BIOREMEDIATION OF HEAVY METALS IN TANNERY EFFLUENTS

Abstract

Human health has suffered significant impairment, alongside various environmental issues, due to the mismanagement of waste, particularly wastewater generated from tanning processes. Despite ongoing efforts to mitigate tannery wastewater (TWW) pollution, terminal treatment remains predominant. This chapter provides a concise discussion on key methods of TWW treatment, including coagulation and flocculation, biological treatment, adsorption, membrane filtration, and advanced oxidation processes. Electrochemical treatment is specifically emphasized for its superior performance, environmental friendliness, and high efficiency. Integrated or combined treatment approaches, offering improved performance and multifunctionality, are recommended considering the challenging physico-chemical conditions of TWW. However, comprehensive studies on method combinations and cost-effectiveness are still necessary. This work highlights the persistent presence of chromium residue in treated sludge and elevated salinity in effluent, offering feasible solutions. Future directions for TWW treatment should prioritize multifunctionality, recycling, and intensification.

Keywords: Tannery wastewater, Decontamination, Treatment, Electrochemical methods, Recycling.

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I. INTRODUCTION AND BACKGROUND

Waste is produced across various sectors of society, including industries, agriculture, transportation, construction, and by consumers. These discarded pollutants encompass a wide range of waste materials. Industrial activities generate different types of waste, with the quantity and toxicity of waste generated varying depending on the specific industrial processes involved. Research has shown that microorganisms hold significant potential for mitigating environmental pollution, particularly in addressing water pollution resulting from industrial activities. Current studies indicate that leather industries are among the most significant sources of pollution, leading to adverse environmental impacts due to their waste emissions.

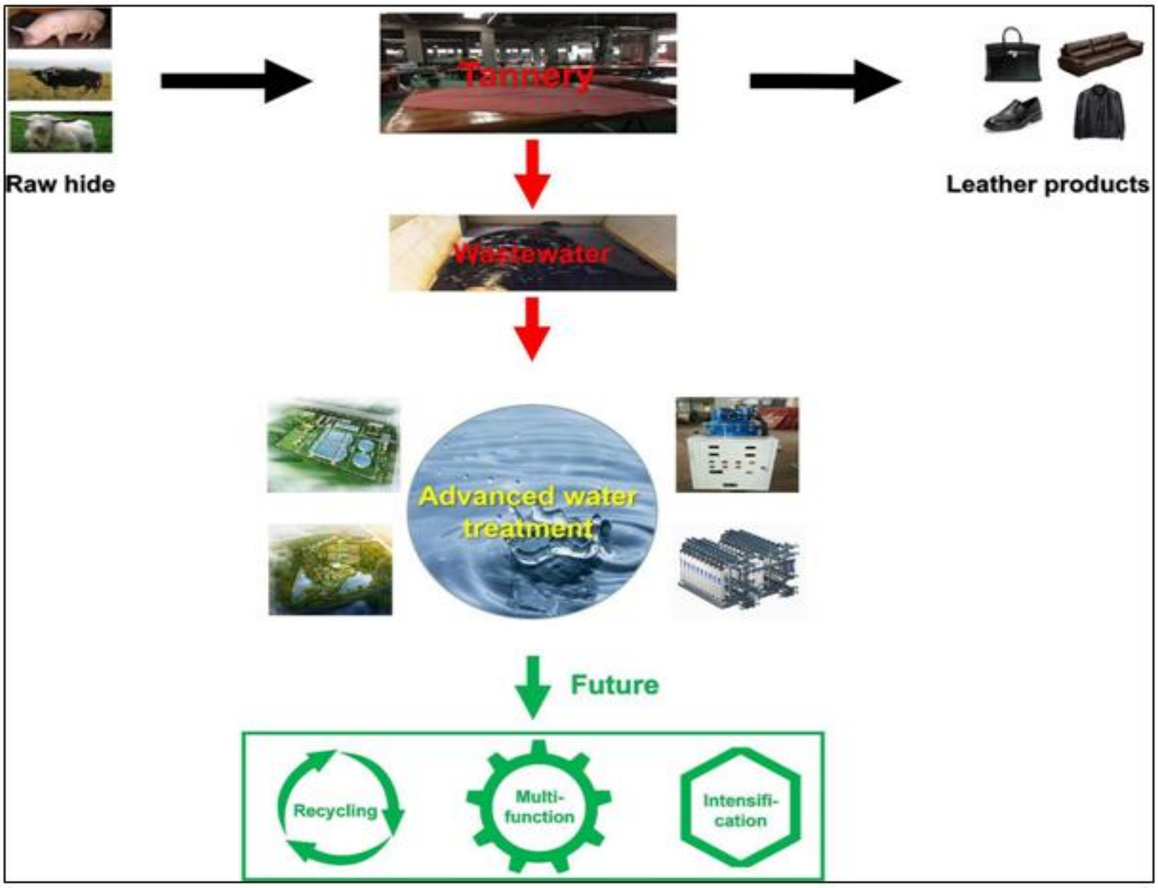

Figure 1: Schematic representation of the tanning process and TWW treatment (Zhao et al.2022)

The discharge of tannery effluents into land and water leads to soil and water pollution, respectively. Aquatic environments suffer adverse effects from heavy metal discharge, particularly from tannery effluents, resulting in lasting negative impacts on nearby flora and fauna in both terrestrial and aquatic regions.

Processing waste from the manufacturing units comprises of wastewater from industries, mainly including sanitary wastes of employees, water discharged from washing factory floor and relatively uncontaminated heating and cooling water. (1)

Among all industrial wastes, tannery effluents have been ranked as the highest pollutants, especially consisting of large contributors of chromium pollution. (2)

Out of all the effluents released from tanneries, chromium stands out as one of the most perilous pollutants (3). Chromium compounds have been linked to mutations that can result in cancer, as well as to inhibition of enzymes and nucleic acid biosynthesis (4). Additionally, chromium exposure can lead to various health issues such as diarrhea, eye irritation, kidney failure, ulcers, and the development of skin and lung cancer (5).

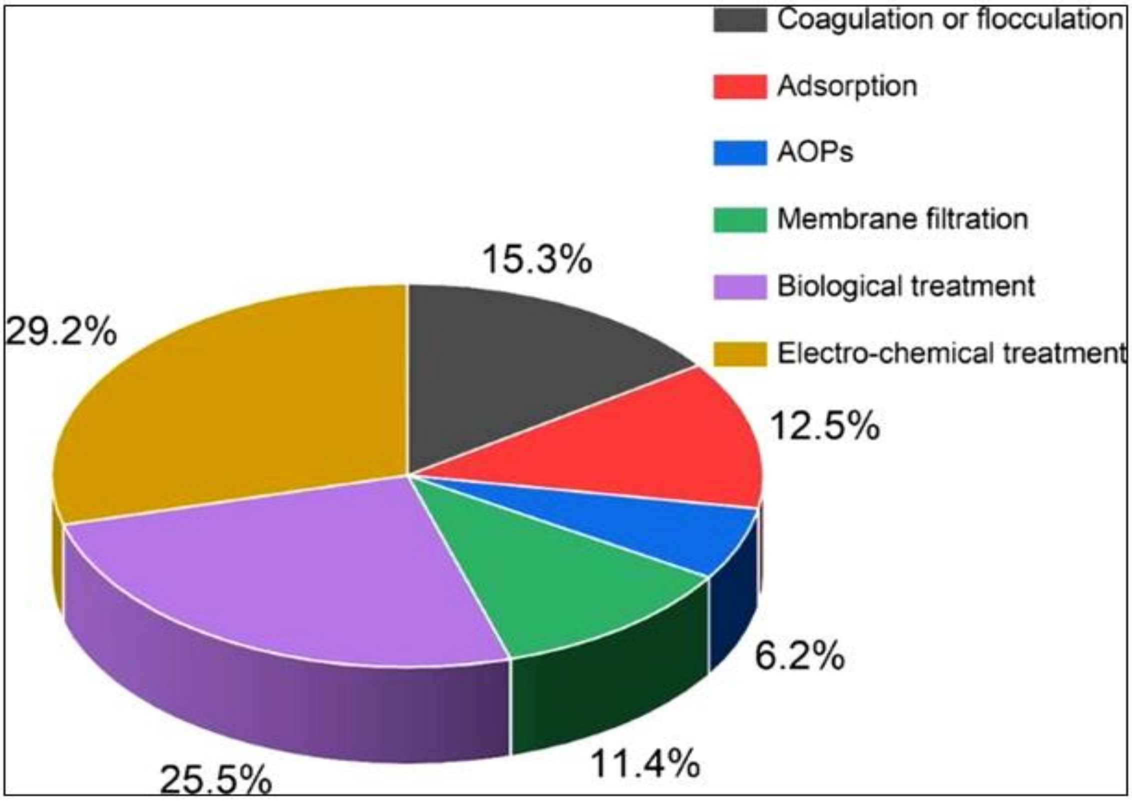

Figure 2: Proportions of various methods utilised in TWW treatment (Zhao et al.2022)

Wastewater treatment encompasses several conventional methods, including advanced oxidation processes, membrane filtration, electrocoagulation, electrochemical methods, ozonation, various adsorbents, and precipitation. However, precipitation is generally less favored due to its cost inefficiency and propensity to cause secondary pollution. Therefore, a combination of physical and chemical methods is typically employed for treating tannery effluents. While these integrated approaches are often efficient, they may not always be cost-effective in terms of energy and chemical consumption, and they can generate substantial quantities of sludge, posing challenges for waste disposal. As a result, there have been numerous efforts to explore the effectiveness of biological methods in treating tannery wastewater. Among them methods like Treatment with biomass and Biochar, Microbial Fuel Cell, Activated Sludge, Bio-Electrochemical Systems, Wetland Systems, Microbial Consortiums, Phytoremediation, Aerobic Digestion, Membrane Bio-Reactors, and Enzymes have been proven to be both cost effectives as well as eco-friendly methods. Certain bacterial species, including Pseudomonas, Arthrobacter, and Bacillus, have demonstrated efficacy in bioremediating tannery wastes (7,8). Since microorganisms possess enzymatic systems capable of oxidizing organic compounds, bioremediation emerges as a highly cost-effective, efficient, and environmentally friendly approach for reducing pollutants in various effluents. Chromium remediation investigations have involved a range of organisms such as Pseudomonas species, Aeromonas species, Bacillus species, Micrococcus species, and Microbacterium species (9). Previous studies on certain Common Effluent Treatment Plants (CETPs) have indicated the presence of Cr (IV) exceeding permissible concentrations. Research has shown that treating effluents with a consortium of Sulphate Reducing Bacteria (SRBs), including Desulphovibrio desulfuricans, D. vulgaris, and D. gigas, which utilize Cr(VI) as an electron acceptor for sulfate reduction, effectively removes both Cr(IV) and sulfates from the solution (Singh, Rajesh et al., 2010).

II. CHARACTERISTICS OF TANNERY WASTE WATER

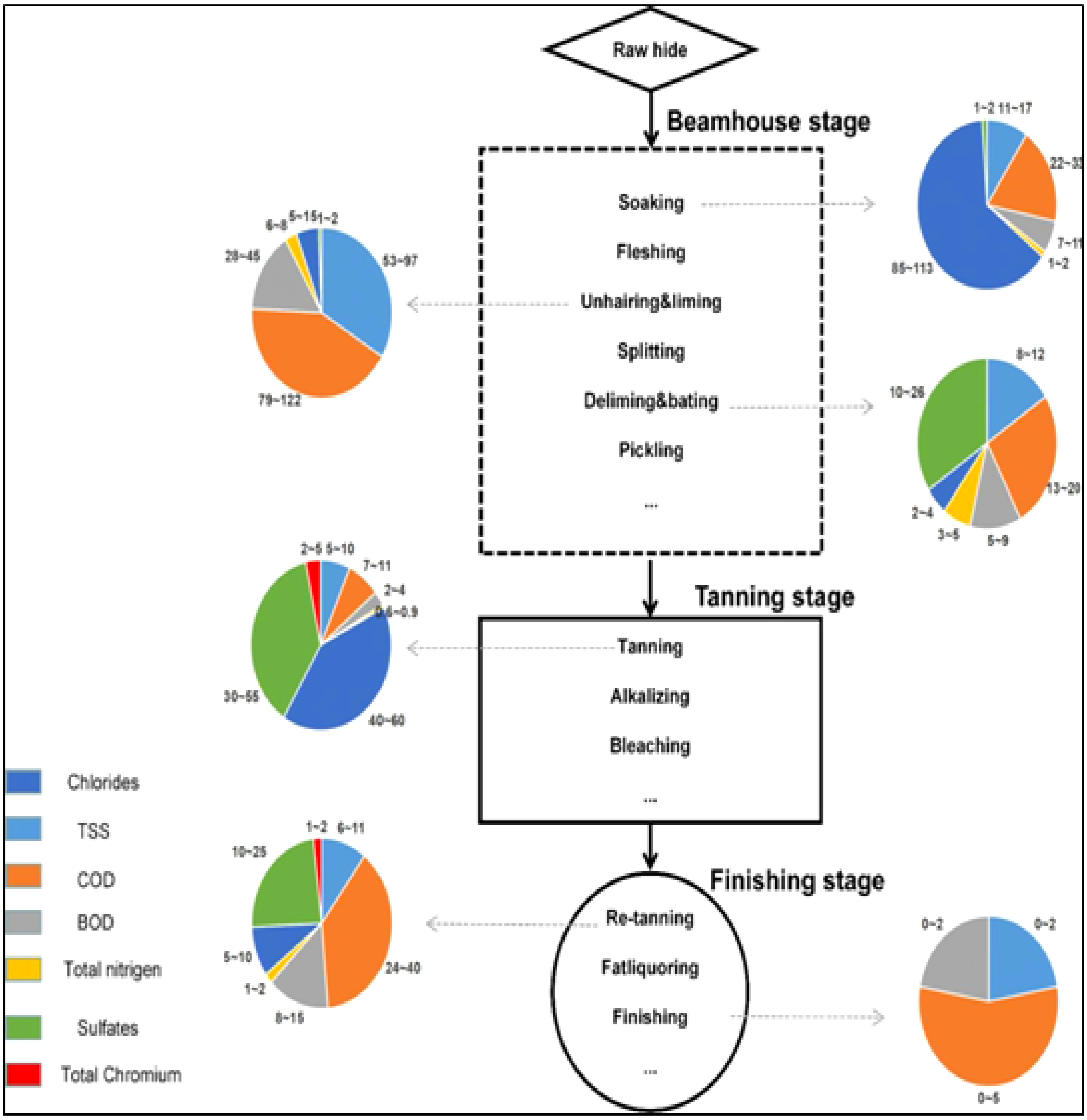

Figure 3: Schematic of leather processing and pollution profile of main processing steps (not all processes are included due to diverse product standards, and the unit of pollution load is denoted as kg per ton hide) (Zhao et al.2022)

The leather manufacturing process can be divided into three main stages: the beamhouse stage, tanning stage, and post-tanning and finishing stage, as illustrated in the figure below. Additionally, the pollution profile of the primary leather processing steps is depicted in Figure 2. Typically, the conventional wet-end process contributes to approximately 90% of the total pollution load in a tannery.

Sulphates, phenolics, chromium salts, sulphonated oils, acids, and alkali are utilized in the conversion of collagen within raw hide or skin into durable commercial leather. Consequently, tannery effluent becomes heavily contaminated, characterized by a dark brown hue and elevated levels of BOD, COD, and TDS. Moreover, significant concentrations of chromium, phenolics, and sulphates persist (10, 11). Although efforts are made to treat tannery effluents prior to discharge into water bodies, challenges remain as high levels of BOD, COD, TDS, chromium, phenolics, and sulphates persist above permissible thresholds due to the absence of effective treatment methods (12). Worldwide, it is evident that treated water fails to meet required quality standards. For instance, in certain treatments of Tannery Wastewater (TWW), Total Suspended Solids (TSS) ranged from 250 to 35,200 mg/L, BOD ranged from 250 to 2960 mg/L, and total chromium content ranged from 4.5 to 15 mg/L (13, 14, 15, 16). This highlights the pressing need for globally-improved technologies capable of efficiently managing tannery wastewater, ensuring both safety and efficacy.

1. **Flocculation and Coagulation**: Aluminum, silicon, calcium, and iron-based compounds have traditionally been employed as common inorganic flocculants and coagulants to mitigate COD, TSS, color, and the concentration of various pollutants prior to further treatment of Tannery Wastewater (TWW) (17, 18, 19, 20). Additionally, proteins and polymers have been developed as organic coagulants or flocculants (21,22). Notably, there has been increasing interest in the use of electrocoagulation (EC) for TWW treatment, with aluminum and iron electrodes being the most commonly selected options (23,24,25). In the EC process, hydroxide coagulants are generated from anode dissolution and water splitting under an applied electric field (26, 27). Ions such as NH4+, SO2−, Cr(III), etc., can be precipitated and separated from TWW towards the anodic and cathodic zones via ion exchange membrane (28). The combination of EC and ED for TWW treatment is anticipated to yield superior performance (29). Although flocculation and coagulation appear to be straightforward and effective in eliminating both organic and inorganic pollutants, they are not optimal as the primary treatment method for Tannery Wastewater (TWW). This is mainly attributed to the complex composition of TWW, which frequently requires substantial amounts of flocculants or coagulants to attain satisfactory decontamination results, resulting in considerable rises in treatment expenses. Moreover, the resulting sludge from treatment is expected to have elevated concentrations and toxicity, posing the risk of pollution transfer if not properly disposed of.

2. **Biological Treatment**: Biological degradation is widely employed in treatment plants for Tannery Wastewater (TWW) due to its cost-effectiveness and additional functionalities such as denitrification and dephosphorization. Figure 4 illustrates a typical biological treatment for TWW, involving the utilization of diverse microbes. This process encompasses various biological techniques including activated sludge digestion, biofilm production, sedimentation, coagulation, anaerobic sludge digestion, biological filtration, and others.

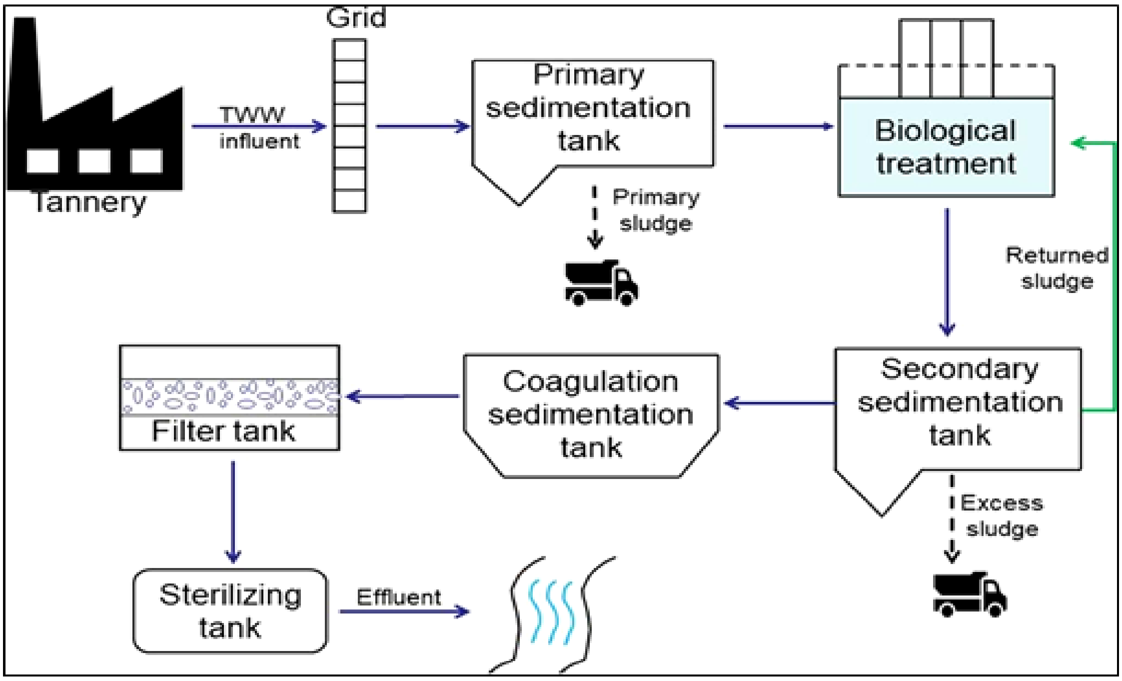

Figure 4: Flow Chart of Biological Treatment (Zhao et al.2022)

The effectiveness of biological treatments hinges on the growth and physiological activity of commonly occurring microorganisms. However, due to the presence of high salinity coupled with toxic substances, Tannery Wastewater (TWW) often exerts adverse effects on these microorganisms (30, 31). To enhance efficacy, researchers have identified and cultivated bacteria, archaea, and fungi with heightened tolerance for salinity and heavy metals (32-36). In the realm of TWW treatment, research has predominantly focused on assessing the overall detoxification efficiency of various microbial strains. For instance, a study investigated the detoxification efficiency of four fungal strains immobilized on nylon mesh, observing removal performances after 120 hours with 82.52% COD, 86.19% color, 99.92% Total Cr, and 95.91% Total Pb (37). Further investigations with similar comparative analyses have potentially expanded the range of effective microorganisms for biological treatment of Tannery Wastewater (TWW) [38-41]. Some studies have shown that significant removal of Chemical Oxygen Demand (COD), Total Chromium (Cr), and color, along with reduced biotoxicity, can be achieved in the aeration lagoons of Common Effluent Treatment Plants (CETPs) during the treatment of industrial TWW, demonstrating the capability to address challenging wastewater conditions. Specifically tailored active sludge or biofilm cultures for TWW treatment have been developed to decrease microbial acclimation time in CETPs, resulting in considerable improvements in effluent quality [42-46]. Additionally, anaerobic digestion has emerged as an alternative method for pollutant removal and bio-gas production, given the nutrient-rich composition of TWW. Technologies such as up-flow anaerobic sludge blanket reactors (UASBs), membrane bioreactors, non-aerated bio-film, and packed bed reactors have been utilized for denitrification, dephosphorization, anammox, detoxification, and biogas production through the cultivation of tolerant microbial species. Despite the inhibitory effect of Tannery Wastewater (TWW) on enzymatic reactions for anaerobic digestion, these technologies have been successfully employed [47-52]. Additionally, combined oxic-anoxic biological treatments or facultative ponds have been developed to achieve more comprehensive removal of multiple pollutants, with the optimization of treatment process configurations playing a crucial role in their full-scale application [53-55]. Constructed wetlands (CWs) are another widely utilized biological treatment for TWW, known for their energy efficiency, ease of operation, and environmental friendliness [56]. Constructed wetlands (CWs) can be viewed as miniature ecosystems comprising plants, microorganisms, and aquatic animals, where physical, chemical, and biological decontamination processes occur simultaneously [57,73]. Attention must be given to selecting tolerant plants, microorganisms, and packing materials for specific CWs that are fed with Tannery Wastewater (TWW), considering various CW types such as vertical flow, horizontal subsurface flow, and surface flow [58]. For example, a pilot-scale constructed wetland in Venezuela, planted with carefully chosen phragmites, demonstrated remarkable removal rates, including high COD (82%) and NH4+ and N removals (96%), as well as nearly complete removal of Cr from the outflow [59]. Hybrid constructed wetland systems, such as those combining horizontal subsurface flow with free water surface flow or subsurface vertical flow with horizontal and vertical flow, have also been closely examined. These systems have exhibited excellent properties for denitrification, dephosphorization, and detoxification [60,61]. A novel approach known as floating treatment wetlands (FTWs), inoculated with selected bacteria, has been developed as an alternative to conventional constructed wetlands (CWs), where plants typically grow on substrates like gravel, sand, or porous soil. This innovative method aims to achieve satisfactory improvement in effluent quality (62). Further research on the design of CWs for Tannery Wastewater (TWW) treatment has demonstrated that all types of CW systems hold promise for the removal of multiple target contaminants (63-67).

A comprehensive study has summarized practical experiences, including guidelines on plant selection, substrate choice, and operational parameters (68).Biological treatments have thus far been the most widely adopted methods for TWW treatment, despite challenges such as the difficulty in isolating and acclimating tolerant species, the time-consuming nature of the process, and issues related to non-biodegradable pollutants. Additionally, the fluctuating volume of TWW presents a significant challenge for the operation of biological treatment systems. Another emerging concern is the construction of biological treatment plants scaled for TWW treatment, which often requires significant land usage. However, future restrictions on land development are anticipated to make securing adequate land increasingly challenging.

3. **Membrane Filtration:** In the last two decades, there has been significant advancement in technologies such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). Figure 5 illustrates a simplified membrane process where contaminated water permeates membranes under propulsive forces like pressure, flow, and concentration gradient, while simultaneously various contaminants are rejected. Membrane filtration has become a key focus in treating Tannery Wastewater (TWW). Ceramic membranes are extensively employed in TWW treatment, with continuous efforts aimed at exploring more cost-effective and efficient ceramic membrane materials. The utilization of raw materials such as Boehmite [69], natural clay [70], pozzolan [71], and perlite [72, 73] for membrane production and their application in TWW treatment has been documented. Furthermore, organic membranes have been investigated. For instance, Velu et al. developed a polyether sulfone ultrafiltration membrane that achieved an 80–90% reduction in BOD and COD during TWW treatment [74].

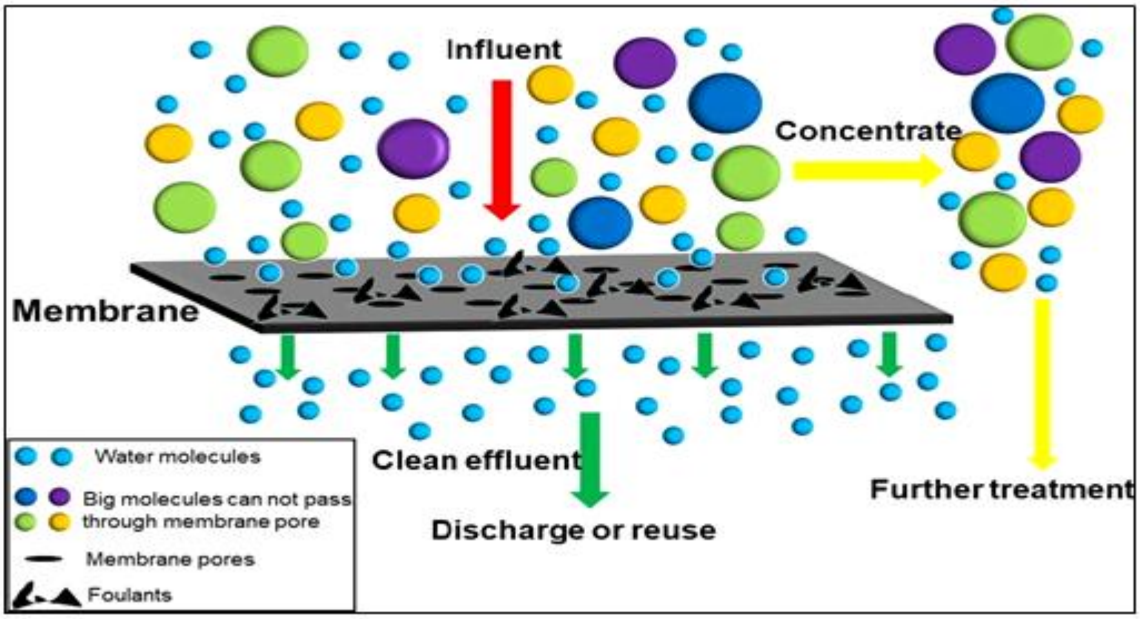

Figure 5: Simple schematic diagram of membrane filtration process (Zhao et al.2022)

Membranes equipped with adsorbents for pollutant removal have displayed impressive performance in various studies. For example, a study devised versatile layered double hydroxides (LDHs)/polyacrylonitrile (PAN) membranes, achieving over 99% removal of Cr(III) in synthetic Tannery Wastewater (TWW) [74]. Researchers have also combined nanofiltration and reverse osmosis (RO) processes, achieving around 78% permeate recovery with low Total Dissolved Solids (TDS) in a pilot plant. The reclaimed water from the membrane system was effectively reused in tannery processes [75]. Attributes such as higher selectivity and lower operating pressure are sought-after in membrane fabrication. A review introduced the concept of "loose nanofiltration," which appears promising in meeting these requirements. This type of membrane is expected to exhibit high permeation of salts and small organic molecules, rendering it suitable for resource separation/recovery and potentially valuable for treating highly saline TWW and recycling valuable substances [76]. Additionally, there has been a proposal to integrate membrane processes into specific stages of the tanning cycle instead of treating mixed TWW collectively. This approach is particularly beneficial when there is a need to recycle valuable resources and maintain strict control over effluent quality at each tanning stage [77].

4. **Electrochemical Treatment:** In recent years, electrochemical treatment of wastewater has garnered significant attention due to its effectiveness in eliminating various organic and inorganic contaminants. Indeed, electrochemical treatment has been utilized as both pre and post-treatment processes. However, its widespread application has been hindered by high energy consumption and facility costs. Considering that Tannery Wastewater (TWW) typically exhibits high salinity and contains numerous metal ions, which enhance the conductivity of the wastewater, further advancement and adoption of electrochemical treatment are anticipated.

During electrochemical treatment processes of wastewater, the removal of organic contaminants typically occurs through electrooxidation (EO). EO can be broadly classified into three categories [84]: 1. Direct electron transfer (DET), which involves the oxidation of organic pollutants directly on the anodes. While DET processes are prevalent in most EO systems, they usually do not lead to complete mineralization of pollutants and are therefore considered to have a minimal impact on pollutant degradation. However, certain contaminants, such as perfluorinated compounds, have been observed to decompose efficiently only when DET is the initial step [85, 86]. Mediated oxidation (MO) relies on the catalytic formation of anodic reactive oxidant species (ROS) to decompose contaminants.

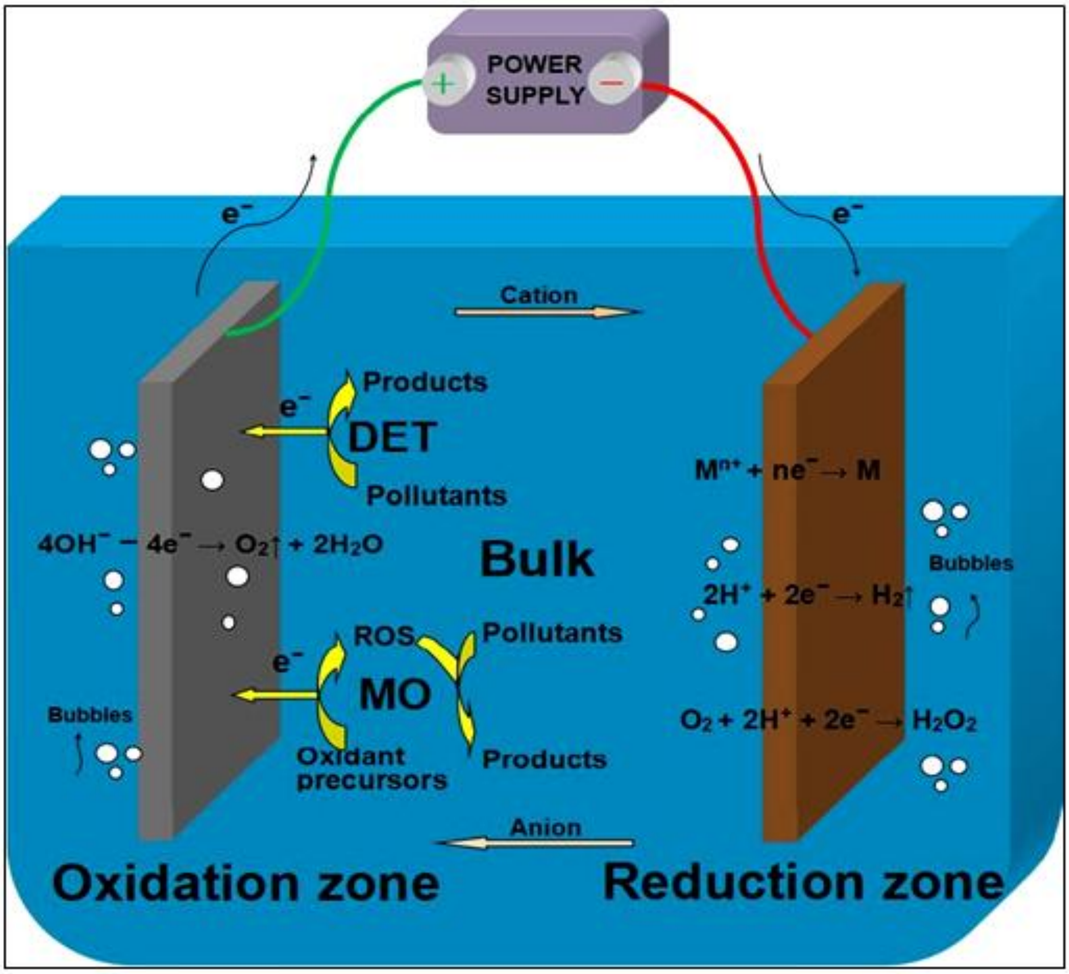

Figure 6: Schematic of a typical electrochemical reactor. DET direct electron transfer, MO mediated oxidation, ROS reactive oxidant species, Mn+ metal ions. (Zhao et al.2022)

III. PROBLEMS TO BE SOLVED AND POTENTIAL SOLUTIONS

Although the treatment methods previously discussed significantly improve the quality of effluent from tanneries, thus reducing their detrimental environmental impact, Tannery Wastewater (TWW) treatment plants still face challenges. The most pressing and contentious issues revolve around chromium (Cr) pollution and high salinity. Efforts are made to minimize the Cr content in the effluent of TWW treatment plants. However, the primary concern arises from the treatment by-products generated through various advanced segregation methods. By-products, such as Cr-containing sludge, foulants, and adsorbents, pose significant hazards and are often inadequately managed. To cut costs, these by-products are typically disposed of in landfills, which can lead to serious pollution transfer. Recycling or reusing these by-products is a preferable approach, considering the value of Cr for industrial production. However, it must be acknowledged that identifying facile and cost-effective recycling or reusing methods is challenging, especially given their reliance on the specific methods adopted in TWW treatments. Rather than engaging in labor-intensive efforts to eliminate Cr pollution during wastewater treatment processes, it is argued that reducing discharge at the source is a more logical and cost-effective approach.

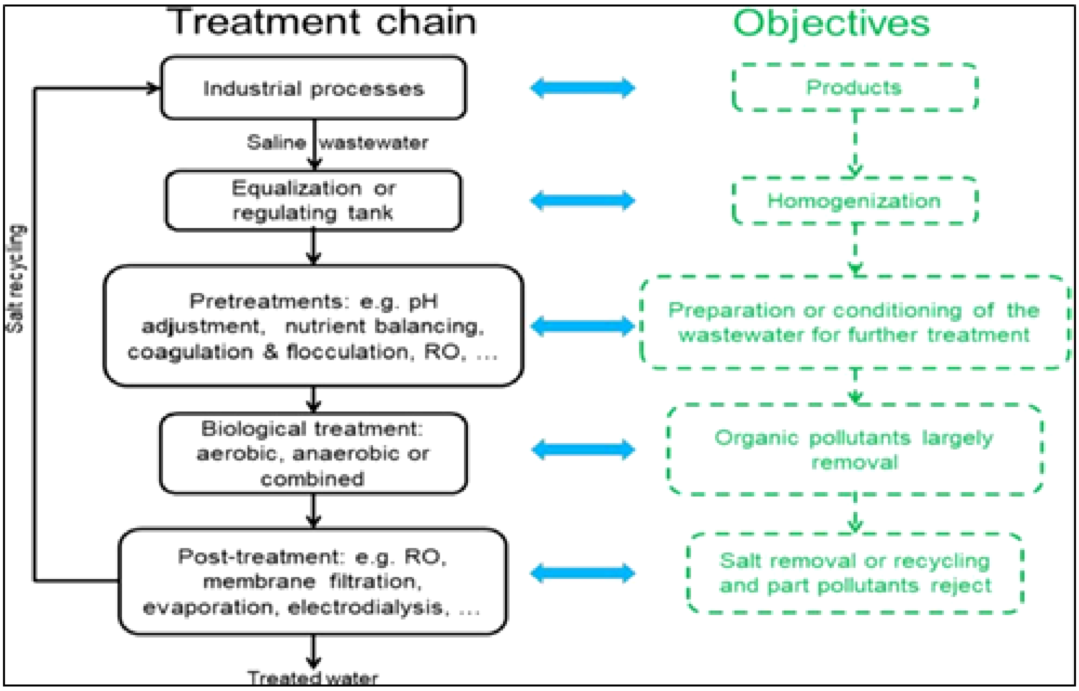

Figure 7: Simplified diagram of wastewater treatment and desalination chain (biological treatment + RO), adapted from [83] (Zhao et al.2022)

Consequently, the adoption of a cleaner production strategy in tanning operations has been proposed as a promising approach. This strategy aims to minimize or entirely eliminate chromium (Cr) discharge into effluent, thereby mitigating a range of challenges related to Cr disposal in subsequent wastewater treatment processes. Examples of cleaner production strategies include enhancing Cr uptake during the tanning process, termed as "Cr exhaustion" [78], employing innovative Cr tanning agent carriers [79], utilizing Cr-free syntans [80], embracing non-chrome metal tanning agents, and incorporating chrome-free organic tanning agents [81-83]. These low-risk strategies have the potential to alleviate the burden of Cr segregation in wastewater treatment processes and may even eliminate Cr pollution at its source.

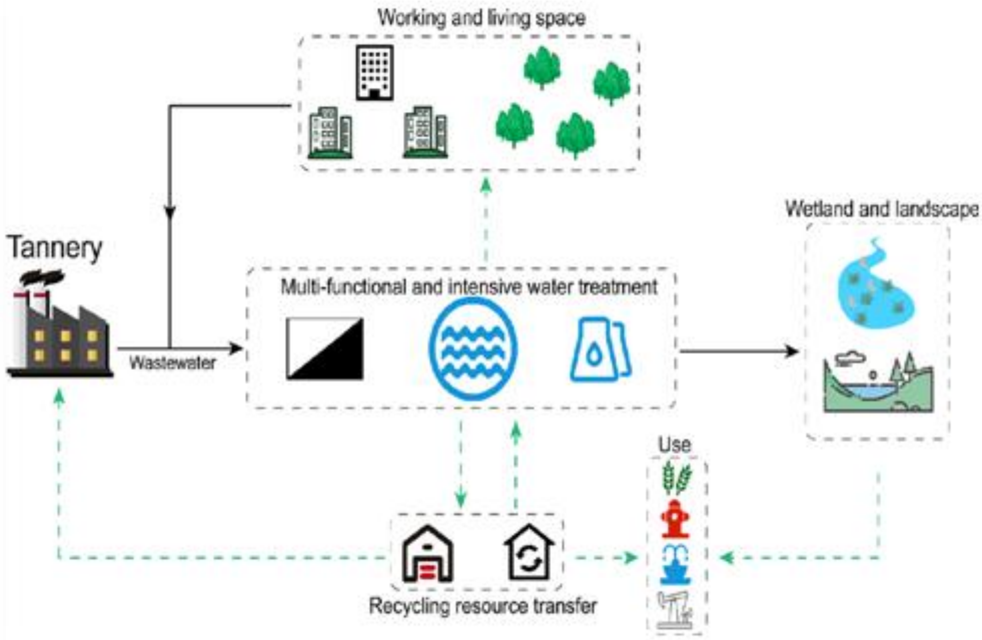

Figure 7: Proposed plan for TWW treatment in the future (Zhao et al.2022)

IV. TWW TREATMENT FOR THE FUTURE

Presently, the challenges facing Tannery Wastewater (TWW) treatment extend beyond the removal of harmful pollutants to include the need for cost-effectiveness. As previously discussed, individual methods alone cannot achieve complete decontamination of TWW while also being cost-saving. Hence, wastewater treatment plants should carefully consider recommending integrated or combined methods. For example, combining Advanced Oxidation Processes (AOPs) with the activated sludge process could enhance the biodegradability of wastewater, potentially reducing operation time and pollution load in the aeration tank, thereby increasing treatment capacity. Similarly, the combination of electrocoagulation with electrooxidation can degrade organic pollutants and precipitate inorganic pollutants simultaneously, simplifying TWW treatment steps. Furthermore, future TWW treatment plants should consider resource recycling, given the global trend towards "Carbon Neutrality." rephrase the following paragraph without changing its technical terms and meaning in the field of wastewater treatment, there is a growing emphasis not only on optimizing and upgrading treatment processes, but also on promoting extensive recycling and reuse of valuable resources. The substantial quantities of recycled water, salts, metals, etc., recovered from Tannery Wastewater (TWW), have the potential to generate economic returns, thereby contributing to the long-term self-sustainability of treatment plants. Additionally, with the increasing demand for intensified land use in many countries, future wastewater treatment plants should be designed to be as compact as possible. This entails a reduction in the popularity of step-by-step treatment processes involving space division, particularly as land use regulations become more stringent. Consequently, there is a greater need for the advancement of physical and chemical treatment methods such as membrane filtration, electrochemical oxidation, etc. These methods are expected to play a more significant role in the future, given their higher hourly treatment capacities compared to conventional biological treatment, thereby facilitating a significant reduction in the land footprint of treatment plants.

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