Treatment of Chrome Tannery Wastewater by Biological Process - A Mini Review

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Abstract—Chrome tannery wastewater causes serious environmental hazard due to its high pollution potential. As a result, rigorous treatment is necessary for abatement of pollution from this type of wastewater. There are many research studies on chrome tannery wastewater treatment in the field of physical, chemical, and biological methods. In general, biological treatment process is found ineffective for direct application because of adverse effects by toxic chromium, sulphide, chloride etc. However, biological methods were employed mainly for a few sub processes generating significant amount of organic matter and without chromium, chlorides etc. In this context the present paper reviews the characteristics feature and pollution potential of wastewater generated from chrome tannery units and treatment of the same. The different biological processes used earlier and their chronological development for treatment of the chrome tannery wastewater are thoroughly reviewed in this paper. In this regard, the scope of hybrid bioreactor - an advanced technology option has also been explored, as this kind of treatment is well suited for the wastewater having inhibitory substances.

Keywords—Composite tannery wastewater, biological treatment, Hybrid bioreactor, Organic removal.

I. INTRODUCTION

With the rapid industrialization coupled with urbanization and population growth, environmental pollution becomes a crucial problem around the world. Therefore, it is extremely necessary today to check the environmental pollution, caused by the industrial activity up to a permissible limit. Wastewater produced by chrome tannery has a great potential for environmental pollution in many ways. Among all the industries, leather manufacturing is water intensive and therefore it is critically called water polluting industry [43], [25], [31]. Unlike the other industries at present environmental regulation is very strict in major leather producing countries and extensive treatment of the effluent is very much essential [16]. Two methods are mainly used for tanning of raw Hide/Skin vegetable tanning and chrome tanning, but chrome tanning is followed in about 80% cases [37]. Generally, chrome tanning process is carried out in a sequential batch mode in Drums and Paddles [41], [32].

The process is divided into four distinct divisions, i.e. beam house, tannining yard, post-tanning, and finishing operations. Various wastewater streams are generated from different operations like soaking, liming & unhairing, fleshing, deliming & bating, and pickling & tanning. The wastewater is generated under batch mode showing variable characteristics and differential treatability [32]. Most of the wastewater streams from chrome tanning operation contain significant amount of organic substances, making them amenable for biological treatment. In contrary, wastewaters from the rest sources contribute large quantity of inorganic substances, necessitating for physical and/or chemical treatment. In view of disintegrated nature of chrome tanning industry, segregation of individual waste streams is not feasible and not economically viable. As a result, the organics laden combined wastewater is contaminated with inorganic species like chloride, sulphide, chromium etc., which are inhibitory for the microbial growth. Hence, the performance of the biological system does not reach to the satisfactory level. On perceiving this critical issue, the present paper overviewed the status of biological treatment in case of chrome tannery wastewater.

II. CHROME TANNING PROCESS AND ITS WASTEWATER GENERATION

The first operation of the tannery unit is beam house process by which the Hides/ skins are dressed for tanning. It is divided into different sub-processes consisting of soaking, liming & unhairing, deliming, and bating. Soaking is intended for the removal of the salt, impurities etc. and for increasing the moisture content, which is lost during the curing operation. Liming & unhairing is accomplished for removal of epidermis layer and inter-fibre substances (mainly soluble protein and fats) providing the room for the tanning agent. This fundamental process is conducted in paddle operation, which is followed by another task called fleshing. Thereafter ‘Deliming & Bating’ is carried out in drum operation. Deliming is practiced for the removal of the lime added during liming & unhairing and also for lowering the pH. Bating, a special operation is also followed along with deliming for the production of good quality leather to be used in Bag making. Proteolytic enzyme is used for this purpose in the same drum operation. Pickling & chrome tanning is accomplished after deliming and bating in the same drum. Pickling & chrome tanning is considered as the most polluting process in tanning operation due to presence of untreated Basic chrome sulfate (BCS) in the form of Cr(III), the typical process flow-sheet for chrome tanning operation are sequentially presented in Fig. 1

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There is a wide variation of composition in terms of organic carbon, nitrogen, and other toxic substances in the wastewater generated from chrome tannery unit. It depends upon the size of the tannery, chemicals used for a specific process, the amount of water used and type of the final product [45]. The average consumption of water from tanneries is in the range of (25 – 80) m³ per ton of processed raw material [31]. A variety of chemicals is used for processing of hides/skins, which make the wastewater very complex in nature [11]. Salt (NaCl) is used mainly for the preservation of fresh Hides/Skins from immediate decomposition and, the excess amount of salt needs to be removed before further processing. This is done by washing operation, i.e. Soaking process, using almost 400% of water/kg of hides/skins. Sodium sulfide (Na₂S) is then added to the same water or to a different float of water during Liming & Unhairing process [12]. Liming and unhairing wastewater is mostly biodegradable in nature [32]. The skins/hides from liming and unhairing operation are undergone for Fleshing to remove the attached flesh matrix. Thereafter, Deliming and Bating operation is followed, which generates wastewater, characterized by high amount of nitrogen mainly in form of organic nitrogen and trace amount of inorganic nitrogen [34]. In tanning operation of skins/hides basic chrome (III) sulphate [Cr (H₂O)₅(OH)SO₄] is used as the tanning agent both in single bath and/or double bath process. In hides / skins maximum uptake of chromium is 60-70 % on the surface area basis [12]. So, un-reacted trivalent and hexavalent chromium are present in very high concentrations in most cases.

III. POLLUTION POTENTIAL OF CHROME TANNERY WASTEWATER

The major concern of pollution in chrome tannery unit is envisaged for chromium, which prevals in two states - trivalent (Cr³⁺) and hexavalent (Cr⁶⁺). It is already observed from earlier research work that 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. Several in-vitro studies revealed that high concentrations of chromium (VI) in the cell could lead to DNA damage, causing several genotoxic insults of human body. Besides this, Chromium salts (chromates) also cause allergic reactions in human body. In tanning process about 25–30% of the total chrome used emanate through the wastewater. It can be reused by means of direct recycling of chrome-tanning float and chrome through precipitation [32]. In general, composite tannery wastewater is slightly alkaline in nature ranging between pH 7.5-8.5. It is because of mixing of acidic liquor from pickling & chrome tanning effluent with alkaline liquor from beam house operation. The physico-chemical characteristics of tannery wastewater are represented by dark color, high content of organic matters, suspended solids, organic & inorganic nitrogen, and total chromium. In addition, tannery wastewater also contains high amount of sulphate, sulphide, chloride etc. A typical characteristic profile of tannery wastewater is shown in Table I. These show that the COD values are considerably high in the range of (500-11500) mg/L, Total Kjeldal Nitrogen (organic plus inorganic nitrogen) ranges between (200–550) mg/L, whereas total chromium (Cr⁶⁺ plus Cr³⁺) concentration is in the range of (5-140) mg/L. It is also observed that major fraction of nitrogen is available as NH₃-N. A very low amount of phosphorus also comes out with wastewater. The biodegradable content is below 50% of the total dissolved organic substances, which indicates slow biodegradability. Moreover the total dissolved solid (TDS) is very high in comparison to total suspended solid (TSS), presumably on account of inorganic constituents. The combined effect of these pollutants in composite tannery effluent leads to drastic depletion of DO level in fresh water followed by death of fish population, excessive growth of water hyacinth and development of chromium toxicity. In general, the constituents present in tannery wastewater makes the biological treatment difficult and more complicated due to its low biodegradability.

The composite tannery effluent in most cases, does not meet the desirable standard in respect of COD, ammonia and sulphide/sulphate and chromium [17]. There are many research studies, investigated on Tannery wastewater treatment, especially in the field of physical or chemical methods or combination of both [33], [39], [44], [46], [42], [2]. Biological processes are usually employed as a secondary treatment option after primary treatment to remove the major portion of dissolved organics and nutrients from tannery wastewater. The major biological treatment processes investigated in the field of composite tannery wastewater treatment are Activated sludge process, Sequential batch reactor, Up flow anaerobic sludge blanket etc. [38], [17], [29], [19], [25].
IV. BACKGROUND OF BIOLOGICAL TREATMENT OF CHROME TANNERY WASTEWATER

Conventional treatment of tannery wastewater involves separation of tanning yard and soaking wastewater from the remaining streams, chemical treatment of tanning yard wastewater for precipitation of chromium, physical treatment of chloride laden soaking wastewater and finally biological treatment of the rest. References [38] and [29] reported some important research findings in activated sludge process for the treatment of composite chrome tannery wastewater. They determined the kinetic coefficients (k, Ks, Y, Kd) from treatability study using pre-treated tannery wastewater and applying Monod kinetics. This performance study indicates almost 80% removal can be achieved in ASP system. Apart from aerobic process, anaerobic methods were also practiced in treating chrome tannery wastewater, because it is a high-strength wastewater [21], [48]. All these studies highlighted the scope of high-strength wastewater like from chrome tannery in presence of different sulfur species. The effects of different sulfur species in anaerobic process were investigated and some crucial measures for removing sulphur toxicity were suggested.

Reference [17] modified the activated sludge process into Sequential Batch reactor for treating tannery wastewater by introducing alternative anoxic, anaerobic, and aerobic phases in the same aeration tank. The concept of SBR was thought on account of some toxic/inhibitory substances including chromium in the chrome tannery wastewater. It was believed that because of cyclic exposure of biomass in SBR to different phases, those were capable of functioning in extreme environment [27], [19]. In addition, SBR was found much efficient for nitrification and denitrification as compared to ASP in the case of tannery wastewater. In the field of anaerobic treatment process upflow anaerobic sludge blanket (USAB) reactor was successfully used by several researchers [28], [15]. Reference [28] employed USAB reactor in soak liquor, which contains very high amount of biodegradable substance and also very high salinity. Subsequently, the combination of USAB reactor and aerobic post treatment i.e. comprehensive treatment improved the performance.

Reference [9] developed some new innovative processes especially with integrated growth (combined attached and suspended growth) such as Sequential Batch biofilm reactor. Similarly, [4] used Hybrid upflow anaerobic sludge blanket reactor (HUASB) in the field of anaerobic treatment of chrome tannery wastewater. The suspended growth biomass along with attached growth increased solid retention time (SRT) value of the reactor, which in turn enhanced the efficiency of treatment. Moreover, the presence of attached growth increases the resistance to the toxic load and hence ensures requisite biomass concentration in the reactor. As a result, the reactor becomes small and therefore this can be used in low to medium scale tannery units where the space is low. Reference [32] treated combined liming & unhairing, fleshing and deliming & bating wastewater using an aerobic hybrid bioreactor system containing suspended and attached growth biomass. The successive development of biological processes applicable for the treatment of chrome tannery wastewater is shown in Fig. 2.
temperature 30°C under initial COD ranging between 1500 – 2900 mg/L resulted 60% COD removal at an optimum pH 7 and 65% of TKN operated at 25°C and DO concentration around 0.5 mg/L. Although biological treatment by activated sludge process is regarded as conventional technology for tannery wastewater treatment in terms of organic carbon and nitrogen removal, the applicability and usefulness, may suffer due to its special characteristics. The special characteristics are basically envisaged by the presence of biological inhibitors such as chromium, sulphide. chlorides etc. in excess concentration [22], [29], [5], [10], [45]. Reference [22] indicated that 70% of Cr(III) was removed in an activated sludge continuous-flow system at the influent level of (40 ± 5) mg Cr(VI)/L. Reference [5] reported almost 84% removal of Cr(VI) from an influent substrate concentration of 10 mg/L in a bench scale ASP. Nitrification process also appeared to be relatively sensitive to Cr (VI), which was on account of considerable inhibitory effect of Cr(III) on Nitrosomonas. Chromium III caused 50% inhibition on ammonia uptake rate at a concentration of 85 mg/L [29].

**V. PERFORMANCE OF BIOLOGICAL PROCESSES IN THE TREATMENT OF TANNERY WASTEWATER**

**A. Activated Sludge Process**

The treatment of tannery wastewater by activated sludge process, has been reported by many researchers both in lab and pilot scale [38], [13], [9], [17], [29], [27], [19], [25], [35]. In case of pre-settled tannery wastewater, the BOD removal efficiency was more or less 80% under 12 hrs detention time at a pH 7 and with MLVSS concentration of 3500 mg/L [25]. Later on, [24] determined various kinetic coefficients for settled tannery wastewater in a bench scale ASP reactor applying Monod’s kinetics. Similar study was performed on Activated sludge process to examine its efficiency for removal of nitrogen and organic matter from pre-treated and settled tannery wastewater through the combined nitrification and denitrification process [29]. Influent total nitrogen and COD concentrations varied from 927.0 mg /L to 2,140.0 mg/L and 9,583.0 mg/L to 13,515.0 mg/L respectively, whereas influent ammonium nitrogen varied from 149.0 mg /L to 178.0 mg /L. Sulphide and Total Chromium concentrations were in the range of 466.3–794.0 mg /L and 23.3–42.5 mg/L, respectively throughout the operation. This study revealed significant total nitrogen and COD removal efficiencies in the range of 82–98% and 95–98% respectively.

Reference [10] treated composite tannery wastewater in a batch ASP reactor using mixed microbial consortium collected from common effluent treatment plant (CETP). The study resulted 60% COD removal at an optimum pH 7 and temperature 30°C under initial COD ranging between 1500 - 6500 mg/L. The optimum COD removal efficiency was recorded as 74%. Reference [5] also studied the ASP system for treating synthetic and real life tannery wastewater containing COD and Chromium concentration of 4640 and 80 mg/L respectively. The study showed maximum COD and Cr 6+ removal efficiency of 65 % and 84 % respectively.

Reference [23] reported a performance study on ASP at Pallavaram CETP for treating composite tannery wastewater. This study showed 85-90% COD removal efficiency for the initial concentration in the range of (432–1617) mg/L. Reference [22] established that the total COD and TKN concentrations in the pre-treated tannery wastewater varied in the range of 2020–2790 mg COD/L and 203–276 mg N/L respectively. The removal efficiency was recorded almost 85-87% of COD and 65% of TKN operated at 25°C and DO concentration around 0.5 mg/L. Although biological treatment by activated sludge process is regarded as conventional technology for tannery wastewater treatment in terms of organic carbon and nitrogen removal, the applicability and usefulness, may suffer due to its special characteristics. The special characteristics are basically envisaged by the presence of biological inhibitors such as chromium, sulphide. chlorides etc. in excess concentration [22], [29], [5], [10], [45]. Reference [22] indicated that 70% of Cr(III) was removed in an activated sludge continuous-flow system at the influent level of (40 ± 5) mg Cr(VI)/L. Reference [5] reported almost 84% removal of Cr(VI) from an influent substrate concentration of 10 mg/L in a bench scale ASP. Nitrification process also appeared to be relatively sensitive to Cr (VI), which was on account of considerable inhibitory effect of Cr(III) on Nitrosomonas. Chromium III caused 50% inhibition on ammonia uptake rate at a concentration of 85 mg/L [29].

**B. Sequential Batch Reactor**

Nowadays, the sequencing batch reactor (SBR) is regarded as an alternative of activated sludge process (ASP), which operates in fill and drawing mode for treating biodegradable organic substances. However, the process is not a novel one still there should have further research study on SBR for the treatment of Chrome tannery wastewater. Reference [17] studied the scope of treatment of tannery wastewater containing chromium in a Sequential Batch reactor. SBR was found much efficient for nitrification and denitrification as compared to ASP in case of tannery wastewater. On account of cyclic exposure of biomass to different phases and concentration gradients allowed for their selection and enrichment, those are capable of functioning in extreme environment [27], [19]. The results from the SBR demonstrated that chromium addition had nominal influence on the microbial function. Moreover, it was observed that nitrification and denitrification rates, at the same chromium concentration, were higher in the SBR reactor than in batch ASP reactor. Several research studies confirmed that up to 180–190 mg/L of total chromium concentration, no inhibition can take place in nitrification and denitrification along with organic carbon removal compared to ASP reactor. Similar studies on SBR for the treatment of tannery wastewater [27], [19] reported the removal of 80–82% COD, 78–80% TKN and 83–99% NH$_3$-N at a 12-h cycle. With regards to effect of salt concentration on biological treatment process; [27] showed that highly saline effluents can be biologically treated in an aerobic SBR acclimated with halophilic bacteria. Optimum removal efficiencies of 95%, 93%, 96% and 92% on COD, PO$_4^{3-}$, TKN and SS, respectively were obtained under 5 days hydraulic retention time (HRT), at an organic loading rate (OLR) of 0.6 kg COD m$^{-3}$d$^{-1}$ and salt content of 34 g/L NaCl. Thus the earlier studies clearly revealed that the SBR technology was a reliable tool for tannery wastewater
treatment due to its flexible operation compared to ASP reactor. Reference [9] conducted a performance study on real life sample in a lab scale innovative Sequential batch biofilm reactor (SBBR) followed by ozonation. The combined process for treating tannery wastewater was evaluated on the basis of COD and nitrogen removal. In comparison to conventional SBR systems, the main advantages of envisaged in biofilm attachment, thereby higher biomass concentration and solid retention time (SRT). The COD and nitrogen removal efficiency were achieved as 85-90% and 98% respectively from the initial concentrations ranging between 3700-3900 and 220-260 mg/L respectively. Hence the COD and nitrogen removal efficiency could be much better in comparison to conventional ASP system for the treatment of Tannery wastewater [27], [19], [11].

C. Wetland and Ponds

Wetland or ponds can be an alternative option in tannery wastewater treatment where long piece of land is readily available. There is no external energy source needed for operating the wetland system [6]-[8], [40]. Although this process is extremely slow and unable to produce a good quality of effluent, it facilitates carbon sequestration and thus reduces global warming. Reference [7] investigated the survival of various plant species in subsurface horizontal flow constructed wetlands fed with tannery wastewater. Five pilot units were vegetated with Canna indica, Typha latifolia, Phragmites australis, Stenotaphrum secundatum and Iris pseudacorus, and the sixth unit was kept as a non-vegetated control. Their study also implies that those plant species were able to remove the organics form wastewater. Two different hydraulic loading rates viz. 3 and 6 m³d⁻¹ were employed in this experiment. COD was reduced by 41–73% for an initial organic loading in the range of (332–1602) kg ha⁻¹ d⁻¹ and in case of BOD₅, the degree of removal varied in the range of 41–58% for initial organic loading of 218 – 780 kg ha⁻¹ d⁻¹.

In addition, nutrient removal also occurred to a low extent. Two plant species viz. Phragmites australis and Typha latifolia were capable to grow up satisfactorily under tannery wastewater environment. In spite of high removal of organic content from the influent wastewater, during 17 months of operation, no significant deviation in performance was observed for these units. Another study by [40] confirmed that Wetlands performed as the ‘kidney of nature’, with an excellent ability to remove pollutants. Furthermore, it was observed that the concentration of trace metals (Pb, Zn and Fe) in wastewater were reduced by 25–45%, while total Cr was reduced by 95%, from the source point to the discharge site i.e. on the course of the 40 km long wastewater carrying canal in East Calcutta Wetland.

D. Upflow Anaerobic Sludge Blanket

Though the aerobic process is cost effective and reliable in nature producing stable end products, it often suffers from trouble particularly in treatment of high-strength wastewater. Under such circumstance, anaerobic treatment was traditionally followed before aerobic treatment in case of tannery wastewater. During last few decades, the technologist preferred anaerobic treatment of tannery wastewater over aerobic process due to its low sludge production and energy recovery. The anaerobic treatment of tannery wastewater is widely performed by Up flow Anaerobic Sludge Blanket (UASB) reactors [28], [15]. However, the anaerobic process for composite tannery wastewater treatment is susceptible to various inhibitions because of sulphide/sulphate [48], [15], [21], [28]. There are several research studies conducted for tannery wastewater treatment particularly with soak liquor due to presence of significant amount of biodegradable matters.

The anaerobic digestion of tannery soak liquor was investigated using a UASB where COD removal reached 78% at an OLR of 0.5 kg CODm⁻³ d⁻¹, at an HRT of 5 days and a TDS concentration of 71 g l⁻¹. Reference [28] demonstrated that the combination of the UASB with an aerobic post-treatment could enhance the performance of the overall treatment process and in a specific case, the overall COD removal efficiency reached to 96%.

Reference [15] explored the possibility of adopting upflow anaerobic sludge blanket (UASB) in tannery wastewater treatment as a low cost biological method. Anaerobic treatment was conducted through two stage UASB reactors connecting in series, each with volume of 94L. Five different hydraulic retention times (HRT), viz. 24, 18, 12, 8 and 5 h were maintained throughout the experimental study. This study revealed that COD and BOD₅ removal efficiency ranged between 40% and 70% in UASB reactor 1 and between 50% and 80% in UASB reactor 2 after 14 weeks. It was also observed that UASB reactors operating with 12 h HRT were ideal and 8 h HRT would be a minimum acceptable limit for tannery wastewater intended for disposal into the municipal sewer. It was also observed that HRT of 12 h also attained acceptable TSS levels in the effluent according to discharge standards. Reference [47] presented a comparative assessment of the cost analysis and quality of treatment between upflow anaerobic sludge blanket (UASB) and activated sludge process (ASP). It was perceived that the ASP reactor was superior in all respects. Moreover, total costs, including capital and operation and maintenance costs, for the UASB and ASP plants were INR 4.24/Litre and INR 3.36/Litre, respectively. Moreover, the treated UASB effluent had higher BOD₅ & COD and considerable amount of other objectionable constituents, like chromium (Cr) and sulfide, as compared to the ASP effluent. The outcomes of this study are totally reverse to the conventional philosophy of the efficacy of anaerobic processes especially for tannery wastewater treatment.

E. Hybrid Biological Process

Biological treatment of composite tannery wastewater with ASP, SBR, UASB, constructed wetland /ponds etc has already been conducted. Most of the biological systems consist of either suspended or attached growth biomass which degrades the carbonaceous content mainly, but not up to a satisfactory level. Besides this, there are some untreated nutrients and
chromium in the effluent. In view of above constraints, a novel biological method was innovated over last couple of decades in case of tannery wastewater. This unique reactor system hybrid bioreactor has been developed with the introduction of an attached growth phase within a classical suspended growth system. In hybrid bioreactor aerobic, anoxic and also anaerobic environment can be created by employing suspended and attached growth biomass simultaneously under different configurations to enhance both the carbonaceous and nitrogen removal efficiency [32], [3]. This system was incorporated in the line of the upgradeation of ASP to enhance the treatment capacity by introducing static media or moving bio carriers since 1980. In early 90's [26] developed a mathematical model of hybrid bioreactor considering simultaneous activity of activated sludge process and bio-film system. Thereafter, a few research studies were performed on hybrid bioreactor system [20], [14], [1], [18], [30]. Hybrid bioreactor can also be a viable option for composite tannery wastewater treatment.

Reference [32], studied on a shaft-type hybrid bioreactor treating segregated wastewater of achrome tannery unit firstly under suspended growth and then under hybrid system containing 5 mm sized tyde tube beads. The continuous study was also conducted under suspended growth and under hybrid system having 10–30 g/L of beads. Results of this study indicated that, maximum COD removal was 70.9% under an organic loading rate of 5.250 kg/day/m³. Reference [4] also treated the tannery wastewater using fixed biofilm upflow anaerobic sludge blanket (HUASB) reactor over a period of 370 d, at two distinct hydraulic retention times (HRT) viz., 60 and 70 h. The bioreactor attenuated the advantages of both fixed film and conventional upflow sludge blanket system. The reactor was operated at two different organic loading rate (OLR) of 2.74 kg COD m⁻³ d⁻¹ and 3.14 kg COD m⁻³ d⁻¹, at a HRT of 70 and 60 h, respectively.

VI. CONCLUSION

Biological process is the best option for the treatment of wastewater containing carbonaceous and nitrogenous organic matter (COD and TKN). The conventional biological method like ASP, SBR, UASB etc. have certain limitation towards the treatment of composite tannery wastewater because of various inhibitory substances. Out of biological methods, aerobic process is observed to be more cost effective and consistent in performance than anaerobic process. Wetland is also found a good and alternative treatment option for tannery wastewater treatment in case of sufficient land availability. SBR technology is also promising in the field of chrome tannery wastewater showing COD and TKN removal efficiency appreciably higher than conventional ASP system. The Upflow anaerobic fixed film reactor can also treat chrome tannery wastewater efficiently because of attached growth biomass that withstand toxic/inhibiting environment by virtue of long residence period. Based on this research finding it can be concluded that the introduction of an attached growth phase in a classical suspended growth system i.e. Hybrid bioreactor may be a viable option for the treatment of composite tannery wastewater. In this system, carbonaceous oxidation, nitrification and denitrification as well as chromium removal are possible within a single reactor.

REFERENCES

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