# Bioremediation of Heavy Metals in Tannery Effluents

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## Abstract:

Human Health has been severely impaired in addition to causing various environmental problems, due to the mismanagement of various waste especially waste water produced by tanning processes. Terminal treatment is still in dominant condition, even after putting in constant efforts to alleviate the pollution of Tannery Wastewater (TWW). This Chapter is related to a brief discussion on main methods of TWW Treatment such as coagulation and flocculation, biological treatment, adsorption, membrane filtration, advanced oxidation process. Electrochemical Treatment has been discussed specifically due to it's better performance as well as environment friendliness and high efficiency. Integrated or combined treatment methods with better performance and multifunction may be recommended considering the harsh physico-chemical conditions of Tannery Wastewater. However the fact still prevails that comprehensive studies on escalation of methods combination and cost-effectiveness are needed. It has been tried to highlight in this work that the residue Cr in treated sludge and high salinity in effluent still remains, and feasible solutions related to that has been provided. The perspective of realizing multi-function, recycling, and intensification should be the developing direction of future TWW treatment.

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

## Introduction and Background:

Wastes are generated by all sectors of our society like industries, agriculture, transportation, construction and consumers. Discarded pollutants consist of wastes. Different processes generate industrial wastes, and the amount as well as toxicity of waste released varies with its own specific industrial process. It has been studied that microorganisms processes great potential to control environmental pollution, especially the water pollution caused by industries. It has been studied that in the present scenario leather industries have been the most polluting sources, and results in imparting negative impacts in the environment due to the pollution caused by it.

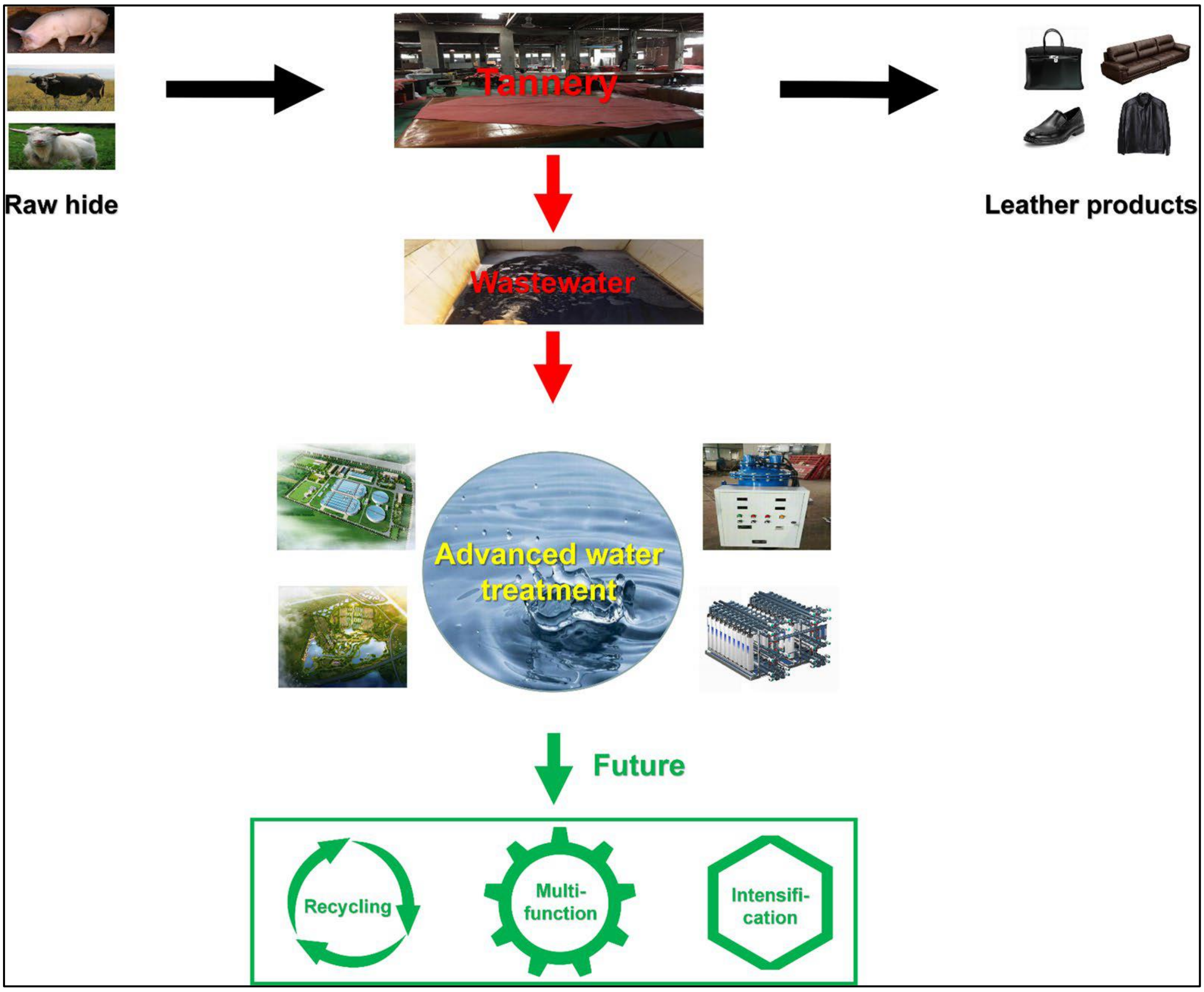


**Figure 1: Schematic representation of the tanning process and TWW treatment (**Zhao et al.2022)

Soil and water pollution are caused due to the discharge of tannery effluents in land and water respectively. Aquatic environment is adversely effected by the discharge of heavy metals, especially tannery effluents, in-turn causing negative and everlasting impact on the nearby flora and fauna, both in terrestrial and aquatic region.

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)
Among all effluents discharged from tanneries, chromium is one of the most hazardous pollutants (3). The compounds of Chromium can cause mutations leading to cancer, also hinder enzymes and nucleic acid biosynthesis. (4). It can also cause diarrhoea, eye irritation, kidney failure, ulcers, skin and lung cancer. (5)

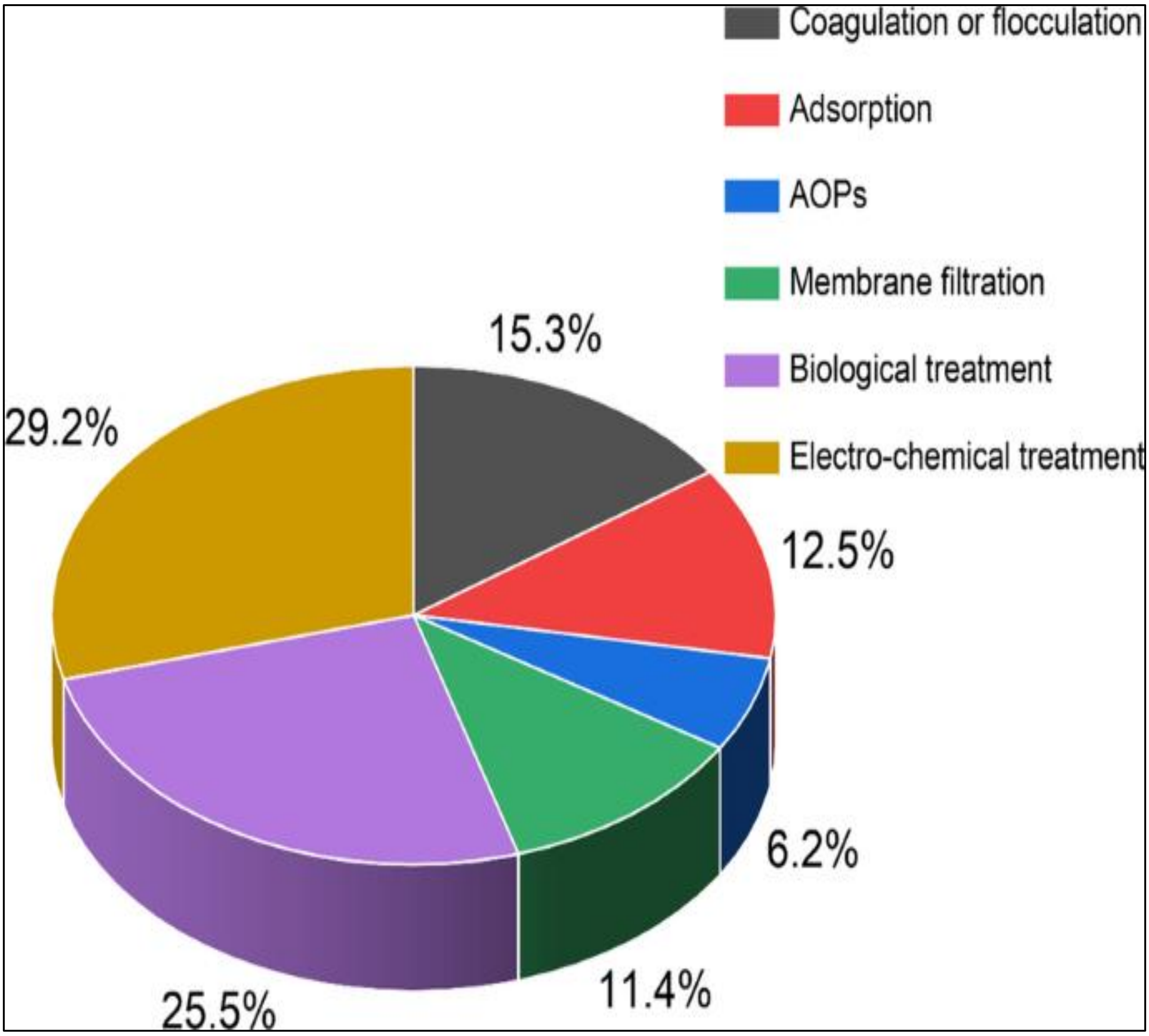


**Figure 2: Proportions of various methods utilised in TWW treatment (**Zhao et al.2022)

The treatment of wastewater involves several conventional methods such as advanced oxidation processes, membrane filtration, electrocoagulation, electrochemical methods, ozonation, several adsorbents, and precipitation. Yet the process of precipitation in less preferred due to its cost affectivity and reason for causing secondary pollution. It can thus be considered that various physical as well as chemical or both methods are being used for the treatment of tannery effluents. Although these integrated methods can be considered to be efficient but they, may not be considered as cost effective in terms of both energy anf chemical consumption, also these methods produce a large quantity of sludge, which renders problematic waste disposal. (6) To find a solution to this problem, there have been several trials to check the efficiency of biological methods in tannery waste water treatments. 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 species of bacteria have been found effective for the bioremediation of tannery wastes, like, *Pseudomonas, Arthrobacter, and Bascillus*. (7,8). As micro-organisms are gifted with enzymes system for the oxidation of organic ingredients, therefore, Bioremediation is an extremely cost effective, efficient and eco-friendly strategy for the reduction of pollutants present in any effluents. Chromium remediation studies have been carried out with a variety of organisms like, *Pseudomonas species, Aeromonas species, Bascillus species, Micrococcus species, and Microbacterium species*. (9) Earlier studies containing some Common Effluent Treatment Plant (CETP) showed the presence of Cr (IV) exceeding the limiting concentration. Studies have proved that if effluents are treated by a consortium of Sulphate Reducing Bacteria (SRBs) (*Desulphovibrio desulfuricans, D. vulgaris, and D. gigas*), that utilize Cr(VI) as an electron acceptor for sulphate reduction; that both Cr(IV) and sulphates get removed frpm the broth. (Singh, Rajesh & Kumar, Anil & Kirrolia, Anita & Kumar, Rajender & Yadav, Neeru & Bishnoi, Narsi & Lohchab, Rajesh. 2010).

## Characteristics of Tannery Waste Water

The entire leather manufacturing process can be basically divided into three parts: beamhouse stage, tanning stage, and post–tanning and finishing stage illustrated in Figure below. Figure 2 also demonstrates the pollution profile of main leather processing steps, and the conventional wet-end process usually accounts for nearly 90% of the total pollution load in a tannery.

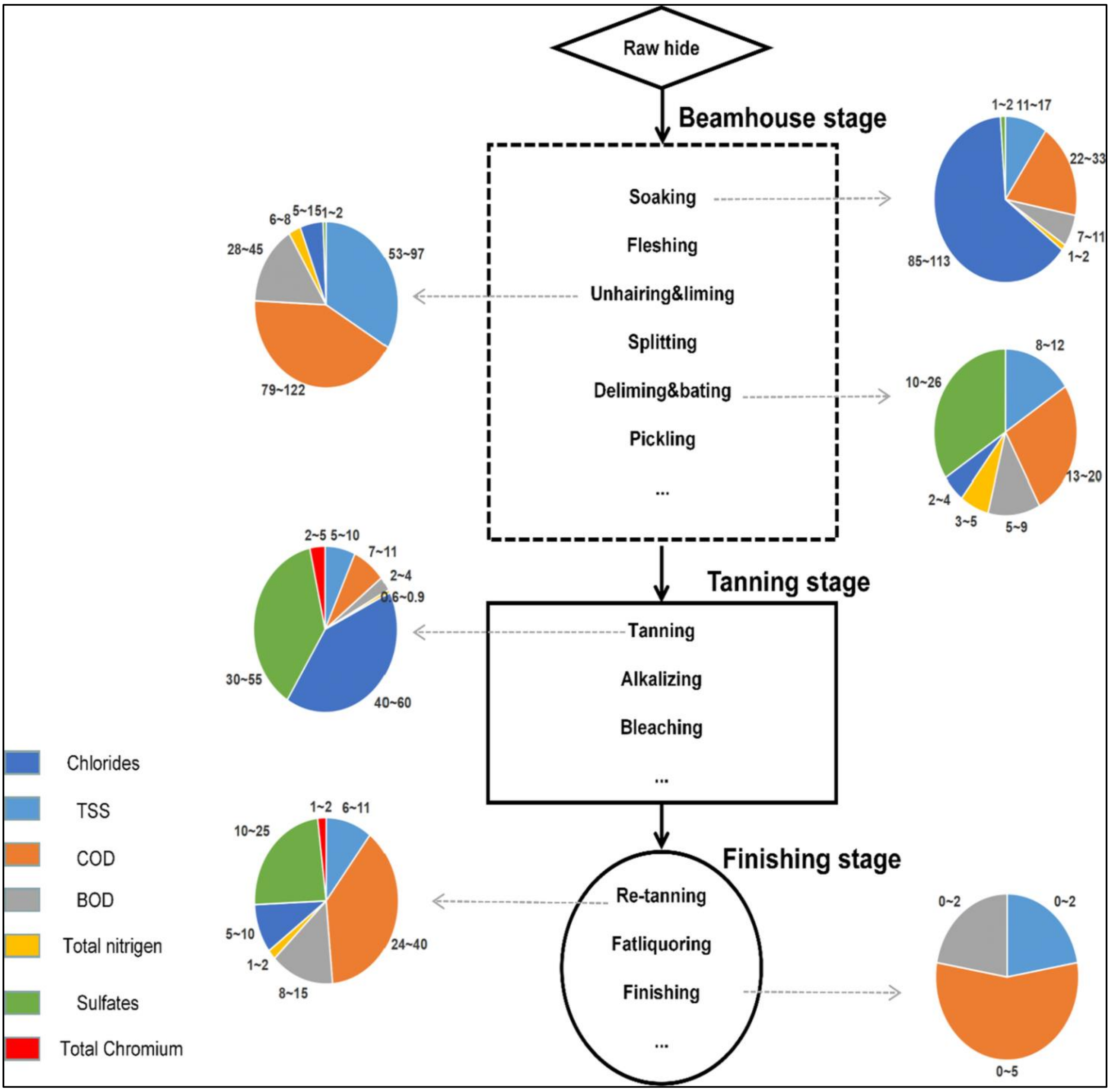


**Figure 3: Schematic of leather processing and pollution profile of main processing steps (not all processes are included due to diverse product standards, and the unit of pollution load is denoted as kg per ton hide) (Zhao et al.2022)**

Sulphates, phenolics, chromium salts, sulphonated oils, acids & alkali are used to convert collagen in raw hide or skin into long lasting commercial leather. The hide or skins immobilize the leather products and the tannery effluent is highly contaminated which is dark brown in color and high BOD, COD, TDS values. Also high concentrations of chromium, phenolics, and sulphates are also found. [10, 11]. The tannery effluents are being treated before releasing it into water bodies but high BOD, COD, TDS values are found and chromium, phenolics and sulphates are still above the stipulated values because of lack of productive treatment method [12]. Around the world it has been observed that treated water could not meet the required quality parameters. For example TSS content ranging from 250 to 35,200 mg/L, BOD content ranging from 250 to 2960 mg/L, total Cr content ranging from 4.5 to 15 mg/L were still detected in some TWW treatments [13,14,15,16]. The fact should be highly noticed that there still exist some plants without enough safety or efficiency in handling TWW, leading to the urgent need for advanced technologies with better performance worldwide.

**Flocculation and coagulation**: Aluminium, silicon, calcium, iron-based compounds were widely used as typical inorganic flocculants and coagulants to reduce COD, TSS, colority and the concentration of many pollutants before further TWW treatment [17, 18, 19, 20]. Proteins, polymers were also developed as organic coagulant or flocculant [21,22]. It is noteworthy that electrocoagulation (EC) process has been raising interest for TWW treatment, in which aluminium and iron electrodes were most extensively selected [23,24,25]. During EC, hydroxide coagulants are formed from anode dissolution and water splitting under an applied electric field [26, 27]. Ions such as $NH_4^+$, $SO_4^{2-}$, Cr (III), etc. can precipitated and segregated from TWW towards anodic and cathodic zones via ion exchange membrane [28]. Combination of EC and ED for treatment of TWW with better performance would be expected [29]. Though it looks facile and have the effect of removing both organic and inorganic pollutants, flocculation and coagulation is not suitable to serve as the main process for TWW treatment. The reason is clear: the harsh quality of TWW usually requires massive addition of flocculants or coagulants to obtain a relatively satisfactory decontamination performance, however this would make the treatment cost increase a lot. Furthermore, the treated sludge is supposed to characterize with high concentration and high toxicity, and might cause pollution transfer without proper disposal methods.

**Biological Treatment**: Biological degradation is extensively used in TWW treatment plants, since it reduces the cost and also has functions like denitrification and dephosphorization. One typical biological treatment for TWW could be seen in Fig. 4. It is a process where diverse microbes are use. The biological processes involve techniques such as activated sludge digestion, biofilm production, sedimentation, coagulation, anaerobic sludge digestion, biological filtration etc.

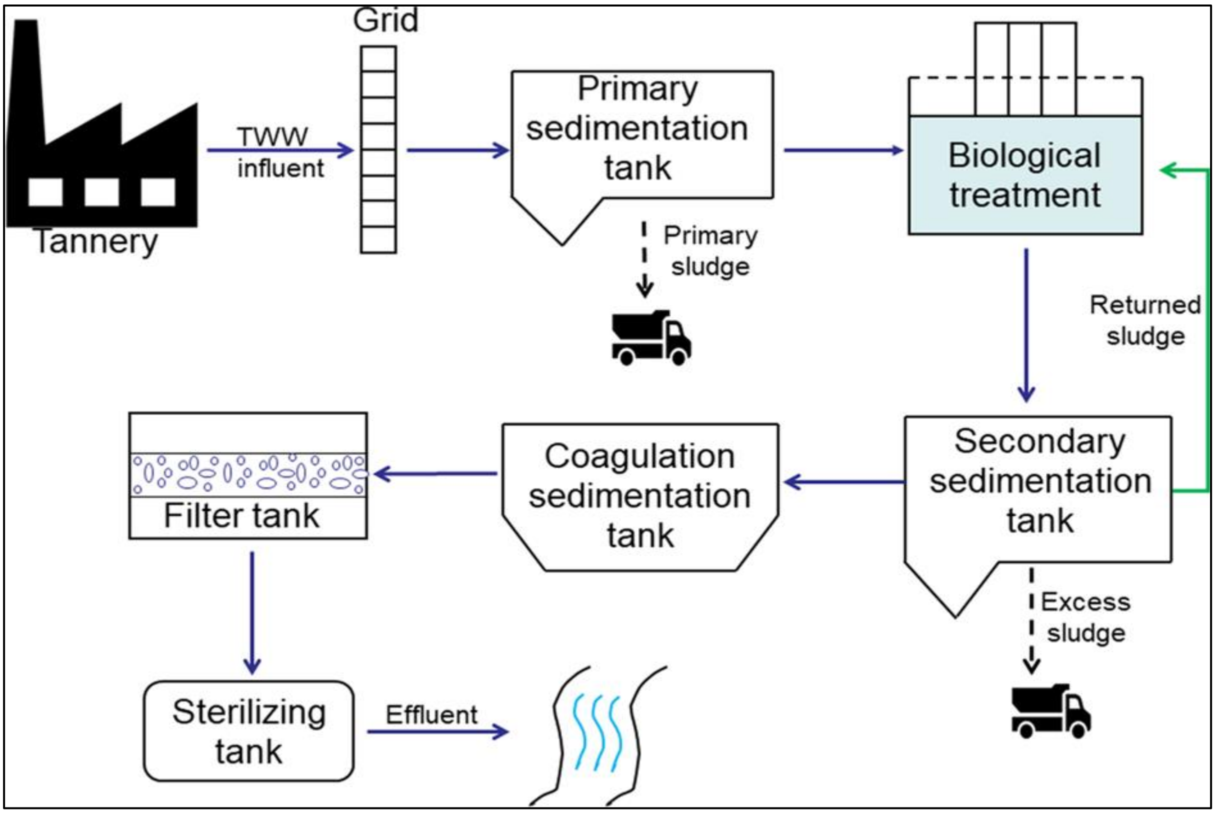


**Figure 4: Flow Chart of Biological Treatment (**Zhao et al.2022)

The efficiency of biological treatments depends upon the growth and physiological activity of common microorganisms. Because of high salinity with toxic substances, TWW often has adverse effects on common micro-organisms [30, 31]. To improve the efficacy, researchers identified and cultured some bacteria, archeae, fungi which have high tolerance for salinity and heavy metals [32-36]. In TWW treatment, research mostly concentrated on the overall detoxification efficiency of diverse microbes. A study compared detoxification efficiency of four fungal strains immobilized on nylon mesh, and after 120 h, removal performance with 82.52% COD, 86.19% color, 99.92% Total Cr, 95.91% Total Pb were observed [37]. Other studies with similar comparisons potentially offered more options of effective micro-organisms for TWW biological treatment [38-41]. Some investigations demonstrated that COD, Total Cr, color could be largely removed together with reduced biotoxicity in aeration lagoons of common effluent treatment plants (CETPs) during the treatment of industrial TWW, providing the ability to address harsh wastewater. The special active sludge or bio-film dedicated for treating TWW were cultured with the purpose of decrement of microbe,s acclimation time in CETPs, so that the quality of effluent got considerable improvement (42-46). Anaerobic digestion becomes an alternative way for pollutant removal together with bio-gas production as TWW contains abundant nutrient substance. Technologies such as up-flow anaerobic sludge blanket reactors (UASBs), membrane bioreactors, non-aerated bio-film, and packed bed reactor were thereby exploited for denitrification, dephosphorization, anammox, detoxification, and bio-gas production through cultivation of tolerant microbial species, despite the fact that TWW generally has an inhibition effect on enzymatic reactions for anaerobic digestion [47-52]. Combined oxic-anoxic biological treatments or facultative ponds were also developed for a better and multi-target pollutants removal, where the optimization of oxic-anoxic treatment process’s configuration played an important role for full scale application [53-55]. Another biological treatment widely utilized for TWW treatment is constructed wetlands (CWs), which is characterized with energy saving, handy operation and environmentally sound [56]. CWs may be considered as some small ecosystems consisting of plants, microorganisms, aquatic animals, where physical, chemical, biological decontamination processes co-exist [57,73]. Same attention must be paid to tolerant plants as well as micro-organisms and packing stuffs for certain CW fed with TWW, as CW's types such as vertical flow, horizontal subsurface flow, surface flow etc. (58). A pilot-scale constructed wetland planted with specially choosing phragmites in Venezuela showed high COD (82%) and $NH4^{+}$, N removals (96%), and almost complete Cr removal in the outflow [59]. Hybrid constructed wetland systems, i.e., horizontal subsurface flow combined with free water surface flow or subsurface vertical flow combined with horizontal flow and vertical flow were also under careful inspection, which exhibited the excellent properties for denitrification, dephosphorization, and detoxification [60,61]. A novel floating treatment wetlands (FTWs), inculated with selected bacteria was also designed, varying from the conventional CWs with plants growing on gravel, sand, porous soil etc., to achieve satisfying amelioratiob of effluent quality (62). More studies about designing CWs for TWW treatment have shown that all the CWs system exhibit promising prospect for multiple target contaminant removal (63-67). Summary of one substantial work contains practise experience such as how to select plants, substrate, operation load etc. (68) Til now biological treatments have been the most extensively adopted methods for TWW treatment, still problems such as inconvenient isolation and acclimation of tolerant species, time consuming and non-biodegradable pollutants also caused problems. In addition, the sudden changes in TWW volume are another great challenge for the operation of biological treatments. One another issue also gradually emerged nowadays: construction of a biological treatment plant that has TWW wastewater treatment scale usually requires a certain land occupation, however this would be more and more difficult because limit of land exploitation in the future would become stricter.

**Membrane filtration:**

Technologies of microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) have been actively developed in the past two decades. A simplifed membrane process is illustrated in Fig. 5. Polluted water permeates membranes under propulsive forces such as pressure, flow, concentration gradient, etc. with various contaminants rejected simultaneously. Membrane filtration has been a hot spot domain for TWW treatment. Ceramic membranes are very commonly used in TWW treatment, and efforts to explore cheaper and more efficient ceramic membrane materials have been taking all the time. Boehmite [69], natural clay [70], pozzolan [71], perlite [72, 73] as raw materials for manufacturing and utilization of membranes, in TWW treatment have been reported. Organic membranes were also another kind of materials under inspection. Velu et al. devised the polyether sulfone ultrafiltration membrane that achieved 80–90% reduction in BOD and COD in TWW treatment [74].

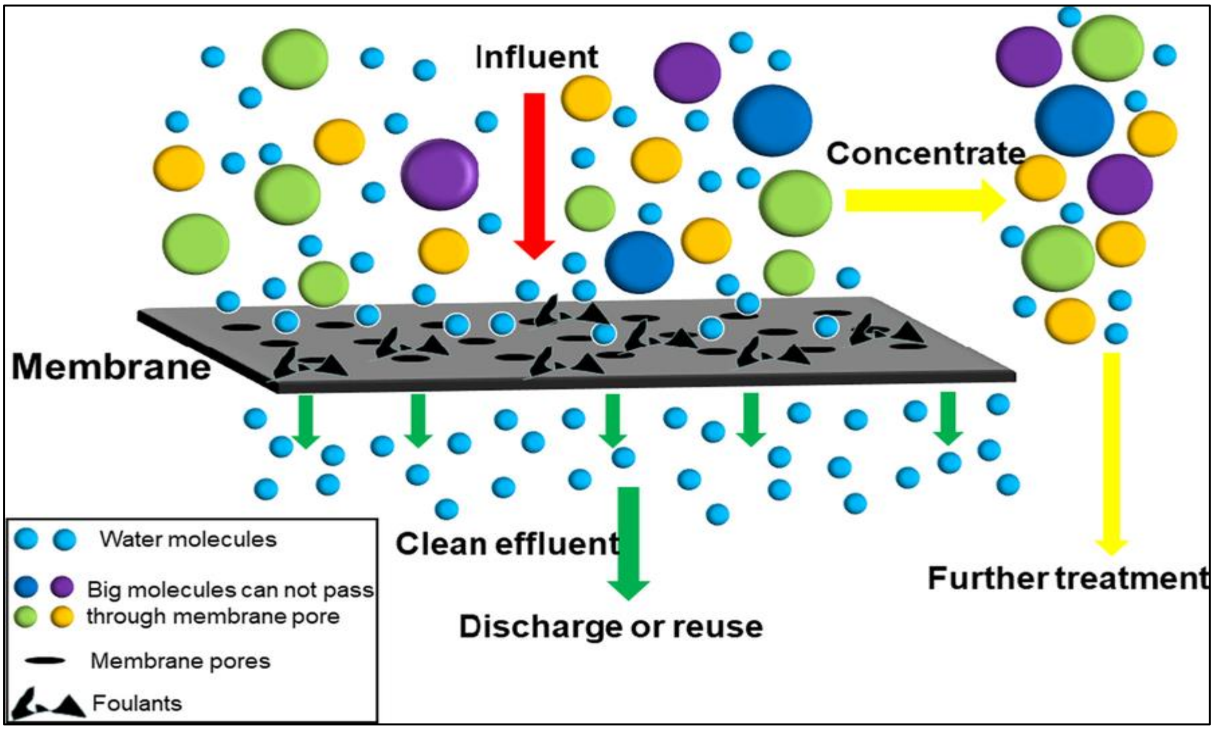


**Figure 5: Simple schematic diagram of membrane filtration process (**Zhao et al.2022)

Membranes carrying adsorbents for pollutants removal showed outstanding performance in several studies. One versatile layered double hydroxides (LDHs)/polyacrylonitrile (PAN) membranes were smartly weaved, which achieved more than 99% Cr (III) removal in synthetic TWW [74]. Researchers also combined nano-filtration and RO processes, where about 78% permeate recovery with low TDS was achieved in one pilot plant, and the water recovered from the membrane system was successfully reused for tannery process [75]. Higher selectivity and lower operating pressure are ideal characteristics for membrane fabrication; a review briefly introduced the novel "loose nanofiltration" appearing to satisfy this. This kind of membrane is supposed to possess high permeation of salts and small organic molecules, which therefore could be used for resource separation / recovery and may be potential for highly saline TWW treatment and valuable substances recycling [76]. Membrane process integrated in some steps of tanning cycle instead of together treatment of mixed TWW was put forward, especially when valuable resources needed to be recycled and the quality of effluent in each tanning step was under strict control [77].

**Electrochemical treatment:**

Electrochemical treatment of wastewater has been an active research spot in recent years due to its prominent performance in removal of a range of organic and inorganic contaminants. In fact, electrochemical treatment of wastewater has already been used as pre or post treatment process, however the high energy consuming and facilities cost inhibited its scale-up application. Given that TWW usually contains high salinity and much metal ions that could improve the conductivity of the wastewater to be treated, further promotion of electrochemical treatment is to be expected.

Organic contaminants are usually removed via electrooxidation(EO) during electrochemical treatment processes of wastewater. EO could be basically classified to three categories [84]: (1) One is direct electron transfer (DET) of organic pollutants adsorbed on anodes. The DET process could occur in most EO systems, but generally could not serve as the final mineralization approach hence considered negligible impact on pollutants' degradation. Nevertheless, some special contaminants such as perfluorinated compounds were proved to be efficiently decomposed only when DET as the trigger step [85,86]. Mediated oxidation (MO), which depends on anodic reactive oxidant species (ROS) catalytic formation to decompose contaminants.

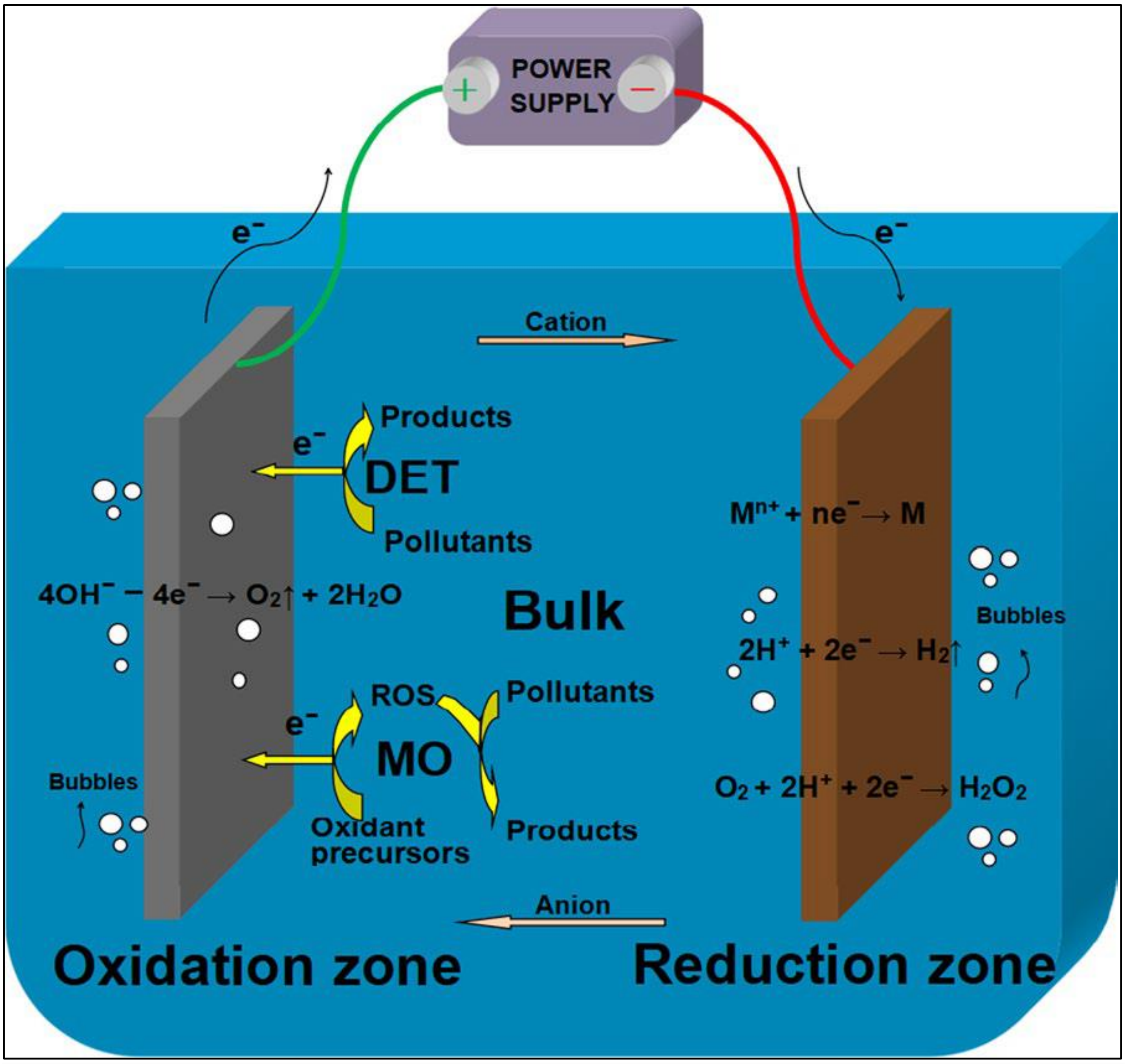


**Figure 6: Schematic of a typical electrochemical reactor. DET direct electron transfer, MO mediated oxidation, ROS reactive oxidant species, Mn+ metal ions. (**Zhao et al.2022)

**Problems to be solved and potential solutions:**

Even though these treatment methods discussed before could greatly ameliorate the quality of effluent from a tannery hence greatly reduce its harmful environmental impact, there still remains some problems baffling TWW treatment plants. The most concerned and controversial problems are Cr pollution and high salinity. The content of Cr in effluent of TWW treatment plants is reduced as much as possible, however the real threat

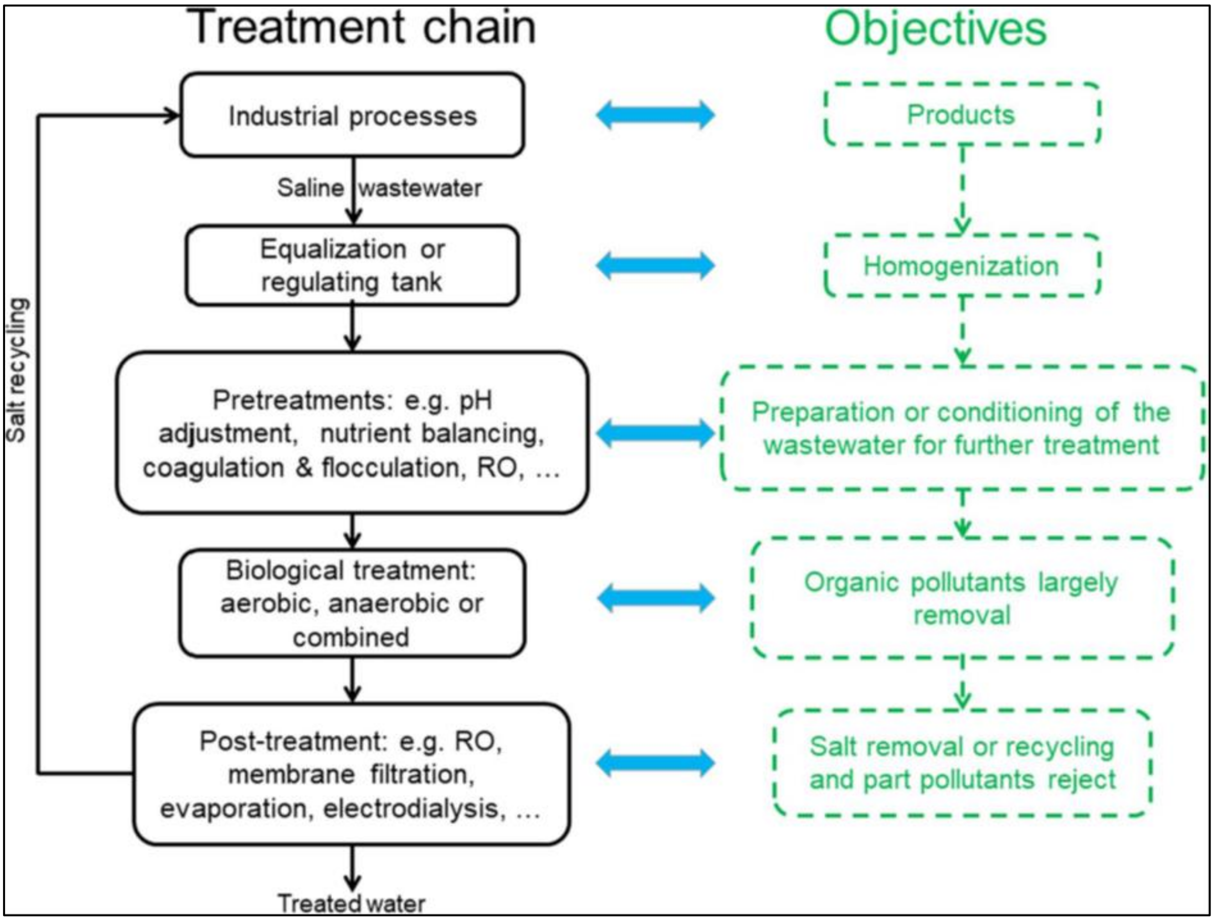


**Figure 7: Simplified diagram of wastewater treatment and desalination chain (biological treatment + RO), adapted from [83] (**Zhao et al.2022)

comes from the treatment by-products through various advanced segregation methods. By-products after wastewater treatment, including Cr-containing sludge, foulants, adsorbents, etc. are extremely hazardous yet usually in poor management. To save cost, these by-products are always transferred to landfill treatment, which might cause serious pollution transfer. Compared to conventional landfill, recycling or reuse of these by-products is a rather desirable way as Cr has much value for industrial production. Nevertheless, it should be acknowledged that the facile and cost-effective recycling or reusing methods seems hard to find, even worse is that these methods are highly dependent on the adopted methods in TWW treatments. Rather than the laborious trials to eliminate the Cr pollution in wastewater treatment processes, we believe that reducing discharge at sources is more reasonable and cost-effective.

Therefore, cleaner production strategy in tanning processes has been proposed as one more promising way as it could reduce even eradicate Cr discharge to effluent so that a series of tough problems concerning Cr disposal in subsequent wastewater treatment processes could be solved. Cleaner production strategy such as enhancing Cr uptake in tanning process or "Cr exhaustion" [78], novel Cr tanning agent carriers [79], Cr-free syntans [80], non-chrome metal tanning agents and chrome free organic tanning agents [81-83], etc. are good examples, and these low-risk strategies could relieve the pressure from Cr segregation in wastewater treatment processes even eliminate Cr pollution from root cause.

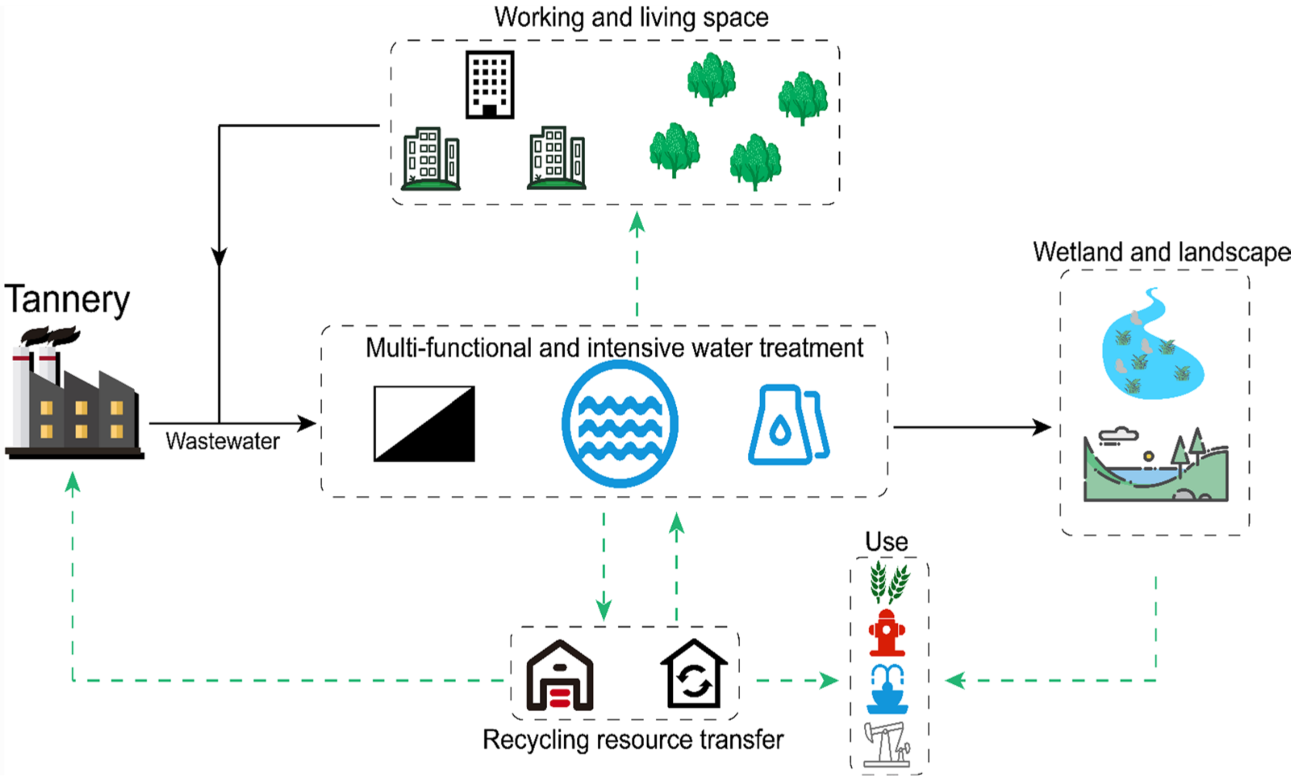


**Figure 7: Proposed plan for TWW treatment in the future (**Zhao et al.2022)

**TWW treatment for the future:**

The challenges for TWW treatment nowadays lie not only in noxious pollutants removal, but also in balance of cost-effectiveness. As mentioned before one single method we discussed in former sections could not achieve TWW totally decontamination as well as cost saving, integrated or combined methods should be carefully recommended in wastewater treatment plants. For instance, AOPs treatment combined with active sludge process could enhance the bio-degradability of wastewater, which might reduce the operation time and pollution load in aeration tank therefore increasing treatment capacity in one sense. Electrocoagulation combined with electrooxidation could simultaneously degrade organic pollutants and precipitate inorganic pollutants, which might be used for TWW treatment steps simplification. In addition, resources recycling would be considered in the future TWW treatment plants since the concept of “Carbon Neutrality” has become a general trend worldwide. Specific to wastewater treatment field, not only the optimization as well as upgradation of treatment processes, but also valuable resources recycling as well as reusing are supposed to be extensively advocated. The considerable amount of recycled water, salts, metal, etc. separated from TWW could bring economic income hence contributing to treatment plant’s long-term self-sustaining operation. What’s more, as the requirement of land use intensification prevails in many countries, wastewater treatment plant should be constructed as compact as possible in the future. This means that step by step treatment processes of space division would be less and less popular, especially when land use permission gets stricter. The physical and chemical treatment methods such as membrane filtration, electrochemical oxidation, etc. should be therefore well developed and play a more important role in the future, since the hourly treatment capacity of these methods could be enough large compared to conventional biological treatment so that the land occupation of treatment plants could be greatly reduced.

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