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Full Length Research Paper

Tannery wastewater treatment using two-stage anaerobic sequence batch reactor (ASBR) at mesophilic and thermophilic phase

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Industrialization has resulted in the formation of huge amount of waste products, which are released into the environment in the form of wastewater leading to environmental pollution and deterioration. Tannery effluent is among one of the dangerous pollutants of the industry. Most of the leather industries in Ethiopia discharge their effluent partially or without any treatment to the nearby water bodies. This creates a serious effect on aquatic biota and surrounding environment due to its high organic loading and chromium content. To minimize the effect of tannery wastewater, it should be treated before the effluent is discharged to the environment. Therefore, the main objective of the study was to use a two-stage laboratory scale Anaerobic Sequence Batch Digester (Reactor) in order to investigate the treatment potential of composite tannery wastewater at mesophilic and thermophilic phases. Two-Stage Anaerobic Sequence Batch Digester was used because it has a conducive environment for micro-organisms at a different temperature. Four sets of conditions were investigated; 1) mesophilic to mesophilic; 2) thermophilic to thermophilic; 3) mesophilic to thermophilic; 4) thermophilic to mesophilic, respectively. The Hydraulic Retention Time (HRT) of the hydrolysis/acidification was between 2 and 3 days and greater than 7 days in acetogenesis /methanogenesis. The Organic Loading Rate (OLR) was wide-ranging between 9.58 to 10.28 kg COD/m^3-day throughout the study. The removal efficiency of COD, TN, NO^3-N, S^{-2} and SO$_4^{-2}$ of all digesters were in the range of 57-70, 38-51, 44-61, 90-96 and 57-71%, respectively. While the concentration of NH^{4+}-N showed an increment from the influent by 22-31% in all digester. Generally, treatment of composite tannery wastewater by two-stage ASBR shows significant removal of pollutants at thermophilic - thermophilic phase especially S-2.

Key words: Anaerobic sequential batch reactor, composite tannery wastewater, removal efficiency, organic loading rate.

INTRODUCTION

Water is a source of life and regarded as the most essential of natural resources. Furthermore, existing

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freshwater resources are gradually becoming polluted and unavailable due to human or industrial activities. The increasing contamination of water systems with thousands of industrial and chemical compounds has become one of the most important environmental problem (Kumar and, Lee, 2012).

Industrialization is causing more demand than ever for the dwindling supply of water, which makes water crisis on a global scale. Wastewater is generated and dispersed in large amounts such that one out of six people (1.1 billion) has no access to safe drinking water and two out of six people (2.6 billion) lack adequate sanitation (WHO and UNICEF, 2004). This a contributor to a wide range of health problems and disorders in humans.

During the last century, a huge amount of industrial wastewater was discharged into rivers, lakes and coastal areas. With a rapidly expanding human population and a growing trend of industrial development added with limited technological advancement, problems related to the management of industrial waste have become a major problem in Ethiopia (Seyoum et al., 2004). The leather industry sector is one of the fast-growing economic sectors in Ethiopia. Currently there are 19 functioning leather tanneries with 20 new leather industry facilities in the planning stages (Abadi, 2000). According to EPA (2003) in Ethiopia, there are more than 20 tanneries. Accumulation of large volumes of dried-sludge in treatment compound has become common (Seyoum et al., 2004). This has immediate public health implications, which are manifested as frequent outbreak of major epidemic diseases and contributes to climate change as it releases greenhouse gases; methane and carbon dioxide (Dida, 2010).

The industrial strategic development plan of Ethiopia gives great emphasis to improve export-led products to join the international market in large-scale such as leather products. However, in Ethiopia, most of the leather industries discharge their effluent without any treatment to nearby rivers (EPA, 2003; Seyoum et al., 2004). This creates a serious effect on aquatic biota and the surrounding environment. For developing countries such as Ethiopia, the cost is a major issue. One of the many economically, as well as environmentally friendly strategies suggested, is to design a protocol that can treat hazardous tannery wastewater with biological system. The purpose of this study was to investigate the potential of a two-stage laboratory scale Anaerobic Sequence Batch Reactor (ASBR) for the treatment of composite tannery wastewater at a different temperature.

MATERIALS AND METHODS

Description of the study area

Modjo is a town in the central rift valley of Ethiopia, named after Modjo River. It is Located in East Shewa Zone of the Oromia Regional State (Figure 1). It has a latitude and longitude of 8°39′N 39°5′E with an elevation between 1788 and 1825 m above sea level with tropical rainfall climate (Richard, 1968).

In this study, materials such as amber glass bottle, influent feeding tanker, rubber hose, gas kit maker, hose, incubator, furnace, peristaltic pumps, measuring cylinder, analytical balance, air compressor, beaker, 100 ml syringe, scissors, desiccators, iron wire, iron rings and standings, clamps, hot plate, crucibles, plastic bags, stopper, burettes, pipettes, controlling valves and water bath were used. Analytical equipment such as a spectrophotometer, AAS, DO meter, pH meters and thermometer were used. All apparatus was properly washed first with soap solution and then with 1 normal nitric acid, finally washed with distilled water, and allowed to dry on hot air oven.

Different chemicals were used in the study. The chemicals used were polyvinyl alcohol, sulfuric acid, sulfide reagent 1, sulfide reagent 2, potassium hydroxide, sodium hydroxide, nitraver 5 nitrate, hydrochloric acid, nitrogen persulfide, hydrogen peroxide, potassium sulfate, total nitrogen reagent A, B and C, boric acid, Nessler reagent and copper sulfate.

Sample collection and preparation

Tannery wastewater samples were collected from Modjo Tannery, central Ethiopia using different size plastic bags every seven days for three months (12 days). The samples were collected from three different effluent lines which included the sulfur line; chrome line and general wastewater line and a composite sample was prepared by combining the three stream samples proportional to their respective volumes. Every 7-days, 20 L composite sample was collected and transported to the research lab at the Centre for Environmental Sciences, Addis Ababa University and stored at 4^0C in the refrigerator until added to the digester for treatment.

Experimental set up of the two-stage anaerobic sequential batch reactor (ASBR)

Two parallel anaerobic digestions, consisting of four ASBRs in series, were tested. The two reactors in the first system were operated at the same temperature of 35°C (mesophilic), respectively; the two reactors in the second system were at the same temperature of 55°C (thermophilic) respectively, the two reactors in the third system were at two different temperatures of 35 and 55°C (mesophilic to thermophilic), respectively and the two reactors in the fourth system were at two different temperatures at 55 and 35°C (thermophilic to mesophilic), respectively. Each reactor had a total liquid volume of 2.8 L. Totally, eight reactors were prepared to observe the treatment potential of composite tannery wastewater. The objective of the first stage reactor was to have a good solid settlement of composite tannery wastewater, reduce the effect of shock loadings and improve the stability of the two-phase system in an effort to improve the performance of the second stage reactors. The first stage reactor was fed the composite tannery wastewater from Mojo Tanner and the second stage reactor was fed composite tannery wastewater from the first stage reactor.

In the first stage, the first hose was stretched up to the bottom of the solution enabling decanting of all the solution to the second stage while the second hose was placed above the solution. In a similar manner, the second phase reactors had also two hoses at the top. The first hose was immersed to half-height of the reactor and used for filling of the solution from the first stage and decanting the solution while the second hose was above the solution with a plastic bag at the top to collect unwanted gas from the digester and control the temperature (Figure 2). In the experiment, each treatment was run in triplicates. The system was adapted from

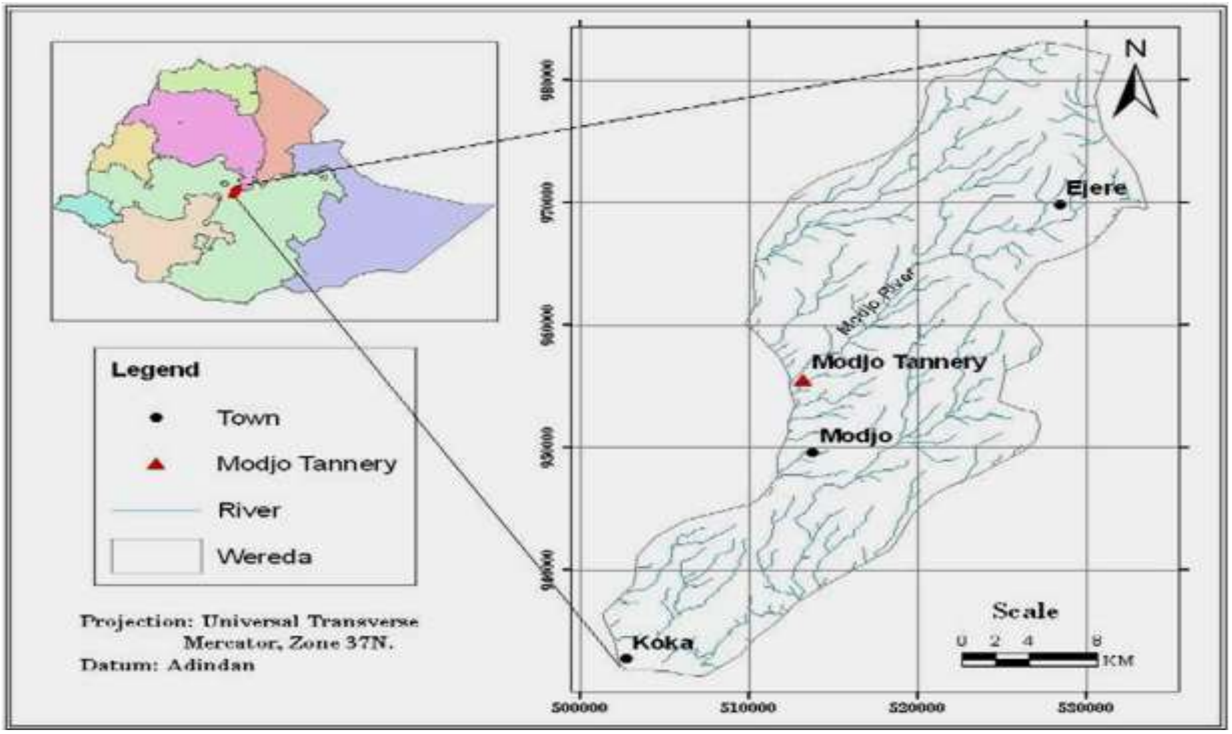

Figure 1. Map of the study area.

Figure 2. The two-stage ASBR set up.

Dugba and Zhang (1999). Table 1

Operation of the ASBR

The study was conducted for 90 days (3 months) in two different operational phases. The first phase at the startup period of the ASBR was operated for 30 days. This time was assigned for the accumulation of biomass. During this period, the digester was operated in 24 h cycle mode, whereas 20 h was given for the reaction phase (T_R) and 3 h given for settling (T_S). To have a good biomass, the supernatant was manually decanted from the uppermost of the reactor for 30 minwith the help of pump drivers (PD 5206) at a speed of 606 rpm. Batch feeding was performed mechanically through the top of the reactor at the beginning of the next cycle for 30 min at the same speed as the substrate was

Table 1. The general description of phase1 operational cycle.

Phase	Cycle period
Fill and mixing	30 min
React	20 h
Settle	3 h
Decant	30 min
Total cycle time	24 h

Table 2. General description of phase 2 operational cycle.

Phase	Cycle period
Fill and mixing	30 Min
React	46 h
Settle	1 h
Decant	30 Min
Total cycle time	48 h

Table 3. Characterization of composite wastewater in terms pollutant levels.

Parameters	Composite tannery wastewater
pH	9.49± 1.15
COD	11,980± 1033.71 mg/l
TN	1150± 131.26 mg/l
NO^{-}_{3}-N	320± 22.13 mg/l
NH4+-N	256 ± 72.13 mg/l
S-2	232± 28.44 mg/l
SO4-2	600± 74.55 mg/l
TN%	1.53

decanted.

During the second phase (Table 2), the ASBR was operated for 60 days (2 months) with a different cycle time from the first phase. The reactors were operated at 48 h cycle mode, where 46 h was given for the reaction period (T_R), 1 h for settling (T_S) and the remaining 1 h was for fill and decants, operated in the same way as in the first phase.

The total cycle time (t_c) is the sum of all the four phases as presented in Equation 1

$$T_C = T_F + T_R + T_S + T_D \quad (1)$$

Where, T_C = total cycle time; TF = total fill time; T_R = total react time; T_S = total settled time and T_{D} = total decant time.

Chemical analysis

Chemical Oxygen Demand (COD), total nitrogen (TN) nitrate-nitrogen (NO_3^--N), ammonium-nitrogen (NH_4^+-N), sulphides (S^{-2}), and sulphate (SO_4^{-2}) were determined colorimetrically using spectrophotometer (DR/2010 HACH, Loveland, USA) according to HACH instructions. Percent of removal efficiency (% RE) for each parameter was determined by the following Equation;

$$\% \, RE = \frac{Ci - Cf}{Ci} \, X \, 100 \quad (2)$$

Where, Ci = Initial parameter concentration; Cf = Final parameter concentration.

RESULTS AND DISCUSSION

Characterization of Modjo Tannery composite wastewater in terms of pollutant levels

The average value of chemical oxygen demand (COD), total nitrogen (TN), nitrate nitrogen (NO_3-N), ammonium nitrogen (NH_4^+-N), sulfide (S^{-2}) and sulphate (SO_4^{-2}) of the feedstock are presented in Table 3. The average COD and NO^{-}_{3}-N, content of the composite tannery wastewater was 11,980 and 320 respectively which is similar to those of Seyoum (2004) and Taddese (2010) and the average NH_4^+-N and S^{-2} was 256 and 232 mg/l respectively and similar with those of Hanna (2010) and Andualem (2008). The average value TN% in this study were 1.53.

The temperatures in this study were controlled at mesophilic condition (35 ± 2°C) and at thermophilic condition (55 ± 2°C). The pH of each digester was maintained between 6.02 to 7.66. As described in Table 3, the average pH of composite wastewater was 9.49. In the first week of the startup period, the pH of the substrate in the reactor was between 9.49 to 8.13; in the next weeks the pH decreased. This may be due to the formation of acids by acidogenic bacteria during the incubation period. Generally, the average value of the pH in the first stage reactor in this study was 6.02± 0.51 almost similar to the value 5.7 to 5.8 reported by Kasapgil et al. (1995). Acidity plays a crucial role in the breakdown of organic matter because pH affects the solubility of compounds which indirectly affect the accessibility by bacteria (Vieno et al., 2006). Extremely high or low pH levels are able to kill bacteria; deposition of organic matter occurs due to lack of degradation (Haandel and Lettinga, 1994). The pH of the second stage reactor in this study ranged from 6.30 to 7.66 and with an average pH of 7.26 ± 0.3 over the duration of the study. According to Gerardi (2002), suitable pH range for organic matter degradation is a range of 7 to 8.

Characteristics of the effluent

Evaluating the removal efficiency of pollutants

The performance of the whole system was evaluated to assess the removal efficiency of the characterized pollutant. The analysis test result of two-stage ASBR

Table 4. Removal efficiency of the Two Stage ASBR system.

Parameter	Effluent from D_1	Effluent from D_2	Effluent from D_3	Effluent from D_4	%Removal efficiency of D_1	%Removal efficiency of D_2	%Removal efficiency of D_3	%Removal efficiency of D_4
COD	5100	3550	3850	4650	57.42	70.36	67.86	61.18
TN	710	562	587	621	38.26	51.13	48.95	46
NO^-_3-N	177	124	143	165	44.68	61.25	55.31	48.43
NH_4^+-N	376	332	348	369	-	-	-	-
S_2^-	21	9	16	18	90.94	96.12	93.10	92.24
SO_4^{-2}	254	172	181	203	57.66	71.33	69.83	66.16

Concentration is in mg/L.

system is shown in Table 4.

The result in this study shows the average COD after anaerobic digestion of composite tannery wastewater was 5100, 3550, 3850, and 4650 mg/l, respectively from D_1 to D_4. Considerable removal efficiencies for COD were achieved (57.42, 70.36, 67.86 and 61.18%, respectively recorded from D_1 to D_4).

The main reason for good removal of COD could be related to maintenance of optimum environmental conditions like temperature and pH required for anaerobic acetogenic and methanogenic bacteria. According to Metcalf and Eddy (1991), environmental factors that affect biological organic matter removal are pH and inhibitory substances. pH level less than 6.8 affects biological organic matter removal while pH around neutral makes enables optimum performance to occur. Another factor could be related to the uptake of a substantial amount of organic matter by methanogenic and sulfate reducing bacteria. Moreover, in the mesophilic range, the bacterial activity and growth decrease by one half for each 10°C drop below 35°C (Hulshoff, 1995). Thus, for a given degree of digestion to be attained, the lower the temperature, the longer is the digestion time (Messay.and Mekibib (2017). Bacterial growth is sensitive to temperature because the high temperature can increase the fluidity of the phospholipid bilayer which leads to cell lysis. However, bacteria are known to have higher enzymatic activity at the higher temperature because of increased thermal energy (Meabe et al., 2013). The growth rates of thermophilic methanogens are 2-3 times higher than those of the mesophilic ones (Van Lier et al., 1993; Mladenovska and Ahring, 2000).

The TN before AD was 1150 mg/l and 710, 562, 587 and 621 mg/l after anaerobic digestion of composite tannery wastewater from D_1 to D_4. The removal efficiency of the digesters from D_1 to D_4 was 38.26, 51.13, 48.95 and 46%, correspondingly. The reduction of nitrogen in the effluent might have occurred due to the assimilation (followed by cell wastage) or the oxidation of ammonium into nitrite and nitrate by nitrifying bacteria (Metcalf and Eddy, 2003). The other factors might be associated with inhibition of nitrification by excessive COD loading. This can be attributed to the depletion of dissolved oxygen caused by heterotrophic organisms which utilized the organic matter present in the wastewater. Although COD levels up to 60-80 mg/L can be tolerated by nitrifying bacteria, it has been shown that COD levels above 60 mg/L can lead to as little as 50% nitrification (Wild et al., 1971). The optimum pH and temperature condition for the nitrification process were in the range of 6.5 to 8.6 and 20-30°C respectively (Grunditz and Dalhammar, 2001). The pH and the temperature of the reactor were 8.17 ± 0.18 pH units and 23°C, respectively. These were in the normal range of nitrification processes.

The average NO_3^--N after anaerobic digestion of composite tannery wastewater was 177,124,143 and 165 $mg.L^{-1}$ and the removal efficiency was 44.68, 61.25, 55.31 and 48.43%, respectively from D_1 to D_4. Nitrate was converted to gaseous nitrogen by denitrifying bacteria with optimum temperature and other driven parameters. The abundance of highly efficient denitrifying bacteria in system could be directly related to the removal efficiency of the system. In wastewater, denitrification is most effective at pH values between 7.0 and 8.5 and the optimum is around 7.0 (Metcalf and Eddy, 1991). Denitrification favors a temperature range of 35 –50°C. It also occurs with the temperature range of 5–100°C) at a slower rate. The biological activity will decrease by a factor of about 3 with an associated temperature drop of 15ºC (Levenspiel, 1972). Therefore, the environmental condition at thermophilic phase favors the removal of NO_3^--N. D_2 (thermophilic-thermophilic) removes high amount of NO_3^--N and D_1 (mesophilic – mesophilic) removes the lowest amount of NO_3^--N.

The removal efficiency of S_2^- was 90.94, 96.12, 93.10, and 92.24% recorded from D_1 to D_4. Further in this study, the removal efficiency for SO_4^{-2} was 57.66, 71.33, 69.83 and 66.16%, respectively from D_1 to D_4. Sulfate reduction in anaerobic system could be related to the use of acetate and hydrogen by sulfate reducers which reduces sulfate to hydrogen sulfide. Like methanogens, some sulfate reducers can oxidize H_2 and acetate and thus may compete with methanogens for these substrates (Rinzema and Lettinga, 1988). Thermodynamic and

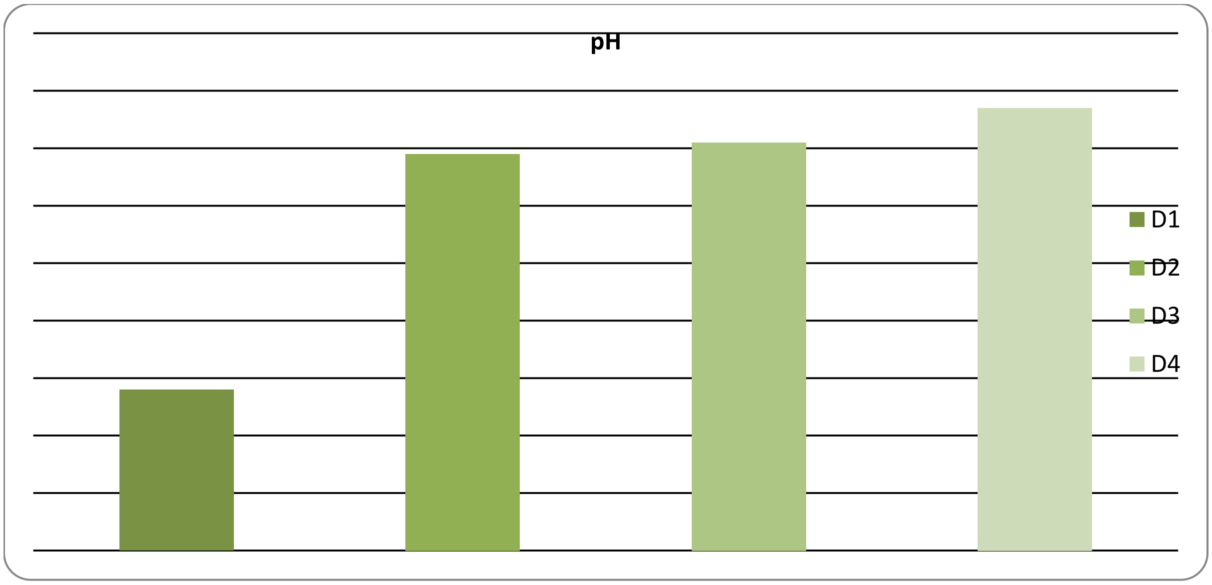

Figure 3. Average ph of the effluent.

Monod-kinetic data shown that sulfate reducer generally have higher growth rates and higher affinity for substrates than acetogenic and methanogenic bacteria. Therefore, sulfate reducing bacteria out-competes acetogenic and methanogenic bacteria (Oude Elferink et al., 1994).

Another reason for sulfide removal in anaerobic reactor could be its use as sulfur source by anaerobic bacteria. Methanogenic bacteria use ammonia and sulfide as nitrogen and sulfur sources respectively. Although un-ionized sulfide is toxic to methanogens at level exceeding 150–200 mg/L (Rinzema and Lettinga, 1988; Speece, 1983), the concentration of sulfide in anaerobic SBR effluent was 232 mg/L which was far lower than the limit. Therefore, it favored methanogens to use sulfide as sulfur source to synthesis of new biomass.

The concentration of NH_4^+-N increased in all digesters; it was observed that the feed NH_4^+-N was slightly lower than the effluent NH_4^+-N which indicates that there is no reduction of NH_4^+-N during AD. Kheradmand et al. (2010) found the similar observation of increase of NH_4^+-N concentration in effluent than feed by 8.7 to 31.6%. In addition, Bohdziewicz et al. (2008) observed increase in NH_4^+-N concentration treating leachate. Similarly, this experiment calculates the increase of concentration by 23%. The increase of NH_4^+-N concentrate ion is mainly due to ammonia production by degradation of protein and amino acid of leachate.

pH of the effluent

The average pH value of the effluent for each digester in this study is summarized in Figure 3.

Determination of pH plays an important role in the wastewater treatment process. The average pH value of the effluent varied from 7.18 to 7.67. The minimum and maximum pH accepted values for slurry was 6.0 and 8.5, respectively (Fokhrul, 2009). In addition, William (1998) reported that the values lie in the range of the pH of the compost 6 to 7.

Conclusions

The current study demonstrated that two stage anaerobic sequential batch reactor has a great potential for treating composite tannery wastewater under thermophilic-thermophilic condition and used as wastewater management option. Moreover, this system of managing wastewater significantly contributes towards resource-recovery and pollution management around tannery industry.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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