IJSGS FUGUSAU VOL. 8(2) JULY, 2022 WEBSITE: https://fugus-ijsgs.com.ng

IJSGS

ISSN: 2488-9229
FEDERAL UNIVERSITY
GUSAU-NIGERIA

INTERNATIONAL JOURNAL OF SCIENCE FOR GLOBAL SUSTAINABILITY

A Perspective on the Catalytic Treatment of Tannery Wastewater

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Received on: April, 2022 **Revised and Accepted on:** June, 2022 **Published on:** July 2022

ABSTRACT

Tanning is the transformation process which converts animal hides and skin into stable products called leather. The process generates highly turbid, colored and foul smelling wastewater. The tannery wastewater contains highly recalcitrant chemical pollutants such as chromium-complexed collagen, surfactants, sulfides chlorides and salts. In addition, tannery wastewater often contains high concentration of biological oxygen demand (BOD), chemical oxygen demand (COD) suspended solids (SS), and nitrogen, many researchers reported that several conventional methods namely, coagulation-flocculation precipitation, oxidation, ion exchange and membrane filtration, ozonation have been utilized to treat tannery wastewater. These methods have associated shortcomings such as high cost of reagents, generation of toxic sludge, and low efficiency. Not only that, organic pollutants are also transferred from one phase to another and may even become more toxic than the compounds. In the recent times, heterogeneous photocatalysis which involved the use of metal oxide such as CdO, CuO, ZnO, SiO_2 and Fe_2O_3 have received considerable attention among researchers due to capability to degrade numerous organic contaminants into carbon dioxide and water. Among the aforementioned metal oxide nanocatalysts, ZnO nanoparticles have emerged as the candidate in photocalysis due to its low cost, availability, excellent chemical stability, non toxicity and high initial rate of activities. Similarly, ZnO nanoparticles are relatively cheap and have the ability to absorb a wide range of UV spectrum compared to other metal oxides. The review highlighted all these details and simultaneously provided insights into directions for further investigations.

Keywords: tannery wastewater, treatment, techniques, catalysis

1.0 INTRODUCTION

The contamination of water resources from different point and non-point sources such as industrial activities, agricultural practices, climate change coupled with the geometrical population growth has raised serious environmental and health concern across the globe, thus demanding urgent action (Abdel *et al.,* 2015). In fact, large volume of untreated wastewater containing thousands of organic and inorganic pollutants are generated and discharge on daily into the limited water bodies on daily basis (Jahan *et al.*, 2014). Specifically, industrial tanning transformation process which converts animal hides and skin into stables products called leather generated highly turbid, colored and foul smelling wastewater (Hayelom and Adhena. 2014). The tannery wastewater contains highly recalcitrant chemical pollutants such as chromium-complexed collagen, surfactants, sulfides chlorides and salts (Islam *et al.,* 2014). In addition, tannery wastewater often contains high concentration of biological oxygen demand (BOD), chemical oxygen demand (COD) suspended solids (SS), nitrogen, conductivity, sulfides/sulfates and chlorides among others (Wosnie and Wondie, 2014). It should also be mentioned that the volume of wastewater produced per ton of raw hide is around 30-35m^3 per day (Abdel *et al*., 2015). The review therefore seeks to survey an updated literature on the treatment processes for wastewater with emphasis to catalytic photo-degradation approach adopted. Importance was also placed on the areas requiring further research expansion.

2.0 TANNERY ACTIVITIES AND TANNERY WASTEWATER

Tanning is a chemical process that converts animal and skin into stable products called leather. This transformation of hides into leather occurred in the presence of tanning agents. However, the process produced highly offensive turbid, colored and foul smelling wastewater (Heyelom and Adhena 2014). The tanning of raw hide/skin can be categorized into: vegetable tanning and chrome tanning (Jahan *et al*., 2014). The quality of tannery wastewater differ from tannery depending upon the size of the tannery, chemicals types for specific process amount of water used and type of final product obtained (Abdel *et al.*,2015). The pollution of tannery effluent can be determined by measuring the level of chemical oxygen demand (COD), biochemical oxygen demand (BOD), suspended solids (SS), chromium and sulphides (Yusuf *et al*., 2015). In general, tannery wastewater is alkaline in nature with pH above 7.5 and contains high amount of slowly biodegradable organic substances, suspended solids, considerable amount of nitrates and total chromium (Abdel *et al.*, 2015). Other constituents in tannery wastewater include high amount of sulfides/sulfates, chlorides and other metals (Yusuf *et al*., 2015). There are three main kinds of tannery wastewater with each having its properties. Firstly, wastewater contains high amount of sulfides/sulphates, have high level of pH, however, it is chrome-free. In the second case, the wastewater may emanate from the tanning and re-tanning, and usually contains high chromium content and in most cases acidic. In the third case, soaking and other general wastewater originating from post-tanning operations have low Chromium content (Yusuf *et al*., 2015).

The technique of skin tanning is a complex and multi-task process that involved technological process. This process includes beam house operation, tannery operations, post-tanning and finally finishing of leather (Chowdhary *et al*., 2017). Unfortunately, tannery industry generates toxic and complex wastewater associated with foul odor (Singh *et al.*, 2016). Acids alkalis chromium salts, tannin solvents, sulfides, dyes, auxiliaries, and a host of chemical compounds are used for the conversion of raw or semi-pickled skins into commercial products. Most of these chemicals are not completely absorbed by the skin instead remained in the wastewater (Hasssen and Belay, 2017). For instance, the present commercial chrome tanning process generated about 50-70 chromium related constituent (Abdel *et al.*, 2015). Additionally, synthetic tannin (syntan), oils and resins are often added to soften the leather at varying doses (Nithya and Sudha, 2016). One of such refractory groups of chemicals in tannery wastewater was derived from tannis (Singh *et al.*, 2016). Syntans are complex chemical structures, composed of an extended set of chemical such as phenol ethylene, formaldehyde and melamine-based syntans, and acrylic resins (Hassen and Belay, 2017). Among syntans, the ones based on sulfonated naphthalene's and their formaldehyde played a primary role, depending on the volumes and quantity used during the leather formation process (Abdel *et al.*, 2015). Furthermore the beam soaking, liming and de-liming process usually resulted into discharge of wastewater that is highly rich in sulfides, lime and chromium, chlorides, sulfates, and protein in the environment. Tannery wastewater also contains suspended solids, dissolved lime, sodium sulfide, high ammonium nitrogen and organic matter (Yusuf *et al*., 2015). Unhearing and fleshing wastewater contains fatty fleshing matter in suspension (Jahan *et al.*, 2014). The de-liming liquors constituents responsible for high BOD load (Yadav *et al.*, 2016). The wastewater from neutralization, re-tanning, dyeing and fat liquoring sections contains volatile organic compounds (VOC) (Hassen and Belay, 2017). Organic pollutants (protein and lipid components) originated from skins constituents about 60 out of which 30 were lost during the working cycle (Abdel *et al.*, 2015). Furthermore, tannery wastewater is characterized by high contents of organic compounds which usually leach from the skin (such as fats and proteins), inorganic compounds including chromium, sulphide, nitrogen (Chowdhury *et al*., 2013). It should be noted that 30% of the organic pollutants which comprises protein and lipid components are basically from raw skins which are lost during the working cycle and possibly introduced and remained in the wastewater during processes (Jafrano *et al.*, 2013). Similarly, acids, bases and salts of chromium (iii),sulphide and many other compounds used during the conversion process of raw skin.

3.0 ANTHROPOGENIC EFFECTS OF TANNERY WASTEWATER

Tannery wastewater has strong potentials to cause soil and water pollution (Jahan et al and Yadav *et* *al*., 2016). Tannery activities uses more than 250 chemicals for leather production and release a complex mixture of toxic organic chlorinated phenols, toxic Cr (VI), and other toxic pollutants such as sulphides, phenolic compounds others include magnesium, sodium, potassium, cadmium compounds, cobalt, copper, antimony, barium, lead, selenium, mercury, zinc arsenic, nickel, formaldehyde resins, pesticides residues, mineral salts, dyes and solvents like grease and oils (Jahan *et al.*, 2014). Exposure to pollutants in such wastewater can lead to different health challenges. For instance, high concentration of chromium (VI) have caused carcinogenic, mutagenic and destruction of aquatic environments (Hassen and Belay, 2017). It can cause a temporary effects such as dizziness, headache, irritation of eyes, skin or lungs, allergic reactions, poisoning of liver, kidney or nervous system or collapse due to lack of oxygen (Abdel *et al*., 2015). Tannery wastewater is ranked as the highest pollutants containing effluents among all the industrial wastewater samples (Chowdhury *et al.*, 2013).

4.0 TANNERY WASTEWATER TREATMENT TECHNIQUES

Several conventional methods namely, coagulation-flocculation (Ukiwe *et al.*, 2013) precipitation (Khan and Malik, 2013), oxidation (Rabbani *et al.*, 2013), ion exchange and membrane filtration (Thiagarajan *et al*., 2013), ozonation (Abdel *et al.*, 2015). have been utilized to treat tannery wastewater, these methods are associated with many shortcomings such as high cost of reagents, generation of toxic sludge, low efficiency. Not only that, organic pollutants are also transferred from one phase to another and may even become more toxic than the compounds (Pawar *et al.*, 2014). In the recent times, heterogeneous photocatalysis which involved the use of metal oxide such as CdO, CuO, ZnO, SiO_2 and Fe_2O_3 have received considerable attention among researchers due to capability to degrade numerous organic contaminants into carbon dioxide and water (Pawar *et al.*, 2014). Among the aforementioned metal oxide nanocatalyst, ZnO nanoparticles has emerged as the candidate in photocalysis due to its low cost, availability, excellent chemical stability, non toxicity and high initial rate of activities (Matinise *et al.*, 2017). Not only that, ZnO nanoparticles are relatively cheap and have the ability to absorb a wide range of UV spectrum compared to other metal oxides (Snehal *et al.*, 2016). Zinc oxide is classified as a n-type semiconductor with a wide band gap of 3.37ev, large excitation binding energy of 60 meV and often characterized with high thermal and mechanical stability at room temperature (Kumaran and Muraleedham, 2017; Suresh and Narayanan, 2013).

Zinc oxides nanoparticles can be synthesized by physical and chemical method such as solvothermal reduction (Hassan, 2015), solgel technique (Abdel *et al.*, 2015). Hydrothermal (Yilmaz and Unal, 2016), flame spray pyrosis (Joel *et al*., 2016), thermal evaporation, chemical vapor deposition, photo deposition (Perumal *et al*., 2014), however, these methods are costly, generate toxic by- products, require complex synthesis procedure and in most cases required high energy (Maltinise *et al.*, 2017). Nowadays, the synthesis procedure and in most plants extracts to replace expensive commercial and toxic reducing agents such as $NaBH_4$ or $LiAlH_4$ have been globally recognize as a route to reduce environmental pollution due to availability of plant material (Maltinise *et al.*, 2017), use of less toxic chemicals, eco-friendly nature and one step synthesis of nanoparticles (Manokori *et al.*, 2016). Furthermore, ZnO nanoparticles prepared via physical and chemical methods often have high band gap energy and low surface area thus responsible for its low photocatalytic activity in the visible region (Hassan *et al*., 2015). Concerted efforts are ongoing towards the reduction of the band gap of ZnO and extend its absorption threshold to visible region (Kumaran and Muraleedhran. 2017). One of such approaches is doping of ZnO with metals, non metals, or metal oxides or in most cases both, to prevent electron hole recombination (Snehal *et al.*, 2016). Doping is the anchoring of foreign materials (impurities) onto the ZnO framework in order to suppress electron-movement from valence band to conduction band (Kumaran and Muraleedharam, 2017).

4.1 Other Treatment Procedures

Several other traditional methods such as physicochemical neutralization process (Chowdhury *et al.*, 2013), chemical process (Cesaro *et al.*, 2013) and/or biological processes (Lucyna and Przywara, 2017) have been successfully employed to treat tannery wastewater at the classical and industrial scales. However, it has been widely reported by researchers that treatment of tannery wastewater to get rid organic compounds via these conventional methods was not achieved to the desired results (Yadav *et al.*, 2016). Different other treatment techniques such as coagulation-flocculation (Ukiwe *et al*., 2013), precipitation (Khan and Malik, 2013), oxidation (Rabbani et al.. 2013), ion exchange and membrane filtration`(Thiagarajan *et al*., 2013), ozonation (Abdel *et al*., 2015), among others have been utilized to remove toxic organic and inorganic contaminants in tannery wastewater. These methods have associated shortcomings such as high cost of reagents, generation of toxic sludge, low efficiency, not only that organic pollutants were also transferred from one phase to another without actual decomposition to harmless substances such as CO_2 and H_2O (Jahan *et al*., 2014). In view of the aforementioned, well-known wastewater treatment methods are improved or new solutions are sought, in many cases, to achieve the required end results, advanced oxidation process, involving heterogeneous photocatalysis which utilize the high oxidation potential of generated hydroxyl radicals have been recognized as an alternative method to decompose calcitrant organic pollutants in the tannery wastewater prior to the discharge into the environment (Abdel *et al.*, 2015). The degradation efficiency of the organic compounds and concentration of the organic radicals that can mineralize the target organic compounds into CO_2 and H_2O is an issue of concern for the purpose of assessing process reliability (Cesaro *et al.*, 2013).

5.0 HETEROGENEOUS PHOTOCATALYSIS AND THE PROCESS CHEMISTRY

Photocatalytic technology where a noncatalyst harnesses UV radiation from sunlight or artificial light utilize the energy have been widely applied to break down different substances including organic materials, organic acids, estrogens pesticides (Aljuboury *et al*., 2015). Similarly, dyes, crude oil (Udom *et al.*, 2013), microbes (including viruses) and chlorine resistant organism (Lakherwal, 2014). Others are inorganic molecules such as nitrous oxides (NOx) (Zheng *et al*., 2015). Heterogeneous photocatalysis can be combined with other technologies such as precipitation or filtration to remove metals and other toxic constituent in water (El-dafrawy, 2016). Due to this universal applicability, photocatalysis with nanopartices as catalyst have been applied to reduce air pollution for self-cleaning surfaces in addition to wastewater purification and splitting (Asim *et al*., 2013). Photocatalysis is defined as the sequential chemical reaction (oxidation/reduction) which occurs under light irradiation in the presence of oxygen and semiconductor metal oxide (Asim *et al.*, 2013). Catalyst is activated upon the absorption of a photon of light which has the energy equal or higher than the band gap energy of the metal oxide in question (Zheng *et al.*, 2015). The absorption of light by a semiconductor metal oxide caused excitation of an electrons and charge separation from valence hand to the conduction band and sequentially generate hole (h) and electron (e-) in the former and the later respectively (Meenakshi and Sivasamy, 2017). The photogenerated electrons and holes diffused onto the surface and react with the hydrophilic and nucleophilic substances absorbed on the photocatalyst surface respectively the activated and unstable products (Singh *et al.*, 2013). Depending on molecules adsorbed on the photocatalyst surface and their reaction rates with electron .the reaction usually produces more than two intermediates due to the interaction between organic pollutants and photogenerated species (El-dafrawy, 2016). This is because the shelve the electron-hole pairs before recombination is short in the order of nanoseconds, however the recombination slowed the reduction and oxidation reactions on the photoctalyst surface. This phenomenon quantitatively reduced the effective interactions between the pollutants and free radicals and consequently affects the overall performance of the catalyst concentration of hydroxyl radicals via interaction between the photogenerated holes and water molecules adsorbed on the catalyst surface. Furthermore the presence of adsorbed oxygen molecules near the photogenerated electrons often resulted to the generation of superoxide radical ($-O_2$) and reduces the possibility of electron-hole recombination rate (Zheng *et al.*, 2015). These superoxide radical anions either oxidize the pollutant molecules directly or generate further hydroxyl radicals (Menakshi and Sivasamy, 2017). Hydroxyl radicals are well-known for being oxidize in the interface as adsorbed species on the photocatalyst surface or to diffuse away onto the bulk of the solution and promote oxidation reactions with pollutants therein (Kazeminezhad and Sadollahkhani, 2017). It is worthy to note that, depending on other applied experimental conditions, hydroxyl or superoxide radicals, holes, hydrogen peroxide and oxygen molecules played a crucial role at different levels in the photocatalytic experimental set up (Giri and Golder, 2014). The higher removal of nearly 100% for all the studied heavy metals within 60 min. of process time was recorded. This applies that the synthesized material played a dual role as catalyst and nano-adsorbent. ZnO doped with ZnO nanoparticles under visible light can be very reliable in this regard. The authors observed higher photocatalytic activity for Sr^{2+} doped ZnO than that of pure ZnO. The sample with mole ratio of Sr/ZnO 1:2 calcined at 60^0C for 7h exhibited the maximum activity with the cationic methylene blue degraded faster than anionic methyl orange due to the accumulation of more negative charges on the surface of the catalyst (Menakshi and Sivasamy, 2017).

6.0 CONCLUSION AND FURTHER WORKS

The presence of high concentration of aromatic compounds and heavy metals in tannery effluents represents a serious challenge and causes considerable ecological imbalances. The tannery wastewater is highly turbid, coloured and have odoriferous smell with pH value of about 7.5. Exposures to tannery effluents have been reported to cause several diseases such as skin diseases, kidney problem, disruption of endocrinesin humans and aquatic lives. coagulation-flocculation precipitation, oxidation, ion exchange and membrane filtration, ozonation have been utilized to treat tannery wastewater, these methods have associated shortcomings such as high cost of reagents, generation of toxic sludge, low efficiency, not only that organic pollutants were only transfer from one phase to another and may even become more toxic than the compounds. Additionally, conventional methods such as activated carbon adsorption, chlorination, reverse osmosis, ion exchange and membrane filtration, and precipitation, photo light, electrodialysis among others utilizes expensive reagents, generation of toxic sludge, unable to mineralize and degrade complex aromatic compounds. These shortcomings hinder the effective use of conventional wastewater treatment methods in the treatment of tannery wastewater. Specifically, hetrogeneous photocatalysis involving the use of metal oxide such as CdO, CuO, ZnO, SiO_2 and Fe_2O_3 have received considerable attention among researchers due to capability to degrade numerous organic contaminants into carbon dioxide and water. Among the aforementioned metal oxide nanocatalyst, ZnO nanoparticle have emerged as the best candidates in photocalysis due to its low cost, availability, excellent chemical stability, non toxicity and high initial rate of activities. However, among many other issues that are still unresolved and require further investigations are the mechanism of interaction of photons with wastewater versus catalyst, establishing the main radicals responsible for activity initiation, the light range required to achieve optimum activity with specific catalyst and the effect of metal oxides co-doping the ZnO. Addressing these parameters will enhance the overall process efficiency for the interest of industrial advances.

ACKNOWLEDGEMENT

The authors are grateful to the Tertiary Education Trust Fund (TETFUND) for Research Funding to Federal University Gusau under the Batches 4 and 5 of the Institutional Based Research (IBR) Scheme.

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