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## Biological Treatment of Tannery Wastewater - A Review

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### ABSTRACT

Tannery wastewaters are highly complex and are characterized by high contents of organic, inorganic and nitrogenous compounds, chromium, sulfides, suspended solids and dissolved solids. Treatment of tannery wastewater is carried out by physical or chemical or biological or combination of these methods. This study reviews various biological treatment methods applied for tannery wastewater. Characteristics of wastewaters from different tanneries and various methods for treating these tannery wastes are discussed. It was noted that the Chemical Oxygen Demand (COD) removal efficiencies and process capacities were affected by the variations in organic loading rates, presence of chromium and sulfides. The review shows that all aerobic processes have a similar level of COD removal, but the highest COD removal efficiency at a high organic loading rate was observed in anaerobic reactors. Upflow Anaerobic Sludge Blanket Reactor (UASB) exhibited better performance for treating high strength tannery wastewater effectively, compared with conventional reactors. Both aerobic and anaerobic processes are employed for the treatment of tannery wastewater. From the review it can be concluded that physical/chemical processes combined with biological process is the better option for the treatment of tannery wastewater.

**Key words:** Tannery, wastewater treatment, UASB, SBR, membrane bio reactor, chromium, sulfide

### INTRODUCTION

Tanning is one of the oldest industries in the world. During ancient times, tanning activities were organized to meet the local demands of leather footwear, drums and musical instruments. With the growth of population, the increasing requirement of leather and its products led to the establishment of large commercial tanneries. Two methods are adopted for tanning of raw hide/skin viz., vegetable tanning and chrome tanning. The production processes in a tannery can be split into four main categories: (1) Hide and skin storage and beam house operations, (2) tanyard operations, (3) post-tanning operations and (4) finishing operations.

Tanneries are typically characterized as pollution intensive industrial complexes which generate widely varying, high-strength wastewaters. Variability of tannery wastewaters are not only from the fill and draw type operation associated with tanning processes, but also from the different procedures used for hide preparation, tanning and finishing. These procedures are dictated by the kind of raw hides employed and the required characteristics of the finished product. Tanning industry also has one of the highest toxic intensity per unit of output (Khan *et al.*, 1999). During tanning process at least about 300 kg chemicals are added per ton of hides (Verheijen *et al.*, 1996). Tannery effluent is among one of the hazardous pollutants of industry. Major problems are

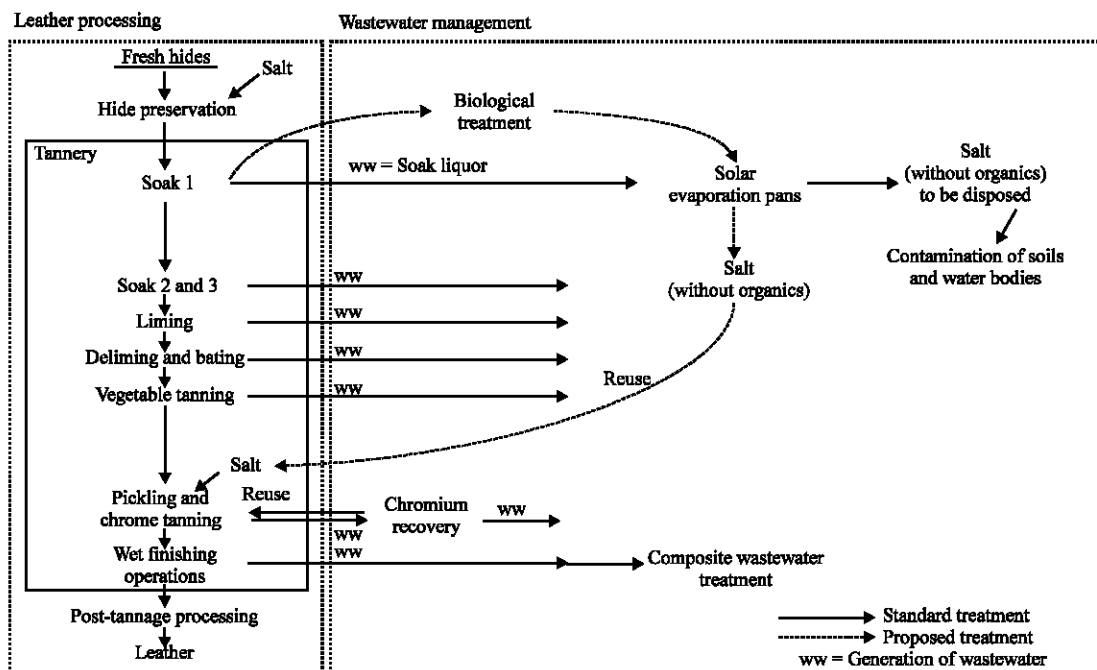


Fig. 1: Simplified leather production chain and management of the effluents associated (Lefebvre *et al.*, 2005)

due to wastewater containing heavy metals, toxic chemicals, chloride, lime with high dissolved and suspended salts and other pollutants (Uberoi, 2003). Tanneries generate wastewater in the range of 30-35 L kg<sup>-1</sup> skin/hide processed with variable pH and high concentrations of suspended solids, BOD, COD, tannins including chromium (Nandy *et al.*, 1999). The growth of industrialization has encroached even to small townships and villages along with all ills of pollution. The tanning process and the effluents generated have already been reported in literature (Sreeram and Ramasamy, 2003; Stoop, 2003) and an overview is presented in Fig. 1. In this review the characteristics of tannery wastewater are discussed and an effort has been made to give a brief idea of an approach to tannery wastewater treatment, particularly discussing and highlighting in brief the biological methods.

### TANNERY WASTEWATER CHARACTERISTICS

The characteristics of tannery wastewater vary considerably from tannery to tannery depending upon the size of the tannery, chemicals used for a specific process, amount of water used and type of final product produced by a tannery. Tannery wastewater is characterized mainly by measurements of Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), suspended solids (SS) and Total Dissolved Solids (TDS), chromium and sulfides etc. Typical characteristics of tannery wastewater are given in Table 1.

In general, tannery wastewaters are basic, have a dark brown colour and have a high content of organic substances that vary according to the chemicals used (Kongjao *et al.*, 2008). The tannery wastewater is characterized by substantial organic matter content and high SS content, resulting in an average total COD concentration of 6200 mg L<sup>-1</sup> and a SS concentration of 5300 mg L<sup>-1</sup>. Very high salinity was reflected by an average TDS concentration of 37,000 mg L<sup>-1</sup>. Total Kjeldah

Table 1: Physico-chemical characteristics of tannery wastewater

Reference	pH	TDS	Total solids	Suspended solids	Volatile		COD	BOD	TKN	Ammonical/	
					suspended solids					nitrogen	Chromium
Apaydin <i>et al.</i> (2009)	7.4	---	---	2890	1280	3700	1470	---	180	---	440
Ganesh <i>et al.</i> (2006)	7.08±0.28	---	10265±1460	2820±140	1505±90	4800±350	---	225±18	128±20	95±55	---
Kongjiao <i>et al.</i> (2008)	7.0-8.7	13,800-19,700	---	600-955	---	4100-6700	630-975	144-170	---	11.5- 14.3	---
Koteswari and Ramanibai (2008)	---	15,152	---	2004	1660	8000	930	---	---	11.2	228
Lefebvre <i>et al.</i> (2005)	7.70	36,800	---	5300	1300	2200	---	270	---	---	---
Leta <i>et al.</i> (2004)	10.72	6810	---	---	---	11153.67	2906	---	162.15	32.87	507.5
Orhon <i>et al.</i> (2000)	7.79	---	---	915	578	2155	---	228	168	50.9	35.8
Ram <i>et al.</i> (1999)	10.5	17,737	18,884	1147	---	3114	1126	---	33.0	83.00	55.00
Szpyrkowicz <i>et al.</i> (2005)	7.7	---	---	---	---	2426	---	370	335	29.3	286
Thanigavel (2004)	8.2-8.5	14750	19775	5025	---	5650	---	---	---	---	---

Except pH all values are in (mg L<sup>-1</sup>)

Nitrogen (TKN), N-NH<sub>3</sub> and PO<sub>4</sub><sup>3-</sup> averaged 273, 153 and 21 mg L<sup>-1</sup>, respectively. Tannery wastewaters are basic and their high organic content can cause considerable environmental pollution (Leta *et al.*, 2004). The pH values of tannery wastewaters range from 7.5 to 10, as shown in Table 1 (Kongjiao *et al.*, 2008; Leta *et al.*, 2004). The influents were characterised by high alkalinity content with a resulting pH value of above 8 due to the chemicals used in leather processing. Influent total nitrogen and COD concentrations ranged from 927 to 2140 mg L<sup>-1</sup> and 9583 to 13515 mg L<sup>-1</sup>, respectively, whereas influent ammonium N varied from 149 to 178 mg L<sup>-1</sup>. Sulfide and total chromium concentrations were in the range of 466.3-794 and 23.3-42.5 mg L<sup>-1</sup>, respectively, during the process feeding stages. It is also observed that tannery effluents are rich in nitrogen, especially organic nitrogen, but very poor in phosphorus. In addition to organic and nitrogen compounds, tannery wastewaters contain sulfide, chromium, which impart high antibacterial activity (Wiemann *et al.*, 1998; Wiegant *et al.*, 1999). Several problems have been encountered during the biological treatment of tannery wastewater because of high toxicity. The inhibition of biodegradation due to the presence of chromium and sulfides demonstrates the antibacterial activity. High concentrations of these constituents make the possible discharge of tannery wastewaters into water bodies problematic, as they cause eutrophication and other adverse environmental effects (Leta *et al.*, 2004). Table 1 shows great variability in the quality of the influent. Great variability was observed with respect to the influent, depending on the type of hides and skins and the region from which they came, at the time of the sampling (Lefebvre *et al.*, 2005).

The BOD<sub>5</sub>/COD ratio was 0.3, which was very low in comparison to domestic wastewater (i.e., 0.5). Therefore, the biodegradability of the influent was found to be low, according to the criteria of Ahn *et al.* (1999). However, BOD<sub>5</sub> is a controversial parameter, when it is applied to tannery wastewater, since it contains many inhibitors of BOD<sub>5</sub> (Ates *et al.*, 1997). The VSS/SS ratio averaged 0.2±0.1, was found to be very low due to the numerous fibres and inorganic particulate (sand, dust) escaping the soak pit. The VSS/(total COD-soluble COD) ratio averaged 1.2±0.7, indicated that every kg of VSS contributed to 1.2 kg of particulate COD. Finally, the COD/N/P ratio averaged 200/22/2 and showed that the waste liquor contained high amounts of nitrogen but lesser amounts of phosphorus. However, no phosphorus deficiency could be identified and this ratio is close to that of domestic wastewater (Lefebvre *et al.*, 2005).

## TREATMENT METHODS

Many conventional processes were carried out to treat wastewater from tannery industry such as biological process (Vijayaraghvan and Murthy, 1997; Wiemann *et al.*, 1998; Farabegoli *et al.*, 2004), oxidation process (Schrank *et al.*, 2003; Sekaran *et al.*, 1996) and chemical process (Song *et al.*, 2004).

**Biological treatment methods:** Biological treatment of wastewater is evaluated as a good treatment method for industrial effluents. Treatment of wastes with bacteria involves the stabilization of waste by decomposing them into harmless inorganic solids either by aerobic or anaerobic process. In aerobic process, the decomposition rate is more rapid than the anaerobic process and it is not accompanied by unpleasant odours, whereas in anaerobic process, longer detention period is required and gives unpleasant odours.

The processes used most frequently for biological treatment of tannery wastewater in CETPs in India are the Activated Sludge Process (ASP) and the Upflow Anaerobic Sludge Blanket (UASB) process (Jawahar *et al.*, 1998; Kadam, 1990; Rajamani *et al.*, 1995). In general, ASP-based treatment is considered to be energy-intensive and expensive from an operation and maintenance point of view. On the other hand, anaerobic processes claim to offer several advantages, especially under tropical climatic conditions (Rajamani *et al.*, 1995, 1997). However, a comprehensive comparison of the relative merits of tannery wastewater treatment by these two processes with field data has not yet been performed. As such, it is imperative that experience and knowledge gained through the operation of full-scale treatment plants treating tannery effluents employing both ASP and UASB processes is properly utilized.

High variability in the organic content (reflected by COD concentration) and salinity (reflected by TDS concentration) of the soak liquor might make the proper operation of a biological treatment plant uneasy, causing important disturbance in the equilibrium of the microbial community. Yet, looking forward to applying the process at an industrial scale, a decision was taken not to artificially change the influent characteristics in order to make it more homogeneous. This choice resulted in frequent changes in the environmental conditions in the bioreactor. The results of some tannery wastewater treated by biological methods are given in Table 2.

**Aerobic biological treatment methods:** Biodegradation of tannery wastewater using activated sludge process has been reported by many research workers (Jawahar *et al.*, 1998; Murugesan and Elangoan, 1994; Eckenfelder, 2002; Tare *et al.*, 2003). The performance of activated sludge process is affected by many factors. Various parameters of importance relating to growth of microorganisms and substrate utilization on which the operation of the reactor is based include mean cell residence time, Mixed Liquor Volatile Suspended Solids (MLVSS) concentration, hydraulic detention time, i.e., aeration time, food to microorganism (F:M) ratio and the dissolved oxygen the reactor. All these studies indicate a BOD<sub>5</sub> removal of 90 to 97% for the tannery effluent concluding activated sludge process as highly useful for the purpose.

An ASP was used for the treatment of tannery wastewater. It was operated continuously for 267 days. Settled tannery wastewater was used as influent to the aeration tank. A removal efficiency of above 90 and 80% for BOD<sub>5</sub> and COD was obtained when the ASP is operated at an MLVSS concentration of 3500 mg L<sup>-1</sup> keeping an aeration time of 12 h (Haydar *et al.*, 2007).

A Common Effluent Treatment Plant (CETP) based on activated sludge process was employed for the treatment of tannery effluent. A significant reduction in COD and BOD levels were

Table 2: Summary of studies on the treatment of Tannery wastewater

Reference	Organisms	COD (%)	BOD (%)	Chromium	Sulfide
Calheiros <i>et al.</i> (2008)		63.78	51.44%		
Di Iaconi <i>et al.</i> (2002)		97			
Ganesh <i>et al.</i> (2006)		80-82			
Goltara <i>et al.</i> (2003)		90			
Haydar <i>et al.</i> (2007)		80	90%		
Lefebvre <i>et al.</i> (2005)	Halophiles	95			
Lefebvre <i>et al.</i> (2006)		78			
Leta <i>et al.</i> (2004)		98			
Mandal <i>et al.</i> (2010)	<i>Thiobacillus ferrooxidans</i>	69	72%	5	
Mazumder <i>et al.</i> (2008)		70.9			
Onyancha <i>et al.</i> (2008)	<i>S. condensata</i> <i>R. hieroglyphicum</i>			>75	
Vankar and Bajpai (2008)	<i>Trichoderma</i> sp.			97.39	
Rajasimman <i>et al.</i> (2007)	Mixed	46-85	65-93%		
Ramteke <i>et al.</i> (2010)	<i>Escherichia coli</i> <i>Vibrio</i> sp. <i>Pseudomonas</i> sp.	98.46 87.5 96.15	90%		
Ryu <i>et al.</i> (2007)		75			
Sekaran <i>et al.</i> (1996)		81	85%		100%
Shakoori <i>et al.</i> (2000)	Bacterial strain			87	
Sivaprakasam <i>et al.</i> (2008)	<i>P. aeruginosa</i> B. <i>flexus</i> <i>E. homiense</i> <i>S. aureus</i>	80			
Song <i>et al.</i> (2003)		60-75			
Song <i>et al.</i> (2004)		32		77	
Srivastava <i>et al.</i> (2007)	<i>Acinetobacter</i> sp.			90	
Srivastava and Thakur (2006)	<i>Aspergillus</i> sp. <i>Hirsutella</i> sp.			70	
Sumathi <i>et al.</i> (2005)	Biological waste (coir pith)			94	
Thanigavel (2004)	Mixed	89.5			
Vidal <i>et al.</i> (2004)		80	99%		
Wang <i>et al.</i> (2007)	<i>A. thiooxidans</i>			99.7	

achieved during the course of treatment in CETP. A reduction of 98.46, 87.5 and 96.15% in bacterial counts especially in pathogens like *Escherichia coli*, *Vibrio* sp. and *Pseudomonas* sp. were observed after treatment. Pathogens were not detected in the dried sludge. Complete elimination of fecal streptococci was observed in treated effluent. Around 10.8% of microbial isolates from the effluent showed ability to reduce chromate >90%. In treated effluent chromium level was 5.48 mg L<sup>-1</sup>, which exceeds the statutory limit of Indian standards (Ramteke *et al.*, 2010).

When the organic load rate is lower than 2 g COD/L per day, the biological oxygen demand (BOD<sub>5</sub>) efficiency was 99%, whereas the Chemical Oxygen Demand (COD) was around 80% for the diluted unhairing wastewater after being treated by an Activated Sludge (AS) system. The AS system was fed for 180 days with diluted unhairing effluent. The operation strategy increased the Organic Load Rate (OLR) from 0.23 to 2.98 g COD/L/d while the HRT was variable until operation day 113. Results show that the reactor operation was stable until 2 g COD/L per day. For higher

values, the system was less efficient (COD and BOD<sub>5</sub> removal rate lower than 40%) and the relation of food/micro-organisms (F/M) was higher than 0.15. Biomass evaluations through oxygen utilization coefficients show that the specific oxygen uptake rate decreased from 1.11 to 0.083 g O<sub>2</sub>/g MLVSS per day, in the same way the endogenous oxygen coefficient decreased from 0.77 to 0.058 per day. The reduction of biomass activity could be attributable to the inorganic compound content (ammonia and chloride) in the unhairing effluent (Vidal *et al.*, 2004).

Mazumder *et al.* (2008) used a shaft-type hybrid bioreactor for treating composite wastewater of chrome tannery. In this reactor, the sample was treated under suspended growth and then hybrid system with 20 g L<sup>-1</sup> of 5 mm tyre tube beads in batch mode. The continuous study was made under suspended growth and hybrid system with 10-30 g L<sup>-1</sup> of beads. The maximum COD removal was 70.9% under a loading rate of 5.250 kg/day/m<sup>3</sup>. The overall removal rate ranged from 0.0824-0.1004/h for the hybrid system.

Salinity of tannery wastewater makes it difficult to be treated by conventional biological treatment. Salt tolerant microbes can adapt to these saline conditions and degrade the organics in saline wastewater. Tannery saline wastewater obtained from a Common Effluent Treatment Plant (CETP) near Chennai (southern India) was treated with pure and mixed consortia of four salt tolerant bacterial strains viz., *Pseudomonas aeruginosa*, *Bacillus flexus*, *Exiguobacterium homiense* and *Staphylococcus aureus* which are isolated from marine and tannery saline wastewater samples. Experiments with optimized conditions and varying salt content (between 2 and 10% (w/v) were conducted. Salt inhibition effects on COD removal rate were noted. Comparative analysis was made by treating the tannery saline wastewater with activated sludge obtained from CETP and with natural habitat microbes present in raw tannery saline wastewater. Salt tolerant bacterial mixed consortia showed appreciable biodegradation at all saline concentrations (2, 4, 6, 8 and 10% w/v) with 80% COD reduction in particular at 8% salinity level the consortia could be used as suitable working cultures for tannery saline wastewater (Sivaprakasam *et al.*, 2008). The COD removal results for tannery saline waste stream by natural habitat strains as well as activated tannery sludge proved they were not suitable and that specialized consortia (salt tolerant) were needed for efficient treatment. The identified salt tolerant bacterial consortia is considered as a suitable working culture for efficient biodegradation of tannery saline wastewater.

The pre denitrification-nitrification process found to be efficient for simultaneous removal of nitrogen and organic substrates from tannery wastewaters. Normally chromium III will not cause process inhibition during performance operations. A pilot wastewater treatment plant consisting of a pre denitrification-nitrification process was constructed and operated for 6 months. Up to 98% total nitrogen and chemical oxygen demand and 95% ammonium nitrogen removal efficiencies were achieved in the system. The average effluent ammonium nitrogen ranged from 8.4 to 86 mg L<sup>-1</sup>, whereas the average effluent for nitrate nitrogen ranged from 2.9 to 4.4 mg L<sup>-1</sup>. The average values of denitrification and nitrification rates determined by nitrate and ammonium uptake rates (NUR and AUR) were 8.0 mg NO<sub>3</sub>-N [g volatile suspended solids (VSS)]<sup>-1</sup> h<sup>-1</sup> and 5.4 mg NH<sub>4</sub>-N (g VSS)<sup>-1</sup> h<sup>-1</sup>, respectively, demonstrating that the treatment processes of the pilot plant were effective. Further studies of the effect of chromium III on AUR showed 50% inhibition at a concentration of 85 mg L<sup>-1</sup>, indicating that this metal was not causing process inhibition during performance operations (Leta *et al.*, 2004).

A study on the biological nitrogen removal from tannery wastewaters without a preliminary chemical-physical phase or an external carbon source for denitrification were carried out by Szpyrkowicz *et al.* (1991). They reported the removal of nitrogen by biological means from

wastewaters consisting of up to 90% chrome tannery and 10% domestic sewages. Experiments were carried out over a 6 month period in a pilot plant of the modified Ludzack-Ettinger configuration. The COD utilization coefficient was 12.5 mg COD for 1 mg of denitrified N. No inhibition of the process was induced by Cr or by  $S^{2-}$  present in the raw wastewaters. A negative effect on the denitrification rate resulted from a high ratio between the quantity of oxygen returned with the mixed liquor and the inlet COD.

Hypersaline wastewater (i.e., wastewater containing more than  $35 \text{ g L}^{-1}$  Total Dissolved Solids (TDS) generated from soak pit of the tannery industry was difficult to treat using conventional biological wastewater treatment processes. The characterization of the soak liquor showed that this effluent is biodegradable, though not easily and highly variable, depending on the origin and the nature of the hides. A lab-scale SBR was used to treat this soak liquor seeded with halophilic bacteria and the performance of the system were evaluated under different operating conditions with changes in hydraulic retention time, organic loading rate and salt concentration. The changes in salinity appeared to affect the removal of organic matter more than the changes in hydraulic retention time or organic loading rate. Despite the variations in the characteristics of the soak liquor, the reactor achieved proper removal of organic matter, once the acclimation of the microorganisms was achieved. Optimum removal efficiencies of 95, 93, 96 and 92% on COD,  $\text{PO}_4^{3-}$ , TKN and SS, respectively, were achieved with a HRT of 5 days, an organic loading rate of  $0.6 \text{ kg COD/m}^3/\text{d}$  and  $34 \text{ g NaCl/L}$ . The organisms responsible for nitrogen removal appeared to be the most sensitive to the modifications of these parameters (Lefebvre *et al.*, 2005).

The wastewater, produced after the oxidation of sulfide compounds coming from the beam house section of a tannery, contained average COD and ammonium concentrations of  $550$  and  $90 \text{ mg L}^{-1}$ , respectively. Goltara *et al.* (2003) used a Membrane Sequencing Batch Reactor (MSBR) to treat this wastewater. The MSBR was operated for a period of 150 days, with no sludge removal during the whole period of operation. The biomass concentration inside the reactor varied considerably, with maximum values close to  $10 \text{ g L}^{-1}$  at the end of operation. Low biomass yield values were achieved probably due to the low Feed/Microorganisms (F/M) ratio. An important accumulation of organic matter in the reactor was noticed, although the COD effluent was not affected due to the permeation through the membrane. Removal efficiencies close to 100% in ammonium and 90% in COD were achieved and the Total Nitrogen removal efficiency ranged from 60 to 90% (Goltara *et al.*, 2003).

The feasibility of treating tannery wastewater containing chromium, an inhibiting compound, was studied by Farabegoli *et al.* (2004). The maximum chromium concentration tolerated by microorganisms is determined through aerobic and anoxic batch experiments and the biomass inhibition process is analyzed in a lab scale sequential batch reactor at higher chromium concentrations. It was observed that the chromium addition had less influence on the denitrification bacteria than on the nitrification bacteria. In addition, it was observed that nitrification and denitrification rates, at the same chromium concentration, were higher in the SBR reactor than in batch experiments with unacclimated biomass. Experimental results confirm that sequencing batch reactors are able to produce a more resistant biomass, which acclimates quickly to inhibiting conditions.

Thanigavel (2004) employed an inverse fluidized bed bioreactor for the treatment of tannery wastewater. A low density particle (Polypropylene-Density,  $890 \text{ kg m}^{-3}$ ) was used. From the result it was found the maximum COD removal of 91.3% was achieved in this reactor at a bed height of 80 cm and for the air flow rate of  $64.4 \text{ cc/s}$ .



SBR coupled with respirometry is a cost-effective and a clear alternative to the conventional biological system for the treatment of tannery wastewater (Ganesh *et al.*, 2006). The removal efficiencies are much higher than the continuous aerobic systems. Such enhanced performance with SBR over conventional activated sludge process is perhaps due to the enforced short-term unsteady state conditions, which facilitates the required metabolic conditions for treatment of wastewater. The removal of COD by degradation is stoichiometric with oxygen usage. Measurement of oxygen uptake rates and corresponding COD uptake rates showed that a 12 h operating cycle was optimum for tannery wastewater. A plot of OUR values provided a good indication of the biological activity in the reactor. At a 12-h SBR cycle with a loading rate of 1.9- 2.1 kg m<sup>-3</sup> d<sup>-1</sup>, removal of 80-82% COD, 78-80% TKN and 83-99% NH<sub>3</sub>-N were achieved. These removal efficiencies were much higher than the conventional aerobic systems. A simple method of COD fractionation was performed from the OUR and COD uptake rate data of the SBR cycle. About 66-70% of the influent COD was found to be readily biodegradable, 10-14% was slowly degradable and 17-21% was non-biodegradable. The oxygen mass transfer coefficient, K<sub>La</sub> (19±1.7 h<sup>-1</sup>) was derived from respirometry. It was observed that with the exception of high organic load at the initial feed the oxygen transfer capacity was in excess of the OUR and aerobic condition was generally maintained. Simultaneous nitrification-denitrification was observed in the SBR during the feed period as proved by mass balance (Ganesh *et al.*, 2006).

**Anaerobic biological treatments:** The combination of the UASB with an aerobic post-treatment enhanced the performance of the overall wastewater treatment process and the COD removal efficiency. However, for effective operation, the system had to be operated at very low OLRs, which affects the economic viability of such a process. The anaerobic digestion of tannery soak liquor was studied by using a UASB. COD removal reached 78% at an OLR of 0.5 kg COD m<sup>3</sup>/d, a HRT of 5 days and a TDS concentration of 71 g L<sup>-1</sup>. The combined anaerobic/aerobic treatment system reached 96% (Lefebvre *et al.*, 2006).

Banu and Kaliappan (2007) made an attempt to treat the tannery wastewater by using hybrid upflow anaerobic sludge blanket reactors. The advantage of this reactor is both the fixed film and upflow sludge blanket treatment. The reactor was operated for 370 days at two different hydraulic retention times viz., 60 and 70 h. The average concentration of COD and tannin in the influent tannery wastewater used were 14000 mg L<sup>-1</sup> and 1987 mg L<sup>-1</sup>, respectively. The reactor performed to its maximum at an organic loading rate of 2.74 kg COD/m<sup>3</sup>/d and 3.14 kg COD/m<sup>3</sup>/d at a HRT of 70 and 60, respectively. They also found that increase in OLR beyond 2.74 kg COD/m<sup>3</sup>/d and 3.14 kg COD/m<sup>3</sup>/d caused a gradual decrease in COD removal efficiency. The degradation of inhibitor substance such as tannin during the anaerobic digestion were also found and it was in the range of 65-91% at a HRT of 70 h.

The retention of the active methanogenic biomass in the form of biofilm supported by a carrier has been developed to treat tannery wastewater ensures with minimum particle washout independent of the HRT. The bio film colonisation on two types of micro carriers was compared. Porous polyurethane foam material was found to be more suitable than Raschig rings as a micro carrier in the UAFBR reactor. Both SEM and TEM techniques were used to study the morphology of the bio film, the location of bacteria in the bio film and species diversity and community structure. In an Upflow Anaerobic Fixed Biofilm Reactor (UAFBR), which retains a high concentration of accumulated biomass the effects of major process variables such as hydraulic retention time, organic loading rate and temperature on chemical oxygen demand removal and

methane yield performances of the reactor were evaluated. This aids the conversion of COD and results in a reduction in effluent suspended solids. COD removal (60-75%) and methane yield ( $0.36 \text{ m}^3 \text{ CH}_4/\text{kg COD}_{\text{rem}}$ ) remained stable across a range of organic loading rates and under conditions of temperature shock. The COD removal and methane yield is higher than the values reported previously (Song *et al.*, 2003).

A long-term study was conducted to identify the process of biological sulfate reduction in anaerobic two-stage pilot plants treating tannery wastewater. Influence of quality and quantity of wastewater on sulfate removal in both stages of the pilot plant was simultaneously tested. In the first stage, desulfurization increased with higher feed flow but the desulfurization then decreased in the second stage. The concentration of sulfate in the influent had a significant influence on the desulfurization in both stages of the pilot plants. The removal of sulfate in the first stage was approximately 30%, whereas in the second stage the desulfurization decreased with higher concentrations of sulfate in the influent. Operational parameters were adjusted in order to restrict the biological sulfate reduction to the first stage. Compared to pH 5 or 6 in the influent, a pH of 7 most increased biological sulfate reduction in the first stage. No significant influence on COD removal or volume of gas was observed. For three pilot plants operated parallel to each other, no significant difference in desulfurization was noticed (Genschow *et al.*, 1996).

Rajasimman *et al.* (2007) studied the performance of a single UASB reactor for treating both the solid (generated from fleshing) and liquid wastes. In this study, UASB reactor was operated at different organic loading rates ranges between  $5\text{-}12 \text{ kg/m}^3/\text{d}$ . From the results it was observed that the COD removal efficiency of 46-85% and BOD removal efficiency of 65-93% were achieved. The gas production was in the range of 2-15 L for the given organic load.

**Wetlands:** Wetlands planted with *Typha latifolia* proved to be tolerant to high organic loadings and to interruptions in feed during biological treatment of tannery wastewater under long-term operation. Two expanded clay aggregates (Filtralite, MR3-8-FMR and Filtralite, NR3-8-FNR) and a fine gravel-FG were used as substrate for the constructed wetland units plus one unit with FMR was left as an unvegetated control. The systems were subject to three hydraulic loadings, 18, 8 and  $6 \text{ cm d}^{-1}$  and to periods of interruption in the feed. The relationship between the substrate, plant development and removal efficiency, especially of organic matter, was investigated. Organic loadings up to  $1800 \text{ kg BOD}_5/\text{ha/d}$  and  $3849 \text{ kg COD}/\text{ha/d}$  were applied leading to mass removals of up to  $652 \text{ kg BOD}_5/\text{ha/d}$  and  $1869 \text{ kg COD}/\text{ha/d}$ , respectively. The three different substrates were adequate for the establishment of *T. latifolia*, although the clay aggregates allowed for higher plant propagation levels. The units with FNR and FMR achieved significantly higher COD and  $\text{BOD}_5$  removal when compared to the FG and to the unplanted units (Calheiros *et al.*, 2008).

**Membrane Bio-Reactors (MBR):** MBR was used to treat tannery wastewater. Bench scale membrane bio-reactors were operated to investigate the treatment efficiency of tannery wastewater with high organic and nitrogen contents and the optimum operating conditions. The optimum ratio of the volume of anoxic denitrification tank to aerobic nitrification tank was 50%. Under these conditions, the effluent COD and Total Nitrogen were  $160$  and  $54 \text{ mg L}^{-1}$ , respectively, which satisfied the effluent limits for the tannery wastewater. It was also observed that supplementation of phosphorus to maintain COD: P ratio of 100:1 is needed to achieve the best performance (Chung *et al.*, 2004).

Munz *et al.* (2007) carried out the treatment of tannery wastewater in a pilot scale membrane bioreactor with Powdered Activated Carbon (PAC). The addition of powdered PAC to analyze improvements in effluent quality and in the filtration process, improvements in COD removal are found to be low, but not negligible and greater than the PAC adsorption effect alone. The results refer to a pilot plant monitoring stretched over a period of 594 days: 380 without PAC, 123 with a PAC concentration of  $1.5 \text{ g L}^{-1}$  and 91 with  $3 \text{ g L}^{-1}$ . The sludge residence time and hydraulic retention time were maintained between 30 and 90 days and 50 and 100 h, respectively. COD removal stability appeared to increase as PAC concentration increased. No effects were observed on the nitrification processes. The filtration process was evaluated in terms of sludge filterability, fouling rate and fouling reversibility. The fouling rate decreased with an increasing PAC concentration and showed complete reversibility both in presence and in absence of PAC.

The role that tannins play in terms of biodegradability did not appear to be significant in tannery wastewater treatment. The methodology has established the preliminary use of respirometry to examine the biodegradability of a selection of commercial products; the subsequent analysis, by means of spectrophotometric reading and RP-IPC (Reverse-Phase Ion-Pair) liquid chromatography, estimates the concentrations of natural tannins and naphthalenesulfonic tanning agents in the influent and effluent samples. A Membrane Bioreactor plant and a full scale Conventional Activated Sludge Process (CASP) plant is employed in parallel to evaluate the results. The results show that a consistent percentage of the Total Organic Carbon (TOC) in the effluent of the biological phase of the plants is attributable to the presence of natural and synthetic (Sulfonated Naphthalene-Formaldehyde Condensates, SNFC) tannins (17 and 14%, respectively). The titrimetric tests that were aimed at evaluating the levels of inhibition on the nitrifying biomass samples did not allow a direct inhibiting effect to be associated with the concentration levels of the tannin in the effluent. Nonetheless, the reduced specific growth rates of ammonium and nitrite oxidising bacteria imply that a strong environmental pressure is present, if not necessarily due to the concentration of tannins, due to the wastewater as a whole (Munz *et al.*, 2009).

**Combination of biological methods with physical/chemical methods:** Due to the extremely changing quality of the raw wastewater, tan-yard wastewater, the biological pre-treatment could not be stabilized all the time and nitrification was sometimes inhibited. Oxidative treatment distinctly improved the aerobic biodegradability of refractory organic compounds and found to be optimal in the range of a specific ozone consumption of about  $2 \text{ g O}_3/\text{g DOC}_o$  for both batch experiments and continuous operating conditions. Moreover, full nitrification could be established during the subsequent aerobic degradation and the remaining ammonia is completely removed. The biological treatment of the tannery wastewater substreams beamhouse (BH, pre-tanning steps) and tan-yard wastewater (TY, tanning and wet-finishing process steps) and the application of an oxidative treatment by ozone, followed by a second aerobic treatment were investigated. It can be stated that the combined oxidative and biological treatment of BH and TY was effective and ensures the meeting of given COD and ammonia-limits for the direct discharge of this special industrial wastewater into rivers (Jochimsen *et al.*, 1997).

Degradation of leather industry wastewater by aerobic treatment incorporating *Thiobacillus ferrooxidans*, Fenton's reagents and combined treatment was studied by Mandal *et al.* (2010). The sole treatment by Fenton's oxidation involving the introduction of  $6 \text{ g FeSO}_4$  and  $266 \text{ g H}_2\text{O}_2$  in a liter of wastewater at pH of 3.5 and  $30^\circ\text{C}$  for 30 min at batch conditions reduced COD,  $\text{BOD}_5$ , sulfide, total chromium and color up to 69, 72, 88, 5, 100% and *Thiobacillus ferrooxidans* alone

showed maximum reduction to an extent of 77, 80, 85, 52, 89 respectively in 21 d treatment at pH 2.5,  $\text{FeSO}_4$ -16 g  $\text{L}^{-1}$  and temperature of 30°C. The combined treatment at batch conditions involving 30 min chemical treatment by Fenton's oxidation followed by 72 h biochemical treatment by *Thiobacillus ferrooxidans* at batch conditions gave rise up to 93, 98, 72, 62 and 100% removal efficiencies of COD, BOD, sulfide, chromium and color at pH of 2.5 and 30°C. They observed a decrease in photo absorption of the Fenton's reagent treated samples, as compared to the blanks, at 280, 350 and 470 nm wave lengths. This may be the key factor for stimulating the biodegradation by *Thiobacillus ferrooxidans*.

Seawater-induced flocculation of alkaline tannery wastewater can increase the removal efficiency of organic compounds, such as particulate, colloids, colored compound and other dissolved organic compounds. Flocculation through the use of seawater was used as the primary treatment in the onsite tannery wastewater treatment plant. Evaluation of the potential biological treatment was performed by the activated sludge system of suspended micro-organisms using seawater flocculated tannery wastewater. The pH adjustment of the influent wastewater and  $\text{PO}_4$ -P addition after seawater flocculation were the most important operational parameters to enhance the removal efficiency of COD in the activated sludge process. Removal efficiency of COD increases with increase in sludge retention time (SRT). With the pH adjustment and  $\text{PO}_4$ -P addition after seawater flocculation, 75% of COD was removed at the SRT of 15 days. Experimental results demonstrated that seawater flocculation was more effective than the comparable ferric salt flocculation in enhancing the biological treatment during the 110 days of operation (Ryu *et al.*, 2007).

The ozonation of biologically pre-treated tannery wastewater and the influence of the applied specific ozone consumption onto a subsequent biological treatment were investigated by Jochimsen *et al.* (1997). From their study it was found that the partial oxidation of COD is favorable for subsequent biodegradation, whereas further mineralization reduces the effectivity of biological oxidation. The optimal range of subsequent biological treatment was observed at a specific ozone consumption of 1 to 3 g  $\text{O}_3$ /g  $\text{DOC}_0$ . As far as the distribution of molecular weight fractions are concerned, the ozonation leads to a relative increase of the low molecular weight DOC-fraction (<1,000 u), which includes the majority of the residual UV-absorbance at 254 nm.

Biological degradation, carried out in a sequencing batch biofilm reactor, with chemical oxidation, performed by ozone is an innovative tannery wastewater process. Furthermore, it is proved that the combined process is characterized by a very low sludge production. The combined treatment at the laboratory scale with and without ozonation were compared resulting to be very satisfactory only in the latter instance where recorded COD,  $\text{NH}_4$ -N and TSS average removals were 97, 98 and 99.9%, respectively. In fact, the measured specific sludge production resulted unexpectedly much lower than the value reported for conventional biological systems (Di Iaconi *et al.*, 2002).

High sulfide concentration present in treated wastewater may render aerobic biological treatment unsuitable. Hence, it became essential to include sulfide removal unit operation preceding aerobic biological unit. Among the techniques available oxidation of sulfide by air using activated carbon as catalyst gained importance for its removal of COD, BOD and TOC in addition to elimination of sulfide in wastewater. Anaerobic treatment of tannery wastewater in high rate close type reactors leaves sulfide in the range 31-795 mg  $\text{L}^{-1}$ , COD 395-1886 mg  $\text{L}^{-1}$ , BOD 65-450 and TOC 65-605 mg  $\text{L}^{-1}$ . Thus post anaerobic treatment of wastewater was required to meet discharging standard. The effect of  $[\text{S}^{2-}/\text{O}_2]$  ratio and hydraulic loading rate on removal of sulfide and organics in counter current reactor containing activated carbon were found. The percentage

removal of COD, BOD, TOC and Sulfide from anaerobically treated wastewater were 81, 85, 82 and 100%, respectively. The sulphate content of catalytically oxidized effluent was increased by 24% at  $[S^{2-}/O_2]$  ratio of 0.3353 and dissolved solids content was increased by 36% (Sekaran *et al.*, 1996).

Coagulation considerably reduced the concentration of sulfide and improved the anaerobic treatability lead to a reduction in waste disposal costs for tannery industries. Both aluminium sulphate and ferric chloride coagulants provided excellent sulfide removal (>71%), even at a low dose of 50 mg L<sup>-1</sup>. Coagulation at pH 7.5, removed at least 32% of COD, 64% of SS, 77% of chromium, 80% of sulfide and 85% of colour. Incorporation of coagulation prior to digestion resulted in an increased capacity of the digesters and improved digestion performance. An anaerobic digestion was carried out on initial samples and supernatants from the coagulation at a hydraulic retention time of 10 days with a loading rate of 0.33 kg COD/m<sup>3</sup>/day. A methane yield of 0.2 l CH<sub>4</sub>/(g COD removed) was achieved, while COD removal was 77% and COD removal rate was 0.24 kg COD/m<sup>3</sup>/day. The combined system provided a residual COD of less than 760 mg L<sup>-1</sup> and a residual sulfide of less than 200 mg L<sup>-1</sup>. The results also demonstrated that a sulfide concentration in excess of 260 mg L<sup>-1</sup> completely inhibited methane production (Song *et al.*, 2004).

**Chromium removal:** The tannery wastewater with increasing chromium concentrations, caused by poor wastewater management with an average value in the influent was around 2.673±0.32 to 3.268±0.73 mg L<sup>-1</sup> Cr. Investigations are focused on identification of the factors affecting the process performance (Banas *et al.*, 1999).

Chromium in tannery sludge causes serious environmental problems and is toxic to organisms and it was efficiently leached by the acidophilic sulfur-oxidizing *Acidithiobacillus thiooxidans*. The results showed that the pH of sludge mixture inoculated with the indigenous *A. thiooxidans* decreased to around 2 after 4 days. After 6 days incubation in shaking flasks at 30°C and 160 rpm, up to 99% of chromium was solubilized from tannery sludge. When treated in a 2 L bubble column bioreactor for 5 days at 30°C and aeration of 0.5 vvm, 99.7% of chromium was leached from tannery sludge (Wang *et al.*, 2007).

The naturally occurring microbes have enough potential to mitigate the excessive contamination of their surroundings and can be used to reduce the metal concentrations in aqueous solutions in a specific time frame. Microbes are isolated, keeping the natural selection in the view, from the tannery effluent since microbes present in the effluent exposed to the various types of stresses and metal stress is one of them. Investigations include the exposure of higher concentrations of Cr(VI) 1.0 to 4.0 mg l<sup>-1</sup> to the bacteria predominant on the agar plate. The short termed study (72 h) of biosorption showed significant reduction of metal in the media especially in the higher concentrations with a value from 1.0±0.02, 2.0±0.01, 3.0±0 and 4.0±0.09 at zero h to 0.873±0.55, 1.840±1.31, 2.780±0.03 and 3.502±0.68 at 72 h, respectively (Srivastava *et al.*, 2008).

A Gram-positive, chromium (Cr) resistant bacterial strain from effluent of tanneries, grown in media containing potassium dichromate concentration up to 80 mg mL<sup>-1</sup> has the reducing capability Cr (VI). The influencing factors are pH of the medium, concentration of Cr and the amount of the inoculum. A study was conducted to determine the ability of the bacterium to reduce Cr (VI) in the medium before and after introduction of bacterial culture under various conditions containing dichromate 20 mg mL<sup>-1</sup> more than 87% reduction of dichromate ions was achieved within 72 h (Shakoori *et al.*, 2000).

Bacterial strains *Acinetobacter* sp. grown in the presence of minimal salt medium and pentachlorophenol (PCP) as sole carbon source showed higher utilization of PCP and adsorption

of chromium. In sequential bioreactor, tannery effluents treated initially by bacterial consortium followed by fungus removed 90 and 67% chromium and PCP, respectively, whereas in another set of bioreactor in which effluents was treated initially by fungi followed by bacteria removes 64.7 and 58% of chromium and PCP, respectively (Srivastava *et al.*, 2007).

Algae namely, *Spirogyra condensata* and *Rhizoclonium hieroglyphicum* were employed to remove chromium from tannery effluent. The effect of pH and chromium concentration show that *S. condensata* exhibit maximum uptake of about 14 mg Cr(III)/g of algae at optimum pH of 5 whereas *R. hieroglyphicum* had 11.81 mg of Cr(III)/g of algae at pH of 4. Increase of initial concentration of Cr resulted to a decrease in adsorption efficiency. Dilute sulphuric acid (0.1 M) shows good desorption efficiency (>75%). Interference from cations negatively impacted on biosorption of chromium. Immobilized algae on Amberlite XAD-8 in a glass column, gives better recovery of chromium in tannery effluent compared to a batch method with unimmobilized algae. Fourier transform infra red (FT-IR) analysis of the two algae revealed the presence of carboxyl groups as possible binding sites (Onyanha *et al.*, 2008).

Srivastava and Thakur (2006) evaluated the potential of *Aspergillus* sp. for the removal of chromium in shake flask culture at different pH, temperature, inoculum size, carbon and nitrogen source. The maximum chromium was removed at pH-6; temperature-30°C, sodium acetate- 0.2% and yeast extract - 0.1%. *Aspergillus* sp. was applied in 2 L bioreactor for removal of chromium and it was observed that 70% chromium was removed after 3 days.

The tannery effluent carrying hazardous Cr (VI) species due to the oxidation of Cr (III) species is found to pollute the soil and the ground water. Biosorption of the Cr (VI) onto the cell surface of *Trichoderma* fungal species in aerobic condition. Batch experiments were conducted with various initial concentrations of chromium ions to obtain the sorption capacity and isotherms. The results obtained at pH 5.5 of chromium solution were 97.39% reduction by non pathogenic species of *Trichoderma*. It was found that the sorption isotherms of fungi for Cr (VI) appeared to fit Freundlich models. The fungal surfaces showed efficient biosorption for Chromium in Cr<sup>+6</sup>oxidation state. Best results for sorption were obtained at 5.5-5.8 pH, at low or high pH values, Cr (VI) uptake was significantly reduced.

Biological wastes (sawdust, rice husk, coir pith and charcoal) and a naturally occurring mineral (vermiculite) were successfully used to reduce the Cr concentrations in tannery effluent. Batch and column experiments were performed and the adsorption capacities of the substrates were also evaluated using isotherm tests and computing distribution co-efficient. The sawdust exhibited a higher adsorption capacity ( $k = 1482 \text{ mg kg}^{-1}$ ), followed by coir pith ( $k = 159 \text{ mg kg}^{-1}$ ). The biosorbent and mineral vermiculite in columns were found very effective in removing Cr from tannery effluent. About 94% removal of Cr was achieved by a column of coir pith and equally (93%) by a column containing a mixture of coirpith and vermiculite (Sumathi *et al.*, 2005).

## CONCLUSIONS

This review article examines the extent of pollution created by tanneries and the different biological processes available for the treatment and disposal of tannery wastewater. The advanced methods like membrane filtration, oxidation by ozone are being field trials. Biological treatment methods appear to be a better choice for the removal of color and organic content; however, some of the questions are yet to be answered on its process efficiency. This is because of the lack of information on various aspects such as desirable influent COD, optimal level of volatile fatty acids (VFA) concentration in the reactor, the reliable estimates of the bio kinetic constants and their

dependence on the substrate levels. In the field of wastewater treatment, it is generally accepted that anaerobic treatment is less energy-intensive and, hence, preferable to aerobic treatment. This review shows that the anaerobic treatment facility is superior in most respects for the treatment of tannery wastewater. The application of combined process of physical or chemical with biological process to treat tannery wastewater would give satisfactory results compared to individual treatment processes

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