

## Biokinetic Studies in the Treatment of Tannery Effluent

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### Abstract

The present investigation deals with the anaerobic treatment of composite tan liquor with varying organic loads. BOD of the influent and effluent and MLSS of reactor were determined at various detention times to generate data for kinetic coefficients. The kinetic coefficients  $k$  (maximum substrate utilization rate),  $K_S$  (half velocity constant),  $Y$  (cell yield coefficient) and  $K_d$  (decay coefficient) were found to be  $1.66 \text{ day}^{-1}$ ,  $1132 \text{ mg/L}$ ,  $0.05$  and  $0.22 \text{ day}^{-1}$ , respectively. Overall BOD removal rate constant 'K' was found to be  $1.46 \text{ day}^{-1}$ . The studies showed that an optimum BOD influent load of  $0.8 \text{ Kg BOD/m}^3/\text{day}$  with 3 days retention time could be adopted to yield about 97 percent BOD reduction. The Bio-kinetic coefficients were evaluated using modified Monod's equations to study the metabolic performance of the digestion process.

**Keywords:** tannery effluent, biokinetic coefficients

### 1. Introduction

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.

Arumugam (1976) has reported on the recovery of chromium from spent chrome tan liquor by chemical precipitation using lime. Pathe et al. (1995) have studied the properties of chromium sludge from chrome tan liquor and related the sludge volume, sludge settling rate, surface loading rate etc. Archana Shukla and Shukla (1994) have studied the treatment of tannery and electroplating effluents by using lime, NaOH and their mixture in the temperature range of 25 to  $100^\circ\text{C}$ . Guruswamy et al. (1995) conducted study 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%. Elangovan et al. (1995) have conducted experiments on the activated sludge treatment of vegetable tanning waste admixed with 10, 25, and 50 settled sanitary sewage and obtained BOD removal from 87 to 96.

Laboratory scale tests indicated that Biological oxygen demand is generally reduced by reducing the organic material with sodium hypochlorite. However, Cr(VI) is formed in this last process. Hydrotalcite (HT) and its calcination product were applied to separate the Cr(VI) formed. The best material found was calcined hydrotalcite, which removed not only Cr(VI) from the wastewater but also other anions, such as  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{HCO}_3^-$  (Gallegos, Martnez, & Bulbulian, 2005).

Removal of hexavalent chromium by adsorption technique from aqueous solution using neem bark (*Azadirachta indica*), subabul charcoal (SC, *Leucina leucocephala*) has been studied and the results are compared with powdered activated charcoal (PAC). The adsorption efficiency of PAC and SC for Cr(VI) was maximum at pH 2.0 and for NB it was maximum at pH 6.0 (Kulkarni & Shrivastava, 2002).

The reduction of the level of chromium and chemical oxygen demand (COD) in waste chrome liquor from tannery industry has been investigated. The chromium reduction achieved by groundnut shell powder (GNSP) treatment is 63-66% at pH 7.70. The total chromium and COD of the effluent after treatment with GNSP using batch reactor and anaerobic baffled reactor has been reduced to 99.9% and 93% respectively (Kumar, Verma, & Rao, 2002).

*Cupressus female cone* (CFC), an inexpensive plant material is investigated as an adsorbent for the removal of Cr(VI) from synthetic solution as well as from industrial wastewater so as to help its safe disposal. Maximum sorption occurs at the acidic pH range of 0.2-0.5 but it decreases on increasing the pH. Increase in initial concentration of Cr(VI) and sorbent particle size are found to reduce the amount of sorption, demonstrating the role of surface effects on sorption (Murugan & Subramanian, 2003).

In order to assess the transformation process involved in the chemistry of chromium, studies on adsorption kinetics and sequential extraction were conducted on oxisol, andept and alfisol soils using  $K_2Cr_2O_7$  as source of hexavalent Cr and indicated more adsorption in oxisol due to its high kaolinite clay content and also due to its acidic nature (pH: 4.2). Besides adsorption, the added, Cr(VI) was considerably reduced by the presence of organic matter (Thangavel, Rajannan, & Ramasamy, 2002).

Study deals with the characterization of tannery effluent polluted soil in Dindigul city. The pH of unpolluted soil was high, whereas that of polluted soil was low. The electrical conductivity and chloride content was high and it was unfit for the growth of plants. The heavy metals in all the samples were adequate for the growth of plants (Mariappan, Balamurugan, & Rajan, 2001).

Alkalotolerant/alkalophilic actinomycetes NCIM 5080 and NCIM 5142 produce alkaline protease in presence of chromium ions. These properties of isolates are suitable for treatment of tannery effluents which are alkaline and contain chromium and proteinaceous matter. Both the actinomycetes are able to grow in undiluted tannery effluents and remove chromium almost completely and reduce the COD by 70%-80% during growth as well as by pregrown biomass (More et al., 2001).

A laboratory scale experiment was conducted on aerobic digestion of tannery effluent using cowdung as the seed material. The BOD removal of 95.8 per cent was obtained at an optimum organic load of 0.6kg BOD/m<sup>3</sup>/d. Biokinetic coefficients were calculated for the data obtained to study the metabolic performance of the microorganisms (Prakash, 2001).

Adsorption technique has been applied for the removal of hexavalent chromium from aqueous solution using wheat straw dust, saw dust, and coconut jute, and the results are compared with the powdered activated carbon. The high uptake of hexavalent chromium was observed with PAC at pH 2.0, and for the other adsorbents at pH 6.0 (Rao & Bhola, 2000).

The tannery effluent emanating from Common Effluent Treatment Plant (CETP), Unnao (U.P, India) was found toxic in nature, having high BOD, COD, TDS and Cr content (5.88 mg l<sup>-1</sup>), which supported growth of chromate tolerant bacteria. Several chromate tolerant bacteria have been isolated from these effluent and maximum tolerant four strains (NBRIP-1, NBRIP-2, NBRIP-3 and NBRIP-4) were characterized in this study (Shukla et al., 2007).

An attempt has been made to assess vermiculite, a phyllosilicate mineral group with high cation exchange capacity, as an alternative for activated carbon. In this investigation, the adsorption potential of raw vermiculite grade 2 was evaluated using vermiculite clay mineral barrier system for reducing pollutant loads in specially designed treatment system, where the important parameters like flow rate, effluent loading rate required were optimized. Kinetics of chrome-tannery effluent treatment by the activated-sludge system was studied and reported (Jayabalakrishnan & Mahimai, 2007).

Anaerobic digestion of tannery waste in which semi-continuous and anaerobic sequencing batch reactor processes was reported (Zupancic & Jemec, 2010).

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 °C) with 30 mg/L, hexavalent chromium with 0.1 mg/L and pH between 5.5 to 9.0.

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 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 carbon dioxide. 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.

## 2. Materials and Method

Laboratory scale reactors are normally used to determine kinetic coefficients. In such a reactor, detention time ( $\theta$ ) is equal to mean cell residence time ( $\theta_c$ ). The procedure is to operate the unit over a range of effluent substrate concentrations. Hence, several different  $\theta_c$  (at least five) are selected for operation ranging from 1 to 6 days. Using the data collected at steady state conditions, mean values are determined for influent BOD ( $S_0$ ), effluent BOD ( $S$ ), and mixed liquor suspended solids (MLSS) of the reactor (denoted by  $X$ ) to find out the kinetic coefficients.

The experiment was designed and operated on the principle of an anaerobic digestion 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. 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 1 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 waste and removal of the effluent. The digester was carried out at room temperature. 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 0.25 kg BOD/m<sup>3</sup>/day in the beginning. After several displacements of the digester contents and after 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.

Samples from the influent to the reactor and effluent from the final clarifier were simultaneously collected to carry out BOD tests. Samples from the reactor were collected to find out MLSS, dissolved oxygen (DO), pH and temperature. Mean values of  $S_0$ ,  $S$  and  $X$  at various  $c$  were used to find out kinetic coefficients while DO, pH and temperature tests were carried out to ensure favorable environmental conditions in the reactor for biological treatment. All the tests were carried out as per procedures laid down in the "Standard Methods".

The samples of effluents' drawn at various stages were analyzed for pH, influent BOD ( $S_0$ ), effluent BOD ( $S_e$ ), 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 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 0.8 kg/m<sup>3</sup>/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. During the course of study, the reactor temperature fluctuated between 29 to 31 °C, which falls within the suitable temperature range for heterotrophs treating wastewater under aerobic conditions (Rao & Bholá, 2000). The pH of the reactor remained between 6.5 and 8.0 which is a suitable range for biological treatment.

### 3. Results and Discussion

The general characteristic properties of composite tannery effluent are presented in Table 1.

Table 1. General characteristics of composite tan liquor

Parameter	Value
pH	7.8
Alkalinity (as CaCO <sub>3</sub> )	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.

The result indicates that the liquor is basic with pH 7.8. The anaerobic reactor used for the study is shown in Figure 1.

The chromium content has been found to 120 mg/L and the BOD and COD of the effluent have been estimated to be 1360 and 2510 mg/L respectively.

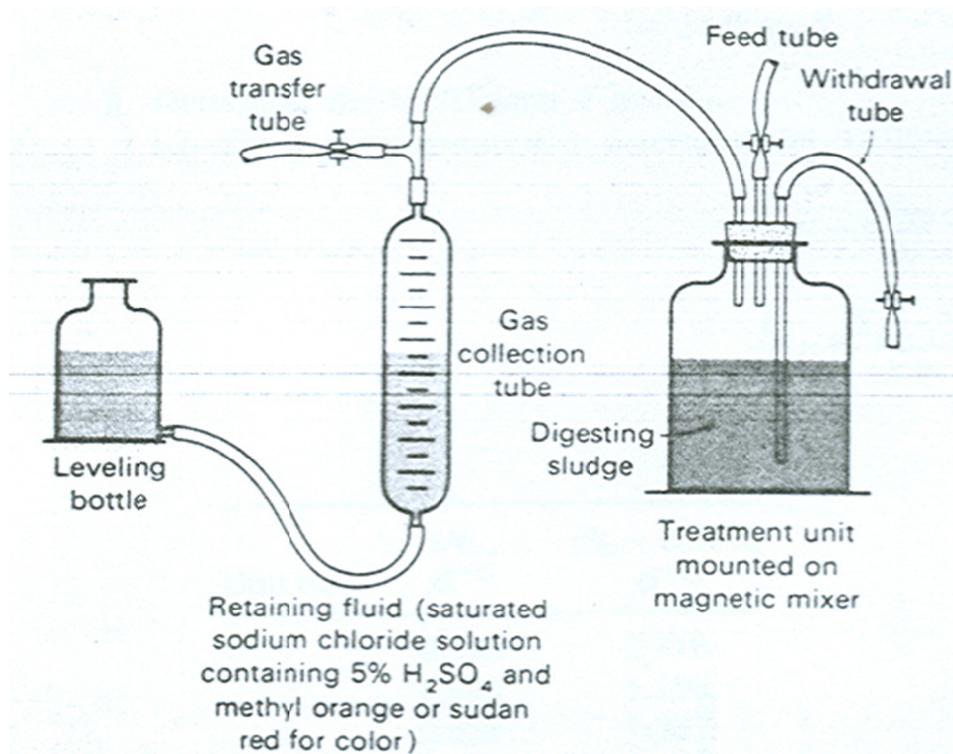


Figure 1. Anaerobic reactor used for the experiment

Table 2. Effect of lime on chromium precipitation

Weight of lime added (g/Lt)	pH	Chromium in filtrate (mg/L)
2.0	8.1	3.64
2.5	8.3	2.89
3.0	8.5	2.66
3.5	8.6	2.00
4.0	8.8	1.64
4.2	8.8	0.90
4.4	8.9	0.40
4.6	9.1	0.54
5.0	9.4	0.66
5.5	9.9	0.68

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/lit. 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<sup>3</sup>/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. The analysis of parameters was done as per the standard procedure given in APHA.

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 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.

Table 3. Anaerobic digestion of composite liquor

BOD load (Kg BOD/m <sup>3</sup> /day)	pH	Alkalinity (asCaCO <sub>3</sub> ) mg/L	Volatile acids (mg/L)	% 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

The results of the treated liquor with maximum and optimum removal efficiency of pollutants are represented in Table 4.

Total Kjeldahl Nitrogen (TKN) and Phosphorous (P) tests were carried out on the settled wastewater collected to check the level of these nutrients for satisfactory biological treatment. The BOD:N:P for the wastewater was found to be 100:21:0.19 as against a recommended value of 100:5:1 (Kulkarni & Shrivastava, 2002). The phosphorous was thus deficient and this deficiency could hamper a satisfactory biological treatment. Therefore, the deficiency was met by adding calculated amount of Potassium Dihydrogen Phosphate ( $\text{KH}_2\text{PO}_4$ ) in the wastewater.

Table 4. Results of the treated liquor

Parameter	Maximum	Optimum
BOD load, Kg BOD/m <sup>3</sup> /day	0.80	0.60
pH	7.6	7.4
BOD reduction (%)	96.9	95.4
Volatile matter reduced (%)	80	76
Alkalinity (as $\text{CaCO}_3$ ) mg/L	1920	980
Volatile acids (mg/L)	236	168
Methane production (%)	34%	28%

Kinetic coefficients of interest for the design of activated sludge process are:

$k$  = Maximum rate of substrate utilization per unit mass of microorganisms, time<sup>-1</sup>,

$K_d$  = Endogenous decay coefficient, time<sup>-1</sup>,

$K_s$  = Half velocity constant, substrate concentration at one-half of the maximum growth rate, mass/unit volume,

$Y$  = Cell yield coefficient, mg/mg (defined as the ratio of the mass of cells formed to the mass of substrate consumed).

„ $k$  value is employed to find out the volume of biological reactors. Greater is the value of  $k$ , smaller will be the size of reactor.  $K_s$  has no direct application in process design. It only gives an idea about the change in the specific growth rate of bacteria with a change in the concentration of the growth limiting substrate.  $Y$  is used to estimate the total amount of sludge produced as a result of wastewater treatment.  $K_d$  is used to find out the net amount of sludge to be handled and hence the size and cost of the sludge handling facilities can be found out from this information.

Data to determine above coefficients were generated by operating the bench scale reactor at different  $\theta_c$ . Mean values of  $S_o$ ,  $S$  and  $X$  corresponding to each are presented in Table 5.

Table 5. Mean values of data for  $k$  and  $K_s$

S.No	$\theta_c$	$S_o$ (mg/L)	$S$ (mg/L)	$X$ (MLSS/L)mg
1	3	446	66	250
2	4	432	62	239
3	5	418	51	166
4	7	410	48	150
5	9	406	47	146

The following linearized equation was used to find  $k$  and  $K_s$  (Gallegos, Martnez, & Bulbulian, 2005).

$$\frac{X\theta_c}{S_o - S_e} = \frac{K_s}{k} \frac{1}{S} + \frac{1}{k}$$

By using the above equation, a graph was plotted with 1/S on the x-axis and X c/(So-S) along y-axis (Figure 2). A linear regression line was fitted to the plotted data. Intercept on the y-axis and the slope of this line were used to find k and Ks. The equation of the fitted line is also shown on the graph.

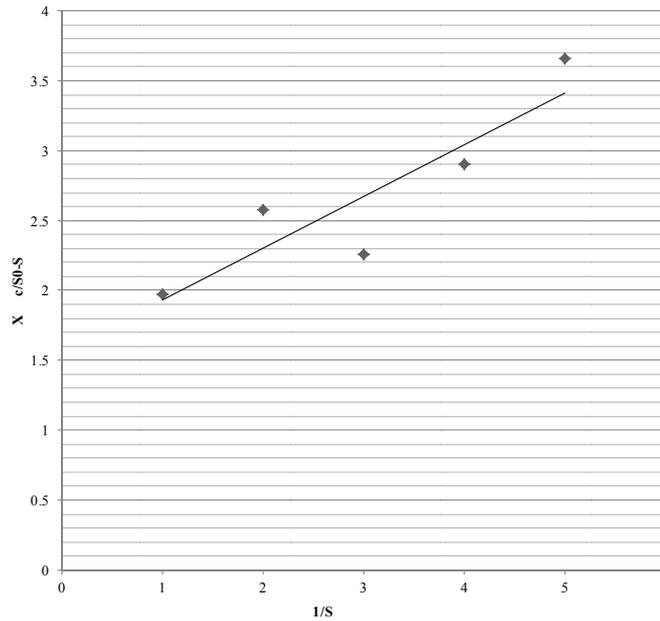


Figure 2. Determination of k and Ks

The following linearized equation was used to find Y and Kd. (Gallegos, Martnez, & Bulbulian, 2005).

$$\frac{1}{\theta c} = Y \frac{So - Se}{X \theta c} - Kd$$

A graph was plotted with 1/θ c along y-axis and (So-S)/X c along x-axis (Figure 3). A linear regression line was fitted to the plotted data. Intercept on y-axis and the slope of the line was used to find Kd and Y.

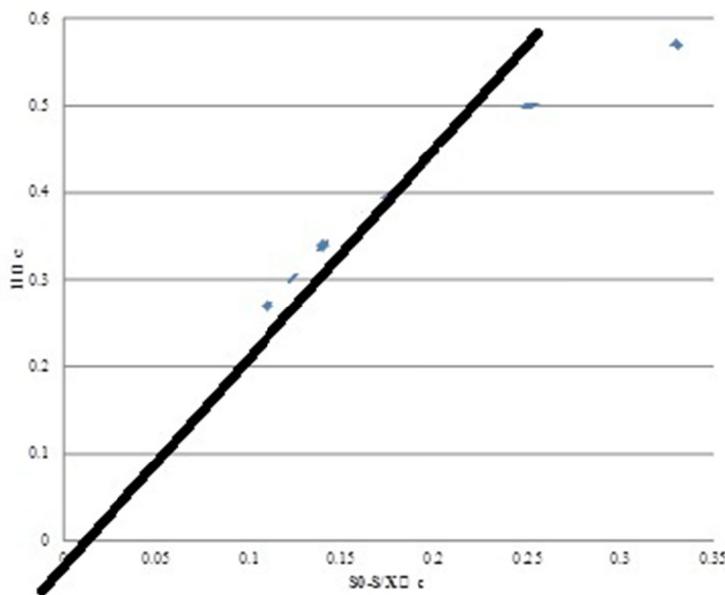


Figure 3. Determination of Y and Kd

The equation of the fitted line is also shown on the graph which was taken from the values shown in Table 6.

Table 6. Mean values of data for Y and Kd

X $\theta$ c	So-S	X $\theta$ c/ So-S	1/S	1/ $\theta$ c	So-S/ X $\theta$ c
750	380	1.973	2.24X10 <sup>-3</sup>	<b>0.33</b>	<b>0.51</b>
956	370	2.58	2.31X10 <sup>-3</sup>	<b>0.25</b>	<b>0.38</b>
830	367	2.26	2.39X10 <sup>-3</sup>	<b>0.20</b>	<b>0.44</b>
1050	362	2.9	2.43X10 <sup>-3</sup>	<b>0.14</b>	<b>0.34</b>
1314	359	3.66	2.46X10 <sup>-3</sup>	<b>0.11</b>	<b>0.27</b>

The values obtained for k, Ks, Y and Kd are presented in Table 7.

Table 7. Bio-kinetic coefficients

Parameter	Value
Substrate Removal Rate Constant, k/day	1.66
Half Velocity Constant, Ks, mg/L	1132
Decay constant, Kd, day <sup>-1</sup>	0.05
Yield Coefficient, Y	0.22
Maximum Specific growth rate of Microorganisms, $\mu_m$ /day	0.368

A comparison of kinetic coefficients for tannery wastewater with other industrial wastewaters would be interesting. Substrate utilization rate „k for tanneries (3.125 day<sup>-1</sup>) is less as compared to others and thus larger volume of biological reactor would be needed to treat tannery wastewater. This less value of „k may be due to specific nature of tannery waste. Decay coefficient Kd is quite low for tannery wastewater when compared with other industrial wastewaters, which indicates larger net sludge volumes resulting from biological treatment. Cell yield coefficient. For this purpose a graph was plotted with „S along x-axis and (So-S)/ $\theta$ c along y-axis. A linear regression line passing through the origin was fitted to the plotted data (Prakash, 2001). The equation of the line is also shown on the graph. Slope of this line gave the value of „K as 1.46 day<sup>-1</sup>. The result was in agreement with the findings of Metcalf and Eddy (2003), who reported that value of K varied from 0.5 to 1.5 day<sup>-1</sup>. The anaerobic digestion of tannery effluents followed a first order kinetics as confirmed from the values shown in Table 8.

Table 8. First order rate constant values

Time (Hours)	Rate constant, k
24	0.0301
48	0.0208
72	0.0204
96	0.0253
120	0.0260
144	0.0245

#### 4. Conclusions and Recommendations

The results of the study lead to the following conclusions.

- (1) By proper maintenance of required alkalinity, the BOD reduction can be increased.

- (2) The maximum BOD reduction was obtained at an applied organic load of 0.80kg BOD/m<sup>3</sup>/day/.
- (3) The CaCO<sub>3</sub> used in the experiment helped to maintain adequate buffer capacity to neutralize free volatile acids.
- (4) The increase of substrate utilization could be attributed to the continuous increase in metabolic activities caused by cell growth.
- (5) The rate constant values confirm a first order reaction.

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