2017-06-30

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Academic Journals

https://doi.org/10.5897/AJEST2017.2305

academicJournals

Vol. 11(6), pp. 339-348, June 2017
DOI: 10.5897/AJEST2017.2305
Article Number: 022BE1B64383
ISSN 1996-0786

http://www.academicjournals.org/AJEST

African Journal of Environmental Science and Technology

Full Length Research Paper

Kinetic analysis of anaerobic sequencing batch reactor for the treatment of tannery wastewater

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Received 22 February, 2017; Accepted 21 March, 2017

A pilot scale anaerobic sequencing batch reactor (ASBR) was operated at different organic loading rate (1.03, 1.23, 1.52 and 2.21 kg.m^{-3}.d^{-1}) in order to determine the chemical oxygen demand (COD) removal and methane production kinetic models. The system was operated at mesophilic temperature. The wastewater was fed using submersible pump in every twenty four hours and agitated with hydraulic pump for fifteen minutes in every one hour. The COD removal efficiencies was found to be between 69-85% and the methane yield was also between 0.17±0.2 and 0.30±0.02 m^3/kg COD removed. In the kinetic studies, modified Stover-Kincannon and second-order models were found to be the most appropriate model for ASBR treating tannery wastewater than first order model. The saturation value constant and maximum COD removal rate found in Stover-Kincannon model were 5.57 and 5.56 kg of COD m^{-3}.d^{-1}, respectively. The kinetic studies of volumetric methane production showed that Michaelis-Menten model was found to be capable of predicting the volumetric methane production in ASBR that treat tanney wastewater.

Key words: Anaerobic sequencing batch reactor (ASBR), chemical oxygen demand (COD) removal, Michaelis-Menten, second order, Stover-Kincannon.

INTRODUCTION

Tanning is almost a wet process that uses about 30 to 40 L of water/kg of hides or skin processed and also discharges about 90% of the consumed water as wastewater (IFC, 2007). The wastewaters, which are discharged without proper treatment, would contaminate surface and ground water as well as soils. A tanning industry can cause groundwater pollution of about 7 to 8 km radius (Mondal et al., 2005). Currently, there are more than 30 tanneries under operation in Ethiopia. These tanning industries generate 11,312 m^3 wastewater per day and disposed to the surrounding water bodies without proper treatments (LIDI, 2010). However, it is characterized by a high load of pollutants which require proper treatment before it would be discharged into the receiving water body. The treatment systems adopted by most industries are frequently considered as

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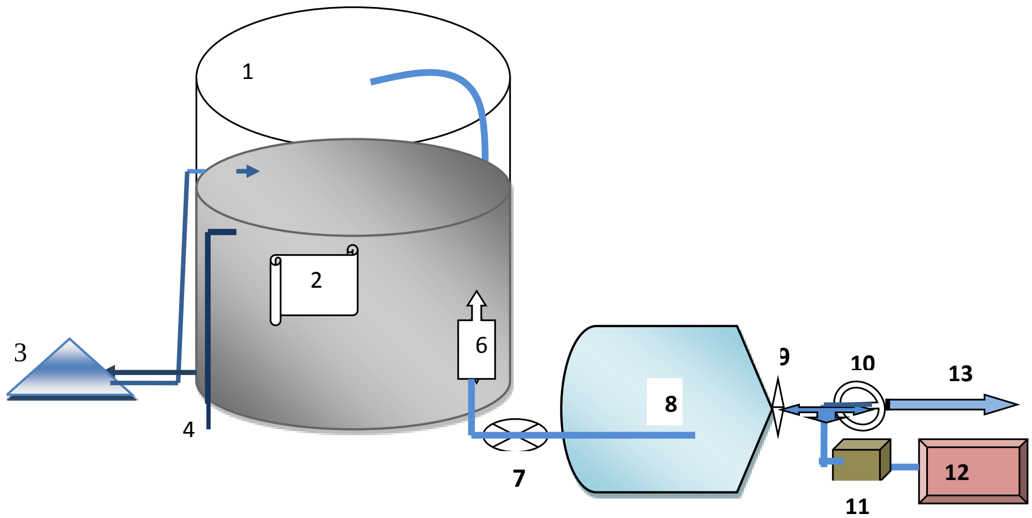


Figure 1. Schematic diagram of the pilot scale anaerobic sequencing batch reactor. ASBR (1); control panel (2); mixing chamber (3); feeding pipe (4); gas pipe (5); gas flow meter (6); moisture trap unit (7); biogas storage bag (8): gas valve (9); gas blower (10); sulfur scrubber (11); generator (12) and gas line to the kitchen (13).

regulatory obligation that increase capital and operational costs and ultimately yield negative economic returns. Compliance to environmental legislations should not necessary lead to the creation of additional costs. It should instead provide a secondary source of income. Anaerobic treatment is considered as sustainable method of reducing pollution from domestic, agricultural and industrial operations (Seghezzo et al., 1998; William and David, 1999). It consumes little energy as no aeration is needed and produces renewable energy in the form of biogas and nutrient rich digestate (Kaparaju and Rintala, 2011). Beside the supply of energy and manure, the anaerobic technology provides the opportunity for the reduction of greenhouse gas emission and mitigation of global warming through substitution of fossil fuel for energy production and chemical fertilizer (Pathak et al., 2009). The total output in CO_2 equivalents might be reduced from 2.4 kg CO_2-eq/kg COD removed for fully aerobic treatment to 1.0 kg for primarily anaerobic processes (Insam and Wett, 2008). In fully aerobic system, up to 1.4 kg CO_2/COD removed is generated from electric power generation, whereas the carbon dioxide generated in the anaerobic processes is greenhouse gas neutral (Insam and Wett, 2008; Kaparaju and Rintala, 2011).

The anaerobic sequencing batch reactor (ASBR) operates in a cyclic batch mode with four distinct phases per cycle. The four phases are: filling, reacting, settling and release (Timur and Oèzturkm, 1999; Zhang et al., 1997). The ASBR systems has been successfully applied in laboratory and pilot scales for treatment of high strength wastewaters including landfill leachate, slaughterhouse wastewater, municipal sludge and dairy wastewaters, and brewery wastewater (Xiangwen et al., 2008).

Mathematical description of biological treatment processes has been performed using process kinetics model. The understanding of process kinetics is important for the design of specific unit and operation of treatment systems, predicting system stability, effluent quality and waste stabilization. The knowledge on kinetics model leads to optimization of performance, a more stable operation and a better control of wastewater treatment operations. There are numerous mathematical models in the literature for describing anaerobic processes such as first order model, second-order, Grau model, Modified Stover-Kincannon, Sundstorm model, Contois model, Chen, Michaelis-Menten type kinetic model, etc (Jafarzadeh et al., 2009; Sandhya and Swaminathan, 2006; Buyukkamaci and Filibeli, 2002). In this study, different mathematical models such as first order model, second order Grau model, Modified Stover-Kincannon, Michaelis-Menten type kinetic model were applied to data obtained from the ASBR operation and kinetic coefficient for the reactors were calculated.

MATERIALS AND METHODS

Experimental set up

The pilot-scale anaerobic SBR was constructed using concrete materials in a cylindrical in shape with a dimension of 4 m in height and 4 m in diameter. Figure 1 shows the schematic diagram of the pilot scale ASBR and the accessories. The total volume was 100 m^3

Table 1. Characteristics of tannery wastewater used at five different OLR.

Parameter	Phase I	Phase II	Phase III	Phase IV
pH	9.64 ±0.46	9.20 ±0.33	9.28 ± 0.311	9.09±0.49
E.C. (mS)	8.76±0.40	8.46±0.39	8.08±0.38	8.43±0.72
TDS (g/l)	7.49±0.36	7.24±0.44	6.81±0.42	7.26±0.68
Salinity (g/l)	9.26±0.57	9.19±0.38	8.91±0.33	9.07±0.71
COD (mg/l)	4221±359	4265± 215	4586± 292	4458± 396
TN (mg/l)	451±47.5	517±112	492.5±89.9	458±58.6
NH_4^+-N (mg/l)	231±45	270±66	255±58	248±44.46
Total phosphorus (mg/l)	22.2±6.8	18 ±4.5	19.3 ±4.16	23.5 ±6.5
Sulfide (mg/l)	93±22.27	126±38.9	123.5±33.8	117.5±29.4
Sulfate (mg/l)	470±75.	390±76.9	520±99.13	469±69

of this 80 m^3 as working volume and the remaining volume for head space. The internal part of the digester was insulated with plastic foam and covered with geo-membrane. Stainless steel tubes were installed 30 cm above the bottom of the digester surface for the circulation of hot water. The top of the digester has two holes. One of the holes was fitted with PVC pipe for biogas outlet to gas flow meter and to the gas storage bag. The other one was fitted with stainless steel tubing extended to the bottom of the digester for hot water circulation. The ASBR was operated under mesophilic condition (31°C) and hot water heated with solar panel was used to maintain the temperature. The wastewater in the ASBR was mixed in every hour using hydraulic pump for fifteen minutes during the day time of the operation.

Operation of the ASBR

The performance of the pilot scale ASBR was evaluated at four different OLRs. The performance of the ASBR was monitored by measuring COD removal efficiency and the biogas production and quality. During the first phase, the reactor was operated at the organic loading rate (OLR) of 1.03 $kg.m^{-3}.d^{-1}$ and constant hydraulic retention time of 4 days. In the second phase, the OLR was increased from 1.03 to 1.23 $kg.m^{-3}.d^{-1}$ by increasing the volume of wastewater from 20 to 23 m^3 and HRT was 3.5 days. In the third phase, the reactor was operated at OLR of 1.52 $kg.m^{-3}.d^{-1}$ and HRT of 3 days by increasing the volume of the inlet wastewater from 23 to 26.5 m^3. Finally, it was operated at OLR of 2.21 $kg.m^{-3}.d^{-1}$ and 2 days by increasing the volume of wastewater from 26.5 and 40 m^3.

Physico-chemical analysis

The characteristics of influent and effluents were determined in terms of chemical oxygen demand (COD), total nitrogen (TN), ammonium-nitrogen (NH_4^+-N), nitrate-nitrogen (NO_3^--N), sulphides (S^{2-}), sulphate (SO_4^{2-}), total phosphorous (TP) and orthophosphate (PO_4^{3-}) colorimetrically using spectrophotometer (DR/2010 HACH, Loveland, USA) according to HACH instructions. Total solid and volatile solid were also measured according to the methods described in standard methods of water and wastewater (APHA, 1998). pH of tannery wastewater was measured using a pH meter (CON, 2700). TDS, EC and salinity were measured using TDS/EC/salinity meter. Sodium, potassium, calcium, magnesium, chromium, copper, iron and lead were determined using an atomic absorption spectrometer (novAA, 400P). Total nitrogen content of the wetland plant samples was determined by Kjeldahl method. Percent of removal efficiency (%) for each parameter was determined by the following equation:

$$\text{RE}(\%) = 100 * \left(\frac{S_{In-}S_{out}}{S_{In}}\right)$$

RESULTS AND DISCUSSION

Characteristics of raw tannery wastewater

Tannery wastewater is characterized mostly in terms of the levels of pH, salinity, organic matter (COD), nitrogenous compounds (TN and NH_4^+), suspended solids (SS) and total dissolved solids (TDS), chromium and sulfides (Jahan et al., 2014). However, these parameters vary significantly from tannery to tannery depending on the size of the tannery, chemicals used for a specific process, amount of water used and type of final product produced.

The mean characteristics of raw wastewaters used in the study are presented in Table 1.

The wastewater was characterized as alkaline with pH value ranging from 9.09±0.49 to 9.64±0.46. It also contain high level of electrical conductivity (8.08±0.38 to 8.76±0.34 mS), total dissolved solids (6.81±0.42 to 7.49±0.36 g/l) and salinity content (8.77±0.72 to 9.26±0.51 g/l). This due to the chemicals used in the soaking and beam house operation. It contained high organic matter and nitrogenous compounds with COD ranges from 4221±359 to 4586± 292 mg/l. The influent had high total nitrogen (TN), NH_4^+-N and sulfate values ranges from 451±47.5 to 517±112 mg/l, 231±45 to 270±66 mg/l and 390±76.9 to 520±99.13 mg/l, respectively; likewise, sulfide and phosphate concentrations ranged from 92.9±23.27 to 127±43.3 mg/l and 18±4.5 to 23.5±6.5 mg/l, respectively. Seyoum (2004) reported higher concentration of TN, ammonium and COD in tannery wastewater.

Table 2. Summary of the performance of anaerobic sequencing batch reactor.

Parameters	Phase I	Phase II	Phase III	Phase IV
OLR (kg.m^{-3}.day^{-1})	1.03±0.09	1.23±0.06	1.52±0.1	2.21±0.23
HRT (day)	4	3.5	3	2
COD removal (%)	81±2.1	79±2.3	76±1.6	69±1.7
COD out (mg/l)	791±149.5	898.9± 122	1101.4±123	1358.3±170
Biogas production (m^3.day^{-1})	26.2±1.6	28.1±1.8	31.8±2.7	36.7±2.8
Methane (%)	70±1.6	68±1.7	64 ±3.0	55±1.9

Performance of the ASBR

The COD removal efficiency and biogas production, methane yield and content of the ASBR are summarized in Table 2. During the first phase of the operation, the COD removal efficiency varied in the range of 78-84%. The average COD removal efficiency and mass removal rate in the single feeding mode were 81±2% and 791±149.5 mg/l, respectively.

In the second phase, the COD removal efficiency varied in the range of 76 to 83% with average removal efficiency of 79±2.3%, while the average concentration of COD was 898.9±122 mg/l in the final effluent. In the third phase, the average COD removal efficiency decreased to 74-79% with average COD concentration of 1101.4±123 mg/l. In the final phase, the COD removal efficiency varied between 67 and 72%. The average removal COD removal efficiency and effluent concentration were 69±1.7% with 1358.3±170 mg/l, respectively. The anaerobic digester showed significant variation in COD removal efficiency with variation of organic loading rate (ANOVA, P<0.05). The results of this study indicate that COD removal efficiency was highest in the first phase of operation and lowest in the final phase of operation. The final phase showed residual COD 31% from the influent. The COD removal efficiencies obtained in this study were higher than the results recorded by Song et al. (2003) from the treatment of tannery wastewater (COD removal of 60 to 75%) using upflow anaerobic fixed biofilm reactor at varying organic loading rate of 0.16 to 3.14 kg m^{-3}d^{-1} and HRT of 16 days to 1 day. The results obtained in phase II (79±2.3%) are comparable with results reported by Lefebvre et al. (2006) for the treatment of tannery soak liquor (COD removal of 78%) at a HRT of 5 days and an OLR of 0.5 kg m^{-3}d^{-1} using up flow anaerobic sludge blanket bed reactor. Other comparable mean COD removal (78.2%) was reported by El-Sheikh et al. (2011) in the treatment of tannery wastewater using two stage UASB reactors. On the other hand, Banu and Kaliappan (2007) found slightly higher COD removal efficiency (86% at OLR of 2.74 kg m^{-3}d^{-1} and HRT of 60 h; 88% at OLR of 3.22 kg m^{-3}d^{-1} for 70 h) in the treatment of tannery wastewater using hybrid upflow anaerobic sludge blanket reactor.

Kinetic model substrate removal

First order substrate removal model

The hydrolysis of organic pollutants was described by first order kinetics model. The mass balance equation for the substrate in the anaerobic system can be described as follows:

$$-\frac{\mathrm{ds}}{\mathrm{dt}} = \frac{\mathrm{Q}}{\mathrm{V}}(\mathrm{S}_{o-}\mathrm{S}_{\mathrm{e}}) - K1Se$$

where, So is substrate concentration in the influent (mg/l): Se is substrate concentration in the effluent; Q is flow rate of influent to reactor (l/d); V is effective volume of the reactor and K1 is first-order kinetic constant (per day). Under steady state conditions, $(-\frac{\mathrm{ds}}{\mathrm{dt}})$=0 and the above equation can be represented in the following form:

$$\frac{\mathrm{S}_{0-}Se}{\theta_{\mathrm{H}}} = K1Se$$

where,θ_{H} = hydraulic retention time.

The value of first-order kinetic constant can be obtained by plotting $\frac{\mathrm{S}_{0-}Se}{\theta_{\mathrm{H}}}$ against Se as given in the above equation. The slope of the line in the plot would represent the value of K_1. The first order model for COD removal is drawn in Figure 2. The value of the k was obtained from the slope of the line that was plotted (So-Se)/HRT versus Se. The graph fit a straight line with regression coefficient of (R^2= 0.83).

This indicates that about 17% of the total variations were not explained in the first order regression model. The k value was determined as 0.99 per day. Isik and Sponza (2005) obtained comparable (k=0.93) value with this study from the treatment simulated textile wastewater using UASBR system

Modified Stover-Kincannon Model

The Stover-Kincannon was developed first for rotating biofilm reactor (Sandhye and Swaminathan, 2006). It is

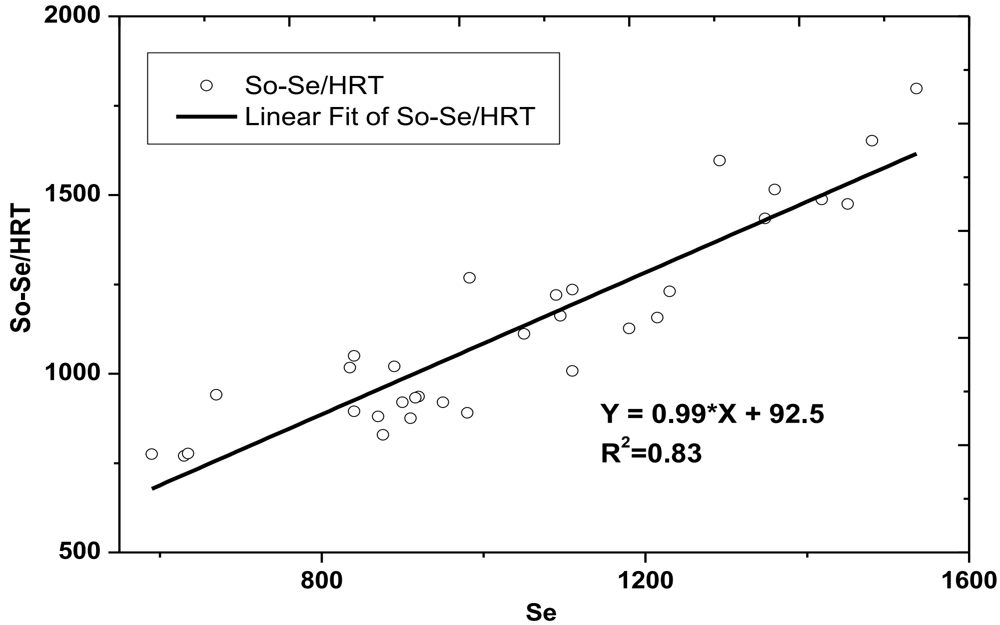


Figure 2. Substrate removal first order model for ASBR.

assumed that the organic loading rate can be correlated with substrate utilization rate using mono-molecular kinetics. The substrate removal rate is defined in two different forms as shown below:

$$\frac{ds}{dt} = \frac{Q}{V}(S_i - S_e)$$

$$\frac{ds}{dt} = \frac{U_{max}\left(\frac{QS_i}{V}\right)}{K_B + \left(\frac{QS_i}{V}\right)}$$

This can be linearized as:

$$\left(\frac{ds}{dt}\right)^{-1} = \frac{V}{q(Si - Se)} = \frac{KB\ V}{Umax\ QSi} + \frac{1}{Umax}$$

Where ds/dt is the substrate removal rate (g/L-day), U_{max} is the maximum utilization rate constant (g/L-day), K_B is saturation value constant (g/L-day), Q is the flow rate (L/day) and V is the effective volume of reactor (L). Since dS/dt approaches U_{max} as the organic loading rate, qSi/V approaches infinity. Figure 3 illustrates the graph drawn between the reciprocal of mass loading removal rate (V/(Q(So-Si) with the reciprocal of OLR to derive the values of Umax and K_B for the anaerobic sequencing batch reactor treating tannery wastewater. The graph fit a straight line with regression coefficient of (R^2= 0.99). This indicates that only less than 1% of the total variations were not explained in the regression model. Hence, the regression coefficient supports strongly the validity of the linearized Stover-Kincannon model. It can be conclude that modified Stover-Kincannon model can be used to describe the performance of mesophilic ASBR treating tannery wastewater in this study. The maximum value for COD removal rate (U_{max}) and saturation constant (K_B) were determined as 5.56 and 5.78 kg of COD m^{-3} d^{-1}, respectively.

The predicted U_{max} was higher than the maximum loading rate (2.21 kg m^{-3} d^{-1}) used in this study. This revealed that ASBR has higher potential in withstanding high strength tannery wastewater. Moreover, the closeness of U_{max} and K_B values obtained indicate that increasing organic loading rates would lead to reduction in the processes efficiency. Senturk et al. (2010) and Ahn and Forster (2000) also made similar conclusion. Table 3 shows the comparison of the value of U_{max} and K_B reported for various types of substrate (wastewater) using the same type of modified Stover-Kincannon model.

The U_{max} and K_B found in this current work were higher than Priya et al. (2009) and Ahn and Forster (2002). They were almost comparable with the values obtained by Kapdan (2005) and Isik and Sponza (2005). On the other hand, the value of both Umax and K_B in this work were lower than that of Senturk et al. (2010), Wanga et al. (2009), Yilmaz et al. (2008) and Sandhye and Swaminathan (2006). The highest U_{max} were obtained for milk permeate waste water in AMBBR system followed by paper mill wastewater, while the lowest was for formaldhyde containing wastewater followed by corrugated paper wastewater. On the other hand, the

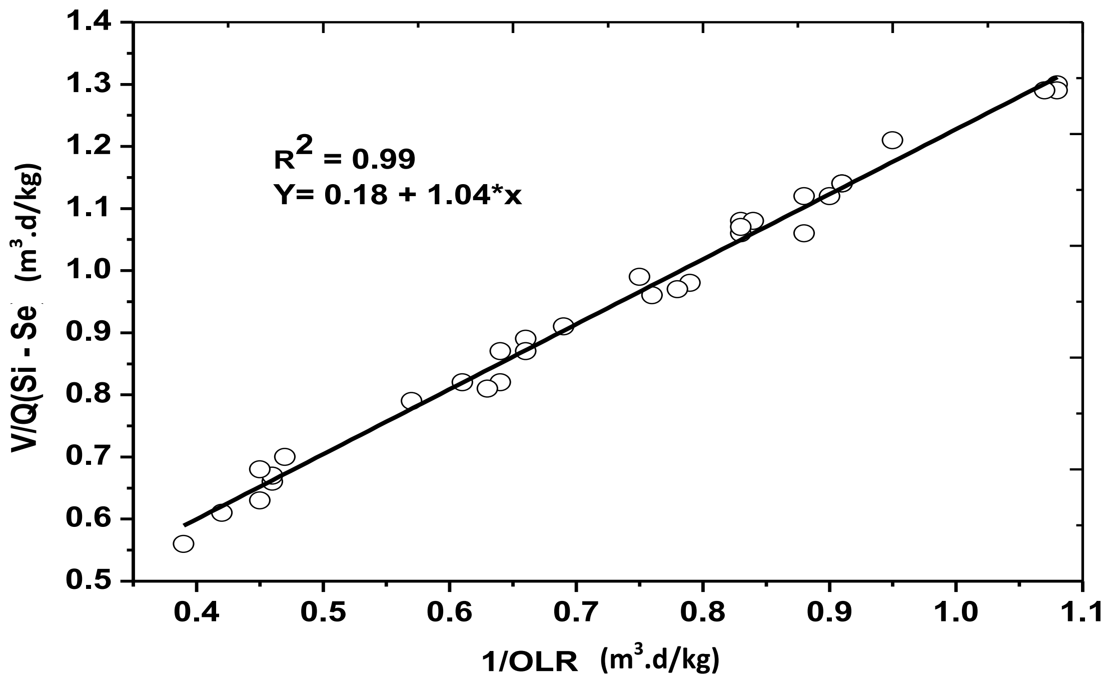


Figure 3. Modified Stover-Kincannon model for ASBR.

Table 3. Comparison of the kinetic parameters obtained for the various substrates.

Types of wastewater	Reactor	U_{max}	K_B	References
Tannery	ASBR	5.56	5.78	This study
Food processing wastewater	Anaerobic contact	22.92	23.59	Senturk et al. (2010)
Formaldhyde containing wastewater	UAFB	3.4	4.6	Priya et al. (2009)
Milk permeate wastewater	AMBBR	89.3	102.3	Wanga et al. (2009)
Paper mill wastewater	AF	86.21	104.15	Yilmaz et al.(2008)
Textile wastewater	UAFB	31.69	45.37	Sandhye and Swaminathan (2006)
Simulated Textile wastewater	UASBR	7.5	8.2	Isik and Sponza (2005)
Synthetic saline wastewater	UASBR	5.3	7.05	Kapdan (2005)
Corrugated paper wastewater	AF	3.86	0.80	Ahn and Forster (2002)

highest K_B was found for paper mill wastewater followed by milk permeates wastewater, while the lowest was from corrugated paper wastewater. The variation of U_{max} and K_B values among different researchers might be attributed to the variation of characteristics wastewater, reactor configuration and microorganisms used in the studies (Priya et al., 2009).

Grau second-order model

The second order model was employed to the experimental results for ASBR system treating tannery wastewater. The general equation of Grau second order kinetics model is shown below:

$$\frac{ds}{dt} = k_2 X \left(\frac{s}{s_o}\right)^2$$

Where, ds/dt is the substrate removal rate (g/L-day), k_2 is kinetic constant (g COD/gVS-day), X is the concentration of microorganisms (gVS/L), S is substrate concentration at any time, and S_0 is the concentration of initial substrate (g/l).

Integrating the above equation within the boundary conditions (S=Si to Se and t=0 toθ_H), and then linearized,

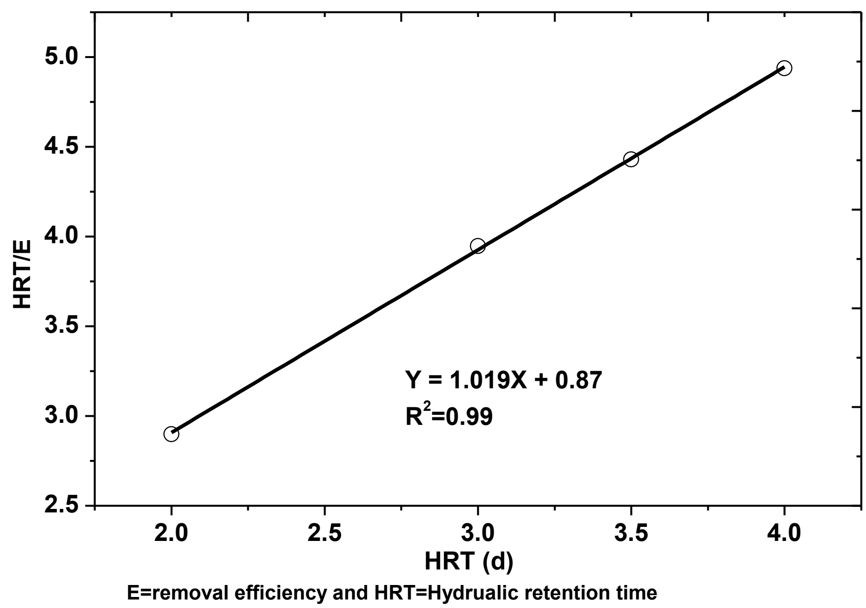


Figure 4. Linear plots of the Grau second-order kinetic model.

the following equation is obtained.

$$\frac{S_0\theta_H}{S_0 - S} = \theta_H + \frac{S_0}{k_2X}$$

Holding the term "S_0/k_2X" in the above equation constant leads to

$$\frac{S_0\theta_H}{S_0 - S} = a + b\theta_H$$

The term $(S_0-S_i)/S_0$ is the substrate removal efficiency (E) and it can be used in the equation as shown below:

$$\frac{\theta_H}{E} = a + b\theta_H$$

If V/Q(Si-Se) is plotted against 1/OLR, K_B/U_{max} is the slope and $1/U_{max}$ is the intercept point of the line. Figure 4 shows the plot of the Grau second-order multi-component model for ASBR.

The value of a, and b were determined from the intercept and slope of the straight lines. The values of a and b were obtained to be 0.87 and 1.019, respectively, with high correlation coefficient (R^2=0.99). This confirms the validity of the application of this model for ASBR treating tannery wastewater. Hence, the formula for predicting substrate concentration in the effluent can be given as:

$$S = S_0\left(1 - \frac{\theta}{0.87 + 1.019\theta}\right)$$

These results show that both modified Stover-Kincannon model and Grau second-order can be applied successfully for modeling of the experimental results of ASBR treating tannery wastewater with high correlation coefficient (R^2=0.99). On the other hand, the first order model appeared to be less successful (R^2=0.83) on predicting substrate removal from tannery wastewater in ASBR system.

Kinetics of methane production

The volumetric methane production rate can be obtained through the expression:

$$r_{CH_4} = \frac{q_{CH_4}}{V}$$

where qCH_4 is the daily methane production (m^3/d) and V is the reactor working volume (m^3).

Figure 5 illustrate the graphical estimation of the concentration of non-biodegradable organic matter (TCOD) based on the relationship between ln (TCOD) and 1/(HRT) (Wang et al., 2009). By the least-square fitting of ln (TCOD) and 1/HRT, an intercept of 0.531 g TCOD L^{-1} (regression coefficient 0.95) was calculated, which corresponds to an infinite HRT. Thus, this can be considered to be the amount of non-biodegradable substrate (Borja et al., 2003; Raposo et al., 2004; Rincon et al., 2006; Wang et al., 2009).

Senturk et al. (2010) and Rincon et al. (2006) found

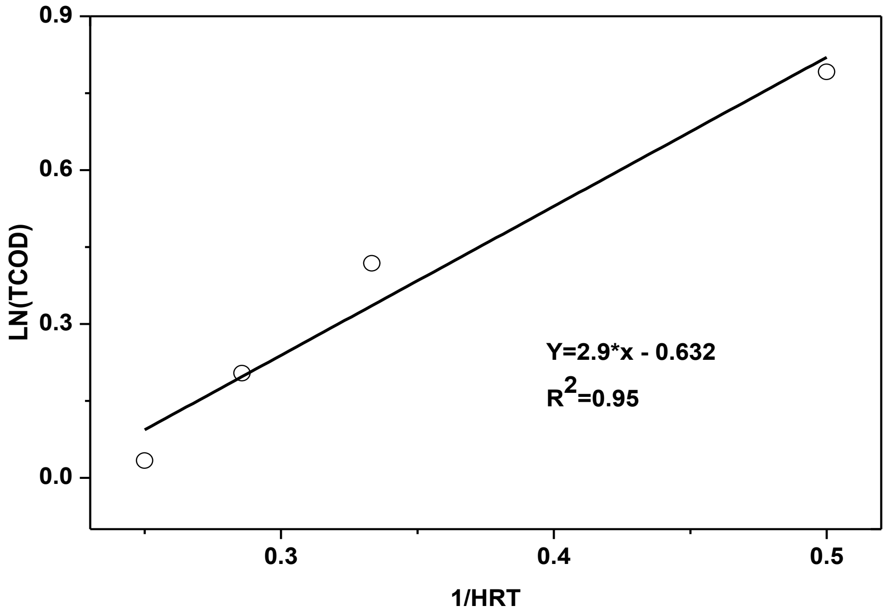
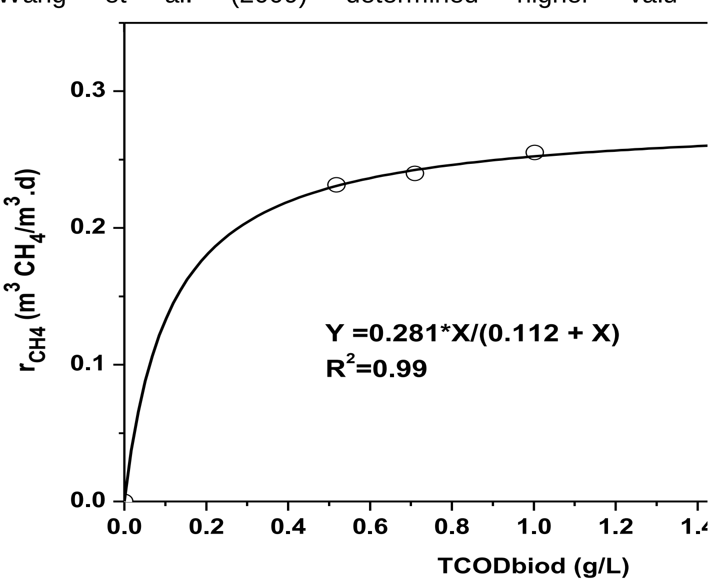


Figure 5. Estimation of the fraction of non-biodegradable soluble organic matter contained in the wastewater used in the study.

Figure 6. Variation of the volumetric methane production rates (r_{CH_4}) as a function of the biodegradable TCOD concentration.

lower non-biodegradable substrate (290 and 92 mg/l) in reactors treating wastewater generated from food processing and protein production from chickpea, respectively. On the other hand, Borja et al. (2003) and Wang et al. (2009) determined higher values of non-biodegradable substrate in the treatment of wastewater generated from Olive Pomace and diary industry, respectively.

The observed values of r_{CH_4} (methane production) plotted as a function of biodegrdabel organic matter concentration (TCOD biod) are shown in Figure 6. As shown in the figure, the observed methane production



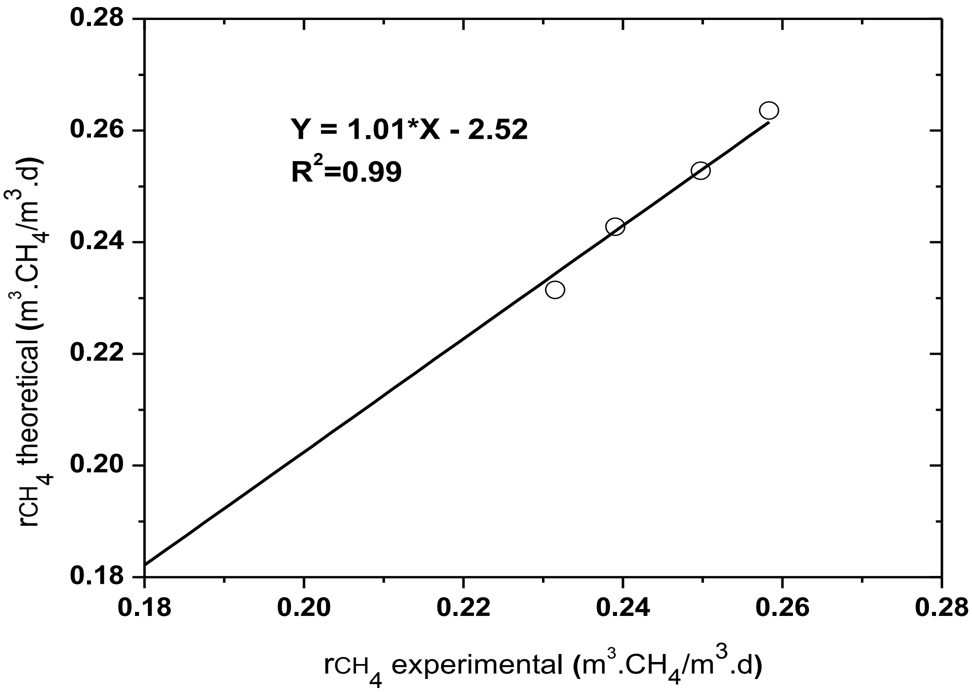


Figure 7. Comparsion between the experimental methane proudction rates and the theoretical value predicted.

values fit Michaelis-Menten type kinetic model, which is a hyperbolic function. By using the origin 8 software, the following kinetic equation was obtained:

$$r_{CH_4} = 0.281TCODb/(TCODb + 0.112)$$

The theortical r_{CH_4}values could be determined using the above equation for the reactor used in this study. The predicted thoertical methane production values were plotted against the observed methane production values. As shown in Figure 7, a linear regression line with a slope of 1.01 and a regression coefficient of the graph 0.99 were obtained. This indicted that the proposed model is capable of predicting the behavior of the ASBR treating tanney wastewater.

Conclusion

The results of this study showed that tannery wastewater could be treated efficiently in anaerobic sequencing batch reactor at different HRT (4, 3.5, 3 and 2 days). The tannery wastewater treatment performance of the ASBR was evaluated at different organic loading rate of 1.03, 1.23, 1.52 and 2.21 kg $m^{-3}.d^{-1}$ and the kinetic analysis for the reactor was performed using the data found in the experiment. The results of the system showed that COD removal efficiency was 69 to 85% and the methane yield was 0.17±0.2 to 0.30±0.02 m^3/kg COD removed. Modified Stover-Kincannon and second-order models were found to be the most suitable model for ASBR (R^2 = 0.99). Michaelis-Menten model was also found to be capable of predicting the volumetric methane production of ASBR that treat tanney wastewater.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

ACKNOWLEDGMENT

This research work was supported by the Swedish International Development Cooperation Agency (SIDA) through Bioresources Innovations Network for Eastern Africa Development (*Bio-Innovate*) program.

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