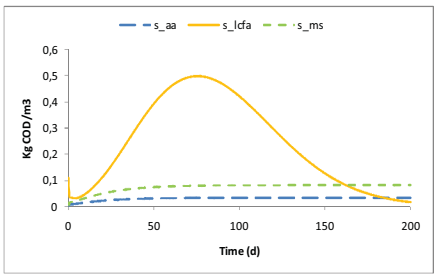


(j) CH_4 concentration

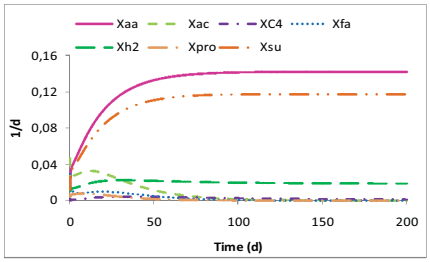
Fig. 1, (a)-(j): Simulation graphs for the influent DO level of 7 mg/L – ADM1-Ox model; The simulation results with zero oxygen influent show the similar behavior



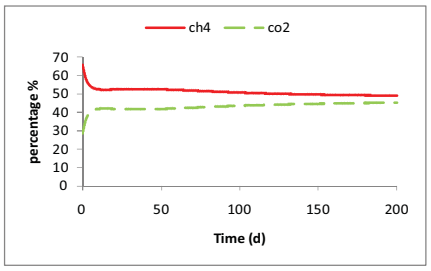
(a) Reactor biomass



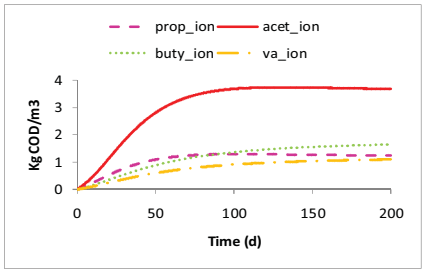
(f) Solubles



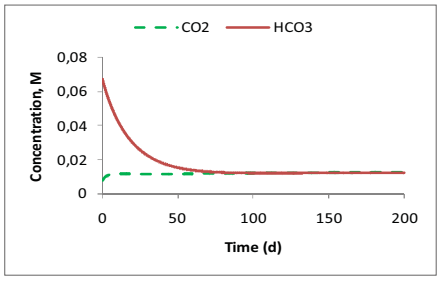
(b) Growth rates



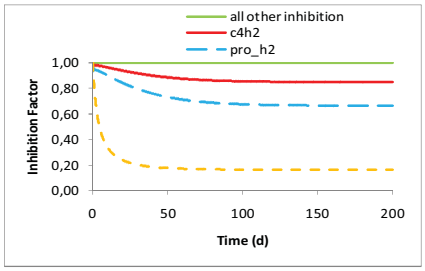
(g) Gas composition



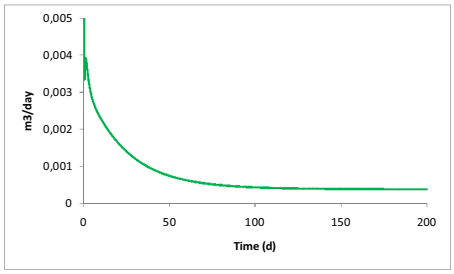
(c) Volatile fatty acids



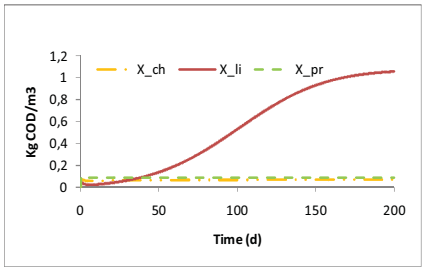
(h) Inorganic carbon



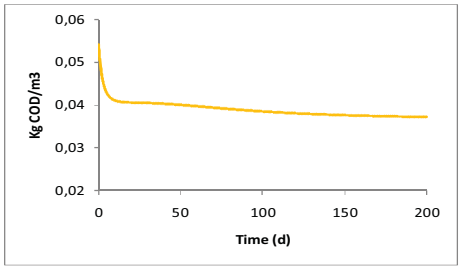
(d) Inhibition factors



(i) Total gas flow



(e) Particulates



(j) CH_4 concentration

Fig. 2, (a)-(j): Simulation graphs after the exclusion of aerobic uptake processes in ADM 1-Ox ; influent DO level of 1 mg/L

5 Conclusions

Effects of free oxygen in anaerobic digestion is modelled mathematically, by expanding the standard ADM 1 model to also account for some known biochemical interactions of oxygen in an anaerobic/methanogenic environment. The developed ADM 1-Ox model is successfully implemented and simulated in the AQUASIM 2.1 simulation tool.

Simulation results are in accordance with common observations and indicate that low oxygen loading conditions such as oxygenated influent streams can not cause detrimental effects in anaerobic digesters, primarily due to the rapid oxygen consuming ability of facultative acidogenic organisms.

Further model expansions and improvements are suggested in order to predict the overall effects of oxygen in more precise details. The initial simulations are found to be a sound basis for planning relevant experimental studies to validate and improve the model.

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7 Nomenclature

aa	- amino acids
ac /act /acet	- acetic, acetate
bu/buty	- butyric, butyrate
ch	- carbohydrates
d	- days (time unit)
DO	- dissolved oxygen
fa/Fa/lcfa	- fatty acids /LCFA
k_{hyd}	- first order hydrolysis rate constant
k_m	- uptake rate constant
k_s	- half saturation constant (Monod)
LCFA	- long chain fatty acids
li	- lipid
pr/pro	- protein
prop	- propionate, propionic
r	- reaction rate
r_{hyd}	- hydrolysis rate
s	- concentration of a soluble component
su/ms	- sugar/monosaccharides
va/val	- valeric, valerate
VFA	- volatile fatty acids
x, X	- concentration of a particulate component/biomass
Xaa	- amino acids degraders (biomass concentration)
Xac	- acetoclastic methanogens
Xc4	- butyrate and valerate degraders
Xfa	- LCFA degraders
Xh2	- hydrogenotrophic methanogens
Xprop	- propionate degraders
Xsu	- sugar degraders (monosaccharide degraders)

Other mathematical symbols used here bear the similar or closely resembling meanings as they do in the standard ADM 1 model [9].

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