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Evaluation and Characterization of Tannery Wastewater 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 pollutionit 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 leatherprocessing. 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, biochemicaloxygen demand (BOD5), chemical oxygen demand (COD), totalsolids (TS), total dissolved solids (TDS), suspended solids (SS), chlorides and sulfides were determined. All the parametersincluded 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, Tannerywastewater, Sulfides, Chlorides.

I. 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 [1,2,3]. Ho wever 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 commercialgoods, 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 retanningprocedures, 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 fromtannins [7]. Syntans are characterized by complex chemicalstructures, because they are composed of an extended set of chemical such as phenol, naphthaleneformaldehyde and melamine-based Syntans, and acrylic resins [8, 9, 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 [10]. 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 [9]. 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, andchloride etc.lime liquors are highly alkaline. This stream contains suspended solids, dissolved lime, sodium sulfide, high ammoniacal 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 fromneutralization, retanning, dyeing and fatliquoring sectionscontribute little pollution load [11]. Solvents and this leads to the emission of volatile organic compounds (VOC) [12]. An average of 30–35m^3 of wastewater is produced per ton of raw hide. However, wastewater production varies in wide range (10–100 m^3 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 thephysico-chemical properties of polluted water discharged from tannery, viz., pH, chloride, sulfide BOD5, COD, alka linity, 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.

II. 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 **4ºC** 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,sulfidesand 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 H_2SO_4 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 $CaCO_3$= (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 N$K_2Cr_2O_7$(potassium dichromate), spatula of mercuric sulphate $HgSO_4$ and 3.5 ml of CODacid were added and kept in COD reactor for 2hrs at 150ºC. 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 O_2×1000ml

Determination of biochemical oxygen demand (BOD)

Biochemical oxygen demand (BOD) of an effluent is the milligram of oxygen required to biologically stabilize one liter of that effluent (by bio-degradation of organic compounds with the help of micro-organisms) in 5 days at 4ºC.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.

Procedure

1. Take 5 litres of distilled water, aerated for 3.5 hours, added nutrients 1 ml nutrient for 1 litre aerated distilled water (FeCl, $CaCl_2$, PO_4, $MgSO_4$, domestic water), aeration for 30 minutes.

2. BOD bottle (300 ml), add sample, fill the bottle with aerated water, put the lid (avoid air bubbles), keeping BOD incubator at 20ºC for 5 days, after 5 days take the bottle and add 2 ml $MnSO_4$, 2 ml alkaliazide iodide and 2 ml conc. H_2SO_4. Shake the bottle well (yellow colour) take 200 ml sample add starch solution as indicator (purple colour) titrated with 0.025 N sodium thiosulphateend point colour change from purple to colorless. In blank filled the bottle with aerated water without the sample and follow the procedure [15].

Calculation

BOD5= (blankvaluetitratedvalue) ×300/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 o f a known volume of sample are total solids. Total solids include Total suspected solids (TSS) and Total dissolved solids (TDS).

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 105ºC) for at least 1 hour, desiccator, take final weight of dish [15].

Calculation

Total solid (mg/l) = (final weight-initial weight) ×1000×1000 / volume of sample

Determination of total dissolved solid

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 105ºC) for at least 1 hour, desiccator, take final weight of dish [15].

Calculation

Total dissolved solid (mg/l) = (final weight-initial weight) ×1000×1000 / 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.

Procedure

Take sample (10 ml to 50 ml), add 2 ml of hydrogen peroxide (H_2O_2), add 2 ml K_2CrO_4 (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].

Calculation

Chloride (mg/l) = (A-B)×N. of silver nitrate×35.45×1000/ 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 and excess of a standard iodine solution and the excess back titrated with a standard thiosulfate solution.

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].

Calculation

Sulfide (mg/l) =(BV-TV) x N. thiox400/
VolumeofsamplexN. ioden
BV= blank value
TV= titrated value

Results and discussion

Characteristics of tannery wastewater

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 andbating pickling, chrome tanning and retanning are listed in table 1 and 2 respectively.

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

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 demand (BOD)

BOD is measure of the content oforganic substances in thewaste water which are biologically degradable withconsumption of oxygen.

Usually indicated as 5-day Biochemical oxygen demand (BOD). This is the amount ofoxygen in milligrams per litre(O_2) (mg/l) that consumed bymicroorganisms in5 days at 20ºC for oxidation of thebiologically degradable substances contained in the water. The results of present study revealed that BOD level from different tanning processes viz. soaking, liming and unhairing, deliming and bating pickling, chrome tanning and retanning is given in Fig. 3and 4 indicating high organic load surpassed legal limitset 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.

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, deliming and bating pickling, chrome tanning and retanning is given in Fig. 5 and 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.

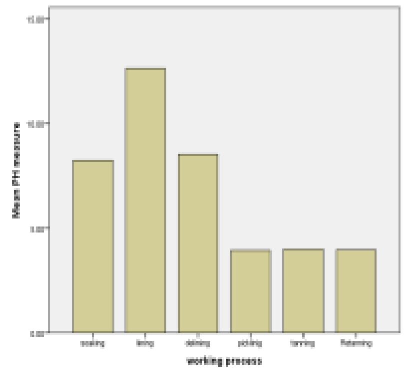

Fig.1: pH of different tanning processes of Modjo Tannery

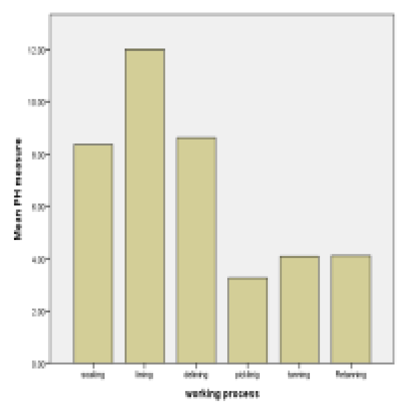

Fig.2: pH of different tanning processes of Batu tannery

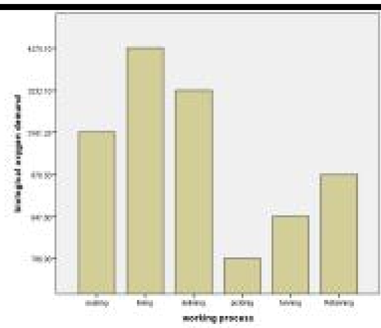

Fig.3:Biological oxygen demand of different processes Batu tannery

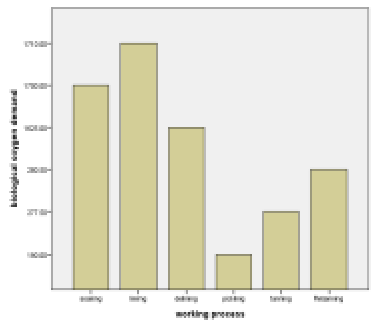

Fig.4: Biological oxygen demand of different processes of Modjo tannery

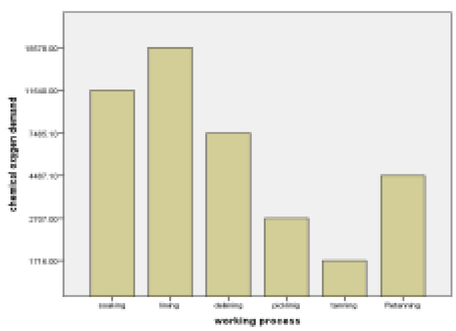

Fig.4: Chemical oxygen demand of different processes of Batu tannery

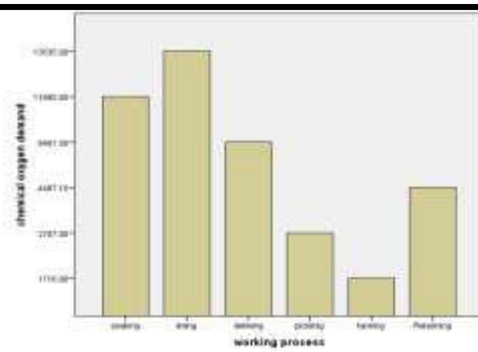

Fig.6: Chemical oxygen demand of different processes of modjo tannery

Determination of Total Solids (TS)

The results of present study revealed that TS level from different tanning processes viz. soaking, liming and unhairing, deliming and bating pickling, chrome tanning and retanning is given in Fig.7 and 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.

Determination of Total Suspended Solids (TSS)

The results of present study revealed that TSS level from different tanning processes viz.soaking, liming and unhairing, deliming and bating pickling, chrome tanning and retanning is given in Fig. 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 [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 3) 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 hydrogensulfide will beliberated if they are exposed to a low pHenvironmental, and if they aredischarged into stream containing iron, black precipitates will be formed. Sulfides may be toxic to stream organisms or to organisms employedin biological treatment systems. The results of present study revealed that sulfide level from liming and unhairing process is given in Table 3 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 Table 3.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 Table 1and 2.

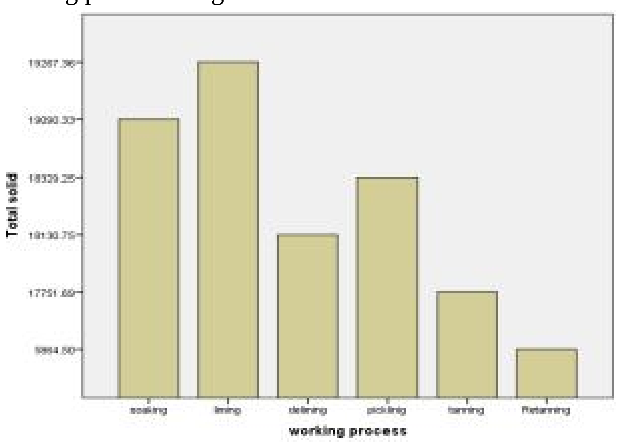

Fig.7: Graphical representation of total solid of different tanning processes of Batu Tannery

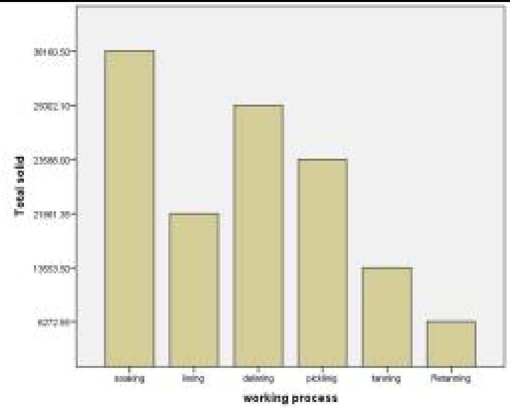

Fig.8: Graphical representation of total solid of different tanning processes of Modjo Tannery

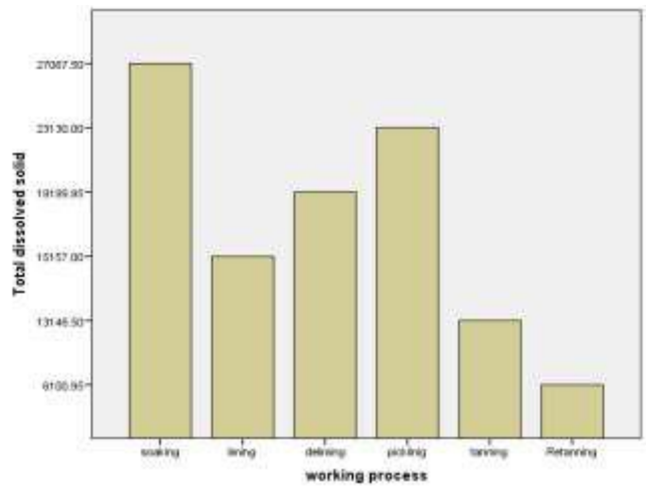

Fig.9: Graphical representation of total dissolved solid of different tanning processes of Batu Tannery

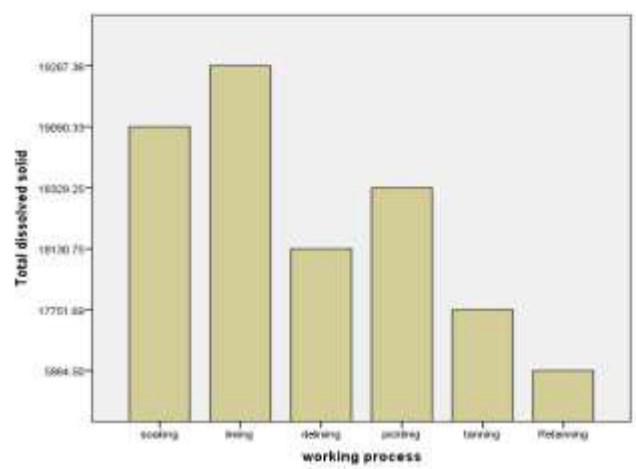

Fig.10: Graphical representation of total dissolved solid of different tanning processes of modjo Tannery

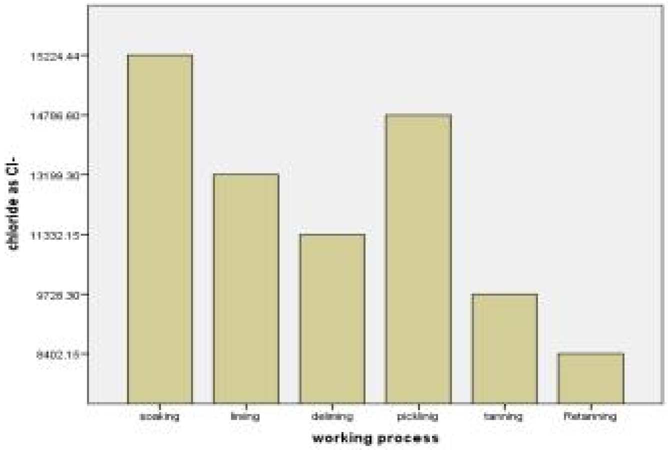

Fig.11: Graphical representation of Chloride of different tanning processes of Batu Tannery

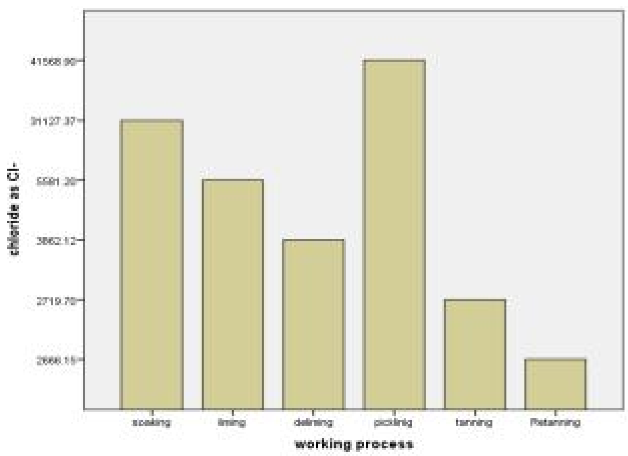

Fig.12: Graphical representation of Chloride of different tanning processes of Modjo Tannery

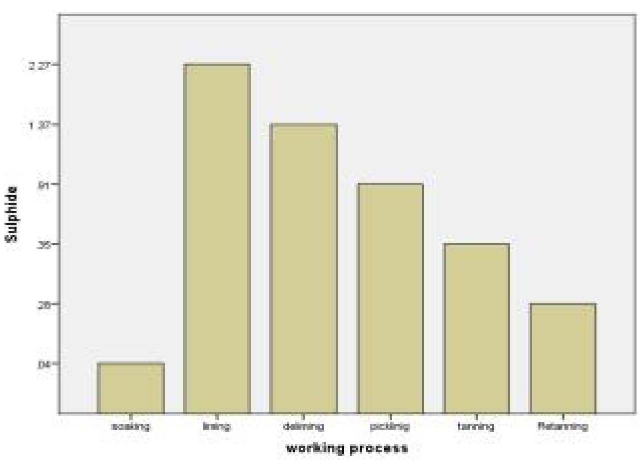

Fig.13: Graphical representation of Sulfide of different tanning processes of Batu Tannery

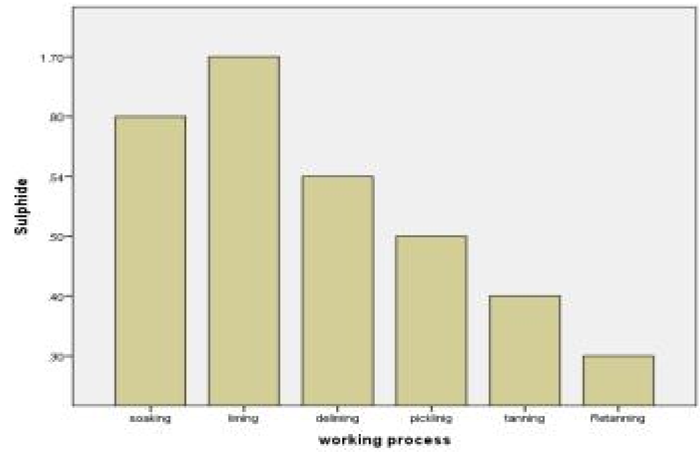

Fig.14: Graphical representation of Chloride of different tanning processes of Modjo Tannery

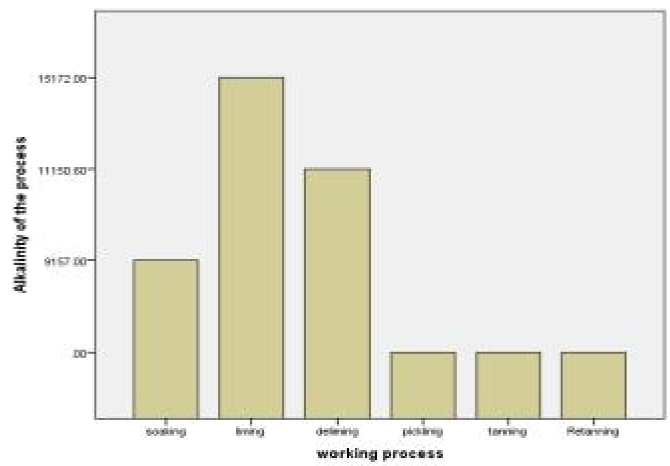

Fig.15: Graphical representation of Alkalinity of different tanning processes of Batu Tannery

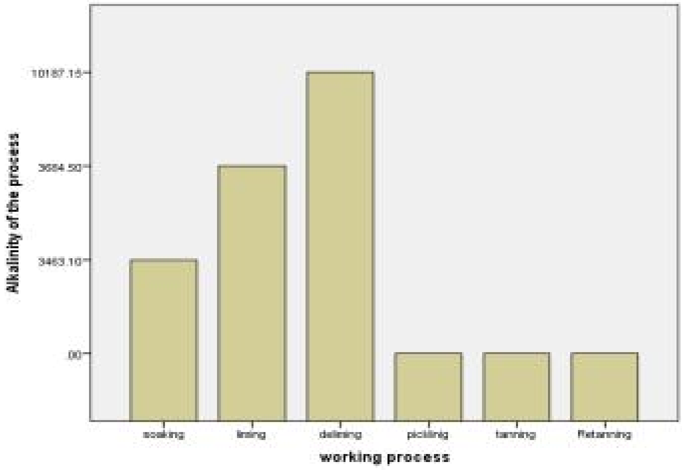

Fig.16: Graphical representation of Alkalinity of different tanning processes of Modjo Tannery

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 Table 1 and 2 Maximum Cr concentrations was show at both tanneriesrespectively.This indicates that the concentration is above permissible limit ofEPA (0.1mg/l).

Conclusions

The processing of hides and skins into leather is carried out in an aqueous medium m 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 highlyWith 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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