

Evaluation and Characterization of Tannery Waste Water in Each Process at Batu and Modjo Tannery, Ethiopia



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Abstract

The leather industry is suffering from the negative impact generated by the pollution it causes to the environment. Nearly 70% of the pollution loads of BOD, COD, and Total Dissolved Solids (TDS) are generated from soaking, liming, deliming, pickling and tanning and retanning processes. There is an enormous pressure from the various pollution control bodies to regulate and minimize the amount of pollution generated from the leather processing. The need for use of alternative to chemical methods to combat pollution problem have become necessary to protect the industry and to comply with the environmental norms. In the present study, effluent samples were collected from Batu and Modjoa tannery in Ethiopia. The effluent samples were collected from all stages of processing viz., soaking, liming, deliming, pickling, Chrome tanning and Retaining. The physicochemical parameters of the tannery effluent viz. pH, alkalinity, acidity, biochemical oxygen demand (BOD₅), chemical oxygen demand (COD), total solids (TS), total dissolved solids (TDS), suspended solids (SS), chlorides and sulfides were determined. All the parameters included in this study are found to be higher than the prescribed discharge limits for tannery industries. The investigation of the tannery wastewater from different tanning processes gave a number of conclusions. The results indicate that the wastewaters from the tanneries do not satisfy the legal ranges of selected parameters discharge to inland water and to sewer.

Keywords: Alkalinity; Acidity; COD; BOD; Tannery Waste Water; Sulfides; Chlorides

Abbreviations: COD: Chemical Oxygen Demand; TS: Total Solids; TDS: Total Dissolved Solids; SS: Suspended Solids; BOD: Biochemical Oxygen Demand

Introduction

The tanning process aims to transform skins in stable and imputrescible products namely leather. There are four major groups of sub-processes required to make finished leather: beam house operation, tanyard processes, retanning and finishing [1-3]. However for each end product, the tanning process is different and the kind and amount of waste produced may vary in a wide range [2,4]. Traditionally most of tannery industries process all kind of leathers, thus starting from dehairing to retanning processes. However, in some cases only pre-pickled leather is processed with a retanning process. Acids, alkalis, chromium salts, tannins, solvents, sulfides, dyes, auxiliaries, and many others compounds which are used in the transformation of raw or semi-pickled skins into commercial goods, are not completely fixed by skins and remain in the effluent. For instance, the present commercial chrome tanning method gives rise to only about 50-70% chromium uptake [5]. During retanning procedures, synthetic tannins (Syntan), oilsand resins are added to form softer leather at varying doses [6]. One of

the refractory groups of chemicals in tannery effluents derives mainly from tannins [7]. Syntans are characterized by complex chemical structures, because they are composed of an extended set of chemical such as phenol, naphthalene formaldehyde and melamine-based Syntans, and acrylic resins [8-10].

Among Syntans, the ones based on sulfonatednaphthalene's and their formaldehyde condensates play a primary role, for volumes and quantity used in leather tanning industry. The oils cover the greater COD equivalents compared to the resins and syntans. The BOD₅/COD ratio of syntans was also lower than other compounds. A brief description about the wastes generated from a tannery and their impact on the environment would be appropriate to understand the problem associated with it. The beam house operations soaking, liming and deliming lead to discharge of high amount of sulfides, lime, and ammonium m salts, chlorides, sulphate, and protein in the effluent. Consequently, the wastewater is characterized with high amount of BOD and COD. Soak liquor contains, suspended

solids, dirt, dung, blood adhering to hides and skins, and chloride etc. lime liquors are highly alkaline. This stream contains suspended solids, dissolved lime, sodium sulfide, high ammonia cal nitrogen and organic matter. Unhearing and fleshing effluent contains fatty fleshing matter in suspension. The spent deliming liquors carry significant BOD load. The spent bate liquors on account of presence of soluble skin proteins and ammonium salts containing high organic matter. Pickle liquors are acidic and contain high amount of salt.

The spent chrome liquors contain high concentration of chrome compounds and neutral salts. The wastewater from neutralization, retanning, dyeing and fat liquoring sections contribute little pollution load [11]. Solvents and this leads to the emission of volatile organic compounds (VOC) [12]. An average of 30-35m³ of wastewater is produced per ton of raw hide. However, wastewater production varies in wide range (10-100 m³ per ton hide) depending on the raw material, the finishing product and the production processes [2]. Organic pollutants (proteic and lipidic components) are originated from skins (it is calculated that the raw skin has 30% loss of organic material during the working cycle) or they are introduced during processes. The objectives of this study to evaluate the physico-chemical properties of polluted water discharged from tannery, viz., pH, chloride, sulfide BOD₅, COD, alkalinity, T.S.S, TDS and evaluate of tannery wastewater in the different tanning processes viz. soaking, liming and unhairing, deliming and bating, pickling, tanning and retanning processes [13,14].

Materials and Methods

Materials

For the present study effluent samples were collected from tanneries in Batu and Modjo, Ethiopia. The effluent samples were collected from all stages of tanning processing viz., soaking, liming, deliming, pickling, Chrome tanning and Retanning. The effluent was collected in polythene containers of two litres capacity and were brought to the laboratory with due care and was stored at 4°C for further analysis. Chemicals used for the analysis of spent liquor were analytical grade reagents. The physical and chemical characteristics of tannery effluents parameters viz. pH, total alkalinity, COD, BOD₅, total solids (TS), total dissolved (TDS); total suspended solids (TSS), chlorides, sulfide sand chromium were analyzed as per standard procedures [15].

Methods

Determination of pH: The pH is determined by measurement of the electro motive force (emf) of a cell comprising of an indicator electrode (an electrode responsive to hydrogen ions such as glass electrode) immersed in the test solution and a reference electrode (usually a calomel electrode). Contact is achieved by means of a liquid junction, which forms a part of the reference electrode. The emf of this cell is measured with pH meter.

Determination of total alkalinity: The alkalinity of sample can be determined by titrating the sample with sulphuric acid or hydrochloric acid of known value of pH, volume and concentrations. Based on stoichiometry of the reaction and number of moles of sulphuric acid or hydrochloric acid needed to reach the end point, the concentration of alkalinity in sample is calculated. A known volume of the sample (50 ml) is taken in a beaker and a pH probe was immersed in the sample. HCl or H₂SO₄ acid (0.1NHCl in 1000 ml distilled water) added drop by drop until the pH of the sample reached 3.7. The volume of the acid added was noted [15].

Calculation: Alkalinity as mg/l of CaCO₃ = (50000 × N of HCl × ml acid titrated value) / volume of sample taken.

Determination of chemical oxygen demand (COD): The chemical oxygen demand of an effluent means the quantity of oxygen, in milligram, required to oxidize or stabilize the oxidizable chemicals present in one litre of effluent under specific condition. 2.5 ml of the sample was taken in tube, 1.5 ml of 0.25 NK₂Cr₂O₇ (potassium dichromate), spatula of mercuric sulphate HgSO₄ and 3.5 ml of COD acid were added and kept in COD reactor for 2hrs at 150°C. After cooling the sample titrated against FAS (standard ferrous ammonium sulfate 0.1N) and used ferrion as indicator. The end point is reddish brown color. In the blank tube 2.5 ml of distilled water was taken and then follow the same procedure in the sample [15].

Calculation: COD (mg/l) = (blank value-titrated value) × N of FAS × 8000 / volume of sample

8000 mill equivalent wt of O₂ × 1000ml

Determination of biochemical oxygen demand (BOD): Biochemical oxygen demand (BOD) of an effluent is the milligram of oxygen required to biologically stabilize one litre of that effluent (by bio-degradation of organic compounds with the help of micro-organisms) in 5 days at 4°C. If the BOD value of an effluent is high, is high, then that effluent contains too much of bio-degradable organic compounds and so will pollute the receiving water highly.

A. Procedure

a) Take 5 litres of distilled water, aerated for 3.5 hours, added nutrients 1 ml nutrient for 1 litre aerated distilled water (FeCl, CaCl₂, PO₄, MgSO₄, domestic water), aeration for 30 minutes.

b) BOD bottle (300 ml), add sample, fill the bottle with aerated water, put the lid (avoid air bubbles), keeping BOD incubator at 20°C for 5 days, after 5 days take the bottle and add 2 ml MnSO₄, 2 ml alkali azide iodide and 2 ml conc. H₂SO₄. Shake the bottle well (yellow colour) take 200 ml sample add starch solution as indicator (purple colour) titrated with 0.025 N sodium thiosulphate end point colour change from purple to colorless. In blank filled the bottle with aerated water without the sample and follow the procedure [15].

c) Calculation

$BOD_5 = (\text{blank value} - \text{titrated value}) \times 300 / \text{volume of sample}$

Determination of Total solid: The term solid refers to the matter either filtrable or non-filtrable that remains as residue upon evaporation and subsequent drying at a defined temperature. Residue left after the evaporation and subsequent drying in oven at specific temperature 103-105°C of a known volume of sample are total solids. Total solids include Total suspended solids (TSS) and Total dissolved solids (TDS).

A. Procedure

Dry weight of empty dish or crucible (initial weight), add 50 ml sample, keep it in water bath until dry, keep it in oven (103 to 105°C) for at least 1 hour, desiccators, and take final weight of dish [15].

a) Calculation

$\text{Total solid (mg/l)} = (\text{final weight} - \text{initial weight}) \times 1000 \times 1000 / \text{volume of sample}$

Determination of total dissolved solid

A. Procedure

Dry weight of empty dish or crucible (initial weight) take sample and filter with What man No.1, add 50 ml filtrate sample, keep it in water bath until dry, keep it in oven (103 to 105°C) for at least 1 hour, desiccators, take final weight of dish [15].

a) Calculation

$\text{Total dissolved solid (mg/l)} = (\text{final weight} - \text{initial weight}) \times 1000 \times 1000 / \text{volume of sample}$

Determination of total suspended solid: The difference between the total solids and total dissolved solids are suspended solids.

$$TSS = TS - TDS$$

Determination of chloride: Chloride is determined in a natural or slightly alkaline solution by titration with standard silver nitrate, using potassium chromate as an indicator. Silver chloride is quantitatively precipitated before red silver chromate is formed.

A. Procedure: Take sample (10 ml to 50 ml), add 2 ml of hydrogen peroxide (H₂O₂), add 2 ml K₂CrO₄ (potassium chromate indicator), titrate with silver nitrate (0.0141 N), end point formation of reddish yellow colour (yellow to orange). In blank trial take distilled water instead of sample and follow the same procedure above [15].

a) Calculation

$\text{Chloride (mg/l)} = (A - B) \times N. \text{ of silver nitrate} \times 35.45 \times 1000 / \text{volume of sample}$

A = ml titration for sample

B = ml titration for blank

N = normality of AgNO₃

Determination of sulfide: The sulfides in the solution are oxidized with an excess of a standard iodine solution and the excess back titrated with a standard thiosulfate solution.

A. Procedure: Take sample (10ml) in conical flask, add 5 ml zinc acetate (5%), filter through filter paper, take the filter paper and put it in the same conical flask, add 100 ml distilled water. then add 20 ml, iodine solution and 4 ml 6N HCl, add 2 drops of starch as indicator (purple colour will form), titrate against sodium thiosulphate (0.025N), end point the colour change from blue colour to colorless. In the blank test take 100 ml distilled water instead of sample and follow the same procedure above for the sample [15].

a) Calculation:

$\text{Sulfide (mg/l)} = (BV - TV) \times N. \text{ thiox}400 / \text{Volume of sample} \times N. \text{ Ioden}$

BV= blank value

TV= titrated value

Results and Discussion

Characteristics of tannery waste water

Wastewater of each tannery process consists of pollution of varying pH values. Similarly, a large variation exists in every parameter BOD, COD, Chloride, Sulphate, etc. Discharge of these chemicals into wastewater is hazardous for the environment. Analysis of physical and chemical characteristics of the tannery wastewater collected from different tanning processes viz. soaking, liming and unhairing, deliming and bating pickling, chrome tanning and retaining are listed in (Tables 1 & 2) respectively.

Determination of pH

The pH values of both tanneries are in the range 3.25-12.64. Which was very higher value compare to limit set by EPA (6.0-9)? The extreme pH of wastewater is generally not acceptable, as lower pH cause problems to survival of aquatic life. It also interferes with the optimum operation of wastewater treatment facilities. Water with high or low pH is not suitable for irrigation. At low pH most of the metals become soluble in water and therefore could be hazardous in the environment. At high pH most of the metals become insoluble and accumulate in the sludge and sediments. The toxicity of heavy metals also gets enhanced at particular pH [6].

Determination of Biochemical Oxygen

a) Demand (BOD): BOD is measure of the content of organic substances in the waste water which are biologically

degradable with consumption of oxygen. Usually indicated as 5-day Biochemical oxygen demand (BOD). This is the amount of oxygen in milligrams per litre (O₂) (mg/l) that consumed by microorganisms in 5 days at 20°C for oxidation of the biologically degradable substances contained in the water. The results of present study revealed that BOD level from different tanning processes viz. soaking, liming and unhairing, delimiting and bating pickling, chrome tanning and retanning is given in (Figures 1-4) indicating high organic load surpassed legal limit set by EPA (200 mg/l). The presence of organic matter will promote anaerobic action leading to the accumulation of toxic compounds in the water bodies.

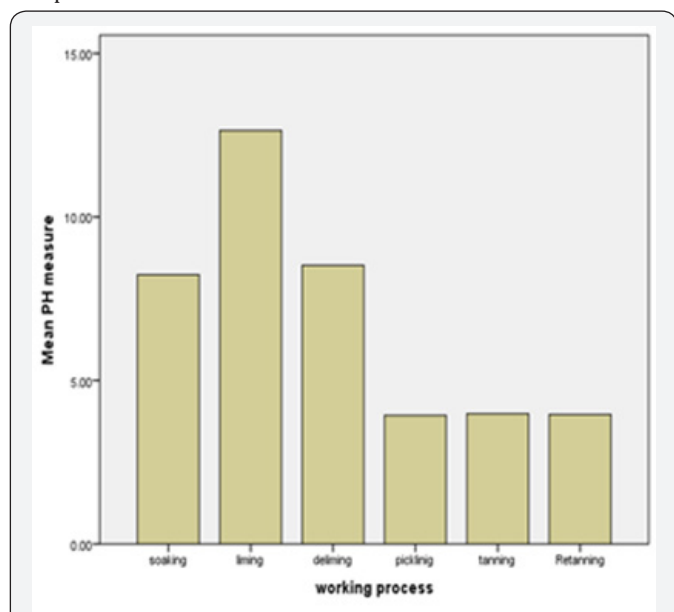


Figure 1: pH of different tanning processes of Modjo Tannery.

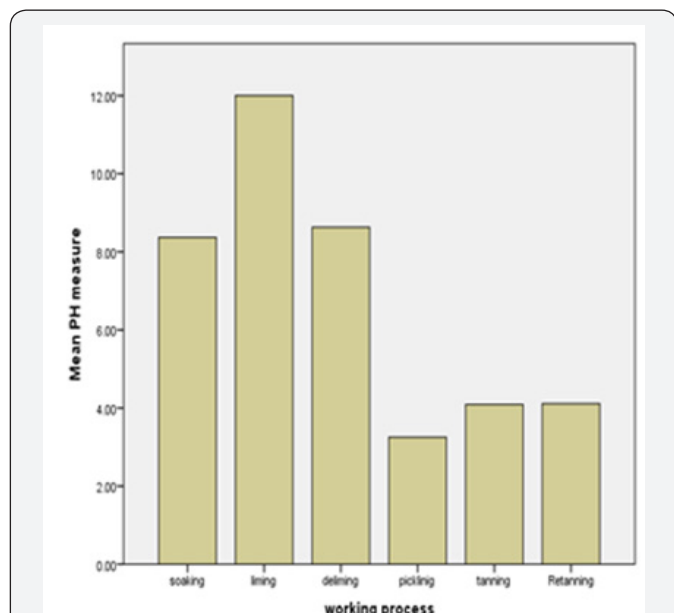


Figure 2: pH of different tanning processes of Batu tannery.

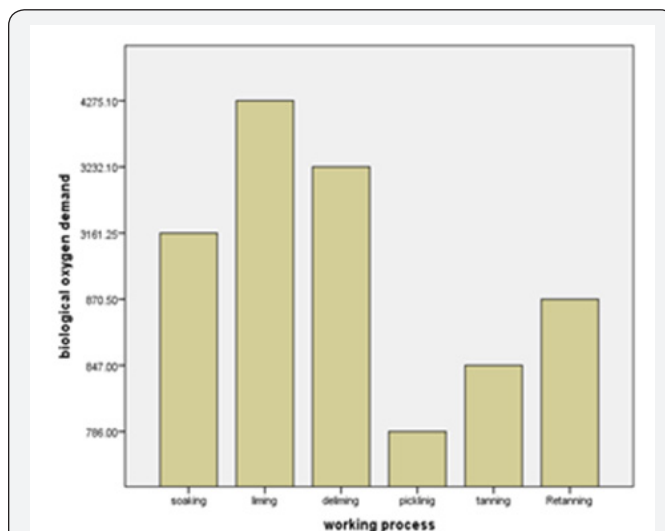


Figure 3: Biological oxygen demand of different processes Batu tannery.

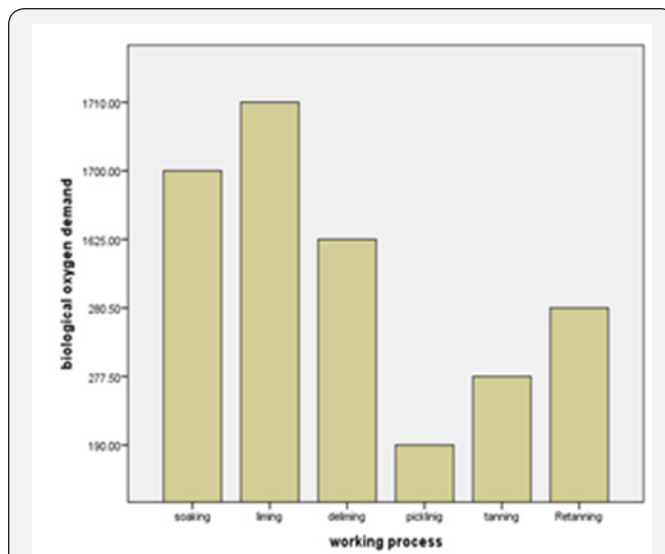


Figure 4: Biological oxygen demand of different processes of Modjo tannery.

Determination of Chemical Oxygen Demand (COD)

Chemical oxygen demand (COD) is quantity of oxygen expressed in milligram consumed by the oxidisable matter contained in one litre of the sample. The test is performed by vigorous oxidation with chemicals and back-titrating the chemical consumed for oxidation. COD is system of measuring the content of organic impurities with oxidizing agents. The results of present study revealed that COD level from different tanning processes viz. soaking, liming and unhairing, delimiting and bating pickling, chrome tanning and retanning is given in (Figures 5 & 6) exceeds the permissible COD level EPA (500mg/l). This indicates that the effluent is unsuitable for the existence of the aquatic organisms, due to the reduction in the dissolved content.

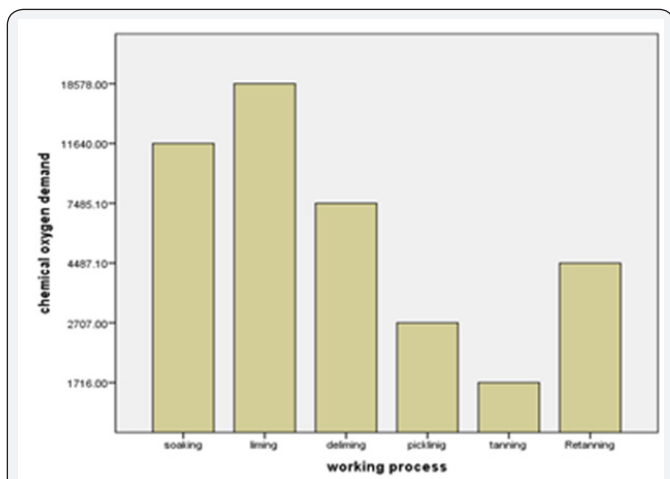


Figure 5: Chemical oxygen demand of different processes of Batu tannery.

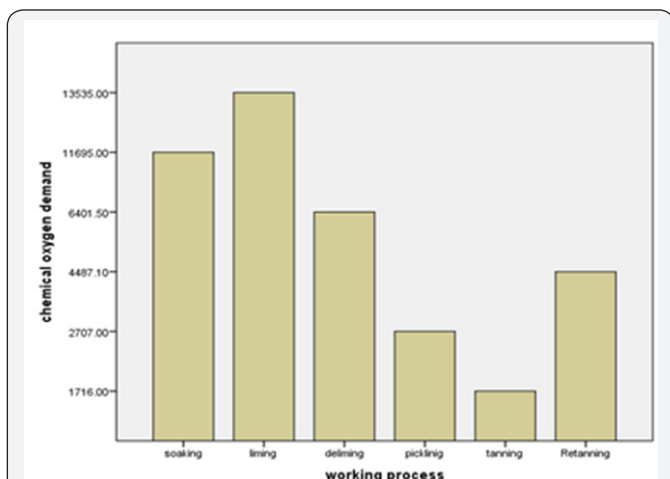


Figure 6: Chemical oxygen demand of different processes of modjo tannery.

Determination of Total Solids (TS)

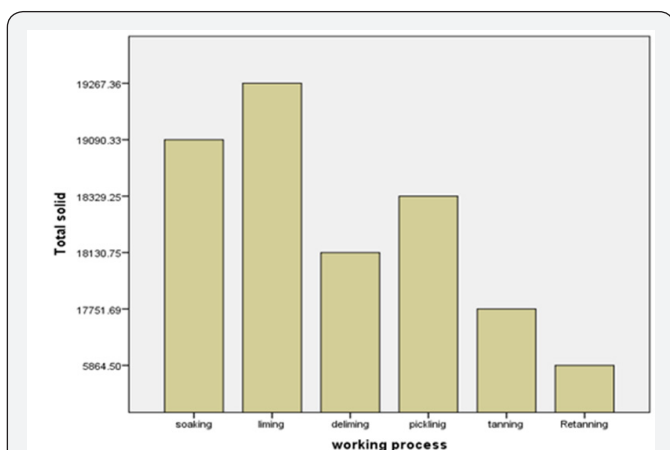


Figure 7: Graphical representation of total solid of different tanning processes of Batu Tannery.

The results of present study revealed that TS level from different tanning processes viz. soaking, liming and unhairing,

delimiting and bating pickling, chrome tanning and retanning is given in (Figures 7 & 8) exceeds the permissible TS level of 110 mg/L. These solid impurities cause turbidity in the receiving streams. The composition of solids present in tannery effluent mainly depends upon the nature and quality of hides and skins processed in the tannery.

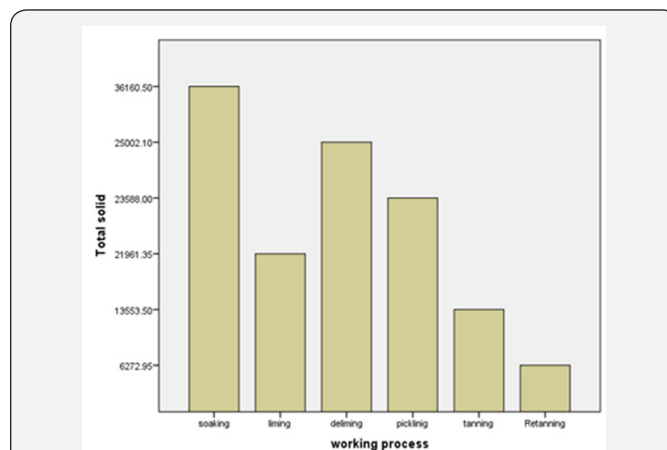


Figure 8: Graphical representation of total solid of different tanning processes of Modjo Tannery.

Determination of Total Suspended Solids (TSS)

The results of present study revealed that TSS level from different tanning processes viz. soaking, liming and unhairing, delimiting and bating pickling, chrome tanning and retanning is given in (Figure 6) and it exceed the permissible TSS level of (20-200) mg/ L. These suspended impurities cause turbidity in the receiving streams. The composition of solids present in tannery effluent mainly depends upon the nature and quality of hides and skins processed in the tannery. High level of total suspended solids present in the tannery effluent could be attributed to their accumulation during the processing of finished leather. Presence of total suspended solids in water leads to turbidity resulting in poor photosynthetic activity in the aquatic system [16-18] and clogging of gills and respiratory surfaces of fishes [19].

Determination of Chloride

The results of present study revealed that chloride level from soaking and pickling, are 19250 mg/ l, 23500 mg/l respectively (Table 2) and the levels exceed the permissible chloride level of 1000 mg/L of effluent discharge into inland surface waters. High levels of chlorides in the tannery effluent could be attributed to the soaking and pickling processes. The chloride content in water sample gives an idea of the salinity of water sample.

Determination of Sulfide

Sulfides are particularly objectionable because hydrogen sulfide will be liberated if they are exposed to a low pH environmental, and if they are discharged into stream containing iron, black precipitates will be formed. Sulfides may be toxic to stream organisms or to organisms employed in biological

treatment systems. The results of present study revealed that sulfide level from liming and unhairing process is given in Table 2 and it exceed the permissible sulfide level of 2 mg/ L. of effluent discharge into inland surface waters [13].

Determination of Total Alkalinity

Alkalinity of water is its acid neutralizing capacity. It is the sum of all the bases. The alkalinity of natural water is due to the salt of carbonates, bicarbonates, borates silicates and phosphates along with hydroxyl ions indeliming & bating process is given in (Tables 1 & 2). The Free State. However the major portion the alkalinity is due to hydroxides, carbonates and bicarbonates. The results of present study revealed that alkalinity level from soaking, liming and unhairing, and deliming process are given in (Figures 9-16).

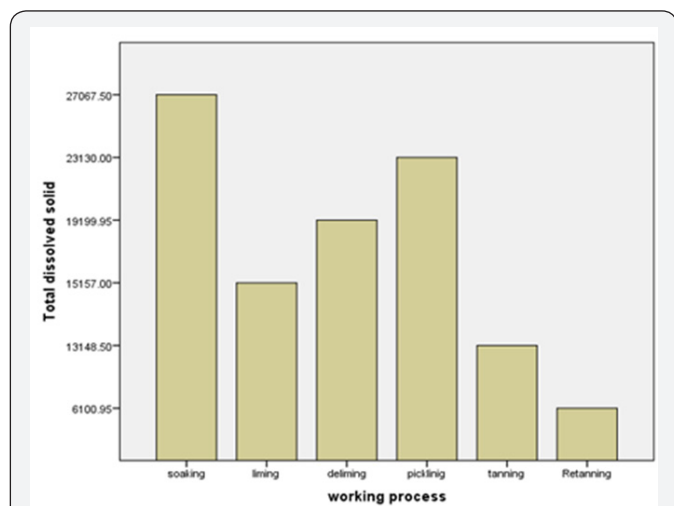


Figure 9: Graphical representation of total dissolved solid of different tanning processes of Batu Tannery.

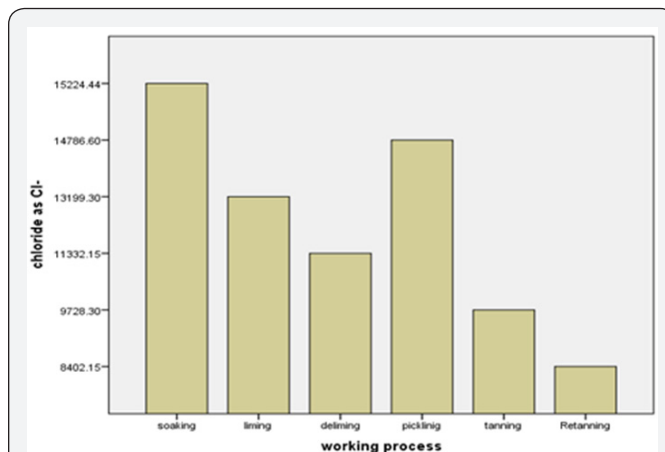


Figure 11: Graphical representation of Chloride of different tanning processes of Batu Tannery.

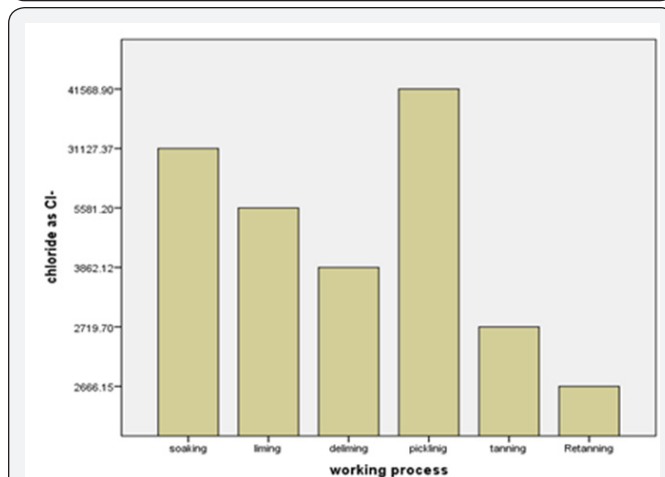


Figure 12: Graphical representation of Chloride of different tanning processes of Modjo Tannery.

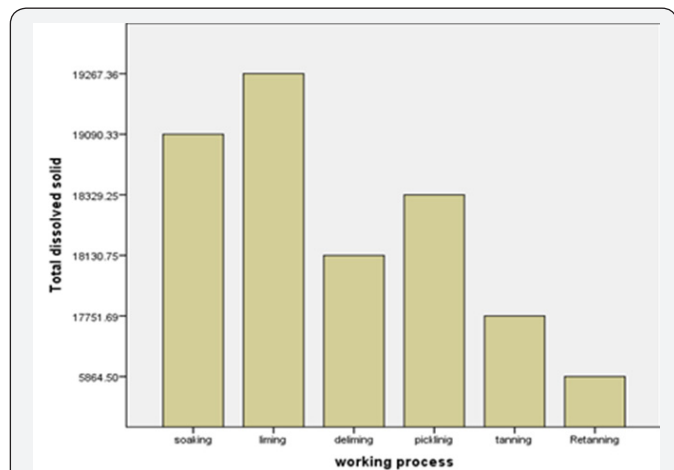


Figure 10: Graphical representation of total dissolved solid of different tanning processes of modjo Tannery.

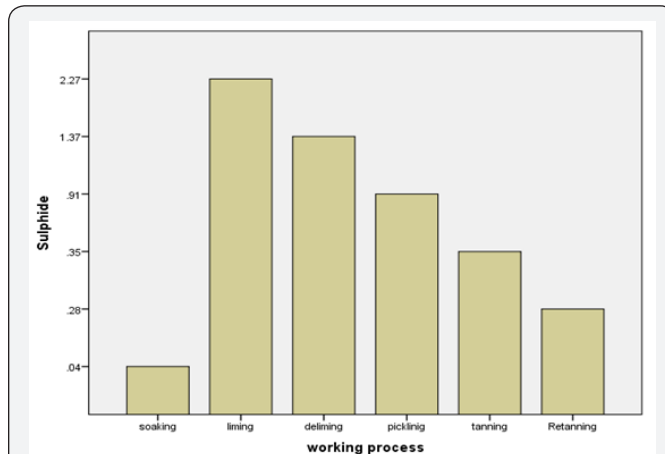


Figure 13: Graphical representation of Sulphide of different tanning processes of Batu Tannery.

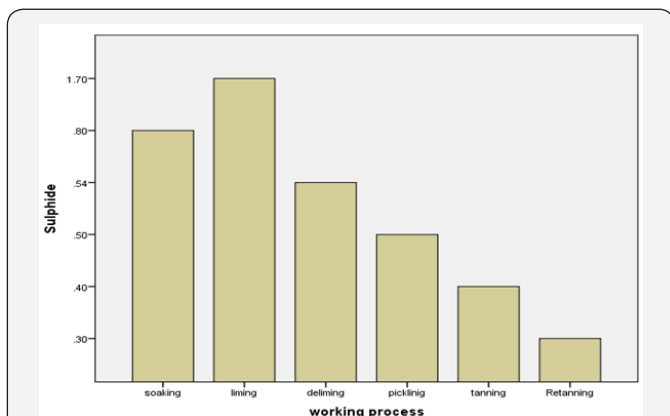


Figure 14: Graphical representation of Chloride of different tanning processes of Modjo Tannery.

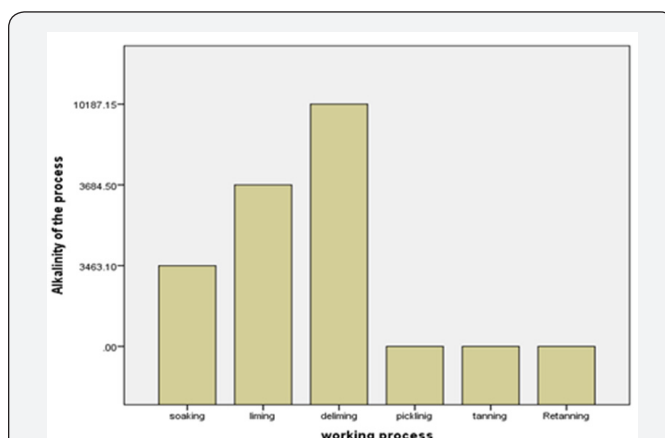


Figure 16: Graphical representation of Alkalinity of different tanning processes of Modjo Tannery.

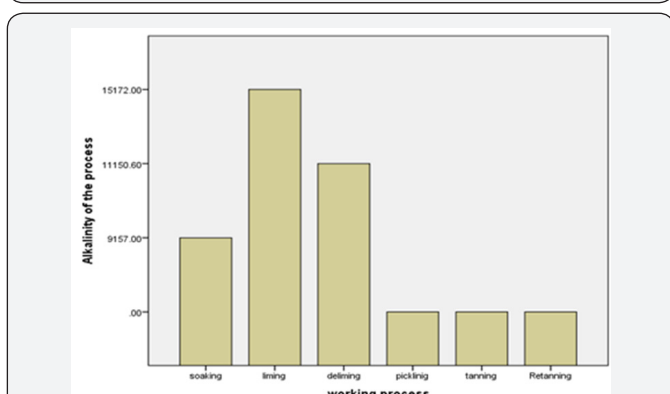


Figure 15: Graphical representation of Alkalinity of different tanning processes of Batu Tannery.

Determination of hexavalent chromium

Cr is one of the most important pollutants released from the tanning industries in the effluent. According to Saritha and Meikandaan (2013) chrome tanning processes originates toxic metals and regular treatment systems are not eligible for the elimination of it. The wastewater generated by tanneries is the major source of Chromium pollution. The chromium (Cr) is well-known to be toxic to living organisms due to their bioaccumulation and non-biodegradable properties. In Tables 1 & 2 Maximum Cr concentrations was show at both tannerierespectively.This indicates that the concentration is above permissible limit of EPA (0.1mg/l).

Table 1: Batu Tannery.

Parameter	Soaking	Liming	Deliming	Pickling	Tanning	Re-tanning
PH	8.37±0.988	12.00±0.707	8.63±0.989	3.25±0.212	4.09±0.141	4.11±0.127
Total alkalinity	9157±5503.5	15172±4282.2	11150.6±7140.8	-	-	-
BOD5	1700±141.42	1710±56.66	1625±66.47	190±36.77	277.5±88.39	280.5±77.075
COD	11640±1484.92	18578±1827.15	7485.1±1808.07	2707±687.34	1716±619.43	4487.1±1121.61
Total solid	36160.5±9772.95	21961.35±1695.4	25002.1±11543.58	23588±12215.97	13553.5±16899	6272.95±8345.2
Total dissolved solid	27067.5±9853.5	15157±1636.24	19199.95±11596.48	23130±12204.6	13148.5±16897.7	6100.95±8342.37
Suspended solid	9093±80.61	6804.35±59.18	5802.6±52.89	458±11.313	405±1.41	172±2.82
Chloride	31127.37±849.05	5581.2±72.14	3862.12±140.89	41568.9±1423.37	2719.7±364.202	2666.15±436.49
Sulphide	0.035±0.0014	2.267±0.583	1.365±0.275	0.905±0.487	0.35±0.125	0.280±0.226
Chromium	-	-	-	-	.006	1.22

Table 2: Modjo Tannery.

Parameter	Soaking	Liming	Deliming	Pickling	Tanning	Re-tanning
PH	8.23±0.68	12.64±0.00	8.52±1.01	3.93±0.38	3.96±0.212	3.98±0.113
Total alkalinity	3463.1±1220.61	10187.15±3392.18	3684.5±1321.61	-	-	-
BOD5	3161.25±147.4	4275.1±568.66	3232.1±626.64	786±124.451	870.5±149.2	847±88.4
COD	11695±1704.13	13535±2354.67	6401.5±1808.072	2707±510.34	1716±456.43	4487.1±8021.61
Total solid	19090.33 ±4679.8	19267.36±4787.2	18130.75±3896.6	18329.25±3987.13	17751.69±3662.77	5864.5±1461.03
Total dissolved solid	23171±4625.15	23380.4±4575.05	20971.8±3268.14	25508.5±5324.54	17963.2±3662.8	7288±1562.17
Suspended solid	7118.9±699.54	6987±572.6	5559±468.97	8670.5±1120.69	768.5±198.3	1926.5±322.76
Chloride	15224.44±102.16	13199.3±244.31	11332.15±183.78	14786.6±152.7	9728.3±70.64	8402.15±49.43
Sulphide	0.8±0.1	1.7±0.23	0.54±0.09	0.5±0.013	0.4±0.08	0.3±0.02
Chromium	-	-	-	-	1.46±0.556	4±1.03

All value except pH are stated Mg/l

Conclusion

The processing of hides and skins into leather is carried out in an aqueous medium and hence the discharged water from pits, drums or paddles containing several soluble and insoluble constitutes the effluents from the tannery. In the present study, investigation of the tannery wastewater from different tanning processes gave a number of conclusions. Results of the analysis showed that the tannery wastewater from different tanning processes viz., soaking, liming and unhairing, declaiming and bating, pickling, chrome tanning and retanning is highly With a disagreeable pH, alkalinity, acidity, total solids, total dissolved solids, suspended solid, chemical oxygen demand, biochemical oxygen demand, chlorides and sulfides. The results of the analysis indicate that the wastewaters from different units of the tannery do not satisfy the legal ranges of selected parameters.

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