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Evaluation of kinetic models for tearing textile dyeing wastewater using UASB reactor

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Abstract*: In this study the wastewaters from textile dyeing industries and tapioca starch (SAGO) were combined for the treatment using two-phase Upflow Anaerobic Sludge Blanket (UASB) reactor. The system was inoculated with seed sludge from the anaerobic digester treating sago wastewater. The processes were carried out by feeding the combined wastewater at different mixing proportion with COD ranging from 5200 to 6320 mgL^{-1}. The maximum COD conversion was about 53.1 % in acidogenic reactor and 88.5 % in methanogenic reactor. The kinetic values obtained for Haldane's model a, b and c is 0.034, 19 and 8031, for modified Stover–Kincannon model is R_{max}=3.48 gL^{-1}d^{-1} and K_B=7.77 gL^{-1}d^{-1} and for Grau second-order model the values of a, and b were found to be 0.197 and 1.42. Among the other three kinetic models, Grau second order model and Stover-Kincannon model were observed to be the preeminent model for predicting the performance of two phase hybrid UASB reactors for treating combined real textile dyeing and sago wastewater with higher average regression coefficients (R^2=0.979).*

Keywords*: Textile dyeing effluent, mixing ratio, sago, kinetic, UASB.*

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I. Introduction

Textile processing industries produce high quantities of effluent with varying composition depending upon the wet process employed (Anderson et al. 1996). The textile industry consumes large amount of water, energy and auxiliary chemicals. In India, an average mill producing 60-104 m of fabric per day is likely to discharge approximately 1.5 million liters per day of effluent and the wastewater is being discharged into natural water bodies without proper treatment. Considering both the volume generated and the effluent composition, the textile industry wastewater is rated as the most polluting among all industrial sectors (Arslan et al. 2002). Important pollutants in textile effluent are mainly recalcitrant organics, color, toxicants and inhibitory compounds, surfactants, chlorinated compounds (AOX), pH and salts. Dyes are synthetic organics and generally have a complex chemical structure. They are xenobiotic in biotic environments and hence show persistence to biodegradation in nature. Apart from the aesthetic deterioration of the natural water bodies, dyes also cause harm to the flora and fauna in the natural environment (Basibuyuk and Forster 1997; Bhunia and Ghangrekar 2008). Physico-chemical methods, such as adsorption, coagulation- flocculation, membrane filtration and advanced oxidation, can be very effective for the removal of the colour in wastewaters but these processes merely transfer the pollutants from one phase to another. On the other hand, biological processes provide a low cost and efficient alternative for simultaneous colour and organic matter removal. However, complete dye degradation in wastewater treatment plants based only on aerobic processes is difficult to achieve since the main mechanism responsible for colour removal is adsorption onto the sludges (Buyukkamaci and Filibeli 2002). Anaerobic treatment is widely employed for treatment of most of the industrial wastewater containing high concentrations of soluble organic matter. The Upflow Anaerobic Sludge Blanket (UASB) reactor system developed by Lettinga and his co-workers in the 1970s has received widespread acceptance and has been successfully used to treat a variety of biodegradable industrial wastewaters. UASB reactors belong to the group of high–rate anaerobic reactors with a sludge bed. Granular biomass with high methanogenic activity and excellent settling properties can be cultivated in these reactors. The limitations of UASB reactors are related to the wash-out of biomass. In the present investigation, textile dyeing effluent and tapioca starch wastewater at various proportions was deployed. Advantage of combining the effluents was the pH gets neutralized, therefore no need of adding caustic/lime. The sago effluent was utilized as co-substrate to enhance the degradation of textile dye-house effluent which is recalcitrant in nature. There is no requirement of separate treatment plant to treat both the industrial effluent. The determination of kinetic constants of a bioprocess is a useful tool to be able to describe and predict the performance of the system. Therefore, the aim of this paper is to determine the process kinetics of anaerobic digestion and to compare kinetics among the models applied for the describing the substrate removal kinetics of UASB reactor treating simulated and real textile dyeing wastewater.

II. Materials And Methods

Biomass

The methanogenic granular sludge with unidentified microorganisms used in this experiment was procured from the anaerobic digester treating tapioca starch effluent at Salem, Tamilnadu, India. Before loading the reactor, granular sludge was clearly washed, filtered through a fine mesh ASTM 16 to reduce all the floating and suspended inorganic mineral contents. The volatile suspended solids content of the sludge was 60000 mg L^{-1} (APHA 2005).

Wastewater

Real untreated wastewater from starch industry and textile dyeing industry was collected at Salem, Tamilnadu, India. Ten samples were collected from each industries for duration of three months and the mean values of the parameters have been tabulated in Table 1. The analysis of the wastewater was carried out according to standard methods (APHA, 2005).

Table 1 Characteristics of wastewaters

S.No.	Parameters*	Textile dyeing effluent	Sago Effluent
1	pH	12.8	4.5
2	Total Suspended Solids	420	640
3	Total Dissolved Solids	3520	1200
4	Chlorides	1520	400
5	Sulphates	180	123
6	BOD	175	2400
7	COD	1600	6000

*All values except pH are in mgL^{-1}

Experimental Setup

A UASB reactor was fabricated to study the operational and performance characteristics. The volumetric ratios of acidogenic (Phase 1) and methanogenic (Phase 2) reactors were 1:4. The first phase was an acidogenic reactor (300 mm inner diameter and 820 mm height) made up of plexi-glass with working volume of 56 L and second phase was a stainless steel methanogenic reactor (350 mm inner diameter and 2400 mm height) with working volume of 230 L. The untreated real effluent was fed into the acidogenic reactor followed by methanogenic reactor. Sampling ports were provided at various heights of reactor approximately 400 mm. On the top of the reactor, GLSS was attached. The Gas-Liquid-Solid Separator (GLSS) was attached at top of the reactor, consisted of an inverted conical funnel at top of the water column for the collection of biogas. In addition to the GLSS arrangement, a packed medium consisting of a PVC spirals with size of 26 mm, surface area 500 m^2 m^{-3} and void ratio 87 % has been provided for a height of 200 mm located at 1770 mm from the bottom of the reactor. These spirals will retain the biomass in addition will give a polishing effect to the effluent. The sludge granules trapped in GLSS and the spirals will return to the reactor as soon as the gas entrapped inside the granules was released. Biogas generated was measured using wet gas flow meter. After stabilizing the reactor, studies were conducted under the steady state conditions. The reactor was operated under room temperature (30±2 ^{0}C) at different Hydraulic Retention Time (HRT) of 36, 30, 24 and 18 h. The pH, TDS and temperature of both the reactors were monitored continuously at every 15 minutes by programmable logic control (PLC).

Analytical procedures

The treated and untreated samples were analyzed for pH, COD and colour removal, VFA and alkalinity as per Standard Methods for Examination of Water and Wastewater (APHA 2005). Colour removal was estimated by monitoring the Optical Density (OD) of the samples using UV-vis spectrophotometer (HITACHI – U2001) at 600 nm. COD was monitored by using closed reflux titrimetric method. VFA and Alkalinity was also estimated by titrimetric method. Before analysis, all samples were filtered through 0.45 mm filters to remove suspended matters.

III. Result And Discussions

Reactor performance

The UASB reactor was operated with real textile dye and sago wastewater with optimum mixing ratio (70:30) (Senthilkumar et al. 2010) at four different HRTs in order to obtain overall kinetic coefficients for different models at steady-state conditions. The results obtained under steady-state condition during the reactor operation at four different HRTs are summarized in Table 2. The COD removal efficiency in acidogenic and methanogenic reactor was in the range of 20-35 % and 76-85 % respectively. Colour removal efficiencies in acidogenic and methanogenic reactor were in the range of 35-45 % and 65-74 % respectively. The overall COD and colour removal efficiency of real wastewaters in two-phase upflow anaerobic sludge blanket reactor was 90.1 % and 85.42 % at 24 h of HRT. The biogas production at 24 h HRT was 312 l/d. The pH in the

methanogenic outlet was in the range of 7.29-7.67. The VFA in acidogenic and methanogenic reactors were in the range of 738-746 mg/l and 56 mg/l respectively and VFA concentration is increased in both reactors when HRT is decreased. The methanogenic reactor effluent alkalinity and VFA/Alk ratio is in the range of 934-966 mg/l and 0.057-0.059 respectively. VFA/Alk ratio is increased when HRT is decreased. Volatile Fatty Acid (VFA)/Alkalinity ratio can be used as a measure of process stability. In this study in methanogenic reactor the pH and VFA concentration was under stable condition according to Snadhya and Swaminathan (2006). The optimal pH could be explained by the neutralization of hydrogen anions being released from the volatile fatty acid together with the carbonates dissociated from the carbonic acid with the bicarbonate alkalinity inside the UASB reactor (Razo- Flores *et al*., 1997). The maximum biogas production of 356 L/d was achieved at 18 h of HRT.

Table 2 Experimental data obtained for Pilot scale two-phase UASB reactor under steady state conditions at four different HRTs.

Parameter	**HRT (h)**							
	4.5	18	6	24	7.5	30	9	36
	Aci.	Meth.	Aci.	Meth.	Aci.	Meth.	Aci.	Meth.
pH	6.32	7.29	6.85	7.52	6.75	7.61	7.14	7.67
Inlet COD (mg/l)	4480	3280	4880	3200	4640	3520	4720	3760
Outlet COD (mg/l)	3280	560	3200	480	3520	720	3760	880
COD removal (%)	32.78	82.92	34.42	85	24.13	79.54	20.33	76.59
Influent Colour λmax @ 600 nm	0.295	0.186	0.295	0.164	0.294	0.177	0.294	0.191
Effluent Colour λmax @ 600 nm	0.186	0.065	0.164	0.043	0.177	0.058	0.191	0.059
Colour removal (%)	36.95	65.05	44.41	73.78	39.79	67.23	35.03	69.11
Biogas (L/d)		356		312		244		225
VFA ($mgCH_3COOH/l$)	744	56	738	56	746	56	746	56
Alkalinity ($mg\ CaCO_3 / l$)	561	946	566	966	542	956	575	934

Haldane model

In order to acquire the kinetic coefficients of Haldane model is plotted in Figure 1 and 2. For acidogenic reactor the values of kinetic coefficients obtained from Figure 1 are K_s=683.51 mg/L and K_i=3865.48 mg/L on account of larger values of K_i, indicating no inhibition effect on the growth rate of microorganisms. The values of a, b and c are found to be 0.013, 50.14 and 34347 respectively with high correlation coefficients of (R^2) 0.96. For methanogenic reactor the values of kinetic coefficients obtained from Figure 2 are K_s=422.43 mg/L and K_i=559.16 mg/L on account of larger values of K_i, indicating no inhibition effect on the growth rate of microorganisms. The values of a, b and c are found to be 0.034, 19 and 8031 respectively with high correlation coefficients of (R^2) 0.962.

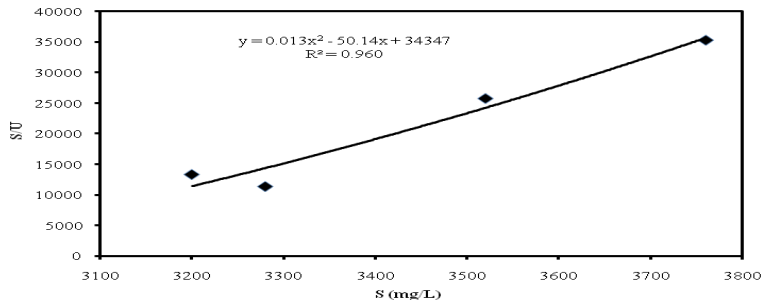

Fig. 1 Determination of half velocity saturation constant K_s and inhibition constant K_i for Haldane model (Acidogenic reactor)

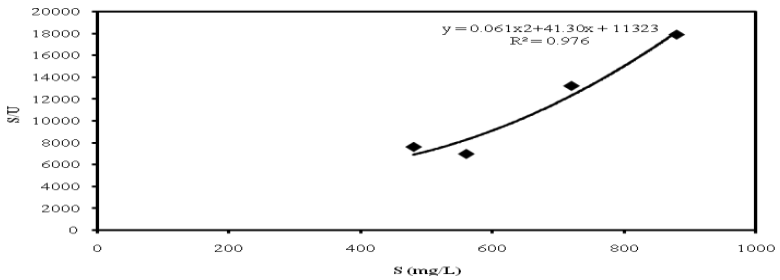

Fig. 2 Determination of half velocity saturation constant K_s and inhibition constant K_i for Haldane model (Methanogenic reactor)

Modified Stover-Kicannon model

The modified Stover–Kincannon model was applied to experimental results from the continuously operated two-phase upflow anaerobic sludge blanket reactor for decolourization and degradation of combined textile dye and sago wastewater, kinetic constants for COD removal were determined at different hydraulic retention times. In addition, there are kinetic models developed for organic substance removal in continuously operated anaerobic reactors (APHA-AWWA, 2005). Figures 3 and 4 indicates the plot of COD loading $V/(QS_0)$ versus COD removal rate $V/[Q(S_0\text{-}S)]$ of acidogenic and methanogenic reactor. From the figure 3 slope and intercept of a best-fit line (R^2 = 0.977), kinetic constants for COD removal in acidogenic reactor were determined as R_{max}=2.36 $gL^{-1}d^{-1}$ and K_B = 30.3 $gL^{-1}d^{-1}$, respectively. From the figure 4 slope and intercept of a best-fit line (R^2 =0.989), kinetic constants for COD removal in methanogenic reactor were determined as R_{max}=3.48 $gL^{-1}d^{-1}$ and K_B=7.77 $gL^{-1}d^{-1}$, respectively. The kinetic values obtained using acidogenic reactor are comparable with the kinetic constant K_B and R_{max} are 12.9 $gL^{-1}d^{-1}$ and 37.7 $gL^{-1}d^{-1}$ obtained by Kapdan (2005) for anaerobic packed bed reactor treating simulated textile dye wastewater.

Therefore the rate expression for COD in acidogenic reactor takes the following form:

$$\frac{Q(S_0 - S)}{V} = \frac{2.36(QS_0/V)}{30.3 + (QS_0/V)} \quad (1)$$

The rate expression for COD in methanogenic reactor takes the following form:

$$\frac{Q(S_0 - S)}{V} = \frac{3.48(QS_0/V)}{7.77 + (QS_0/V)} \quad (2)$$

and effluent COD concentration of acidogenic reactor can be predicted by rearranging the Eq. (1)

$$S = S_0 - \frac{2.36S_0}{30.3 + (QS_0/V)} \quad (3)$$

Effluent COD concentration of acidogenic reactor can be predicted by rearranging the Eq. (2)

$$S = S_0 - \frac{3.48S_0}{7.77 + (QS_0/V)} \quad (4)$$

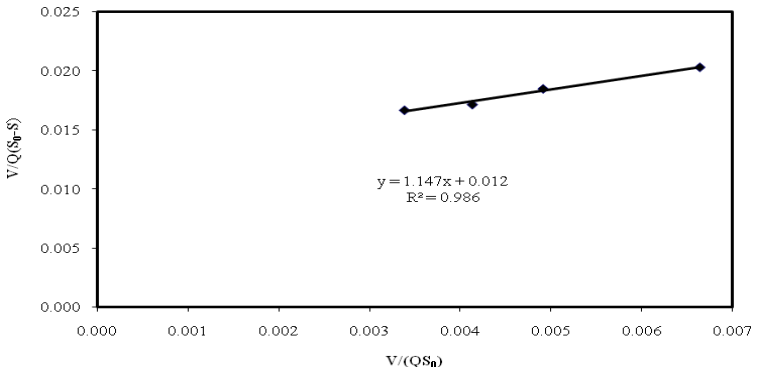

Fig. 3 Stover-Kicannon model plot for acidogenic reactor

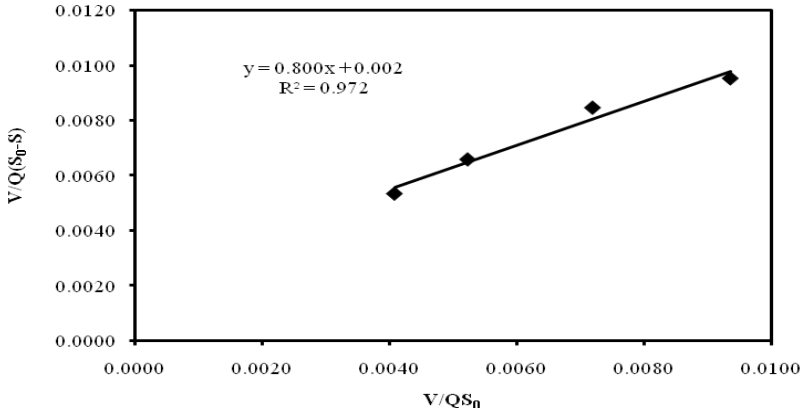

Fig. 4 Stover-Kicannon model plot for methanogenic reactor

Grau second-order multicomponent substrate removal model

In order to determine the kinetic co-efficient (a, b and k_s) was plotted in Figures 5 and 6. From Figure 5 the values of a, and b were found to be 0.841 and 6.936 with high correlation coefficients of (R^2) 0.949. The multicomponent substrate removal rate constant (k_s) for acidogenic reactor was then calculated from the equation $a=S_0/(k_sX)$ as 0.212 per day. From Figure 6 the values of a, and b were found to be 0.197 and 1.42 with correlation coefficients of (R^2) 0.993. The multicomponent substrate removal rate constant (k_s) for methanogenic reactor was then calculated from the equation $a=S_0/(k_sX)$ as 0.418 per day. The values of second order multi-component substrate removal rate constant k_s for both acidogenic and methanogenic reactor was comparable with Isik and Sponza (2005) (k_s=0.337) indicating substrate removal for each unit of microorganism depending on second order substrate removal rate constant (k_s).

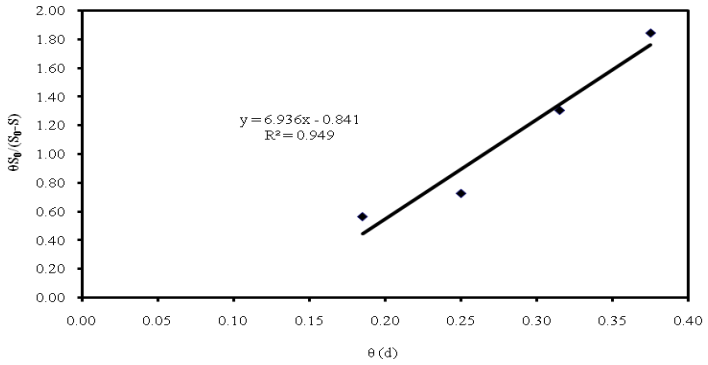

Fig. 5 Determination of kinetic constants (*a*, *b* and k_s) for Grau second order multicomponent substrate removal mode (Acidogenic reactor).

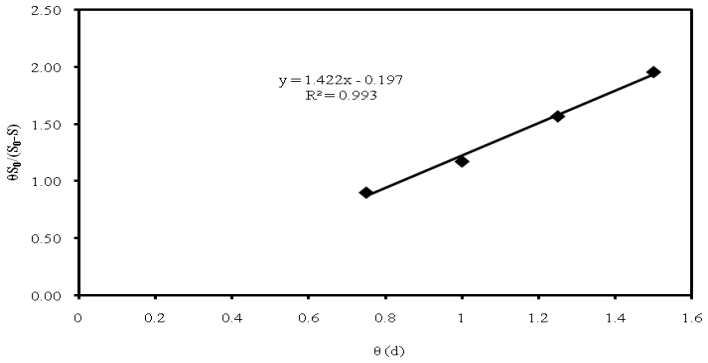

Fig. 6 Determination of kinetic constants (*a*, *b* and k_s) for Grau second order multi-component substrate removal mode (Methanogenic reactor).

Evaluation of the kinetic models

From the engineering point of view, development of kinetic model is a useful tool in designing and optimization of the process by reducing extensive and complex experimental data to simple and convenient mathematical expression. All kinetic coefficients calculated from models are summarized in Table 3 with correlation coefficients. The kinetic data showed that Stover-Kincannon, Grau second order multi-component substrate removal kinetics and Haldane model were more appropriate than the other models for predicting the performance of the pilot scale two phase hybrid UASB reactors when the regression coefficients and kinetic coefficients were compared. The Tables 4a, 4b and 4c summarizes the constants determined from the applicable models in previous studies. In this study, the determined Haldane kinetic coefficients (K_i = 3865.4 mg/L) for acidogenic reactor is comparable with the kinetic coefficient (K_i = 3636.36 mg/L) obtained by Bhunia and Ghangrekar (2008). In case of Grau second order model the k_s (see Table 4a) values obtained in this study was similar to the k_s value (0.337 d^{-1}) found by Isik and Sponza (2005) treating simulated textile dye wastewater using UASB reactor, the k_s value (0.354 d^{-1}) obtained by Bhunia and Ghangrekar (2008) treating synthetic sucrose wastewater using UASB reactor and also the k_s value (0.227 d^{-1}) obtained by Ubay (1994) treating municipal wastewater in UASB reactor. Whereas higher k_s values are obtained by Ozturk *et al*. (1998), Buyukkamaci and Felibeli (2002), Sandhya and Swamynathan (2006), Sandhya *et al*. (2008), Sponza and Ulukoy (2008) compared to the k_s values obtained in this study. The possible reason for differences may be variation in reactor configuration, wastewater characteristics and microorganism used in the study. The modified Stover-Kincannon model suggests that the substrate removal rate is affected by the organic loading rate entering

to the reactor as described. Kinetic coefficients of this model (R_{max}, K_B) were higher than the data obtained in other studies as seen in Table 4b. Whereas the Stover-Kincannon kinetic coefficients R_{max} and K_B (3.48 and 7.77) obtained in this study for methanogenic reactor was similar to the R_{max} and K_B (7.5 and 8.2) obtained by Isik and Sponza (2005). For anaerobic systems a review of common substrate utilization kinetic models was carried out by Kuroda *et al.,* (1993), and also by Mata-Alvarez and Cecchi (1990). The Monod model is most widely used for UASB reactors and for industrial effluents (Castillo *et al*., 1999). In this study, however, it was found that the Monod and Contois model was not appropriate for interpreting the kinetic data of the UASB reactor treating combined real textile and sago wastewater. On the account of poor correlation (R^2<0.3) with the data set Monod and Contois equation have not been considered for the estimation of kinetic constants. The models applied in this study were also evaluated by comparing the predicted COD values with the tentative values obtained from this continuous operation of pilot-scale UASB reactor. The COD values predicted with the Stover-Kincannon and Grau second order models have a high correlation with actual COD concentrations measured from the pilot scale two phase hybrid UASB reactor as shown in Figs. 7 and 8. In conclusion, based on the kinetic studies, it appears that the kinetic coefficients obtained from the anaerobic treatment of combined real textile dyeing and sago wastewater, satisfies with the Stover-Kincannon, and Grau second order multicomponent substrate kinetic models. Although several studies relevant to anaerobic treatment of some industrial wastewaters and also simulated textile dye wastewater were reported in literature, none of the studies contained a kinetic relevant to two phase anaerobic reactors treating a combined real textile dyeing and sago wastewater. As seen in Table 3, a Grau second order kinetic model gives nearly the same correlation as the Stover-Kincannon. This result is not surprising because two models are very similar as mentioned below:
Second order linearized equation could be transformed to the modified Stover-Kincannon model by dividing S_o each part the equation.

$$\frac{V}{Q\times(S_0-S)}=\frac{V}{Q\times S_0}+\frac{1}{k_s\times X} \qquad (5)$$

If Eq. (5) is simulated to the modified Stover-Kincannon model, the following equation can be obtained:
$K_B = R_{max}$
The calculated kinetic parameters from the plots of both models are appropriate to the values calculated from the equations given above. As a result of this similarity, the two models have the same sensitivity in order to obtain the kinetic coefficients from the plots when the regression coefficients of the graphs are compared. The similar results were obtained by Isik and Sponza (2005) and Sandhya et al. (2008).

IV. Conclusions

In this study, the kinetics of UASB reactors treating real textile dyeing wastewater with sago wastewater as co-substrate was investigated using different models such as Monod, Contois, Haldane, Grau second order and modified Stover-Kincannon. On the account of poor correlation (R^2<0.3) with the data set Monod and Contois equation have not been considered for the estimation of kinetic constants. Among the other three kinetic models, Grau second order model and Stover- Kincannon model were observed to be the preeminent model for predicting the performance of two phase hybrid UASB reactors for treating combined real textile dyeing and sago wastewater with higher average regression coefficients (R^2=0.979). The results of kinetic studies obtained from pilot-scale experiments can be used for estimating treatment efficiency of full-scale reactors with the same operational conditions.

Table 3 Kinetic parameters of UASB reactor treating textile dyeing and sago wastewater

Kinetic models	Kinetic parameters	Values	Regression Coefficient (R^2)
Pilot scale Acidogenic reactor			
Haldane	a	0.013	0.96
	b	50.14	0.96
	c	34347	0.96
	μ_{max} (per day)	0.019	0.96
	Ks (mg/L)	683.5	0.96
	Ki (mg/L)	3865.4	0.96
Grau second order	a (per day)	0.841	0.949
	b (dimensionless)	6.936	0.949
	k_s (per day)	0.212	0.949
Modified Stover-Kincannon	K_B (g (l per day))	6.06	0.955
	R_{max} (g COD (l per day))	41	0.955
Pilot scale Methanogenic reactor			
Haldane	a	0.034	0.962
	b	19	0.962
	c	8031	0.962
	μmax (per day)	0.0526	0.962
	Ks (mg/L)	422.43	0.962
	Ki (mg/L)	559.16	0.962
Grau second order	a (per day)	0.197	0.993
	b (dimensionless)	1.422	0.993
	k_s (per day)	0.418	0.993
Modified Stover-Kincannon	K_B (g (l per day))	16.12	0.983
	R_{max} (g COD (l per day))	23.17	0.983

Table 4a Comparison of Grau second order kinetic constants

Model	Substrate	Reactor Type	Influent COD (mg/L)	HRT (d)	Kinetic parameters ks	a	b	References
Grau second order	Municipal wastewater	UASB	230-445	0.25-1.0	0.217	0.002	1.346	Ubay (1994)
Grau second order	Landfill leachate	UASB	9000-25000	1.7-2.80	38.5	0.013	1.066	Ozturk et al. (1998)
Grau second order	Simulated textile wastewater	UASB	4214	0.25-4.16	0.337	0.562	1.095	Isik and Sponza (2005)
Grau second order	Textile wastewater	UAFB	1835-3828	0.4-0.99	10.50	0.915	5.14	Sndhya and Swamynathan (2006)
Grau second order	Synthetic sucrose wastewater	UASB	300-4000	0.16-0.33	0.354	0.558	1.043	Bhunia and Ghangrekar (2008)
Grau second order	2,4 DCP	UASB	4000	0.08-0.83	0.26	0.029	0.0113	Sponza and Ulukoy (2008)
Grau second order	Textile wastewater	AHR	1835-3828	2.3-9.1	40.26	0.013	0.007	Sndhya et al. (2008)
Grau second order	Comibned real Textile Dyeing and sago Wastewater	Pilot UASB Acidogenic	3500-6000	0.19-0.38	0.212	0.841	6.93	In this study
		Methanogenic	3500-6000	0.75-1.5	0.42	0.197	1.422	

Table 4b Comparison of modified Stover-Kicannon kinetic constants

Model	Substrate	Reactor Type	Influent COD (mg/L)	HRT (d)	Kinetic parameters Rmax	K_B	References
Modified Stover-Kicannon	Simulated textile wastewater	UASB	4214	0.25-4.16	7.5	8.2	Isik and Sponza (2005)
Modified Stover-Kicannon	Simulated Textile wastewater	UAFB	1835-3828	0.4-0.99	31.69	45.37	Sndhya and Swaminathan (2006)
Modified Stover-Kicannon	2,4 DCP	UASB	4000	0.08-0.83	0.0098	0.01	Sponza and Ulukoy (2008)
Modified Stover-Kicannon	Comibned real Textile Dyeing and sago Wastewater	Pilot UASB Acidogenic	3500-6000	0.19-0.38	2.36	30.30	In this study
		Methanogenic	3500-6000	0.75-1.5	3.48	7.77	

Table 4c Comparison of Haldane kinetic constants

Model	Substrate	Reactor Type	Influent COD (mg/L)	HRT (d) a	Kinetic parameters b	c	Ks	Ki	References
Haldane	Synthetic wastewater	UASB	300-4000	0.16-0.33	0.0005	1.82	304.2	167.2 3636.36	Bhunia and Ghangrekar (2008)
Haldane	Comibned real Textile Dyeing and sago Wastewater	Pilot UASB Acidogenic	3500-6000	0.19-0.38	0.013	50.1	34347	683.5 3865.4	In this study
		Methanogenic	3500-6000	0.75-1.5	0.034	19	8031	422.43 559.2	

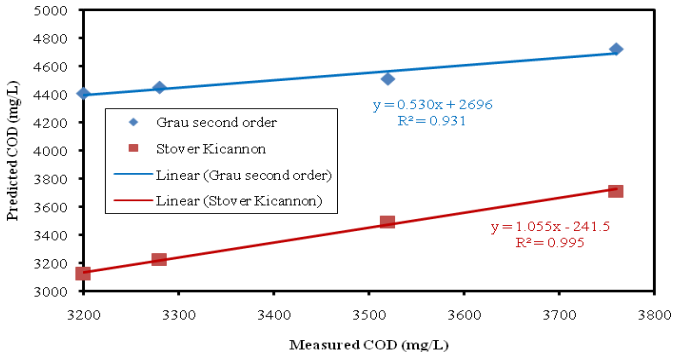

Fig 7 Comparison of the predicted and the actual COD values from the pilot-scale acidogenic UASB reactor.

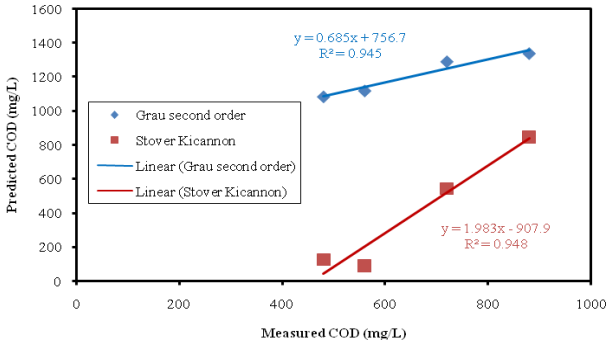

Fig 8 Comparison of the predicted and the actual COD values from the pilot-scale methanogenic UASB reactor.

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