Journal of Engineering Science 06(1&2), 2015, 67-73

JES
an international Journal

BIOLOGICAL TREATMENT OF TANNERY WASTEWATER

J. Khatoon[1] and M. D. Hossain[2]

[1]*Ph. D. Student, Bangladesh University of Engineering and Technology, Bangladesh*
[2]*Professor, Civil Engineering Department, Bangladesh University of Engineering & Technology*

Received: 16 May 2015 Accepted: 24 December 2015

ABSTRACT

In this study, the performance of physic-chemical and biological treatment for tannery wastewater was investigated. Tannery wastewater combined with domestic sewage was used in the reactor. The reactor was operated at different operating conditions by changing the hydraulic retention time, HRT (72 hours and 30 hours) and operation process (extended aeration, oxic-anoxic-oxic cycle). From the results, it was found that oxic-anoxic-oxic cycle gave maximum reduction in BOD_5 (96%), COD (89%) and color (85%) with hydraulic retention time of 30 hours. During anoxic period, 90% removal of ammonia was occurred. Because combining intensive aerobic and anoxic biological treatment can break down nitrogenous compounds. Oxygen demand profile for tannery wastewater was also identified for determination of rate of biodegradation. The biodegradation rate is seed-specific and various operational processes, which results in different organic consumption rate and different pattern.

Keywords: *Ammonia Removal, Anoxic Unit, Biodegradability, Sequential Batch Reactor.*

1. INTRODUCTION

Among all the industrial wastes, tannery effluents are ranked as the highest pollutant (Banuraman and Meikandaan, 2013). Tanneries use 30 to 40 m^3 of water is used for per ton hides/skin processing (Aboulhassan *et al.,* 2008). During tanning process about 300 kg chemical are added per ton hides/skin (Durai *et al.,* 2011).

In Bangladesh, 21,600 m^3 liquid wastes are being generated every day; where yearly waste generation is 485 million m^3 (Mahmood, 2008). Due to the variety of chemicals added at different stages of processing of hides/skin, the wastewater has complex characteristics. Wastewater contain heavy metal, high concentration of suspended and dissolved solids, BOD, COD, Cr, sulphide, toxic chemicals and other pollutants.

The tropical climate of Bangladesh is favorable for rapid stabilization of waste through biological process. Activated Sludge Process (ASP) is a feasible treatment technology for tannery wastewater. In this process, the activity of a microbial species community under controlled operating conditions permits the biodegradation of organic matters and nutrients from wastewater (Hiruy, 2000). BOD_5/COD ratio is the biodegradability index and wastewater can be considered readily biodegradable if it has a ratio between 0.4 and 0.8 (Aboulhassan *et al.*, 2008). SBR (Sequential Batch Reactor) is a modification of ASP. SBR has easy operation, low cost, handling hydraulic fluctuation, no need for settling tank and sludge recycling as well as organic load without any significant variation in removal efficiency (Durai *et al.,* 2011). Mixed liquor remains in the reactor during all cycles, thereby eliminating the need for sedimentation tank.

As tannery wastewater is highly hazardous, only chemical treatment of wastewater demands high cost. Biological treatment combined with physico-chemical process is comparatively cheaper. This study aims to find out the suitable biological treatment process for tannery wastewater.

2. OBJECTIVES

The overall objective of this research work is to develop a suitable technology for biological treatment of tannery wastewater. The specific objectives of the research work are:

a) To assess the characteristics of tannery wastewater,
b) To determine the optimum F/M ratio and detention time for treatment of tannery wastewater,
c) To assess the effect of anoxic condition in treatment system.

3. MATERIALS AND METHODS

3.1 Collection of Wastewater

Different wastewater streams are generated at different times and as a result, the effluent characteristics in the main drain vary significantly. Since no equalization tank is provided, the hourly samples collected over one production cycle are thoroughly mixed in a drum to make a representative tannery sample. Tannery wastes have

* Corresponding Author: khatoon.jahura@gmail.com

deficiency of nutrient for proper biological growth. For this, domestic sewage was added to the tannery sample at 1:4 (domestic: tannery) ratios. BOD_5, COD, DO, pH, color, turbidity, total solids (TS), total dissolved solids (TDS), total suspended solids (TSS), NH_3-N, NO_3-N, NO_2-N, chromium, sulphate, sulphide were determined for identifying influent characteristics following standard methods (AWWA, 1998). Here influent wastewater has biodegradation ratio equals to 0.58.

3.2 Acclimatization of Micro-organism

Tannery wastewater is highly toxic and toxic impurities hinder the microbial activities, raw sample collected from tannery had to be seeded with acclimatized sludge. For acclimatization, textile sludge of 10 liters was kept at rest for 2 hours then sludge was separated from supernatant. Continuous aeration of textile sludge was started and 100 ml. of combined wastewater (tannery + domestic) was added every day. pH was adjusted between 6.0 and 7.0. COD and MLSS were measured from time to time. Continuous aeration was stopped where there was an indication of increasing value of MLSS. This was occurred after 30 days.

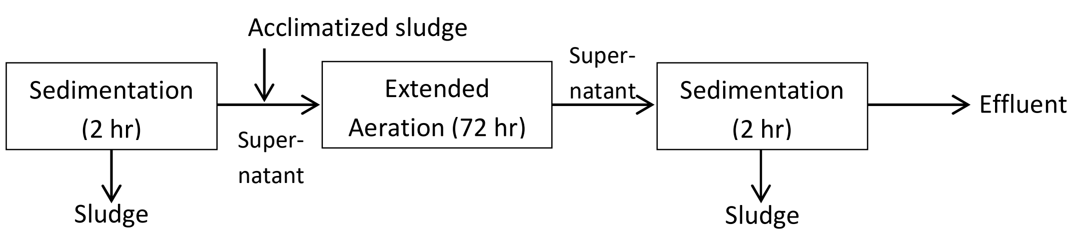
For first case (extended aeration activated sludge process), acclimatized sludge was mixed with combined raw sample in proportion of 200:800, 300:700 and 400:600 to make 1L mixed sample. These proportions resulted in maintaining F/M ratios to 0.28, 0.22 and 0.18 respectively.

Acclimatized sludge was added prior to aeration for all the three cases. Both acclimatized sludge and combined raw sample were settled for two hours and settled sludge was mixed with the supernatant portion of combined raw sample. But for third case, settled sludge was mixed with the supernatant portion left after alum coagulation.

3.3 Experimental Set-up

In order to examine the appropriate technology, ASP has been studied using two cases. Two cases are illustrated in Figure 1.

Flow chart: Case 1



Flow chart: Case 2

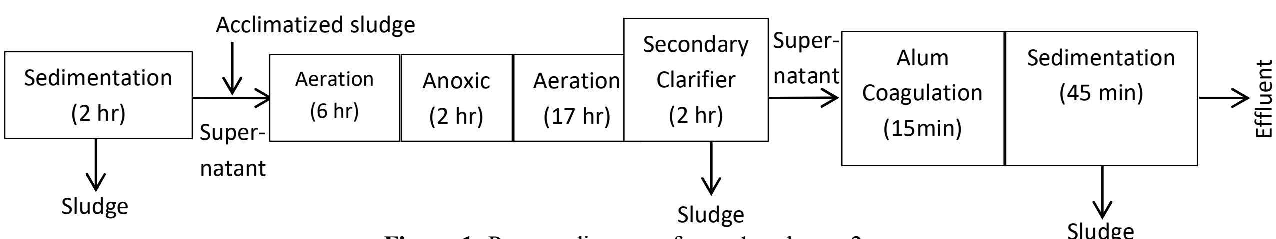


Figure 1: Process diagram of case-1 and case-2

Two equal-volume imhoff cones were used for two cases. The samples in cone were continuously aerated with diffuser. The rising velocity of air bubbles creates sufficient agitation in the waste. This agitation makes the contact between food and micro-organism, resulting in a higher rate of bacterial growth which has great importance in the biological treatment of tannery effluent. Due to presence of high quantity of ammonia in tannery wastewater, there is necessity for anoxic unit to remove ammonia in the form of nitrogen gas through denitrification. DO was 0.2 mg/l during anoxic period.

In case – 2, coagulation was conducted after secondary sedimentation. For coagulation, alum ($Al_2(SO_4)_3.18\ H_2O$) was used as coagulant because alum reduced COD and color. Used alum dose was 50 mg/l for the case of oxic-anoxic-oxic cycle. pH was kept between 6 & 7. DO (Dissolved Oxygen) of the reactor was maintained above 0.2 mg/l which is required for satisfactory biological treatment.

4. THEORETICAL APPROACH OF BIODEGRADATION

The concept biodegradability, expresses the capability of an object to undergo biodegradation (manifested by the decrease in the *COD*). The *COD* has got biodegradable and non-biodegradable components as follows:

$$COD = COD_{In} + COD_{sI} + COD_{sB};\ \text{with } COD_{sB} = S \quad (1)$$

The subscripts *In*, *sI* and *sB* denote respectively the insoluble, soluble inert and soluble biodegradable constituents of *COD*. The subscript *S* symbolizes substrate concentration in well-known Monod equation. In a biodegradable process, the insoluble and inert constituents of the *COD* remain unaffected and the oxygen demand is related to the biodegradable constituent, COD_{sB} only.

$$\Delta COD = \Delta COD_{sB} = \Delta S \text{ with } \Delta COD = COD_0 - COD \text{ and } \Delta S = S_0 - S \quad (2)$$

Where the subscript *0* stands for the parameter at the time t = 0.

The biodegradability, α_0, of an effluent could be defined as follows:

$$\alpha_0 = COD_{sB,0} / COD_0 = (COD_0 - COD_\infty) / COD_0 \quad (3)$$

Where the subscript ∞ stands for the parameter at the time $t \to \infty$. In a biodegradation process, COD_{sB} is assumed zero at the time $t \to \infty$. Conventionally a *BOD/COD* ratio is taken as a measure for the biodegradability of an effluent. The BOD of an effluent is defined as the oxygen demand, *OD* (amount of oxygen consumed by 1L of solution), in a biodegradation process in a given time *t*. The *BOD* value increases with the biodegradation time, and conventionally, the criterion for the effluent quality is accepted to be BOD_5 and the biodegradability is defined as

$$\alpha_s = BOD_u / COD_0 = OD_{t\to\infty} / COD_0 \quad (4)$$

Where BOD_u is known as ultimate *BOD* is the oxygen demand for $t \to \infty$.

From the way of definition of biodegradability, it becomes evident that α_0 is a seed-dependent parameter. But if the oxygen demand is measured for the biodegradation with naturally grown or adapted micro-organism, the biodegradability, α_s, defined by Eq. (4) is quite sound for practical purposes. The biodegradability, α_0, defined by Eq. (3) is of theoretical interest and could also be used for practical purposes.

In a biodegradable process, biodegradable portion of the substrate *COD* is partly oxidized (measured as *OD*) and partly included as microbial cell-*COD* growth (measured as *COD* equivalent of ΔX).

$$\Delta COD = \Delta COD_{sB} = \Delta OD + \Delta COD_{cell} \quad (5)$$

For the simplicity of the analysis, the effect of decay rate k_d of microorganism is ignored and the following linear relations are assumed (Rozich and Gaudy, 1992) :

$$\Delta COD_{cell} = O_x \Delta X \text{ and } \Delta X = Y \Delta COD \quad (6, 7)$$

Where *Y* is the yield coefficient; mass of cells produced per unit mass of substrate utilized (mg *X*/mg *COD*), and O_x is the oxidative potential of the biomass. Combining Eq. (5-7), it can be obtained

$$\Delta COD = \gamma \Delta OD \text{ with } \gamma = 1 / (1 - YO_x) \quad (8)$$

The ΔCOD vs. ΔX (Eq. 7) and ΔCOD vs. ΔOD (Eq. 8) data could be fitted to straight lines to give parameters, *Y* and O_x. Thus *Y* can be determined from individual batch kinetic tests for ΔX vs. ΔCOD data using a laboratory fermentator. A rough estimation of *Y* assuming an average value of O_x in the range of 1.42-1.48, however, seems satisfactory for the analysis of biodegradation using two measured values of *COD* at a given time interval and the corresponding oxygen demand (Chili *et al.*, 2008). Then for calculation of the parameter *Y*, the Eq. 8 is rewritten as follows:

$$Y = (1 - \Delta OD / \Delta COD) / O_x \text{ with } O_x = 1.42 - 1.48 \quad (9)$$

For the present analysis, *COD* was measured initially and after an experimental period of 72 hr and 25 hr, *Y* was calculated by the following equation:

$$Y = \left(1 - \frac{OD_{72}}{COD_0 - COD_{t=72}}\right) / O_x \text{ with } O_x = 1.45 \quad (10)$$

Where OD_{72} is the cumulative oxygen demand for time $t = 72$ h. The oxygen demand profile is presented in Figure 2. Similar data available in the literature (Lee and Oswald, 1954; Kessick, 1976; Min *et al.,* 2004) have also been presented in the figure for comparison.

Case - 1 and Case - 2 followed the trend of Lee, Kessick and Minns' COD reduction curve of GGA solution (mixture of glucose and glutamic acid) which COD was 744 mg/l. Pattern of curves were irregular. From this irregular nature, it can be said that the biodegradation rate is seed-specific and various operational processes.

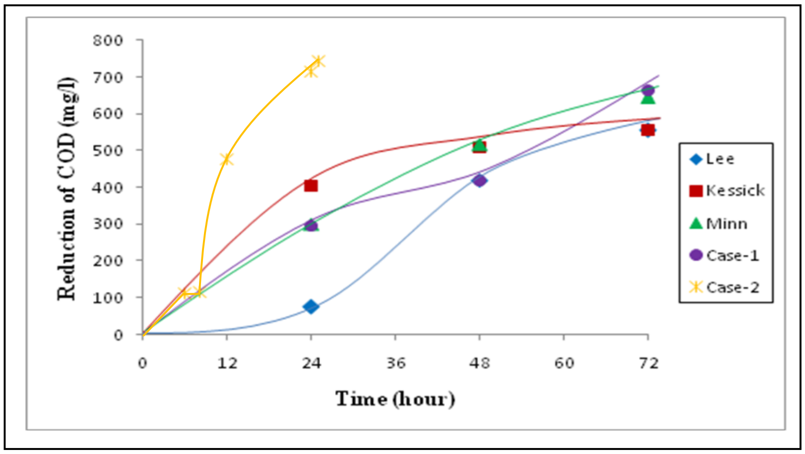


Figure 2: Oxygen demand (*OD*) profile for GGA solution

5. RESULTS AND DISCUSSION

Primary sedimentation (2 hours) resulted in 35% reduction of Color, 28% reduction of COD and 36% reduction of BOD_5 were occurred. Removal percentage of Chromium was 69 and residual value was 15.29 mg/l. Chromium has no effect on biological treatment up to a concentration of 50 mg/l. Influent wastewater characteristics is given in Table 1.

Case-1 was run with physical and biological treatment (extended aeration) procedure. No chemical procedure was applied here. And this setup was chosen as case - 1 because of simplicity of treatment procedure and reduction of cost for not using chemicals. At first, it was not possible to predict correctly which F/M ratio would be suitable for conducting treatment operation. For this, variation of F/M ratio was done in case-1 and results due to variation of F/M ratio is shown in Table 2.

Table 1: Characteristics of combined raw sample

Parameter	Raw sample	Standard value by ECR (Inland surface water)	Parameter	Raw sample	Standard value by ECR (Inland surface water)
pH	8.9	6 - 9	TDS	6436 mg/l	2100 mg/l
EC	7.68 ms/cm	1200	TSS	688 mg/l	150 mg/l
Color	897 Pt-Co unit	150 Pt-Co unit	COD	2212 mg/l	200 mg/l
Turbidity	70 NTU		BOD_5	1285 mg/l	50 mg/l
Chromium	49.325 mg/l	.50 mg/l	NH_3-N	140 mg/l	5 mg/l
Sulphate	4102 mg/l		NO_3-N	462.8 mg/l	10 mg/l
Sulphide	960 mg/l	1 mg/l	NO_2-N	0.235 mg/l	

Table 2: Summary of results in Case - 1

Treatment Steps		Parameters								
		Color (Pt-Co unit)			COD (mg/l)			BOD_5 (mg/l)		
Primary Sedimentation (2 hr)	Inlet		897			2212			1285	
	Outlet		583			1586			812	
	Removal %		35%			28%			36%	
Use of Acclimatized Sludge		F/M 0.28	F/M 0.22	F/M 0.18	F/M 0.28	F/M 0.22	F/M 0.18	F/M 0.28	F/M 0.22	F/M 0.18
Continuous Aeration (72 hr)	Inlet	526	488	453	1506	1457	1312	778	741	625
	Outlet	726	646	663	866	792	829	437	321	386
	Removal %	19%	28%	26%	60%	64%	62%	66%	75%	70%
Final Sedimentation (2 hr)	Inlet	726	646	663	866	792	829	437	321	386
	Outlet	700	601	628	664	553	598	322	232	270
	Removal %	22%	33%	30%	70%	75%	73%	75%	82%	79%

- Percentage calculated from initial value

From the experiment of extended aeration, it is gotten that F/M ratio of 0.22 contributed to the highest removal percentage of wastewater parameters and these values were 28%, 64% and 75% for Color, COD and BOD_5, respectively. But residual values did not meet NEQS. Table-4.1 shows residual color values were increasing with time for every F/M ratio in case-1. This may be occurred due to some internal reaction in longer detention time of 72 hours.

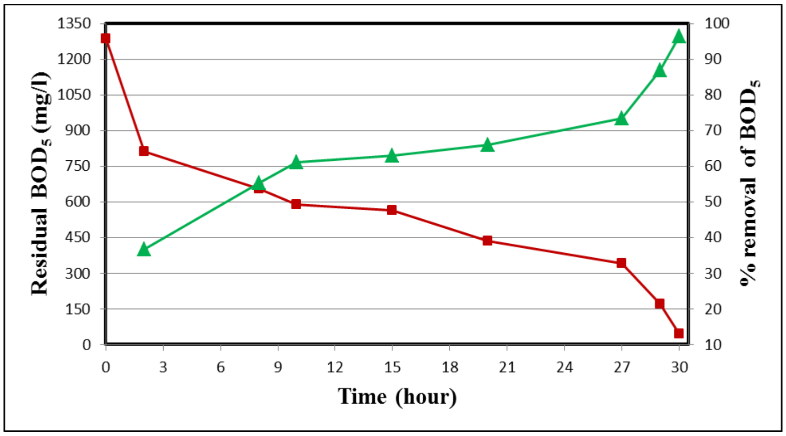


Figure 3: Variation of BOD_5 and % removal of BOD_5 in Case – 2

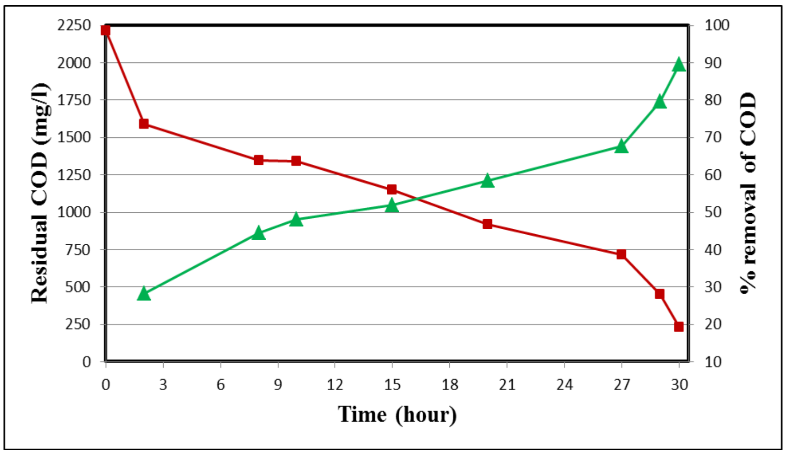


Figure 4: Variation of COD and % removal of COD in Case - 2

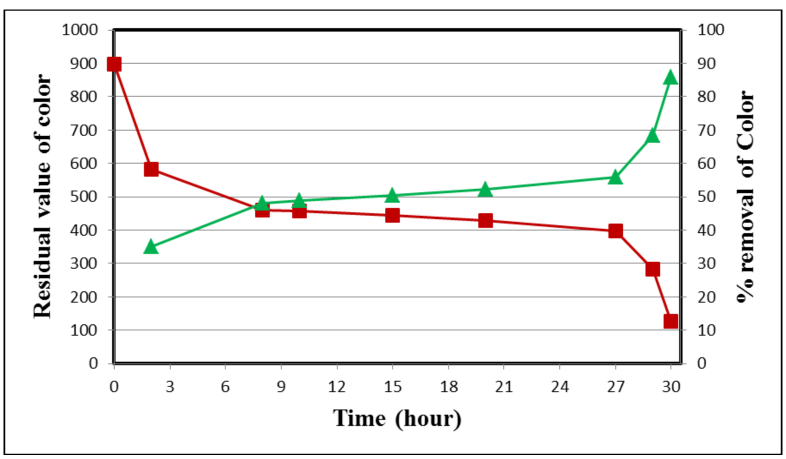


Figure 5: Variation of Color and % removal of Color in Case - 2

Case-2 was operated with F/M ratio of 0.22 and oxic-anoxic-oxic cycle. Because it is necessary to remove ammonia at first due to creation of hinders in oxidization process and oxic-anoxic cycle contributed in ammonia reduction. After oxic-anoxic cycle, color value was still high and alum coagulation was utilized to reduce color. In case-2, after sedimentation in secondary clarifier, color reduction percentage was 68% and residual value was 283 Pt-Co units. But this value was very high. Alum coagulation was used to reduce the residual color value. Used alum dose was 50 mg/l. Sludge volume was small due to using alum after other treatment operations and just before final disposal. This will decrease sludge handling cost. Case-2 gave effluent parameters values within NEQS (specified in ECR'97) which were 126 Pt-Co units for color, 232 mg/l for COD and 46 mg/l for BOD_5. Effects of case – 2 are shown in Figures 3, 4 & 5.

5.1 Ammonia Reduction in Case - 1 and Case - 2

Presence of ammonia creates problems in oxidization of organic matter. In Figure 6, ammonia reduced gradually in Case - 1 and falls to 41 mg/l from 140 mg/l with removal percentage of 70. But in Case - 2, during anoxic period ammonia reduced drastically and reached to 13 mg/l from 140 mg/l with 90% removal. Anoxic period that means oxygen limiting period converts ammonia to nitrogen gas through denitrification. There was increase in pH, which confirms the progress of denitrification. Hence Case - 1 performance in removing ammonia was poor for not existence of anoxic unit.

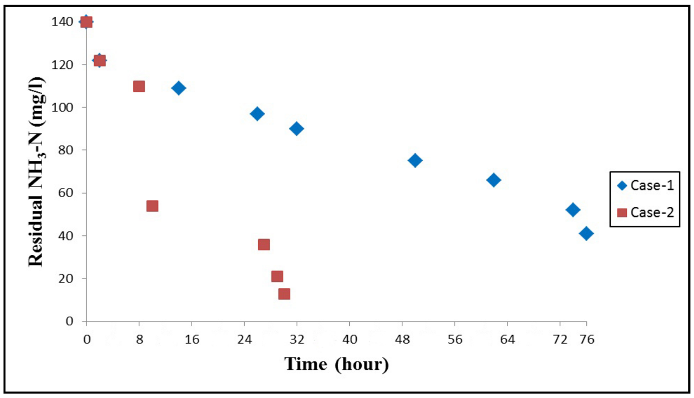


Figure 6: Ammonia reduction in Case-1 and Case-2

5.2 Yield (Y) for Case –2

The correlation between COD reduction and cell concentration (X) was found out by plotting data for COD and X for case-2 and fitting a linear regression line to the plotted data as shown in Figure 7. The correlation obtained may be expressed as:

$$Y \text{ (yield co-efficient)} = 0.6003X + 3370 \quad (11)$$

Equation 11 must be used with caution. The relation cannot be generalized. From Figure 7, yield coefficient was gotten 0.60 that means growth of microorganism was higher. Here decay rate of micro-organism was lower.

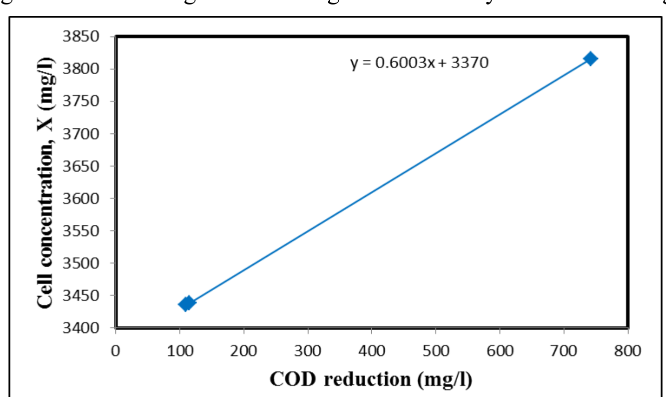


Figure 7: Evaluation of yield co-efficient for Case-2

6. CONCLUSION

The continuous aeration of tannery wastewater was carried out in SBR. Case - 1 i.e. case with extended aeration gave better results for F/M of 0.22 than that of 0.28 and 0.18. High retention time (72 hours) of case - 1 increased color values and reduction percentages of COD and BOD_5 were less. Decrease in hydraulic retention time in case – 2 i.e. case with oxic-anoxic cycle leads to increase in removal percentage of Color(85%), COD(89%) and BOD_5 (96%) and residual values of these parameters met NEQS for Bangladesh. Y (yield co-efficient) was more and micro-organism growth rate was higher for anoxic unit in case - 2. It could also be seen that at 24 hour, COD reduction for case – 2 was greater than that of case – 1. Because case – 2 contained anoxic period which contributed in more NH_4 reduction and due to this incidence, bacteria mainly oxidized organic matter after anoxic period which leads to higher COD reduction.

ACKNOWLEDGEMENT

Authors express their sincere thanks to the Civil Engineering Department of BUET (Bangladesh University of Engineering and Technology) for providing all the facilities and equipment, which enabled them to carry the research work. The authors are grateful to Bangladesh College of Leather Technology (BCLT), the Tannery Association, and other stakeholders for their support in various stages of this study.

REFERENCES

Aboulhassan, M. A., Souabi, S. and Yaacoubi, A., 2008. Pollution Reduction and Biodegradability Index Improvement of Tannery Effluents, International Journal on Environment, Science & Technique, 5, 11-16.

Banuraman, S. and Meikandaan, T. P., 2013. Treatability Study of Tannery Effluent by Enhanced Primary Treatment, International Journal of Modern Engineering Science, 3, 1, 119-122.

Chili, K. Eun, K. C. and In-Soung, C., 2008. Non-steady State Estimation of Biodegradability of Dyeing Wastewater Using Respirometer, Korean Journal of Chemical Engineering, 25(3), 553-557.

Durai, G., Rajasimman, M. and Rajamohan, N., 2011. Kinetic Studies on Biodegradation of Tannery Wastewater in a Sequencing Batch Reactor", Journal of Biotech Research, 3, 19-26.

Hiruy, A. K.., 2000. Developing A Laboratory Scale Sequencing Batch Reactor and Evaluating It's Performance for the Treatment of Tannery wastewater, Environmental Science Program, School of Graduate Studies, Addis Ababa University.

Kessick, M. A., 1976. The Calibration of Closed-end Manometric Biochemical Oxygen Demand Respirometers, Biotechnology and Bioengineering, xviii, 595 – 598.

Lee, E. W. and Oswald W. J., 1954. Comparative Studies of the Dilution and Warburg Methods for Determining BOD, Sewage and Industrial wastes 26 (9), 1097 – 1108.

Mahmood, S. B., 2008. Chemical Treatment Options for Tannery Wastewater, M. Engg. Thesis, Department of Civil Engineering, Bangladesh University of Engineering and Technology (BUET), Dhaka.

Min B., Kohler D. and Logan B. E., 2004. A Simplified Headspace Biochemical Oxygen Demand Test Protocol Based on Oxygen Measurements Using a Fibre Optic Probe, Water Environment Research, 76(1), 29-36.

Rozich, A. F. and Gaudy, A. F., 1992. Design and Operation of Activated Sludge Process using Respirometry, Lewis Publishers, Boca Raton, FL.