



Simulation of Kinetic Model for Anaerobic Digestion of Pharmaceutical Wastewater

Loveth N. Emembolu^{1*}, Joseph T. Nwabanne¹, Elijah C. Onu²
and Innocent O. Obiakor¹

¹Department of Chemical Engineering, Nnamdi Azikiwe University, P.M.B. 5025 Awka, Nigeria.

²Department of Chemical Engineering, Chukwuemeka Odumegwu Ojukwu University, P.M.B 02 Uli, Nigeria.

Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

This work presents the treatment of the pharmaceutical wastewater using anaerobic digestion was carried out in this work. The results obtained showed that most of the wastewater parameters were reduced to acceptable levels after the digestion. The kinetic data was well described by the first-order kinetic equation. The following kinetic parameters: maximum rate of substrate utilization (K), half velocity or saturation constant (K_S), endogenous decay coefficient (K_d), biomass or microbial growth yield (Y) and maximum specific microorganism growth rate (μ_{max}) were determined as 0.0283 day⁻¹, 26.911 mg/l, 0.0112 day⁻¹, 116 mg/mg and 0.0316 day⁻¹ respectively. This suggests that the anaerobic process would require inoculation. The model for the anaerobic digestion in a continuous flow reactor unit under homogeneous steady state was developed using the kinetic data obtained and the subsequent design table developed can be used in the actual plant design for the continuous process.

*Corresponding author: E-mail: ln.emembolu@unizik.edu.ng, lovethemembolu373@gmail.com;

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1. INTRODUCTION

Over the last century, continued population growth and industrialization has resulted in the degradation of various ecosystems on which human life relies on [1]. In African countries, increase in industrialization without same development of treatment facilities adequate for handling industrial wastes has led to the discharge partially treated or raw wastes into the surrounding water bodies [2]. Almost all partially treated raw waste discharged into these water bodies are harmful and so constitute environmental and health hazards for the population. These threaten the availability of clean water needed for drinking and life activities [3].

To combat this ever-growing burden on the aquatic environment, various government bodies have placed strict regulations on pollution discharge with a primary focus on waste [1]. There are many biological and physiochemical treatment methods of removing wastewater pollutants [4]. However, the best environmentally practicable option is the conversion of waste components to energy, fuels and other valuable products, as well as the recovery or salvage of existing materials [5].

Of all these waste treatment techniques, anaerobic digestion is one of the oldest and most commonly used method for treating sludge. This is due to its potential for the stabilization of large volume of sludge at low cost, it zero oxygen requirement, low biomass production, high destruction rate of pathogens and the generation of methane usually burnt for energy [6].

Anaerobic digestion is a microbial decomposition of organic matter into methane. Anaerobic digestion has the advantage of biogas production and can lead to efficient resource recovery and contribution to the conservation of non-renewable energy sources. The stability of the anaerobic microbial ecosystem is very dependent on methanogenic activity, this activity being characterized by slow growth rates of microorganisms and great sensitivity to inhibition processes and environmental conditions such as pH. For a stable digestion to proceed, it is vital that various biological conversions remain sufficiently coupled during the process to prevent the accumulation of intermediate compounds [7].

Pharmaceutical industries produce a wide range of products to be used for humans, animals and plants. In these industries, manufacturing is characterized by five main processes; fermentation, extraction, chemical synthesis, formulation and packaging. Each of these steps may generate waste in form of air emissions, liquid effluents and solid. Waste produced from pharmaceutical industries vary in composition and magnitude depending on various factors such as raw materials used, manufacturing process adopted, process modifications, specific demand of seasonal medicines, etc. Pharmaceutical waste water usually contain high BOD, COD, and electrical conductivity, TDS, TSS, sulphate and phosphate contents. All these constitute high inorganic/organic loading content of the relevant waste water treatment facilities. These inorganic/organic matter when released in excess affect aquatic life. For example, pharmaceutical waste water effluent with COD as high as 800mg/l, disrupts the endocrine system of aquatic life [8].

Anaerobic digestion (both the use of anaerobic fixed film reactor and filter) has reportedly been used to treat pharmaceutical waste water [9]. Various parameters influence the anaerobic digestion of wastewater, the most important of these being; the mass of inorganic substrate, amount of biomass, moisture content of effluent and the reaction temperature [10].

In order to understand the operations of the anaerobic digestion, and hence effectively design a reactor, kinetics is required. Kinetics helps to describe treatment systems and predict their performance using operational parameters [11]. Kinetic analysis results gotten can then be used to estimate treatment efficiency of full scale reactors [12].

A kinetic model of anaerobic digestion of municipal waste was developed using Monod's kinetics of biomass growth and substrate utilization by [13]. This was used to predict the time needed for a batch anaerobic process. A first-order kinetic model for gas generation of the same process was developed by [14]

This work not only involves the description/simulation of a kinetic model for the batch anaerobic digestion of pharmaceutical waste water but also a model for the continuous flow

anaerobic digestion. The effects of substrate and biomass concentration on the efficiency and influent substrate concentration are to be determined in order to get the Hydraulic Retention Time (HRT) and the Solid Retention Time (SRT).

2. MATERIALS AND METHODS

2.1 Wastewater Sample Collection

The pharmaceutical wastewater used for study was collected from AC Drugs Limited located at Chief Edward Nnaji Street, Abakpa Nike, Enugu state, Nigeria. The wastewater was stored in 20-litre air tight containers.

2.1.1 Experimental Procedure

Three metallic fixed dome digester, each with a 25 litre capacity were used for the experiments at the National Centre for Energy Research and Development, University of Nigeria, Nsukka, Nigeria.

18 litres of waste water was poured into each digester to allow for gas collection at the top of the digester. The contents of the digester were stirred thoroughly and then sealed with air tight lids for observation.

On a daily basis, digester content was stirred by carefully shaking the digester to ensure intimate contact of waste water with microbes responsible for converting waste water into biogas. During the digestion, wastewater temperature was recorded periodically.

The pH of the wastewater was taken after every five days to ensure stability of the wastewater slurry. During the retention period, some quantity of the wastewater was collected for COD, BOD₅ and TSS analysis.

Biogas production was measured by downward displacement of water in the measuring cylinder and recorded as the difference between the initial reading at the beginning of each day and the final reading at the end of that same day. This was measure periodically until gas production became negligible. Biogas produced was collected daily via a valve in the cone head of the digester. Microbial load of the waste water slurries were determined every five days, from the point of charging to the end of the retention period.

2.1.1.1 Batch Kinetic Study

The kinetics of the microbial process of anaerobic digestion may be divided into kinetics of growth and kinetics of food (or substrate) utilization [13]. The COD was determined using a time-average cell mass. The limited substrate consumption can be expressed as:

$$-ds/dt = k_1 S \quad (1)$$

Integrating equation (1) with respect to t from S to S₀ and from t to 0 give:

$$S = S_0 \exp(-k_1 t) \quad (2)$$

Where S₀ is the initial influent substrate concentration (mg/l), S is the effluent substrate concentration (mg/l), k₁ is the first order inactivation rate constant (l/day) and t is time (days).

Equation (2) describes an exponential profile of the substrate growth. Rearranging and taking natural logarithm;

$$\ln [S/S_0] = -k_1 t \quad (3)$$

This is a first-order reaction kinetics which is used to verify the kinetics of the process. When the plot of $-\ln(S/S_0)$ against t gives a straight line with appropriate regression coefficient, then the process is said to have followed the first-order kinetics model. The slope of the straight line will be used to evaluate the first-order inactivation rate constant (l/day).

The maximum rate of substrate utilization (K) is obtained by using the relationship between the rate of substrate utilization (U) and the effluent substrate concentration (S_e)

$$1/U = K_s/KS_e + 1/K \quad (4)$$

Where K_s is half-velocity constant (mg/l).

The linear plot of 1/U against 1/S_e is used to determine the constants K and K_s from the intercept and slope of the graph. These values indicate whether there is need for inoculation in the system.

The relation between the mean cell residence time (Θ) and the specific rate of substrate utilization (U) is given by;

$$1/\theta = YU - K_d \quad (5)$$

From the plot of $1/\theta$ against U , the biomass yield (mg/mg) Y and the endogenous decay coefficient (day^{-1}) K_d can be evaluated from the slope and intercept of the linear plot respectively.

The maximum specific rate of micro-organism growth μ_{\max} is calculated using;

$$K = \mu_{\max}/Y \quad (6)$$

Where Y is the biomass yield and K is the maximum rate of substrate utilization

2.1.1.2 Continuous process design model

The characteristics of continuous mode reactor are completely different because the substrate concentration approaches constant under steady state condition [10]. For continuous reactor under steady state, the rate of substrate utilization is:

$$-ds/dt = (S_0 - S)/\theta \quad (7)$$

Where θ is the hydraulic retention time (HRT).

The rate of increase of the biomass concentration X is modelled as a first-order process

$$dx/dt = \mu X \quad (8)$$

Where μ = specific growth rate (day^{-1})

Monod discovered in 1949 that the limiting substrate concentration, S is related to μ as follows

$$\mu = \mu_{\max} \left[\frac{S}{K_s + S} \right] \quad (9)$$

Where μ_{\max} is maximum specific growth rate of micro-organism (day^{-1}).

Combining equations (8) and (9) gives:

$$dx/dt = \mu_{\max} \left[\frac{XS}{K_s + S} \right] \quad (10)$$

Given the biomass yield coefficient as Y , the relationship between the rate of substrate utilization and the rate of biomass growth is given by:

$$dx/dt = -Y(ds/dt) \quad (11)$$

Substituting equation (10) into equation (11) gives:

$$\mu_{\max} \left[\frac{XS}{K_s + S} \right] = -Y(ds/dt) \quad (12)$$

If maximum rate of substrate utilization $K = \frac{\mu_{\max}}{Y}$, then rearranging equation (12) gives

$$-ds/dt = KXS/(K_s + S) \quad (13)$$

For continuous homogenous reactor under steady state will involve incorporating equation (7) into equation (13) to yield

$$S_0 - S/\theta = KXS/(K_s + S) \quad (14)$$

If E is the hydraulic efficiency of the reactor, the substrate concentration at the retention time is

$$S = S_0(1 - E) \quad (15)$$

Substituting this into equation (14) and rearranging gives the hydraulic retention time θ , as

$$\theta = ES_0[K_s + S_0(1 - E)] / KXS_0(1 - E) \quad (16)$$

The solid retention time (SRT) of the anaerobic digestion is given by:

$$\theta_s = X / (-Y ds/dt - K_d X) \quad (17)$$

For continuous homogenous reactor under steady state, equation (17) becomes

$$\theta_s = X / (-Y [(S_0 - S)/\theta] - K_d X) \quad (18)$$

At any particular efficiency E , the SRT becomes

$$\theta_s = X / (Y (ES_0/\theta) - K_d X) \quad (19)$$

3. RESULTS AND DISCUSSION

3.1 Characterization and Proximate Analysis

The physicochemical analysis results before and after the anaerobic digestion is presented in Table 1. During the period of the digestion, the pH increased from 6.21 to 7.21 which are within the World Health Organization (WHO) standard

according to [15]. The temperature was relatively constant at 31 and 32°C. The Total Nitrogen content decreased at the end of digestion from 1.250 to 0.981 mg/l which is quite within the WHO standard [16]. The Biological Oxygen Demand (BOD) at the end of the digestion remained only 31.9 mg/l from its initial value of 364 mg/l though that was still slightly higher than the WHO standard [17]. Also, the Total Suspended Solids (TSS) and the Total Viable Count (TVC) at a concentration of 141 mg/l and 70×10^2 (CFU/100ml) respectively after the given period of digestion also fall short of the WHO standard. If the time of digestion is increased further, it is evident that the BOD, TSS and TVC will still reduce and fall within the acceptable limit. After the treatment, the levels of zinc (0.001 ppm), Chemical Oxygen Demand (COD) 168 mg/l, sulphate (52 mg/l) etc. were all within the acceptable limits of the WHO.

Table 2 is the result of the proximate analysis of waste water slurry obtained after the anaerobic digestion. The moisture content obtained was high. Low ash content indicates loss of some minerals during the digestion process.

3.1.1 Effects of parameters

The effect of temperature with time of the wastewater digestion is presented in Fig. 1. It was observed that during the period of digestion,

the temperature never reached the thermophilic level (between 40 to 55°C) but was always maintained at the mesophilic level (30 to 40°C) and below. This is due to the nature of the wastewater and the fact that anaerobic digestion proceeded much more rapidly at that temperature.

Fig. 2 is a graphical presentation of the variation of the pH with time. For most of the digestion, the pH fluctuating between 6.0 and 9.0 which was within the standard WHO limit. Based on the fact that the pH of the treated wastewater was almost at neutral position, they can be discharged into water bodies without the pH affecting aquatic life adversely.

The effect of time on the cumulative volume of gas produced during the digestion of the wastewater is shown in Fig. 3. It was seen that there was a cumulative increase in the quantity of biogas produced with time during the period up to 54 m³. According to [3], 1.4 KWh of energy can be obtained from 1 m³ of biogas. Therefore about 104.16 KWh of energy can be obtained from this anaerobic digestion process.

3.1.1.1 Kinetics analysis of the anaerobic digestion

Table 3 is the result of the batch experimental data that was used to calculate the kinetic parameters.

Table 1. Characterization of treated and untreated wastewater

Parameter	Concentration		
	Before digestion	After digestion	WHO
pH	6.21	7.21	6.0-9.0
Temperature (°C)	32.0	31.0	–
Total Suspended Solid (TSS) (mg/l)	298.0	141.0	30.0
Calcium hardness/Water hardness/Bicarbonate (mg/l)	96.0	48.0	–
Sulphate (mg/l)	104.0	52.0	200.0
Total Nitrogen (mg/l)	1.25	0.981	4.0
Total Viable Count (Fecal Coliform) (cfu/100 ml)	60×10^3	70×10^2	400.0
Biological Oxygen Demand (BOD ₅)	364.0	31.90	—
Phosphate (mg/l)	72.0	21.00	—
Oil and Grease (%)	20.0	5.10	0.10
Manganese (ppm)	0.00	0.00	0.30
Iron (ppm)	115.10	28.60	0.30
Sodium (ppm)	31.32	27.60	75.0
Potassium (ppm)	2.32	0.5	–
Calcium (ppm)	1.32	0.47	75.0
Magnesium (ppm)	3.11	3.02	–
Aluminium (ppm)	6.32	3.13	–
Chemical Oxygen Demand (COD) (mg/l)	408.0	168.0	200.0
Zinc (ppm)	0.010	0.001	1.00

The first-order kinetics was evaluated by plotting the linear graph of $-\ln(S_e/S_o)$ against t in Fig. 4. From the slope of the plot, the first-order inactivation rate coefficient or the rate constant K, was obtained as 0.0211 day^{-1} . This is the maximum constant rate at which the micro-organisms digesting the available food would do so before they become inactivated. The first-order kinetic plot gave a correlation coefficient (R^2) value of 0.971 indicating that the kinetics of the digestion followed a first-order reaction. [13] obtained the first order rate constant K as 0.183 in the anaerobic digestion of Municipal waste.

The linear plot of $\frac{1}{U}$ against $\frac{1}{S_e}$ was used to calculate the maximum rate of substrate utilization K and the half-velocity constant K_s from the intercept and the slope respectively as shown in Fig. 5. The value of K was 0.0283 day^{-1} while that of K_s was 26.911 mg/l. The correlation coefficient of the linear plot was 0.928. The small value of K suggests that for effective digestion, the reactor used must be very large [18] and an inoculant will be required. The need for

inoculation for the complete digestion of waste water pollutants confirms that the digesting micro-organisms will need high retention time to regenerate after being inactivated [13]. It is noted that in anaerobic digestion, the cell production is relatively low resulting in low sludge generation [19,20].

Table 2. Proximate analysis of the slurry after digestion

Parameter	Percentage
Moisture content (%)	80.0
Total solid (%)	20.0
Ash content (%)	0.05
Fat content (%)	1.0
Nitrogen content (%)	0.196
Protein content (%)	1.23
Fibre content (%)	0.05
Carbohydrate content (%)	17.67
Volatile solid (%)	2.5
Carbon content (%)	10
C/N ratio	5.10
pH	7.26

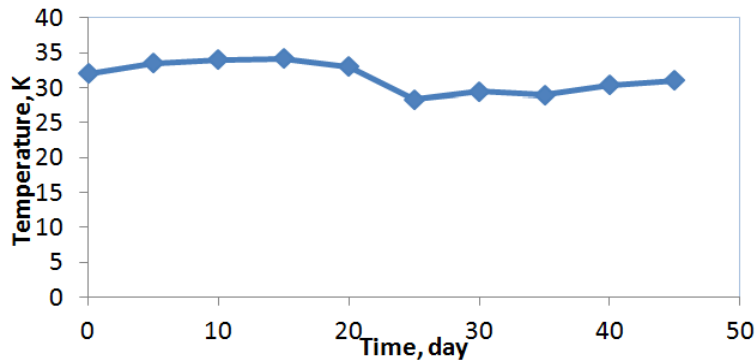


Fig. 1. Effect of time on the temperature

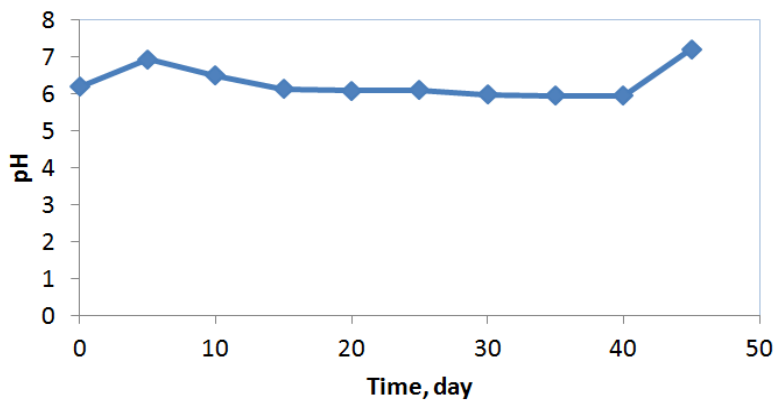


Fig. 2. Effect of time on the pH

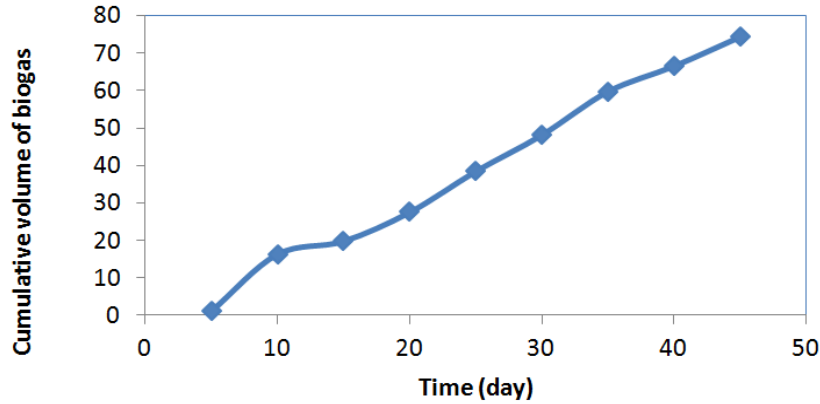


Fig. 3. Effect of time on the cumulative volume of biogas produced

Table 3. Determination of kinetic parameters

T (days)	COD (mg/l)	BOD (mg/l)	TSS (mg/l)	TVC (cfu/100 ml)	So (mg/l)	Se (mg/l)	Xo (mg/l)	Xe (mg/l)	X (mg/l)
0	408.0	364.0	298.0	60×10^3	408.0		298.0		
5	380.0	325.0	284.0	62×10^7		380.0		284.0	291.0
10	364.0	286.4	258.0	70×10^7		364.0		258.0	278.0
15	328.0	215.0	252.0	50×10^7		328.0		252.0	275.0
20	252.0	165.9	238.0	89×10^6		252.0		238.0	268.0
25	248.0	91.0	208.0	90×10^5		248.0		208.0	253.0
30	218.0	48.0	199.0	83×10^4		218.0		199.0	248.5
35	201.0	42.0	176.0	81×10^3		201.0		176.0	237.0
40	190.0	38.5	156.0	79×10^2		190.0		156.0	227.0
45	168.0	31.9	141.0	70×10^2		168.0		141.0	219.5

$$X = \frac{X_o + X_e}{2} = \text{Average TSS cell mass concentration}$$

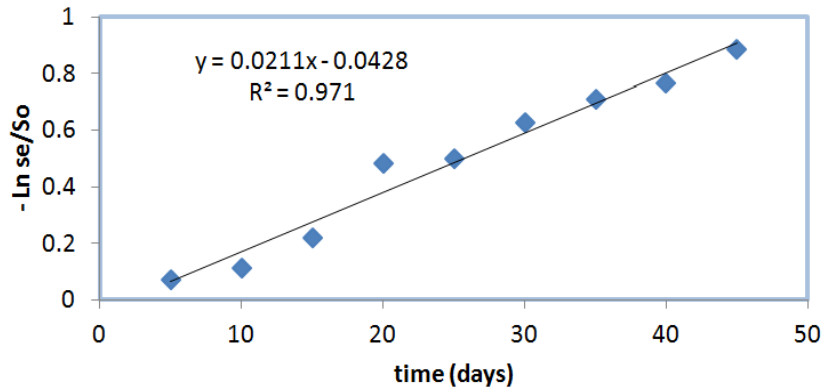


Fig. 4. First order kinetic plot

Fig. 6 shows the plot of the inverse mean cell residence, $\frac{1}{\theta}$ against the specific substrate rate of utilization, U. From the straight graph, the biomass yield, Y was obtained as 1.1168 mg/mg while the endogenous decay coefficient, K_d as 0.0112 day^{-1} . These values are small and so indicates that the net sludge volume obtained

from the anaerobic digestion was small. The coefficient of correlation was 0.9691. The maximum specific growth rate of microorganisms, U_{max} was obtained as 0.0316 day^{-1} . In the anaerobic digestion of Municipal waste, [13] obtained the maximum specific growth rate μ_{max} as 0.053 day^{-1} .

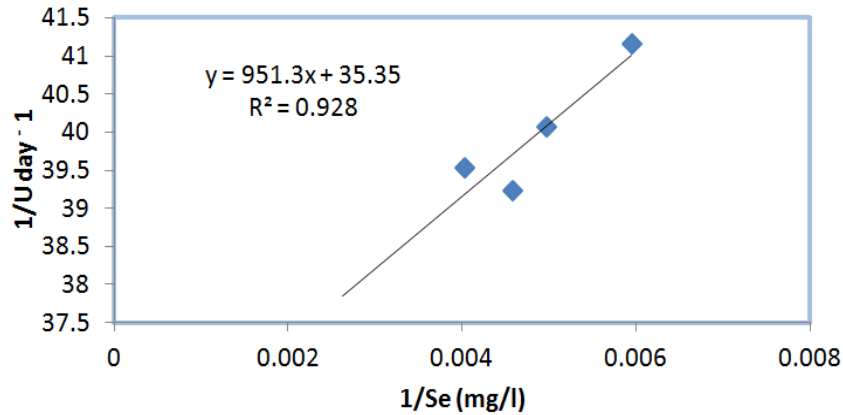


Fig. 5. Plot for determination of K and K_s

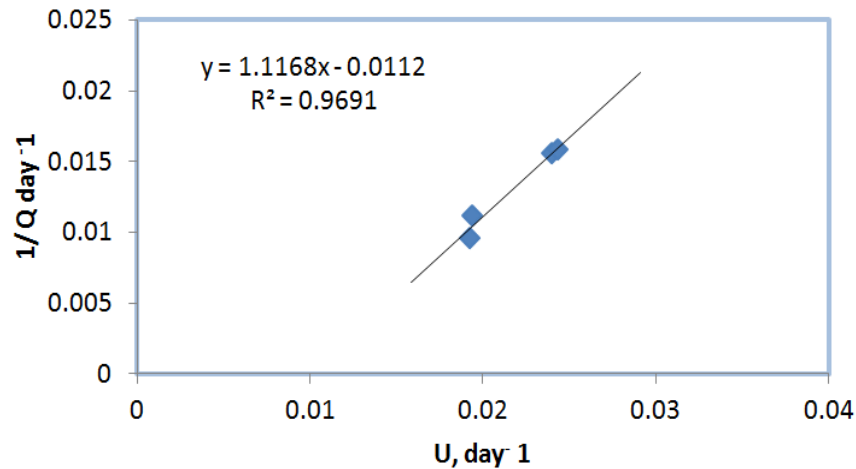


Fig. 6. Plot for determination of Y and K_d

3.1.1.2 Continuous process design model simulation

Considering a homogeneous continuous flow type reactor, the Hydraulic Retention time (HRT) required under steady state to accomplish the anaerobic digestion was calculated for various reactor efficiencies (E) within the range of 70-90%. The influent COD was 408 mg/l and the food to micro-organism ratio (F/M) used were 0.7, 0.75, 0.8, 0.85, 0.9, 0.95, and 1.0 which corresponds to the biomass concentrations of 582.9, 544, 510, 480, 453.3, 429.5 and 408 mg/l respectively. Fig. 7 shows the variation of HRT for the various efficiencies used for the anaerobic digestion. Table 4 shows the values where it was observed that the HRT increased almost linearly with increase in efficiency which is quite expected as per model equation. The HRT was also seen to increase as the biomass concentration decreased and this will

help in the reactor design. Some researchers such as [10] obtained a similar result in the anaerobic digestion of Municipal Solid Wastes.

The Solid Retention Time (SRT) was also calculated for the same steady state homogeneous continuous flow type reactor for the same reactor efficiencies. Fig. 8 and Table 4 also shows the variations of SRT for the various efficiencies used for the anaerobic digestion. It was observed that the SRT increased with increase in reactor efficiency. The plot of the SRT increase represents hyperbolic curve.

The homogeneous continuous flow type reactor was used to determine the effect of different influent substrate concentration on the HRT and SRT values to attain a targeted efficiency under steady state condition for the anaerobic

digestion. The influent concentration was varied in the range of 582.9 to 408.04 mg/l using the calculated difference of 24.98 mg/l. Using two F/M ratios of 0.8 and 1.0, the biomass concentration used was 510 and 408 mg/l respectively and the HRT and SRT were calculated for 70, 80, and 90% reactor efficiency in response to the same F/M ratio of 0.8 and 1.0. Table 5 showed the values obtained while Fig 9 showed the variation of HRT with the influent concentration. From the plot, it is seen that the HRT increased linearly with increase in influent substrate concentration for a particular value of reactor efficiency and biomass concentration. It was also observed that HRT increased with decrease in biomass concentration and increase in reactor efficiency which is expected.

Fig. 10 showed the variation of SRT with varying influent substrate concentration varied in the range of 582.9 to 408.04 mg/l for different values of reactor efficiency of 70, 80, and 90%. The biomass concentration used was 510 and 408 mg/l corresponding to the two F/M ratios of 0.8 and 1.0 respectively. It was observed that with increase in influent substrate concentration, there is a decrease in the SRT. This indicated that as the efficiency is increased, the decrease in SRT becomes more evident. The plot of the HRT is decreasingly hyperbolic in nature in accordance with the model equation. In the treatment of Municipal Solid Wastes using anaerobic digestion, [10] also reported a decrease in the SRT with increase in influent substrate concentration.

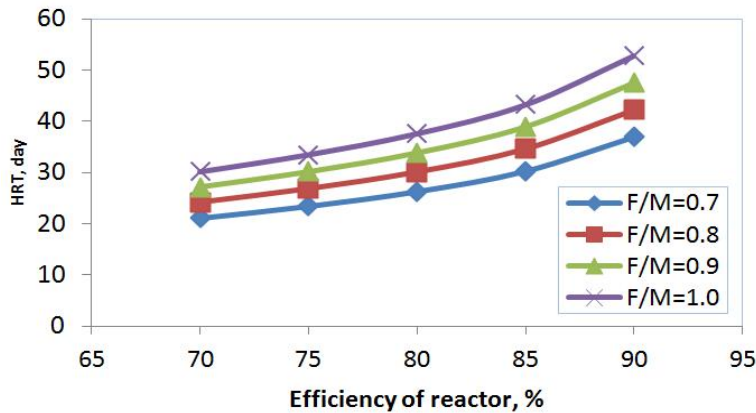


Fig. 7. Variation of HRT with efficiency of the anaerobic digestion

Table 4a and 4b. Variation of HRT and SRT with variation of efficiency of anaerobic digestion

Efficiency (%)	Influent COD, S ₀ (mg/l)	Biomass concentration X (mg/l)						
		F/M =0.7	F/M =0.75	F/M =0.8	F/M =0.85	F/M =0.9	F/M =0.95	F/M =1.0
70	408	582.9	544	510	480	453.3	429.5	408
75	408	582.9	544	510	480	453.3	429.5	408
80	408	582.9	544	510	480	453.3	429.5	408
85	408	582.9	544	510	480	453.3	429.5	408
90	408	582.9	544	510	480	453.3	429.5	408
95	408	582.9	544	510	480	453.3	429.5	408

Table 4b

F/M =0.7	HRT (day)						SRT (day)
	F/M =0.75	F/M =0.8	F/M =0.85	F/M =0.9	F/M =0.95	F/M =1.0	
21.12	22.63	24.14	25.65	27.16	28.66	30.17	67.99
23.44	25.12	26.80	28.47	30.15	31.82	33.49	72.40
26.31	28.19	30.07	31.95	33.83	35.71	37.59	79.56
30.27	32.43	34.59	36.76	38.92	41.08	43.24	93.02
36.94	39.58	42.22	44.86	47.50	50.14	52.78	127.5
54.49	58.39	62.28	66.17	70.07	73.95	77.85	411.8

Table 5. Variation of HRT and SRT with variation of influent substrate concentration of anaerobic digestion

Influent COD, S _o (mg/l)	Reactor efficiency (%)			Biomass, X (mg/l)		HRT (day)						SRT (day)		
	E1	E2	E3	F/M =0.8	F/M =1.0	E1 + X1	E1 + X2	E2 + X1	E2 + X2	E3 + X1	E3 + X2	E1	E2	E3
408.04	70	80	90	510	408	24.14	30.18	30.08	37.59	42.23	53.34	67.98	79.59	127.51
433.02	70	80	90	510	408	25.35	31.69	31.46	39.32	43.78	54.73	66.74	77.44	120.58
458.00	70	80	90	510	408	26.56	33.20	33.10	41.06	45.34	56.68	65.65	76.68	114.83
482.98	70	80	90	510	408	27.76	34.72	34.23	42.79	46.90	58.62	64.64	73.97	109.94
507.96	70	80	90	510	408	28.99	36.23	36.00	44.52	48.46	60.57	63.86	73.97	105.73
532.94	70	80	90	510	408	30.20	37.75	37.11	46.25	50.01	62.52	63.09	71.64	102.02
557.92	70	80	90	510	408	31.41	39.26	38.38	47.98	51.57	64.46	62.39	70.09	98.80
582.90	70	80	90	510	408	32.62	40.78	39.77	49.71	53.13	66.41	61.76	69.08	95.95

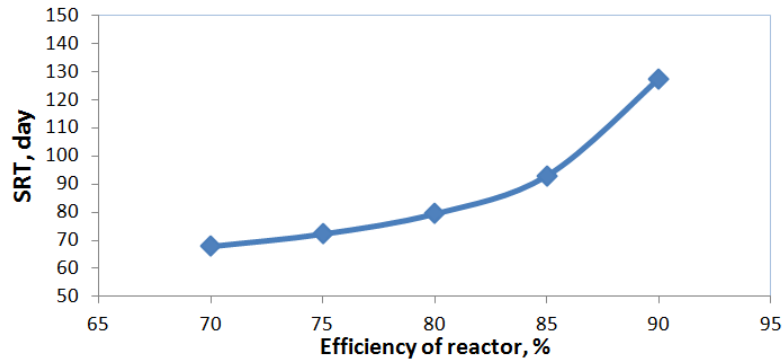


Fig. 8. Variation of SRT with efficiency of the anaerobic digestion

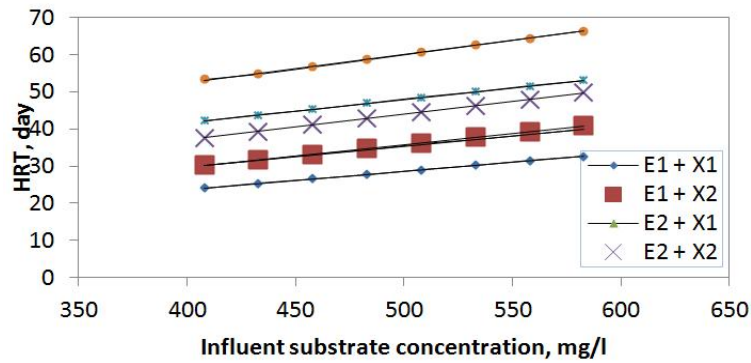


Fig. 9. Variation of HRT of the anaerobic reactor with influent substrate concentration (E1=70%, E2=80%, E3=90%, X1=510mg/l, X2=408mg/l)

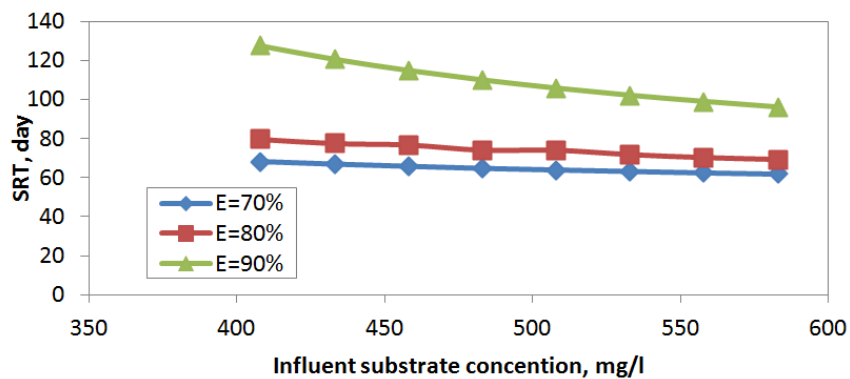


Fig. 10. Variation of SRT of the anaerobic reactor with influent substrate concentration

4. CONCLUSION

The physicochemical analysis showed that most of the parameters of the wastewater were reduced to an acceptable level after the anaerobic digestion. The kinetic study revealed that the digestion followed first order kinetics. Both the maximum rate of substrate utilization

and the maximum specific growth rate of the micro-organisms were small indicating that for effective digestion, the reactor must be large and an inoculator will be needed. For a continuous flow type homogenous reactor under steady state, the models developed here can be employed to estimate the control parameters of the process. The SRT increases with increase in

reactor efficiencies and influent substrate concentration. Equally, the increase in the efficiency of the reactor and the decrease in the biomass concentration results to an increase in the SRT of the digestion. The design process data provided is very useful in the designing of the continuous steady state anaerobic digestion system for treating pharmaceutical wastewater.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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