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MATHEMATICAL MODEL DEVELOPMENT FOR BOD REMOVAL EFFICIENCY IN TANNERY WASTE TREATMENT

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ABSTRACT: *The composite tannery waste water has been treated by anaerobic digestion using varying organic loads. The chromium present has been removed by precipitation method using lime as the agent. The treated water was analysed for pH, influent BOD (So), effluent BOD (Se), mixed liquor volatile suspended solids (MLVSS) before sludge wasting, initial MLVSS and the net growth rate of microorganisms. Results indicated that the BOD and COD removal efficiency decreases with decrease in mean cell residence time θc and decrease with increase in F/M ratios. A maximum BOD reduction of 96.9% was obtained at the BOD loading rate of 0.80 kg BOD/m^3/day and throughout different loading rates and modified Monod's equation was used to determine the biokinetic coefficients. A mathematical model was developed to predict the BOD removal efficiency and the model signifies the prediction with a deviation of ±3.6.*

KEYWORDS: Tannery waste, Biokinetic constants, Mathematical model

INTRODUCTION

The principal raw material is the hide or skin of animals including that of reptiles, fish and birds. The tannery operation involves converting the raw skin, a highly putrescible material, into leather, a stable material, which can be used in the manufacture of a wide range of products. The whole process involves a sequence of complex chemical reactions and mechanical processes. Performing various steps of pre- and post-treatment, generates a final product with specific properties: stability, appearance, water resistance, temperature resistance, elasticity and permeability for perspiration and air, etc .

Leather is an intermediate industrial product, with numerous applications in down-stream sectors of the consumer products industry. For the latter, leather is often the major material input and is cut and assembled into shoes, clothing, leather goods, furniture and many other items of daily use. The tanning of hides and skins also generates other by-products, which find outlets in several industrial sectors such as-dog biscuits and other animal food production, fine chemicals including photography and cosmetics, soil conditioning and fertilizers. The process of making leather has always been associated with odour and water pollution Tanning of animal hides to convert them into leather is an important industrial activity. But the pollution from tanneries has a long-term negative impact on the environmental resources. The liquid waste from tanneries is a dangerous pollutant because it contains organic matter and inorganic pollutants in the solution, in suspension as well as in colloidal dispersion. Hence, there is a need to remove these pollutants before they are

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released to render them harmless. In the past ten years, a number of different anaerobic processes have been developed for the treatment of industrial wastes. Anaerobic digestion is one of the oldest processes used for the stabilization of sludges. It involves the decomposition of organic and inorganic matter in the absence of molecular oxygen. In this process, the organic matter in the mixture of primary settled and biological sludges is converted biologically, under anaerobic conditions, to a variety of end products including methane and carbondioxide. The process is carried out in an airtight reactor. Sludge, introduced continuously or intermittently, is retained in the reactor for varying periods of time. The stabilized sludge, withdrawn continuously or intermittently from the reactor, is reduced in organic and pathogen content and is non-putrescible. As the name anaerobic refers, the anaerobic digestion is carried out by microorganisms that can only live in an oxygen free environment. The decomposition of biowaste occurs in four stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis as shown in figure 1and the bacterial growth for stabilization of organic pollutants is represented in figure 2.

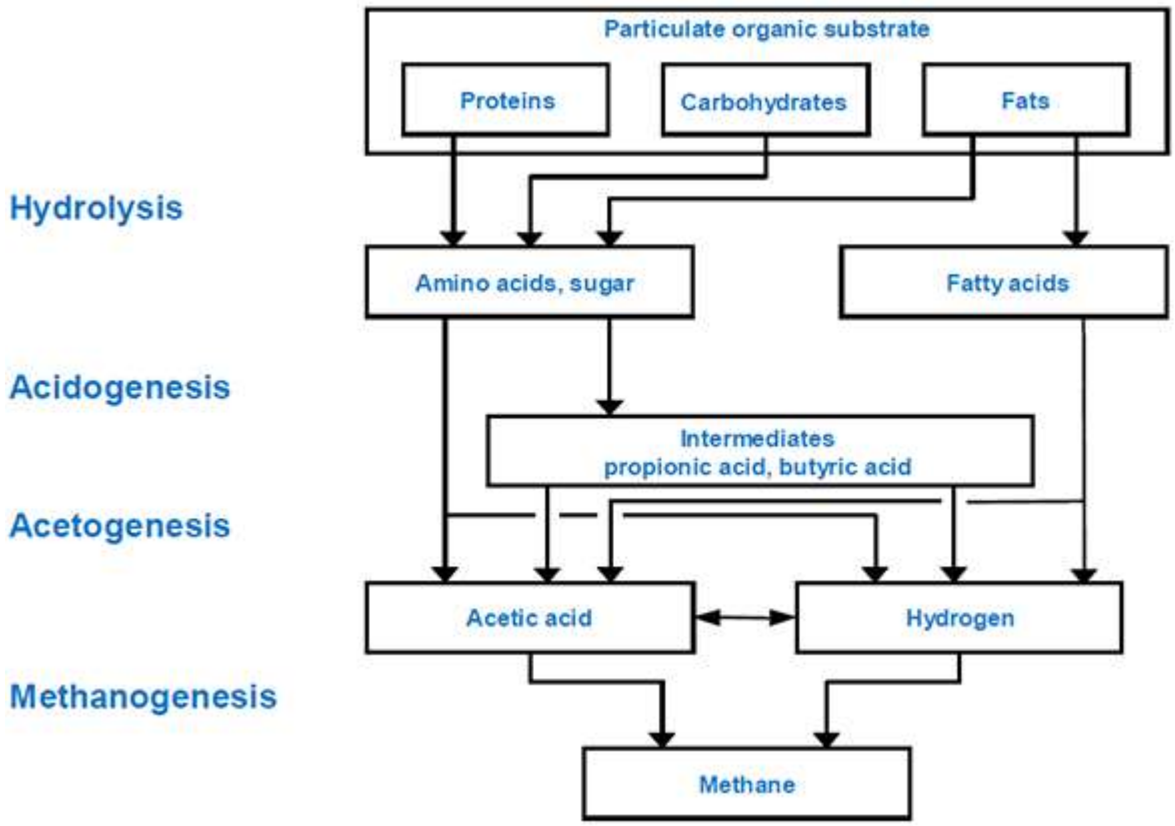

Figure 1: Degradation steps of anaerobic digestion process

$C_6H_{10}O_4 + 2H_2O \rightarrow C_6H_{12}O_6 + 2H_2$
$C_6H_{12}O_6 \leftrightarrow 2CH_3CH_2OH + 2CO_2$
$C_6H_{12}O_6 + 2H_2 \leftrightarrow 2CH_3CH_2COOH + 2H_2O$
$C_6H_{12}O_6 \rightarrow 3CH_3COOH$

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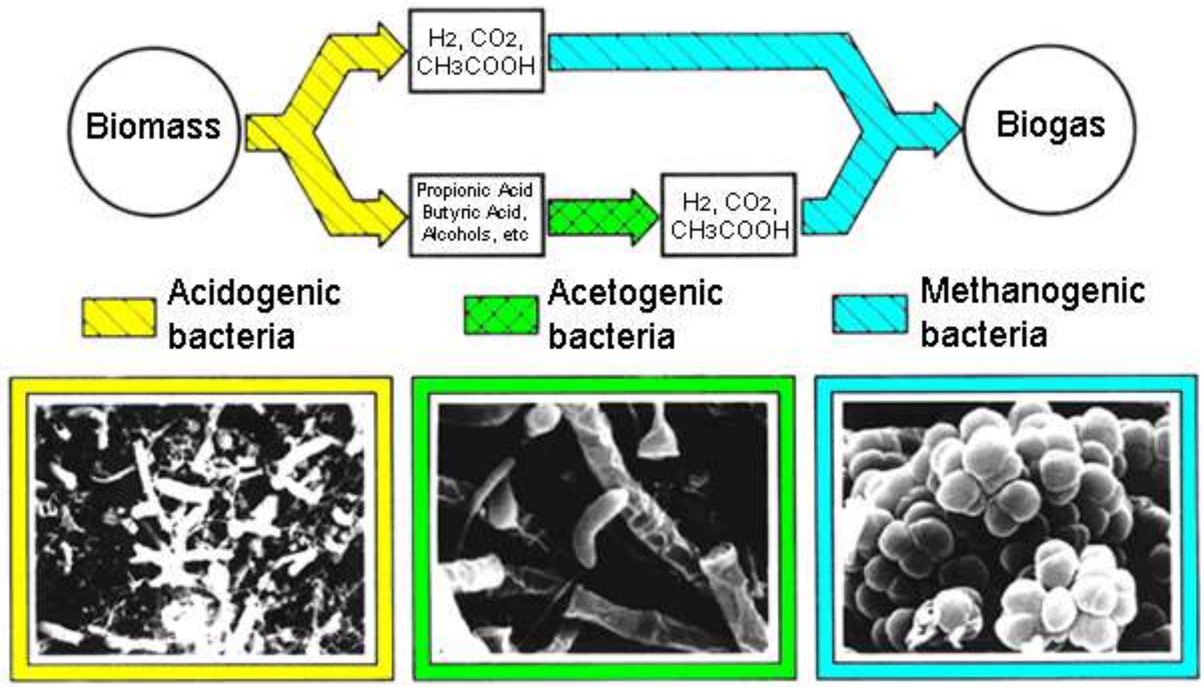

Figure 2. Schematic representation of Bacteial growth

$CH_3CH_2COO^- + 3H_2O \leftrightarrow CH_3COO^- + H^+ + HCO_3^- + 3H_2$
$C_6H_{12}O_6 + 2H_2O \leftrightarrow 2CH_3COOH + 2CO_2 + 4H_2$
$CH_3CH_2OH + 2H_2O \leftrightarrow CH_3COO^- + 2H_2 + H^+$
$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$
$2C_2H_5OH + CO_2 \rightarrow CH_4 + 2CH_3COOH$
$CH_3COOH \rightarrow CH_4 + CO_2$

Environmental pollution has become major concern in developing countries in the last few decades. Major sources of water pollution are the untreated or partially treated industrial effluents. Tanning industry is reputed globally as major industry which contributes to water pollution. The quality of discharged water from tanneries is far from the desired level of acceptance into water ways. A tannery discharges from 21,500-21,950 liters a day, corresponding to 86-88 liters per kg of leather processed. Chromium is known to be highly toxic to the living aquatic organism in the hexavalent state and somewhat less toxic in the trivalent form. The effluents from chrome tanning industry shall meet with the specific tolerance limits for chloride with 1000 mg/L, BOD (5 day at 20^0C)with 30 mg/L ,hexavalent chromium with 0.1mg/L and pH between 5.5 to 9.0.

The study on Hexavalent Chromium removal from Industrial waste water by chemical precipitation method was reported (C.R.Ramakrishnaiah and B.Prathima, 2012). The characterization, kinetics and thermodynamics studies were carried out for the removal of hexavalent chromium from wastewater by FeO-nanoparticles-chitosan composite beads (Liu TY et al. 2012). The separation of the chromium(III) dissolved in a tanning wastewater was studied by means of precipitation with calcium carbonate, reverse osmosis with polyamide membrane and

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adsorption on activated carbon(B.H.Hintermeyer et al.2008). The Electrochemical precipitation of chromium (Cr^{6+}) from an electroplating wastewater was reported (N. Kongsricharoern, C. Polprasert,1995). The Chromium removal from Tannery waste water using chemical and biological Techniques for zero discharge pollution was reported.(Hesham et al.2010). Tanning process using chromium compounds is the most common methods for processing of hides (Sreeram, and Ramasami, 2003). In this process about 60% - 70% of chromium reacts with the hides.

Although actinomycetes constitute a significant component of the microbial population in most environments, their metabolic diversity and genomic characteristics indicate them as well suited agents for bioremoval of metal and organic compounds (Polti *et al*., 2007). Recent studies showed chromium bioremoval by *Streptomyces rimosus* generated from the antibiotic industry (Sahmoune and Louhab, 2008) and biological reduction of chromate by *Streptomyces griseus* (Poopal and Laxman, 2009).

Arumugam has reported on the recovery of chromium from spent chrome tan liquor by chemical precipitation using lime (V.Arumugam 1976). Pathe et al. have studied the properties of chromium sludge from chrome tan liquor and related the sludge volume, sludge settling rate, surface loading rate etc (Pathe P P et al.1995). The treatment of tannery and electroplating effluents by using lime, NaOH and their mixture in the temperature range of 25 to 100^0C have been conducted (Archana Shukla and N P Shukla,1994). A study was conducted on a laboratory scale completely mixed continuous flow activated sludge system to treat settled chrome tannery wastewater and observed that the BOD and COD removal ranged from 84 to 96%.(Gurusamy et al 1995). The experiments were conducted on the activated sludge treatment of vegetable tanning waste admixtured with 10, 25, and 50% settled sanitary sewage and obtained BOD removal from 87 to 96%.(Elangovan et al 1995).

MATERIALS AND METHODS

The experiment was designed and operated on the principle of an anaerobic activated sludge process to evaluate the bio-kinetic parameters, which could be used in the rational design and operation of large-scale anaerobic installations. The reactor was a wide mouthed Pyrex glass bottle of 5 liter capacity as shown in figure 3. The reactor has provision for adding wastes, for removing treated effluent and settled solids and for gas transfer. The gas collection apparatus consisted of a glass bottle of 2 liter capacity and another bottle of I liter capacity for the water displaced from the gas bottle. Care was taken to remove the air from the reactor as well as from the gas collection bottle at the beginning of the experiment and the entire set up was checked for gas leaks. Tubes were connected to the digester to facilitate feeding of the west and removal of the effluent. The digester was kept in water bath at a constant temperature of 35^0C. Cow dung was used as the seed material and fed into the digester to start with. After establishing necessary biota from cow dung sludge, the chromium free composite liquor is fed into the digester daily. The pH of the influent sample was adjusted to pH 7 by adding alkali before feeding. After feeding, the contents in the digester were given thorough mixing by manual shaking. The BOD load was kept at 0.25kg BOD/m^3/day in the beginning. After several displacements of the digester contents and after

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establishing stable conditions of digestion the loading rate was gradually increased. Gas measurements were done once a day. The gas was burnt periodically to confirm the presence of methane which formed a major portion of a gas.

The samples of effluents' drawn at various stages were analyzed for pH, influent BOD (So), effluent BOD (Se), mixed liquor volatile suspended solids (MLVSS) before sludge wasting, initial MLVSS and the net growth rate of microorganisms $\Delta X/\Delta t$ which was obtained from the difference of MLVSS before sludge wasting and initial MLVSS values. The pH was maintained within the optimum range of 6.8 to 7.4 which is favorable for anaerobic bacterial growth. Calculated amount of diammonium phosphate and urea were added to the feed solution as and when required in order to maintain the BOD: N : P ratio at 100 : 2.5: 0.5 which is effective for anaerobic digestion. In anaerobic digestion, biomass is formed having a molecular formula $C_5H_7O_2N$. Cell synthesis requires Nitrogen (amino acid formation) for which Nitrogen (in the form of Urea) rich nutrient is supplied. During cell synthesis, energy in the form of ATP is released for which phosphorus acts sink. The contents in the reactor were continuously mixed with the help of magnetic stirrer. The tannery wastewater was filled up to a volume of 2 litres in the anaerobic reactor and the mixture was mixed daily at frequent intervals. Neither waste feeding nor withdrawal of mixed liquor was done until gas production was noticed. Regular wasting and feeding were continued until a steady state condition was reached. The daily BOD loading rate was kept constant at around 0.3 kg/m^3/day. The daily gas production, the influent and effluent BOD, Mixed Liquor Volatile Suspended Solids (MLVSS) which indicates the concentration of microorganisms in the reactor, pH, volatile acids and alkalinity were recorded at the steady state condition at which the sludge growth and gas production remained constant. The mean cell residence time was varied by operating the reactor at several MLVSS concentrations.

RESULTS AND DISCUSSION

The general characteristic properties of composite tannery effluent are presented in table 1. The result indicates that the liquor is basic with pH 7.8. The chromium content has been found to 120 mg/Land the BOD and COD of the effluent have been estimated to be1360 and 2510mg/L respectively. The results indicate that the effluent has to be treated for an effective removal of chromium before being subjected to biological treatment. Lime was used as the precipitating agent for chromium removal and effect of lime on chrome precipitation is presented in table 2. It was observed that the chromium removal increased with increase in pH and the maximum chromium removal of 99.7% was observed at pH at 8.9 with a lime dose of 4.4 g/L. Further increase in lime has resulted in the decrease of chromium removal due to redissolution of the mixture under such experimental conditions. The results of the anaerobic digestion of the chrome free composite liquor are presented in table 3. The data consists of varying BOD loading rate changes in pH alkalinity, volatile acid, and percentage BOD reduction. It was observed that a maximum BOD reduction of 96.9% was obtained at the BOD loading rate of 0.80 kg BOD/m^3/day and throughout different loading rates, The BOD reduction was more than 94% which could be due to the proper maintenance of alkalinity and volatile acids in the digester. In the beginning of the process, the pH of the effluent was 6.9. As the loading increased gradually the pH increased to 7.6 up to the optimum loading and dropped down slightly to 7.4 at the maximum loading. The

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increase in alkalinity was steady as the loading increased gradually. Side by side there was a production of volatile acids but was not considerable. With the initial pH correction and with proper seeding of the waste, the process of digestion was taken place unhindered, without undue accumulation of intermediate products. There was no possibility for the formation of free volatile acids.

The optimum organic load required for the maximum removal of BOD and COD of the effluent has been determined and it was observed that the BOD and COD removal reached a maximum of 92.8% . During the digestion, the mean cell residence time θc was varied by operating the reactor at varying food to microorganism ratio(F/M) by varying the MLVSS concentration. Results indicated that the BOD and COD removal efficiency decreases with decrease in mean cell residence time θc and decrease with increase in F/M ratios as represented in table 4. The substrate removal constants, namely, half saturation concentration (K_s) and the maximum rate of substrate utilization (k) were determined from the Lawrence and McCarty's modified Monod equation given below:

$$\frac{1}{U} = \left(\frac{Ks}{k}.\frac{1}{S}\right) + \frac{1}{k}$$

S = Substrate (SCOD and NH_4^+-N) concentration at any time in reactor (mg/L), U = Specific substrate utilization rate = $(S_0 - S)/\theta X$ (mg of SCOD or mg of NH_4^+-N/day/mg of MLVSS), θ = Contact time (day), X = MLVSS at any time in the reactor (mg/L), S_0 = Substrate (SCOD and NH_4^+-N) concentration of the influent (mg/L).

The substrate removal kinetics, such as the substrate removal rate constant (k) and the half-velocity constant (Ks) could be evaluated from the intercept and slope of the straight line. The values of the reciprocal of the mean cell residence time θc were plotted against the specific substrate utilization rate (U) .

The sludge growth kinetic constants namely the yield coefficient (Y) and the endogenous decay coefficient (K_d), were determined from the Lawrence and McCarty's modified Monod equation given below:

$$\frac{1}{\theta} = \mathrm{YU} - \mathrm{Kd}$$

The yield coefficient(Y) was determined from the slope of the straight line and the endogenous decay coefficient was obtained from the intercept. The biokinetic coefficients were evaluated using modified Monod's equations and were given in table 5. The rate of substrate utilization was found to be higher at the early stages of digestion throughout the process and the reason for the initial high rate may be due to the adsorption of soluble substrate by the bacteria and extra cellular slime. The subsequent drop in rate following the initial high rate may be interpreted as saturation of adsorption sites. The subsequent rate increase may be attributed to continued increase in metabolic activities caused by cell growth. Adsorption and metabolism occur concurrently.

Multiple Linear Regression

The biokinetic data has been applied into the mathematical model as derived and shown in table 6. Many formulae can be used to ascertain the relationships among more than two variables, the most frequently used method is that of linear equations. The multiple linear regression takes the following form,

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$Y = a+b_1X_1+b_2X_2+b_3X_3+\ldots\ldots+bxXx$ ……..(3)

In the present study, the multiple linear regression technique has been used to predict the BOD removal efficiency using the biokinetic coefficients calculated using modified Monod's equations for the data obtained during anaerobic digestion of effluent.

If X_1 and X_2 are the substituted with biokinetic constants, then,

$X_1 = \frac{k}{Ks}.\frac{V}{Q}.X$ …….(4)

$X_2 = \frac{BOD_{influent}}{BOD_{effluent}}$ ……(5)

Where,

k = Substrate removal rate constant

Ks = Half velocity constant

V/Q = Hydraulic retention period

X = Mixed Liquor volatile suspended solids

Substituting X_1 and X_2 in equation 3, the Y_{eff}, BOD removal efficiency takes the form,

$Y_{eff} = a + b_1\frac{k}{Ks}.\frac{V}{Q}X + b_2.\frac{BOD_{influent}}{BOD_{effluent}}$

The constants a, b_1 and b_2 are determined by the methods of least squares by solving the systems of equations.

$\Sigma Y = a+b_1\Sigma X_1+b_2\Sigma X_2$

$\Sigma X_1Y = a\Sigma X_1 + b_1\Sigma X_1^2+b_2\Sigma X_1X_2$

$\Sigma X_2Y = A\Sigma X_2 + b_2\Sigma X_1X_2 + b_2\Sigma X_2^2$

The constants a, b_1, b_2 are evaluated by taking data of the present study under varying organic loads. The data used for the determination of multiple regression coefficients are presented in table 4. The regression coefficients a, b_1 and b_2 evaluated are given below.

a = 77.5

$b_1 = 0.98$

$b_2 = 0.97$

Hence, the proposed model for the prediction of BOD removal efficiency is given by the following equation.

$Y_{cal} = 77.5 + 0.98\left[\frac{k}{Ks}.\frac{V}{Q}.X\right] + \left[\frac{BOD_{influent}}{BOD_{effluent}}\right]$ …….(7)

The analysis of the data on anaerobic digestion of tannery effluent according to equation 7 has been shown in table 7 which gave a maximum deviation of ± 3.6 as shown in figure 4. Thus the results of the present study on the prediction of BOD removal efficiency using multiple linear regression technique has successfully arrived at a model for anaerobic digestion process of tannery effluent.

CONCLUSIONS

1. The optimum organic load required for the maximum removal of BOD and COD of the effluent has been determined to be 0.8 kg BOD/m^3/day
2. BOD and COD removal efficiency decreases with decrease in mean cell residence time θc and decrease with increase in F/M ratios
3. BOD and COD removal reached a maximum of 97%

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4. The mathematical model developed by incorporating the biokinetic coefficients proved fit with a maximum deviation of ±3.6.

Nomenclature:
K=Substrate removal rate constant(day^{-1}), Ks=Half velocity constant(mg/L), Yc=Yield coefficient, Kd=Decay coefficient(day^{-1}), μ_{max}=Maximum specific growth rate of microorganisms(day^{-1}), θ = Hydraulic retention time(day), θc= Mean cell residence time(d), So=Influent BOD(mg/L), Se=Effluent BOD(mg/L), X= Concentration of microorganisms in the reactor.

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Table 1 General characteristics of Composite Tan liquor

Parameter	Value
pH	7.8
Alkalinity (as $CaCO_3$)	1100
Total solids	22400
Total dissolved solids	20890
Total suspended solids	1510
Volatile suspended solids	810
Chlorides	7600
Sulphates	2840
BOD	1360
COD	2510
Chromium	120
Sulphide	90

All Values except pH are expressed in mg/L

Table 2 Effect of Lime on Chromium Precipitation.

Weight of lime added(g/Lt)	pH	Chromium in filtrate (mg/L)
4.0	9.6	3.64
4.5	9.9	2.89
5.0	10.3	2.66
5.5	10.9	2.00
6.0	11.4	1.64
6.5	11.6	0.90
7.0	11.9	0.40
7.3	12.0	0.54
7.8	12.3	0.66
8.5	12.4	0.68

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Table 3 Anaerobic digestion of Composite liquor.

BOD load (Kg BOD/m^3/day	pH	Alkalinity (as $CaCO_3$)	Volatile acids	% BOD reduction
0.25	6.9	340	40	94.6
0.30	7.0	410	60	95.0
0.35	7.2	560	76	95.2
0.40	7.4	950	110	95.4
0.50	7.4	980	144	96.0
0.60	7.6	1460	168	96.2
0.70	7.6	1880	190	96.8
0.80	7.6	1920	236	96.9

Table 4 Anaerobic digestion of Tan Liquor with varying organic load

Organic load (Kg BOD/m^3/day)	Final BOD (Kg BOD/m^3/day)	MLVSS (mg/L)	HRT (day)	θc (day^{-1})	U (day^{-1})	BOD removal efficiency (%)
0.3	650	2140	3.2	7.8	0.31	83.8
0.4	580	2380	3.6	7.2	0.38	85.5
0.5	500	4390	3.8	7.0	0.40	87.6
0.6	350	3570	3.9	6.2	0.44	91.3
0.7	200	4160	4.1	5.6	0.60	95.0
0.8	180	4450	4.2	5.3	0.68	96.6

Table 5 Biokinetic coefficients

Organic load (Kg BOD/m^3/day)	**k(day^{-1})**	**Ks(mg/L)**	**Kd(day)**	**Y**	**μ_m(day^{-1})**
0.30	1.80	1620	0.064	0.76	1.368
0.40	1.66	1510	0.066	0.68	1.129
0.50	1.23	1380	0.080	0.66	0.811
0.60	1.46	1300	0.076	0.70	1.022
0.70	1.52	1240	0.068	0.66	1.003
0.80	1.58	1210	0.056	0.63	0.995

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Table 6 Application of biokinetic data in the mathematical model

X_1	X_2	Y_{expt}	X_1^2	X_2^2	X_1Y	X_2Y	X_1X_2
7.6	0.461	83.8	57.76	0.212	636.88	38.632	3.5036
9.4	0.690	85.5	88.36	0.476	803.7	58.995	6.486
11.8	1.0	87.6	139.24	1.0	1033.68	87.60	11.8
15.6	1.714	91.3	143.36	2.938	1424.28	156.49	26.74
18.0	3.5	95.0	324.0	12.25	1710.0	332.50	63.0
19. 6	4.8	96.8	420.0	18.6	2042.6	433.2	75.46

Table 7 Results of the Mathematical Model

X_1	X_2	Ycal	Yexpt	Deviation
7.6	0.461	85.3	83.8	1.5
9.4	0.690	87.4	85.5	1.9
11.8	1.0	90.0	87.6	2.4
15.6	1.714	94.4	91.3	3.1
18.0	3.5	98.6	95.0	3.6

Figure 3. Anaerobic Reactor

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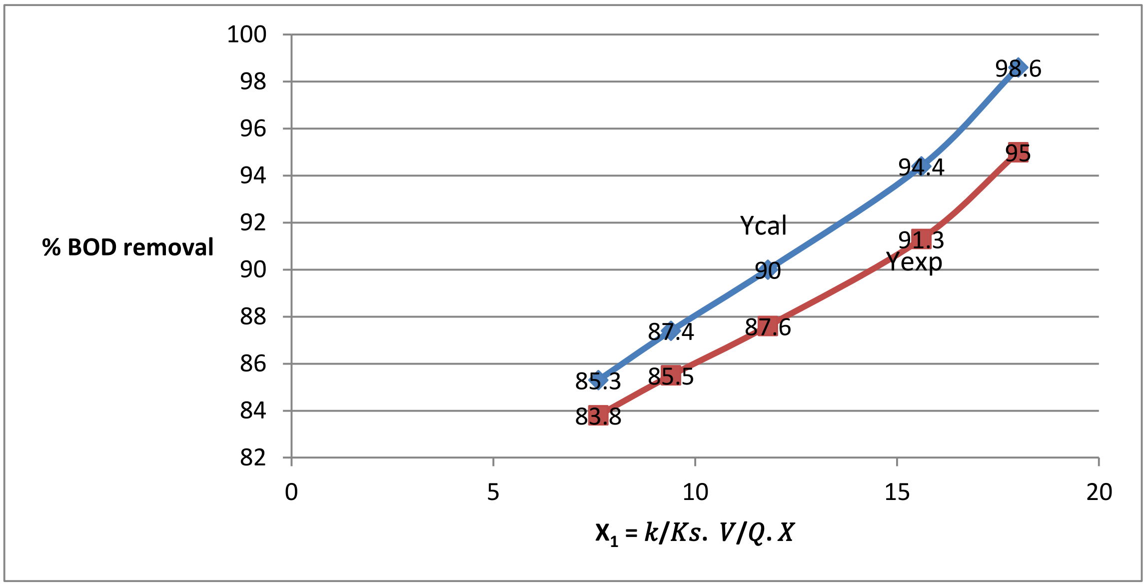

Figure 4. Mathematical model showing Deviation

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