



TANNERY WASTE WATER TREATMENT AND CONTROL

REVIEW PAPER ON TANNERY WASTEWATER TREATMENT AND CONTROL.

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ABSTRACT

The Leather industry in Uganda is of economic importance in terms of providing employment opportunities and revenue. However, leather manufacturing is a water intensive process and generates a significant volume of wastewater mainly characterized by high salinity, high organic loading and pollutants. Therefore, the main objective of this study is to assess the existing wastewater management practices in the leather industry of Uganda focusing on two different tanneries so as to determine their efficiency and effectiveness in addressing potential pollution incidents.

The uncontrolled release of tannery effluents to natural water bodies increases health risks for human beings and environmental pollution. Effluents from raw hide processing tanneries, which produce wet blue, crust leather or finished leather contain compounds of trivalent chromium (Cr) and sulphides in most cases, organic and other ingredients are responsible for high BOD and COD values and represent an immense pollution load, causing technical problems, sophisticated technologies and high costs in concern with effluent treatment. chemicals such as inorganic salts and polymeric organic coagulants are used for primary coagulation, as coagulant aids and for sludge dewatering; lime Ca(OH)_2 and soda ash (Na_2CO_3) are necessary for pH correction and

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water stabilization; caustic soda (NaOH) is used for pH adjustment, powdered activated carbon (PAC) can remove taste and odour compounds and micro pollutants such as atrazine, bentonite aid's coagulation, and ammonium hydroxide is used in chloramination.

INTRODUCTION

This paper provides an overview of the leather processing industry including composition of tannery waste water, tannery operations, its wastewater management practices and associated challenges. The paper also reviews leather production in Uganda, wastewater management practices, the tanning process, characteristics of wastewater from the tanning process and associated emissions, treatment of tannery wastewater, and the environmental impacts of tannery wastewater.

LEATHER TANNING PROCESS

Tanning is the process (Figure 2.1), which converts the protein of the raw hide or skin into a stable material, which will not putrefy and is suitable for a wide variety of end applications [2]. The two main types of tanning are chrome tanning and vegetable tanning. The highly polluting chromium is the most commonly used tanning material producing leather that is more supple and pliable than vegetable-tanned leather, and does not discolor or lose shape in water as drastically as vegetable-tan.[3]

Chromium compounds are applied to protect hides and skins from decay and to make them more durable against moisture and aging [4].

Chromium interacts with fibers in the raw hide/skin during a bathing process, after which the tanned hides are wrung and prepared for finishing. The seven major tanneries in Uganda apply chrome tanning as opposed to vegetable tanning to convert leather into a stable material, in this case wet blue leather.

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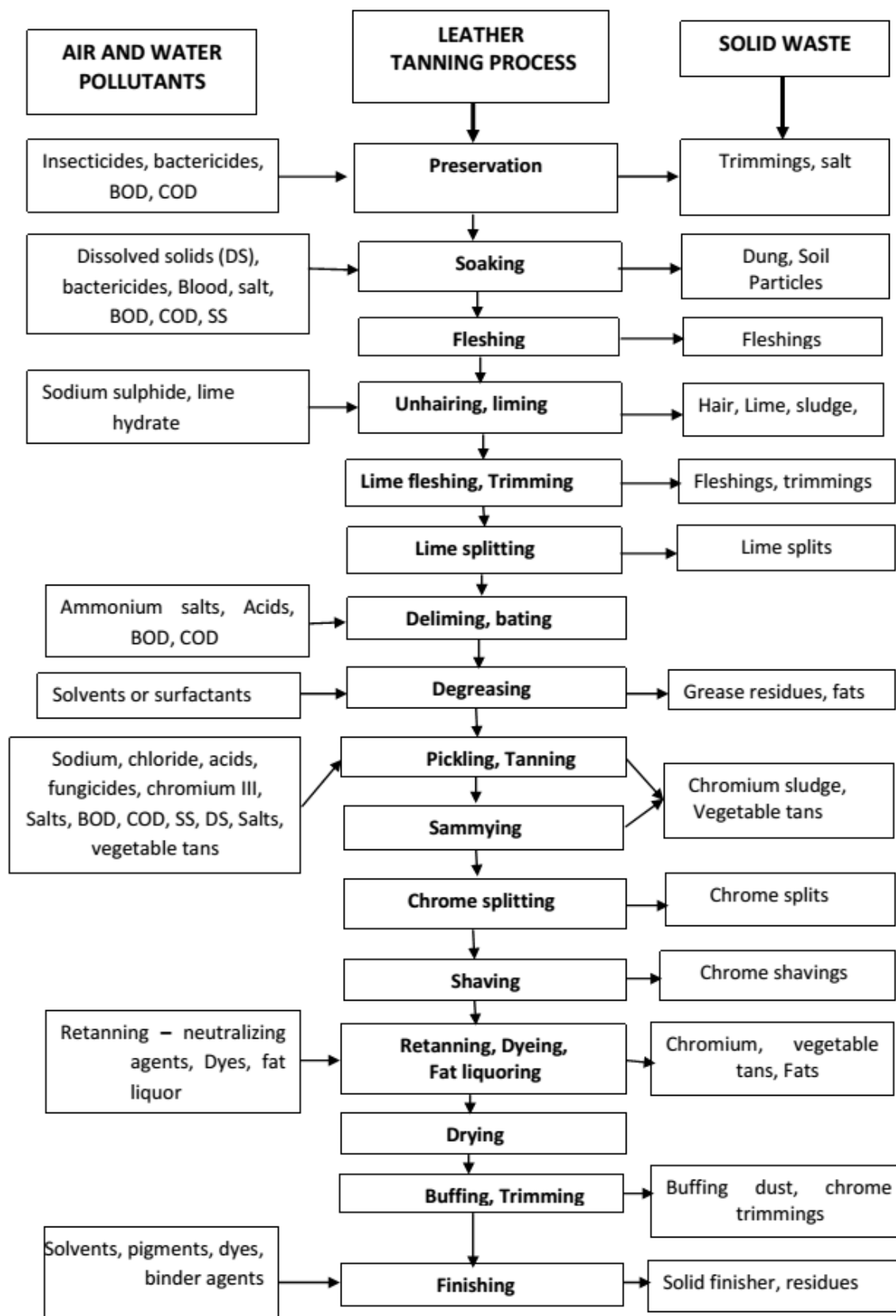


Figure 2.1: Schematic of tanning process, indicating waste streams [1]

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The production process of finished leather in a tannery involves four major groups of sub processes[5]. These include; Beam house operation, Tan yard processes, Retanning and Finishing. However for each end product, the tanning process is different and the quality and amount of wastewater produced may vary in a wide range characteristics.

Below is a detailed description of each of the four leather tanning sub-processes:

Hides and skins storage and beam-house operations:

Upon delivery, hides and skins can be sorted, trimmed, cured (when the raw material cannot be processed immediately) and stored pending operations in the beam house.

The following processes are typically carried out in the beam house of a tannery: soaking, de-hairing, liming, fleshing (mechanical scraping off of the excessive organic material) and splitting. The beam house operations soaking, liming lead to discharge of high amount of sulphides, lime, ammonium salts, and protein in the effluent.

Tan yard operations:

Tan yard operations include; de-liming, bating, pickling and tanning. The two main tanning agents are mineral (trivalent chromium salts) and vegetable (quebracho and mimosa). The tanned hides and skins, once they have been converted to a non-putrescible material, are tradable as intermediate products (wet blue). The tan yard processes generate wastewater that is highly contaminated because of the inorganic salts, chloride, ammonia, chromium and sulphate [6].

Deliming liquors carry significant BOD load whereas bating liquors account for the presence of soluble skin proteins and ammonium salts containing high organic matter. Pickle liquors are acidic and contain high amount of salt and chrome liquors contain high concentration of chrome compounds and neutral salts [7].

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Post-Tanning operations/retanning:

This sub-process involves washing out the acids that are still present in the leather following the tanning process. According to the desired leather type to be produced the leather is re-tanned (to improve the feel and handle of leathers), dyed with water-soluble dyestuffs (to produce even colors over the whole surface of each hide and skin), fat liquored (leathers must be lubricated to achieve product-specific characteristics and to re-establish the fat content lost in the previous procedures) and finally dried. Major water pollutants discharged at this level include; dyes, fats, fat liquor, syntans and chromium. The wastewater from neutralization, retanning, dyeing and fat liquoring sections contribute little pollution load [7].

Finishing operations:

The purpose and art of finishing leather, is to give it a thin, smooth and attractive finish as much as possible without harming its known characteristics such as its look and ability to breathe. The aim of this process is to treat the upper (grain) surface to give it the desired final look. The overall objective of finishing is to enhance the appearance of the leather and to provide the appropriate performance characteristics in terms of color, gloss, and handling, among others. The finishing operation is only carried out by LIU tannery at small scale levels of production[8].

CHARACTERISTICS OF WASTEWATER FROM TANNING PROCESS AND ASSOCIATED EMISSIONS

Tannery wastewater is heavily polluted containing appreciable biodegradable matters as well as inorganic substances like chromium, sulphide, chloride et cetera [9]. Tannery effluent is among the most hazardous industrial pollutants due to its huge organic and inorganic load, which is highly toxic to human life and environment[10, 11]. The characteristics of tannery wastewater

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vary considerably from tannery to tannery depending upon the size of the tannery, chemicals used for a specific processes, amount of water used and type of final product produced by a tannery [11]. In general, tannery wastewaters are basic, have a dark brown colour resulting from the decay of organic matter, suspended solids and have a high content of organic substances that vary according to the chemicals used [11].

The wastewater is highly polluted in terms of Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Ammonia, Suspended Solids, Nitrogen, Electrical Conductivity, Sulphates, Sulphides, and Chromium. Discharge of chromium, Ammonia sulphate, Sodium-sulphide, Sodium chloride into wastewater is hazardous to the environment [12].

COMPOSITION OF TANNER WASTE WATER.

The characteristics of tannery effluent 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[13].The tannery waste contains large number of pathogens and foul smelling liquids containing both alkaline and acidic liquors[14].

i. Solids

The solids to be found in tannery effluent fall into several distinct categories[15],solids are removed from tannery waste mechanically[16]

a. Suspended solids

The suspended solids component of an effluent is defined as the quantity of insoluble matter contained in the wastewater. These insoluble materials cause a variety of problems when discharged from a site; essentially, they are made up of solids with two different characteristics [15].

b. Solids with a rapid settling rate (settleable solids)

Settleable solids comprise material that can be seen in suspension when an effluent sample is shaken, but settles when the sample is left to stand. The majority of these solids settle within 5 to 10 minutes, although some fine solids require more than an hour to settle[15].These solids

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originate from all stages of leather making; they comprise fine leather particles, residues from various chemical discharges and reagents from different waste liquors. The settleable solids content is determined by leaving the shaken sample to settle and then filtering a known volume of the semi-colloidal matter remaining in suspension. After drying and weighing, the quantity of semi-colloidal matter can be calculated. The difference between the suspended solids and this figure is the settleable solids content[15].

c. Semi-colloidal solids

Semi-colloidal solids are very fine solids that, for all practical purposes, will not settle out from an effluent sample, even after being left to stand for a considerable period of time. They can, however, be filtered from solutions. Together with the more readily settleable solids, they thus comprise the suspended solids of an effluent that can be measured analytically [15]. Most of these solids are protein residues from the beam-house operations - mainly liming processes; however, large quantities are also produced owing to poor uptake in vegetable tanning processes, another source being poor uptake during retanning[15].

d. Gross solids

Gross solids are larger than a sampling machine can handle, hence they are not measured. Their presence, however, is clear to see and the dangers they pose are fully recognized. The waste components that give rise to this problem are often large pieces of leather cuttings, trimmings and gross shavings, fleshing residues, solid hair debris and remnants of paper bags. They can be easily removed by means of coarse bar screens set in the waste water flow. If, however, they emerge from the factory, they settle out very rapidly[15].

ii. Chromium compounds

Two forms of chrome are associated with the tanning industry

a. Chrome 3(trivalent chrome, chrome III)

Chromium is mainly found in waste from the chrome tanning process[15]. Despite the thermodynamic stability of Cr (III), the presence of certain naturally occurring minerals, especially MnO₂ oxides, can enhance oxidation of Cr (III) to Cr (VI) [17]. At high pH, Cr (VI) is bio available, and it is this form that is highly mobile and therefore poses the greatest risk of groundwater contamination[17]

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b. Chrome 6⁺ (hexavalent chrome, chromeVI)

Tannery effluents are unlikely to contain chromium in this form. Chrome in the hexa-valent form is very toxic[13, 18] Dichromates are toxic to fish life since they swiftly penetrate cell walls[15].Cr (VI) is a strong oxidant and therefore more reactive than Cr (III)(32)

iii. Turbidity

Turbidity in water is caused by suspended and colloidal matter such as clay, silt, finely divided organic and inorganic matter, and plankton and other microscopic organisms. Turbidity is a measure of the clarity of a water body and is an optical measurement that compares the intensity of light scattered by a water sample with the intensity of light scattered by a standard reference suspension. It is commonly recorded in nephelometric turbidity units[16].

ENVIRONMENTAL PROBLEMS RELATED TO TANNERY WASTE WATER.

Cr(VI) have different health effects and cause for acute toxicity, mutagenic, carcinogenic and high blood pressure for societies using untreated waste water contain large amounts of Cr(VI) discharged from any tannery industry and also affect seed germination of the plants[18] From earlier research work, hexavalent chromium (Cr⁶⁺) is very much carcinogenic for human being upon long time exposure. The toxicity and carcinogenic effect of chromium (VI) is perceived after it enters into living cell[19]. It was also found that the tannery industries are not only discharging Chromium as heavy metal, which is inherent to the tanning process, but also significant amounts of zinc, copper, manganese, and lead as well[20]. Studies revealed that high concentrations of chromium (VI) in the cell could lead to DNA damage, causing several genotoxic inside the human body[19].Chromium salts (chromates) also cause allergic reactions in human body and highly toxic to animals due to its ability to generate reactive oxygen species in cells[21].

Untreated tannery effluent can cause severe ailments to the tannery workers such as eye diseases, skin irritations, kidney failure, gastrointestinal problems[13],rotten-egg smell and headaches, nausea and affect central nervous system[22].

Air pollution and odor is caused by Sulfide in waste water which comes from the decay of organic matter in the form of hydrogen sulfide (H₂S) produced by decomposing microorganisms

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from organic substances that are toxic to algae and other microorganisms. The results of decay of organic substances cause air pollution and odor. In addition, sulfide is corrosive in the form of hydrogen sulfide which causes problems in the environment[22]

Abnormal concentration of sodium chloride and chromium affect ground water quality seriously within a short period where the effluents is discharged on a land or if stored on in unlined bank[23]

TREATMENT METHODS FOR TANNERY WASTE WATER

Tanneries wastewater effluent is treated in many different ways, treatment is necessary due to the wide range of toxic effects on the environment caused by untreated tannery effluents and sludge[24].

The following treatment steps are necessary in tannery waste water treatment; Mechanical treatment, Effluent treatment, Post-purification, sedimentation and sludge handling[24]. Also the extraction of organic compounds from leather industry wastewater has been carried out by different methods [25] such as liquid-liquid extraction and solid-phase micro-extraction [21].

The leather industry wastewater is also treated with various traditional processes such as adsorption on activated carbon, reverse osmosis[26], coagulation by chemical agents[27], ion-exchange on synthetic adsorbent resins, biological method (biodegradation) and electrochemical methods can be used efficient[25]. However, these techniques are non-destructive, thus creating secondary pollutants[28].

Treatment of secondary pollutants is a highly expensive process[25]. To solve this problem, new technologies have been developed for wastewater treatment as well as to improve the mineralization process of most of the organic pollutants present in the wastewater. The new oxidation technology, known as advanced oxidation processes (AOPs) and heterogeneous photocatalysis, appears as an emerging technology for complete mineralization of the organic and inorganic pollutants[29].

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Conventional treatment methods

Many Conventional treatment methods are used to treat waste water from tanneries. These include; biological process, oxidation process and chemical process[30].

Biological process

Biological treatment of wastewater is more favorable and cost effective as compared to other physiochemical methods. Various microorganisms are capable of reducing the content of pollutants significantly by utilizing them as energy and nutrient source in the presence or absence of oxygen[13].

Biological processes are usually prescribed for treating industrial effluents to reduce organic content as they have economic advantages over chemical oxidation. However high concentration of tannins and other poorly biodegradable compounds as well as metals can inhibit biological treatment[31]

Aerobic treatment

Aerobic microorganisms use organic carbon in the effluent and convert it to biomass and carbon dioxide. A large amount of sludge is generated along with high energy consumption in the process [13]. Under aerobic conditions, sulphate can act as an electron acceptor for a group of bacteria that can couple the oxidation of reduced organic or inorganic compounds to the reduction of sulphate for bioenergetic purposes. This process is known as dissimilatory sulphate reduction (Sulphidogenesis) and the bacteria involved are known as the sulphate reducers or sulphate- reducing bacteria. Based on the metabolic capacities, sulphate reducing bacteria can be classified into two categories - those species or genera that are capable of complete oxidation of organic compounds to CO₂ and those that carry out incomplete oxidation, usually to acetate as end-product[32].

The majority of sulphate-reducing bacterial species can also utilize sulphite, thiosulphate, organic sulphur compounds and elemental sulphur as electron acceptors[32]. Advantages of biological wastewater treatment system are; Low capital and operating costs compared to physico- chemical process ,Oxidation of wide variety of biodegradable organic compounds,

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Removal of reduced inorganic compounds, such as sulphides and Ammonia, Total nitrogen removal through nitrification and denitrification [33].

Anaerobic treatment

Anaerobic treatment of wastewater converts the organic pollutants into a small amount of sludge and large amount of biogas (methane and carbon dioxide). The sulfide present in wastewater inhibits the anaerobic treatment. Methanogenic bacteria are inhibited by sulfide, whereas acidifying and sulfate reducing bacteria do not inhibit. Three inhibiting effects of sulfide or sulfide reduction are known: direct toxicity of sulfide, substrate competition between sulfate reducing and methanogenic bacteria and precipitation of trace elements by sulfide[13].

Conventional Activated Sludge Process (ASP) System:

This is the most common and oldest bio-treatment process used to treat municipal and industrial wastewater. Typically wastewater after primary treatment i.e. suspended impurities removal is treated in an activated sludge process based biological treatment system comprising aeration tank followed by secondary clarifier. The aeration tank is a completely mixed or a plug flow (in some cases) bioreactor where specific concentration of biomass (measured as mixed liquor suspended solids (MLSS) or mixed liquor volatile suspended solids (MLVSS)) is maintained along with sufficient dissolved oxygen (DO) concentration (typically 2 mg/l) to effect biodegradation of soluble organic impurities measured as biochemical oxygen demand (BOD₅) or chemical oxygen demand (COD)[34]

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CHEMICAL TREATMENT PROCESS

Several studies have been carried out for the treatment of industrial effluents through coagulation and flocculation process. Coagulation is typically employed as a pretreatment process and thus further treatments such as biological (secondary) and advanced (tertiary) treatment are required in the leather industry in order to meet the proposed tannery effluent standards [15].

The frequently used coagulants in tannery effluent treatment are: Alum, industrial aluminum sulphate ($\text{Al}_2 (\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$); Iron sulphate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), Iron chloride, industrial ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$); Lime: industrial calcium hydroxide $\text{Ca}(\text{OH})_2$ [17, 35].

Coagulation and flocculation

Coagulation-flocculation process is employed in separating suspended solids materials in tannery effluent [36]. The process operates in steps which break down forces, which stabilize charged particles present in the tannery effluent allowing interparticle collision to occur, hence, generating flocs [37]. Coagulants are added to tannery effluent to neutralize the negative charge of suspended particles. Upon neutralization, the suspended particles stick together to form slightly larger particles. For tannery effluent coagulation, rapid mixing of the coagulant is needed to achieve effective collision; this process is followed by a flocculation process where gentle mixing increases the particle size from sub-microfloc to visible suspended solids. Particles are thus bound together to produce larger macroflocs. To prevent destabilizing of macroflocs an attention is given to the mixing velocity and energy [38]

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Advanced treatment methods

The presence of toxic chemicals has warranted the use of advanced treatment techniques for tannery wastewater treatment which are discussed below.

Electrochemical oxidation

Considering the challenges, a comprehensive advance chemical treatment technology was thought for tannery effluent treatment from the real life experiences. Under this circumstances Electrochemical treatment have been emerged as the new technology in the field of tannery effluent treatment [39]. The electrochemical treatment of tannery effluent has been investigate by many researchers[40, 41] in order to improve the performance of treatment by conventional coagulation and flocculation process. It is also fact that application of different electrode materials with different electrolytic properties can affect reactor treatment efficiency[40]. The removal kinetics of organic pollutants as well as nutrients showed very faster removal than biological treatment[41]. Although Nitrogen, Phosphorus, Chromium, Arsenic and other heavy toxic metals are successfully removed by electrochemical treatment, still there is a certain limitation of applying this technology in raw Tannery effluent .However electrochemical technology can successfully be applied in post treatment or final finishing stage[33].

Membrane filtration

Membrane filtration is a physical separation process that uses a semi permeable membrane to remove suspended solids from a liquid stream. Microfiltration (MF) and ultrafiltration (UF), along with nanofiltration (NF) and reverse osmosis (RO), are all examples of pressure-driven membrane filtration [42]. MF/UF are flexible filtration methods, and both are used to remove a variety of particles, pathogens, and microorganisms from process liquids[43] .

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Several studies showed that crossflow microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) and supported liquid membranes (SLMs) can be applied in leather industry for the recovery of chromium from spent liquors [42].

Photo-Fenton process

A combination of hydrogen peroxide and UV radiation with Fe^{2+} or Fe^{3+} oxalate ion, the so-called photo-Fenton process produces more hydroxyl radicals in comparison to the conventional Fenton method (Fe^{2+} with hydrogen peroxide) or to the photolysis [44].

Ozonation

Aiming for decontamination in drinking water production as well as for treatment of strongly contaminated residual waters, the use of ozone in conjunction with UV light as a method of removal of organic material has been technically developed. The O₃/UV process seems at present to be the most frequently applied AOP for a wide range of compounds. This is mainly due to the fact that ozonization is a well known procedure in water technology and that ozonizers are therefore in most cases readily available in drinking water treatment stations. From the photochemists point of view, the absorption spectrum of ozone provides a much higher absorption cross section at 254 nm than H_2O_2 , and inner filter effects by e.g. aromatics are less problematic. There remain, however, many questions related to mechanisms of free radicals production and subsequent oxidation of organic substrates. In fact, the literature contains many conflicting reports on the efficiency of this oxidation method which may be linked to mechanistic problems as well as to the difficult tasks of dissolving and photolyzing ozone with high efficiency. Finally, linked to the problem of quantifying rates of absorbed photons in heterogeneous (gas/liquid) media and to the reactivity of ozone towards most unsaturated organic compounds, procedures for the determination of quantum efficiencies still remain to be worked out. [45]

Ozonation prior to biological treatment is widely carried out in order to achieve high color removal efficiencies and convert refractory organics to biodegradable organic compounds. This decrease of toxicity was observed in parallel with the destruction of the aromatic surfactant homologues of nonylphenol ethoxylate type [44].

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Cyclic Activated Sludge System (CASSTM):

Cyclic Activated Sludge System (CASSTM) as the name suggests is one of the most popular sequencing batch reactor (SBR) processes employed to treat municipal wastewater and wastewater from a variety of industries including refineries and petrochemical plants.

This technology offers several operational and performance advantages over the conventional activated sludge process. The Cyclic Activated Sludge System process performs all the functions of a conventional activated sludge plant (biological removal of pollutants, solids/liquid separation and treated effluent removal) by using a single variable volume basin in an alternating mode of operation, thereby dispensing with the need for final clarifiers and high return activated sludge pumping capacity[34].

The Cyclic Activated Sludge System (CASSTM), incorporates a high level of process sophistication in a configuration which is cost and space effective and offers a methodology that has operational simplicity, flexibility and reliability that is not available in conventionally configured activated sludge systems. Its unique design provides an effective means for the control of filamentous sludge bulking, a common problem with conventional processes and other activated sludge systems[34].

CONCLUSION

The study revealed that wastewater in the leather manufacturing cycle is majorly generated in the beam-house processes which include; soaking, liming, de-liming, pickling and bating. The tanyard processes as well generate wastewater, especially tanning and re-tanning however the dyeing and finishing processes are normally dry. All wastewater generated at the different stages of leather processing contains pollutants such as high concentration of BOD, COD, suspended solids, sulphates, sulphides, EC, pH and Chromium hence the need for its treatment before final discharge into the environment.

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