

Appraisal of Certain Anaerobic Digestion Studies

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ABSTRACT

From time to time, discussions of various concepts related to anaerobic digestion have surfaced in the literature. With due recognition to the complexity of the pathways of anaerobic digestion and participation of a wide variety of microorganisms, there has been a substantial growth of studies to understand, model and improve the process performance. While doing so, the available literature has been utilised to have information pertaining to kinetic parameters, conversion factors, distribution factors, etc. Appraisal of various studies on anaerobic digestion indicated certain inconsistencies in the literature with regard to the use of kinetic parameters and conversion factors. With this in view, the present study focuses on the prevailing inconsistencies in anaerobic digestion studies. On various occasions, the concepts related to anaerobic digestion have been discussed in the literature. For the purpose of this study, the same has been excluded here to avoid repetitions. From the study, it is very evident that use of kinetic parameters from literature in anaerobic systems' modelling needs care and attention.

Key words- stoichiometric coefficients, anaerobic digestion, chemical oxygen demand (COD), kinetic parameters, microorganisms.

1. INTRODUCTION

The importance of anaerobic digestion process is well recognised in the literature. In the last few decades, several studies on anaerobic digestion have shed light on the understanding of the process performance for a variety of wastes. The role of experimental studies has been also remarkable as these provide useful information regarding various kinetic parameters which are essential to the modelling of anaerobic digestion systems. Associated with these developments in anaerobic digestion literature, there have been also inconsistencies which may often be a source of misunderstanding

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and confusion to the research workers, designers and operators of anaerobic digestion systems. From the review of the literature, it has been found that the inconsistencies are apparent in the adoption of the kinetic parameters, use of conversion parameters, and representation of stoichiometric relationships in the modelling of anaerobic digestion process. With this in back ground, the present study addresses some of these issues and provides necessary framework for the future studies on anaerobic digestion. For the purpose of illustrating the prevalent inconsistencies in the literature, the study is organised in the following sections.

2. KINETIC PARAMETERS

Kinetic parameters are essential for defining the rate of biochemical reactions. In the literature, a variety of rate expressions have been provided. Among these, the Monod kinetics is extensively studied and for this reason, only the kinetic parameters pertaining to the Monod kinetics have been considered. As per Monod kinetics, one needs to know a set of five parameters. The parameter k indicates the amount of substrate utilised per unit mass of biomass per day. K_s indicates the half saturation constant, which is the minimum concentration of limiting substrate at which the specific growth rate is 50% of the maximum specific growth rate of microorganisms. Y represents the biomass yield coefficient and is defined as the amount of biomass produced per unit mass of substrate utilized. μ_{max} is the maximum specific growth rate (i.e, maximum rate of change of biomass concentration per unit biomass concentration) and k_d is the rate of decay of microorganisms. Using these kinetic parameters, one can model the correspondence between the substrate and the biomass.

In case of anaerobic digestion, one may not always encounter a single substrate situation. In fact when a complex organic waste contains a variety of substrates, there may be a need to define kinetic parameters with respect to each contributing substrate. Alternatively, all these substrates can be represented in terms of any of these substrates. For example, in the case of anaerobic reactions utilising mixture of volatile fatty acids, i.e., acetic, propionic, and butyric acids etc., some investigators have represented such mixture of volatile acids in terms of a single substrate, i.e., acetic acid (Lawrence and McCarty, 1969). Although, such a representation may lead to much simplification in the modelling of anaerobic digestion systems, it may have its own limitations as will be discussed later in this paper.

Probably the first study to obtain the kinetic parameters for a variety of wastes, i.e., acetic, propionic and butyric acids can be credited to Lawrence and McCarty (1969). Table 1 shows the values of the some of the kinetic parameters documented by Lawrence and McCarty (1969). It is interesting to see that these coefficients have found applications in subsequent studies of Costello et al (1991a, 1991b) and Pavlostathis and Gomez (1991). It is pertinent here to describe the adoption of kinetic parameters of Lawrence and McCarty (1969) by subsequent investigators. In order to see the equivalence and comparison of kinetic constants and their evaluations, it is essential to critically analyse them.

Table 1. Comparison of kinetic constants as per Pavlostathis and Gomez (1991), adopted from Lawrence & McCarty, (1969) for acetic, propionic and butyric acids as substrate

Kinetic Constants ↓	Kinetic constants from Pavlostathis and Gomez (1991), adopted from Lawrence & McCarty(1969)]			Kinetic constants from Lawrence & McCarty (1969)#			Comparison
	Substrate →	Acetic Acid	Propionic acid	Butyric acid	Acetic acid	Propionic acid	Butyric Acid
k, mgCOD/mgVSS.d T=35°C T=30°C T=25°C	8.7 5.1 5.0	7.7 --- 7.8	8.1* --- ---	8.7 5.1 5.0	7.7 --- 7.8	8.3 --- ---	k values for butyric acid are different.
K _s , mgCOD/l T=35°C T=30°C T=25°C	165 356 930	60 --- 1145*	13 --- ---	165 356 930	60 --- 1143*	13 --- ---	Slight difference in K _s value for propionic acid at 25°C.
μ _{max} , d ⁻¹ T=35°C T=30°C T=25°C	0.357 0.275 0.250	0.313 --- 0.358	0.354 --- ---	0.348 ^c 0.275 0.25	0.3234 ^c --- 0.398	0.390 ^c --- ---	values are slightly different for acetic acid at 35°C, but large differences for propionic and butyric acids
Y, mgVSS / mgCOD T=35°C T=30°C T=25°C	0.041 0.054 0.05	0.042 --- 0.051	0.047 --- ---	0.04** 0.054 0.05	0.042** -- 0.051	0.047** --- ---	slight difference in the value for acetic acid at 35°C.
k _d , d ⁻¹ T=35°C T=30°C T=25°C	0.015 0.037 0.011	0.010 --- 0.040	0.027 --- ---	0.019 0.037 0.011	0.01 --- 0.04	0.027 --- ---	difference in the value for acetic acid at 35°C.

- k and K_s values are expressed as equivalent concentration of acetic acid, * - value is not equal to that mentioned by Lawrence and McCarty (1969); ** - unit, mg/mg; ^c - computed values, μ_{max} = k.Y

2.1 Maximum specific substrate utilization rate (k)

The maximum specific substrate utilization rate (k-values) are expressed by Lawrence and McCarty (1969) in terms of equivalent concentrations of acetic acids. The values of k at 35°C for acetic, propionic and butyric acids respectively are reported as 8.7, 7.7, and 8.3 mg COD to CH₄/mg-d, as given in Table 1. To convert these values in terms of mg COD/mg.d, one needs to make use of conversion factors of 1.067, 0.8 and 0.533 as per Table 2 for acetic, propionic and butyric acids respectively.

Table 2. Conversion factor for equivalent methane COD

Substrate	Reactions considered	g methane COD/ mole of substrate consumed	g methane COD/ g (as acetic acid) of substrate consumed
Acetic acid	$\text{CH}_3\text{COO}^- + 2\text{H}_2\text{O} \rightarrow \text{CH}_4 + \text{HCO}_3^-$	$1 \times 64 = 64$	$64/60 = 1.067$
Propionic acid	$\text{CH}_3\text{CH}_2\text{COO}^- + 1/2\text{H}_2\text{O} \rightarrow \text{CH}_3\text{COO}^- + 3/4 \text{CH}_4 + 1/4 \text{CO}_2$	$(3/4) \times 64 = 48$	$48/60 = 0.800$
Butyric acid	$\text{CH}_3\text{CH}_2\text{CH}_2\text{COO}^- + \text{HCO}_3^- \rightarrow 2\text{CH}_3\text{COO}^- + 1/2 \text{CH}_4 + 1/2 \text{CO}_2$	$(1/2) \times 64 = 32$	$32/60 = 0.533$

In the study of Costello et al. (1991b), each substrate has been assigned a rate kinetics and thus, a separate set of kinetic parameters. If one considers the reaction of propionic acid, the value of k to be used should have been in terms of mmol of propionic acid/mg VSS.d. Unfortunately, this is not the case in Costello et al. (1991b) who have used the k in terms of mmol of equivalent concentration of acetic acid/mg biomass.d as explained below in Table 3.

To have a proper understanding of the implications of converting different wastes into equivalent acetic acid concentration, one needs to consider the reactions of acetic, propionic and butyric acids as documented under by Lawrence and McCarty (1969), i.e.

Acetic acid



Propionic acid



Overall reaction

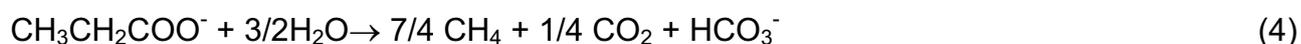


Table 3. Equivalent conversion and comparison of kinetic constants used by Costello et al. (1991) with those from Lawrence & McCarty, (1969) for acetic, propionic and butyric acids as substrate

Constants ↓	Kinetic constants as reported by Costello et al. (1991), adopted from Lawrence & McCarty (1969)			Kinetic constants value As per Lawrence & McCarty (1969) #			Equivalent conversion of values used by Costello et al. (1991) and comparison with those of Lawrence & McCarty (1969)		
Substrate →	Acetic Acid	Propionic acid	Butyric acid	Acetic Acid	Propionic acid	Butyric acid	Acetic Acid	Propionic acid	Butyric acid
k,	0.18 mmol/mg.d	0.16 mmol/mg.d	0.26 mmol/mg.d	8.1 mg/mg.d	9.6 mg/mg.d	15.6 mg/mg.d	0.18x60 = 10.8* mg/mg.d	0.16x60 = 9.6 mg/mg.d	0.26x60 = 15.6 mg/mg.d
K _s ,	2.57 mM	0.53 mM	0.083 mM	154 mg/l	32 mg/l	5 mg/l	2.57x60 = 154.2 mg/l	0.53x60 = 31.8 mg/l	0.083x60 = 4.98 mg/l
Y	2.5 mg/mmol	5.0 mg/mmol	7.5 mg/mmol	0.04** mg/mg	0.042** mg/mg	0.047** mg/mg	2.5/(60x1.066) = 0.039 mg/mg	5.0/(74x1.512)* = 0.045* mg/mg	7.5/(88x1.816) = 0.047 mg/mg
k _d	0.02 d ⁻¹	0.01 d ⁻¹	0.03 d ⁻¹	0.019 d ⁻¹	0.01 d ⁻¹	0.027 d ⁻¹	almost equal	equal	approx. equal

- k and K_s values are expressed as equivalent concentration of acetic acid

* - Value is not equal to that mentioned by Lawrence and McCarty (1969)

** - expressed as mg biological solids produced per mg COD converted to methane

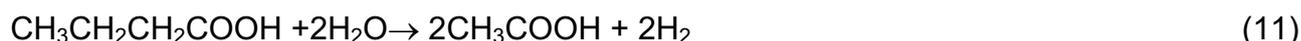
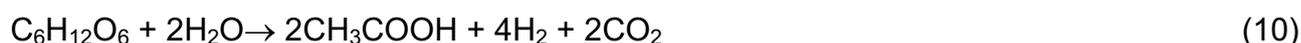
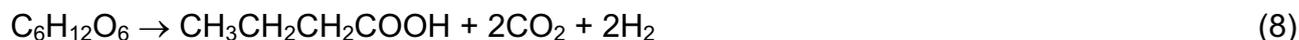
Butyric acid



Overall reaction



and the reactions for glucose degradation as mentioned by Denac et al. (1988), i.e.



It can be seen that in the case of acetic acids only CH₄ is produced while in the case of propionic acid, hydrogen is also produced, which may further contribute to CH₄ formation

as per Eq. (14). This contribution may be as high as 20-30% of the total methane production (Denac et al, 1988). Thus, equivalent representation of other acids (except acetic acid) may not be fully justified. It is interesting to see from the study of Lawrence and McCarty (1969) that ionic reactions do not represent the formation of hydrogen and hence, the H₂-based CH₄ production.

From Table 3, one can also note that in case of acetic acid, the value of k used by Costello et al. (1991b) as 0.18 mmol/mg.d in their model validation is incorrect as its conversion to equivalent acetic acid concentration works out to be 10.8 mg/mg.d which is different from the value of 8.1 mg/mg.d as reported by Lawrence and McCarty (1969).

The values of k mentioned by Pavlostathis and Gomez (1991) are illustrated in Table 1. Although these values are expressed in terms of mgCOD/mgVSS.d, these are truly the values of Lawrence and McCarty (1969) expressed in units of mgCOD to CH₄/mg.d except for butyric acid. Thus, adoption of such values might lead to the errors in computations/predictions in modelling of high rate anaerobic treatment systems due to incorrect use of original values of Lawrence and McCarty (1969).

2.2 Half Saturation constant (K_s)

Similar is the case with the K_s values which are expressed in equivalent acetic acid concentrations. These values can be used only when different volatile fatty acids are expressed in terms of equivalent acetic acid concentration. It is interesting to see that Costello et al (1991b) have not treated different substrates into equivalent acetic acids in their simulations of anaerobic digester's performance (Table 3). If one considers the propionic acid as substrate, the units of K_s must be expressed in terms of mass of propionic acids consumed per litre. Similar is the case in the study of Pavlostathis and Gomez (1991) as can be seen in Table 1.

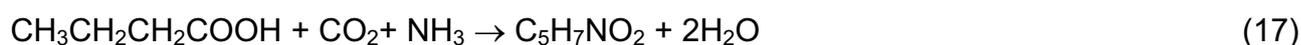
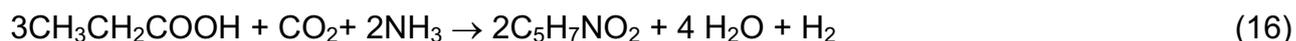
2.3 Maximum specific growth rate (μ_{max})

The parameter μ_{max} is generally expressed as (k.Y). However, one can see the incorrect evaluation of μ_{max} values of Pavlostathis and Gomez (1991), which are different than the computed values as marked with superscript (°) in Table 1.

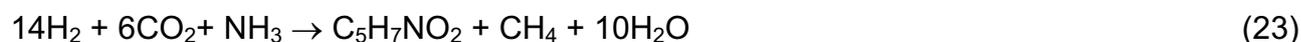
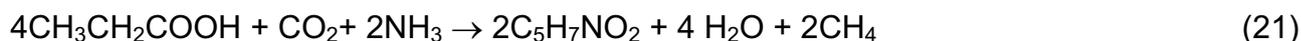
2.4 Biomass yield coefficient (Y)

The value of Y as reported by Costello et al. (1991b) presents another interesting scenario. Contrary to k and K_s values which were expressed in terms of equivalent acetic acid concentration, the biomass yield has been reported in terms of mmol of the actual substrate as can be seen from Table 3.

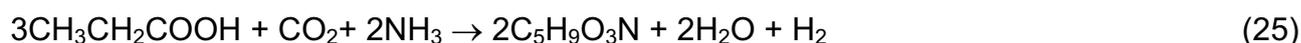
The relative magnitude of specific biomass yields in different volatile fatty acids also presents an interesting situation. Considerations of the following reactions can be used for having certain idea regarding the theoretical yield with respect to synthesis of biomass having cell composition as $C_5H_7NO_2$ or $C_5H_9O_3N$. Following Moletta et al. (1986) approach for synthesis of biomass $C_5H_7NO_2$ from acetic acid, one can have the following possible reactions:



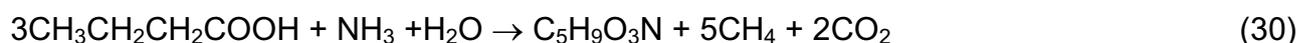
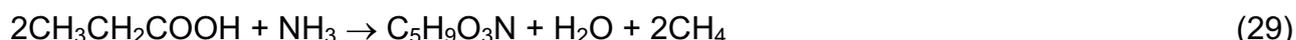
Considering the reactions of glucose degradation and Eqs. 8 to 14, one can write the following overall reactions for synthesis of biomass $C_5H_7NO_2$ from different acids including reaction (19), as given below:



Similarly for a biomass composition $C_5H_9O_3N$, the individual reactions in case of different acids can be written after Costello et al. (1991a) as under:



Considering the reactions of glucose degradation and reactions (15) to (19), one can write the following overall reactions for synthesis of biomass $C_5H_9O_3N$ from different acids, as given below:

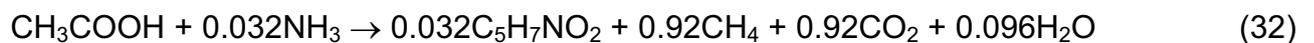


Using reactions (15) to (31), the theoretical yield can be computed. Denac et al. (1988) and Buffière et al. (1995) indicate that yield coefficient in case of butyrate, acetate and H_2

are all equal to 0.029, while in case of propionic acid the yield coefficient is 50% lower, i.e., 0.014. Such variability in yield coefficients is neither observed in Lawrence and McCarty, (1969) nor in the estimation of theoretical yields. Normally, one would expect that the theoretical value of biomass yield from a particular acid is of the same order for butyric and acetic acids in comparison to its experimental yields of acetic or butyric acids. Although such agreement is apparent for the values of biomass yield for different acids in Lawrence and McCarty (1969), the experimental value present another view regarding the relative importance of biomass yield among different acids and thus, one needs to be careful while using these biomass yield estimates.

3. CONVERSION FACTORS

A variety of conversion factors exist in the literature on anaerobic digestion. In the study, focus will be on the conversion factor of 1.3 g COD per g biomass as mentioned by Bryers, (1985), using biomass formula $C_5H_7NO_2$ as basis. If this conversion factor is used in the following reaction of Graf and Andrews (1971) as given below:



From the above reaction, one can see that 1 g acetic acid \equiv $(113 \times 0.032 / 60)$ g biomass + $0.92 \times 16 / 60$ g CH_4 . But as per Bryers (1985), the biomass with composition $C_5H_7NO_2$ has an equivalent COD of 1.3 g COD/ g biomass, Therefore, 1 g acetic acid \equiv $(0.032 \times 113 \times 1.3 / 60)$ g COD + $(16 \times 4 \times 0.92 / 60)$ g CH_4 -COD = 1.0596g COD \approx 1.06 g COD, which is contrary to the reported COD value of 1 g acetic acid as 1.067 g COD.

Similarly, by using a conversion factor of 1.3 in reaction (32), one obtains an equivalent acetic acid COD of 0.979 g COD/g acetic acid. This shows that the COD conversion factor of 1.3 g COD/ g biomass as mentioned by Bryers (1985), is inaccurate. Further, considering the following reactions from the literature (MetCalf and Eddy, 1997):



(1 g cells \equiv $160 / 113 = 1.4159 \approx 1.42$ g COD)



(1 g cells \equiv $160 / 131 = 1.2214 \approx 1.22$ g COD)

From reaction (33), one observes that the theoretically correct conversion factor for biomass $C_5H_7NO_2$ is 1.42 g COD/ g biomass. Use of this conversion factor also leads to correct estimates of 1g acetic acid COD equivalence as 1.067. Although, the average of

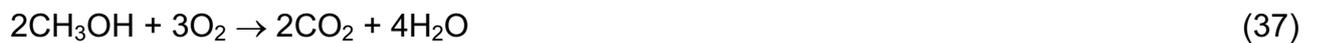
the two compositions of cells lead to 1.33 g COD per g cells, it may not be appropriate to use it arbitrarily in reactions. Further, the value of 1.42 g COD per g cell or 1.22 g COD per g cell shall be used depending upon the representative cell compositions as $C_5H_7NO_2$ or $C_5H_9O_3N$ in reactions under considerations. Thus, the value of 1.3 g COD per g biomass for cell composition $C_5H_7NO_2$ used by Bryers (1985) is questionable.

Similar is the case with the Bhatti et al. (1996), who have mentioned that 1 g TOC is equivalent to 1.4 litres of CH_4 and 1 g Methanol TOC as 2.67 g COD. Considering the conversion reaction of methanol to CH_4 as per Florencio et al. (1995),



From reaction (35), it can be seen that 128 g methanol (4 moles of methanol) \equiv 3 mol methane. As 1 mole methane equals 22.4 litres at standard temperature and pressure (STP), hence, 1g methanol equals $3 \times 22.4 / 128$ L $CH_4 = 0.525$ L CH_4 at STP. As 1 g methanol TOC = $1 \times 12 / 32$ g C/ g methanol = 0.375 g C/ g methanol, and 1 g Methanol TOC = $0.525 / 0.375 = 1.4$ L CH_4 at STP. In fact, 1 g TOC does not have any sense. From the values of Bhatti et al. (1995), it can be seen that 1 g TOC should have been mentioned as 1 g methanol TOC.

Similarly, if one considers the following reactions of acetic acid and methanol for the purpose of computing their COD values,



One can find from reaction (36) that 1 g acetic acid TOC = $1.066 / 0.4 = 2.67$ g COD. Here, a factor 0.4 appears in denominator because 1 mol acetic acid TOC = 2×12 g C and 1 g acetic acid TOC = $2 \times 12 / 60 = 0.4$ g C/g acetic acid. Similarly, from reaction (37), one can find that 1 g Methanol TOC = $1.5 / 0.375 = 4.0$ g COD (1 g Methanol = 1.5 gCOD). Thus, the value suggested by Bhatti et al. (1995) that 1 g methanol TOC = 2.67 g COD, appears incorrect.

4. YIELD COEFFICIENT OF CO_2

Determination of yield coefficients of CO_2 in various reactions of different acids also becomes relevant in modelling of CO_2 production in anaerobic digestion. As the COD of CO_2 is zero, Denac et al., (1988) represented the yield of CO_2 in unit of mole/g COD. Considering the case of acetic acid, one mol acetic acid leads to production of 1 mol of CO_2 ; thus yield is 1 mol CO_2 /1 mol acetic acid = $1 \text{ mol} / 64 \text{ g COD} = 1.5625 \times 10^{-2} \text{ mol/g}$

COD. In the same manner, the yield coefficients in case of propionic acid, H₂ and glucose can be obtained as 8.929×10^{-3} , 1.5625×10^{-3} , and 6.944×10^{-3} respectively in view of different reactions (Eqs. (12), (14) and (10)). Considering the mass balance equation for CO₂ production as given by Denac et al. (1988) in Eq. (15) of their research paper, one can find the incorrect use of some of these yield coefficients; particularly for H₂ and glucose, which has been taken as 1.563 and 9.945×10^{-3} respectively.

5. EQUIVALENT REPRESENTATION

With respect to the kinetic parameters, it was emphasized that different volatile acids can be represented in terms of equivalent acetic acid concentration. Table 4 presents the approach of Lawrence and McCarty, (1969), in which the equivalent representation of different volatile acids has been reported.

Table 4. Acetic acid equivalent conversion of propionic and butyric acid concentrations as mentioned by Lawrence & McCarty, (1969)

Volatile Fatty acids as substrate	Substrate feed concentration (reported value)		
	mg/ l	mg/l as acetic acid	mg COD/l
Propionic acid (T=35°C)	1925.0	$1925 \times 60/74 = 1560.8$ (1560)	$1925 \times 1.512 = 2910.6$ (2910)
Propionic acid (T=25°C)	3715.0	$3715 \times 60/74 = 3012.2$ (3010)	$3715 \times 1.512 = 5617.08$ (5620)
Butyric acid (T=35°C)	2280.0	$2280 \times 60/88 = 1554.54$ (1555)	$2280 \times 1.816 = 4140.48$ (4140)

Note : 1.512 is the COD conversion factor for propionic acid and 1.816 for butyric acid (Dinopoulou et al., 1988). Here the equivalent conversion has molar basis.

In fact, in the representation of Lawrence and McCarty, (1969), the emphasis has been on the equality of moles. Contrary to the conversion values reported in columns 3 and 4 of Table 4, one can see that the number of moles of acetic acid are same as the number of moles of propionic acid. However, this approach may not necessarily represent the equality of COD values. In fact, when the conversion is done, the reciprocal of molecular weight of the propionic to acetic acids, the resulting values tend to give a closer COD matching. Table 5 shows an alternative method for acetic acid equivalent conversion of propionic and butyric acid concentrations.

Table 5. An alternative method for acetic acid equivalent conversion of propionic and butyric acid concentrations as mentioned in Lawrence & McCarty, (1969)

Volatile acid as substrate	Substrate feed concentration (given value)				
	mg/ l	mgCOD /l as acetic acid (a)	mgCOD /l as given acid (b)	f = (b)/(a)	mg COD/l as given acid (a)x f
Propionic acid (T=35°C)	1925.0	$1925 \times 1.066 \times 74/60 = 2530.86$	$1925 \times 1.512 = 2910.6$	1.15	2910.5
Propionic acid (T=35°C)	3715.0	$3715 \times 1.066 \times 74/60 = 4884.23$	$3715 \times 1.512 = 5617.08$	1.15	5616.9
Propionic acid (T=25°C)	3715.0	$3715 \times 1.066 \times 74/60 = 4884.23$	$3715 \times 1.512 = 5617.08$	1.15	5616.9
Butyric acid (T=35°C)	2280.0	$2280 \times 1.066 \times 88/60 = 3564.70$	$2280 \times 1.816 = 4140.48$	$1.16 \approx 1.15$	4099.4

Note: 1.066, 1.512 and 1.816 are the COD conversion factors for acetic, propionic and butyric acids respectively (Dinopoulou et al., 1988).

The computations shown in Table 5 above show that by multiplying with a factor of 1.15, one can have the same COD representation in case of propionic as well as equivalent acetic acid concentration.

CONCLUSION

This paper considers an appraisal of few studies on kinetic parameters for anaerobic digestion and anaerobic systems modelling. The results indicated certain inconsistencies in the literature with regard to the use of kinetic parameters, and conversion factors. With this in view, the present study focuses on the prevailing inconsistencies in anaerobic digestion studies. Use of incorrect kinetic parameters is finding applications even in recent anaerobic system's modelling studies. Such inconsistencies in kinetic parameters will certainly lead to erroneous predictions in modelling and simulations of anaerobic digestion systems. It is believed that the points focussed in this study on inconsistencies prevailing in the literature, will prove useful in better modelling and simulations of anaerobic digestion systems with use of correct kinetic constants.

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