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## STUDY ON ELECTROCHEMICAL ADVANCED OXIDATION PROCESS TO TREAT FISH MEAL INDUSTRY WASTEWATER

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### Abstract

Fish meal processing industries convert the raw fish into a various commercially edible form. The effluent generated from this industry comprises a large amount of pollutants leading to high values of biochemical oxygen demand (BOD), chemical oxygen demand (COD) and total suspended solids (TSS). Mostly traditional biological and chemical methods have been adopted to treat the effluent coming out of the fish meal industry. By using these traditional methods at the end of the process, a large amount of sludge is produced. This sludge should undergo a process like sludge drying, thickening process and finally disposed of in the landfill. Electrochemical advanced oxidation process is an electrochemical method to treat wastewater and it can be performed using different methods such as electro-oxidation, electrochemical advanced oxidation process using Fenton's reagent. In this paper, we used the electro-oxidation method to reduce the polluting loads. A titanium electrode coated with ruthenium dioxide was used as anode, while the cathode was made from stainless steel.

The most complex organic compound could be broken by this electro-oxidation method. We have studied the effects of voltage on COD, BOD and TSS removal by applying the following voltage values: 3.2V with 0.06A, 5.65V with 1.17A, 7.79V with 1.62A. We have also discussed the effect of pH on the pollutants removal. About 88% of COD was removed at 7.79V after four hours of treatment. TSS and BOD removal was found to be 87% and 84% respectively. Overall, the electrochemical advanced oxidation process seems to be efficient in removing the pollutants such as BOD, COD and TSS.

*Key words:* biochemical oxygen demand, chemical oxygen demand, coated titanium anode, ruthenium dioxide, stainless steel cathode, total suspended solids

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### 1. Introduction

Millions of people around the world depend on fisheries for the day to day life. About 14 million are employed by the fishery sector in India and 5.23% of GDP to agriculture comes from this sector (<http://nfdb.gov.in/>). India has a long coastal line of about 8129 km with 3829 fishing villages. Most of the people living in the coastal area will have occupations related to fishes. Total fish production of India is about 8.8 million tons (FAO, 2011) so that India is the major supplier of fish in the world (as per the CIFT, 2008). On average, 60 million peoples are employed by fisheries and aquaculture.

The industry play a major role in culturing, processing, preserving, storing, transporting, marketing, selling and exporting the millions of fish. The effluent produced from this industry comprises a large number of pollutants which, when discharged without treating causes significant threats to the marine system, environment and also to the human health. So, before releasing it into water bodies, the wastewater has to be treated to achieve the standards. Besides high turbidity, strong greenish-yellow color and stinky odor (Afonso and Bórquez, 2002), this wastewater has high concentrations of suspended solids, chemical oxygen demand (COD), ammonia

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and nitrogen due to fat and proteins (Garrido et al., 1998).

The acidic or alkaline nature of the effluent will determine the pH range of effluent. Besides that, it also determines the decomposition of proteins and emission of ammonia (Gonzalez, 1996). Usually, a neutral pH range is observed in seafood processing effluent (Sherly et al., 2015).

There are different methods by which the electrochemical advanced oxidation process (EAOP) can be performed. Few of them are electrochemical oxidation with hydrogen peroxide, electro-Fenton photo electro-Fenton, electro-oxidation process. In electro-oxidation, process pollutants are removed by two methods: 1) direct electrolysis; 2) indirect electrolysis. In direct electrolysis, the pollutants are first adsorbed on the anode surface. Then the pollutant present on the anode surface gets oxidized. The required oxygen is produced from the water. In indirect electrolysis the pollutants are not directly adsorbed on the anodic surface instead they are oxidized by the chemically produced redox reagent. The mechanism of pollutants reaction with the metal electrode is given in Fig. 1. Electrochemical advanced oxidation processes (EAOP) such as electrochemical oxidation with hydrogen peroxide, electro-Fenton, and photo electro-Fenton are powerful transformation technologies for wastewater treatment. Anode and cathode play an important role in determining the oxidation power of these methods to treat refractory organic pollutants (Flores et al., 2018). The electrochemical oxidation process is also used to remove butylated hydroxyanisole from various water matrices (Zhihong et al., 2019). The advanced oxidation process encompasses the treatment of several kinds of synthetic wastewater containing dyes, pesticides, pharmaceuticals and other pollutants (Moreira et al., 2017). Ti/Pt and Ti/PbO<sub>2</sub> anodes are used in the Electro oxidation process to treat the tannery industry effluent. The efficiency of these anodes was checked. It was found that Ti/Pt is the most efficient anode in removing pollutants from tannery effluent, able to remove, for example 0.802 mg COD/h (Rao et al., 2001). The EAOP process the wastewater itself acts as an electrolyte but in some cases, the additional electrolyte such as NaCl is added to enhance the process (Scialdone et al., 2008).

This electrolyte addition depends on the type of wastewater treated. The electrode material plays a very important role in efficient pollutant removal. The electrodes such as IrO<sub>2</sub>, graphite, Pt allow only partial oxidation of organic matters, while electrodes such as SnO<sub>2</sub>, PbO<sub>2</sub>, Boron doped diamond (BDD) allows complete oxidation. Among them, BDD shows high removal efficiency (Panizza and Cerisola, 2004). The research was carried out from June 2019 to December 2019 at the University College of Engineering, Anna University - BIT Campus, Tiruchirappalli, Tamil Nadu, India.

To treat the fishmeal industry wastewater, biological methods have mostly been adopted. Yun et al. (2017) have used natural biodegradation process to treat fish processing wastewater in which TSS, BOD, COD removal percentage were 37%, 81%, 85.5% respectively, the time required to achieve these removals was 8 to 10 days. Riano et al. (2011) have discussed the treatment of fish processing wastewater using photobioreactors at different temperatures. The highest total COD removal was 70%. In a case study made by Colic et al. (2007) at the fish processing industry where they use hybrid centrifugal dissolved air flocculation flotation to treat wastewater. The COD and BOD removal is about 65% on average and they reduced TSS to 20mg/L. The main disadvantage of these biological methods is the production of sludge to overcome this EAOP process has been examined in this paper. Also, EAOP process has successfully used to treat various industrial wastewater such as textile (Silva et al., 2018), tannery (Naumczyk and Kucharska, 2017), food processing (Heponiemi and Lassi, 2012), pulp and paper (Klidi et al., 2018) and various other industries. In this paper we analyzed the performance of EAOP in treating the fish meal industry wastewater.

## 2. Material and methods

We have collected the sample from the fish meal processing industry at Periyapalayam near Thiruvallur district, Chennai for that we have got proper permission from the respective authority of the company to collect the wastewater and to use it for our research activity. The industry processes raw fish of about 20 tons per day. The characteristic of the wastewater has shown in Table 1.

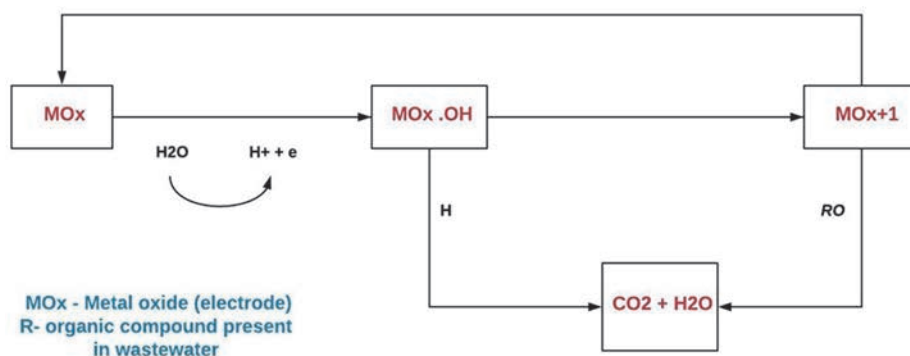


Fig. 1. Reaction mechanism of metal electrode with pollutants

**Table 1.** Characteristics of raw effluent

S.no	Parameters	Unit	Values
1	pH	-	7.23
2	TSS	mg/L	460
3	BOD	mg/L	1252
4	COD	mg/L	2236
5	BOD/COD	-	0.56

The Boron doped diamond (BDD) anode was used to treat paper mill wastewater yielded the COD removal of 85% after three hours of treatment at 1.5A current (Klidi et al., 2018). Different anodes have been used in treating the tannery wastewater Ti/Pt, Ti/PbO<sub>2</sub>, Ti/MnO<sub>2</sub> yielded the COD removal of 62%, 71.3%, 75.8% respectively at 27.6V and 0.5A (Rao et al., 2001). Villanueva et al. (2014) have used the BDD electrode to treat coffee pulping wastewater at a current density of 40 mA cm<sup>-2</sup> which resulted in organic matter removal of 73% to 84%. Mohan et al. (2007) have varied the current densities such as 1, 2, 3, 4, 5 A/dm<sup>2</sup> to treat the textile wastewater and found that the rate of degradation of organic pollutants increased as the current densities increased. Panizza and Cerisola, (2004) have used Ti/TiRuO<sub>2</sub> anode to treat Tannery wastewater at different densities and found that the optimum current density for effective COD removal would be 600 A m<sup>-2</sup>. Ti/BDD and porous Ti/BDD cathodes are used in removing aspirin from pharmaceutical industry wastewater and the results show that Porous Ti/BDD is more efficient in removing Aspirin (Yapeng et al., 2015). The electrocoagulation process showed the COD removal of 85%. And the electrocoagulation followed by coagulation with ferric salt showed the overall COD removal of 98% (Omprakash, 2019) RuO<sub>2</sub> has complex and unique redox chemistry which can act as a versatile catalyst and electrocatalyst in wastewater treatment (Tran, 2016; Venkat et al., 2006). In the present study, RuO<sub>2</sub> is synthesized in the laboratory by thermal decomposition method and this method was preferred because of its simple condition and low cost (Tran et al., 2014). Mohan et al. (2007) have used three different anodes Ti/TiO<sub>2</sub> coated with RuO<sub>2</sub>, Ti/TiO<sub>2</sub> coated with PbO<sub>2</sub>, Ti/TiO<sub>2</sub> coated with SnO<sub>2</sub> to treat textile wastewater and found that anode coated with RuO<sub>2</sub> are more effective than the other two electrodes.

### 3. Experimental setup

In the present study, we have used a lab-scale batch reactor. The titanium electrode (Ti) coated with ruthenium dioxide (RuO<sub>2</sub>) has been used as anode and stainless steel has been used as the cathode. The rods were 300 mm long with a 6.35 mm diameter. Six RuO<sub>2</sub> coated titanium (Ti/ RuO<sub>2</sub>) anodes and six stainless steel cathodes were used in this process. Borosilicate glass reactor of 160 mm diameter and 350mm height has been used as the reactor for the electrochemical oxidation process. The distance between cathode and anodes was 15 mm. The sectional view of the reactor

is shown in Fig. 2a and top view of the reactor is shown in Fig. 2b. In the beginning, the reactor was cleaned using tap water, and later using the distilled water to make sure that no foreign particles were present during the electrical process. The Ti/RuO<sub>2</sub> and stainless steel electrodes used in the process were of high quality. The electrodes were cleaned using cloths before using them in the experiment. We performed the trial run with ordinary tap water to check the working conditions of the electrodes and rectifiers.

Wastewater of volume 5.5 liters was added in the reactor. The experiments were performed for different voltage and current such as: 3.2V and 0.06A; 5.65V and 1.17A; 7.79V and 1.62A, to know the effect of current on the removal of pollutants. We made the alternating current (AC) current to direct current (DC) using rectifiers. The voltage and current were measured using a digital multimeter. The color removal was clearly visible during the process. During the first run of the experiment the COD removal did not increase too much during the 5<sup>th</sup> and 6<sup>th</sup> hour, it is even almost constant. So, we have conducted each run for a period of four hours. In the wastewater sample, initial parameters were tested before the start of each experimental run. During the experiment the wastewater in the reactor was manually stirred and also the temperature was noted at constant time intervals. A volume of 300mL of wastewater was taken from the reactor at the interval of one hour for analysis. Parameters such as COD, BOD, TSS, temperature, pH, were measured in the sample. To investigate the effect of pH, the EAOP process has been conducted for different pH values of 3, 7.2, 9. The pH of the sample was modified using chemical solutions such as H<sub>2</sub>SO<sub>4</sub> and NaOH. For this run, the voltage in the EAOP process is kept as 7.79V. The basic parameters such as COD, BOD, TSS, pH, and temperature have been measured in the laboratory using standard procedures. For pH, the standard pH meter has been used. For temperature, we have used digital TDS meter Model No.651. TSS is determined by gravimetric methods, using filter paper. First, the weight of filter paper has been determined and then the filter paper was washed with sample wastewater. Then the weight of filter paper is noted and finally, TSS is calculated.

For BOD calculation, first two BOD bottles were taken and filled with 10 mL of the sample also water is added to fill up to the brim. Then manganese sulfate and alkali iodide reagent are added in the required proportion. One BOD bottle was kept in an incubator for final BOD determination. From another BOD bottle, the solution was transferred to a conical flask, where was then added sulphuric acid and starch solution and titrated it against sodium thiosulfate. The endpoint was the disappearance of the blue color. From the obtained BOD values the initial DO is determined and after three days the second BOD bottle which was kept in the incubator is taken and the final DO is determined. From initial and final DO the BOD is determined.

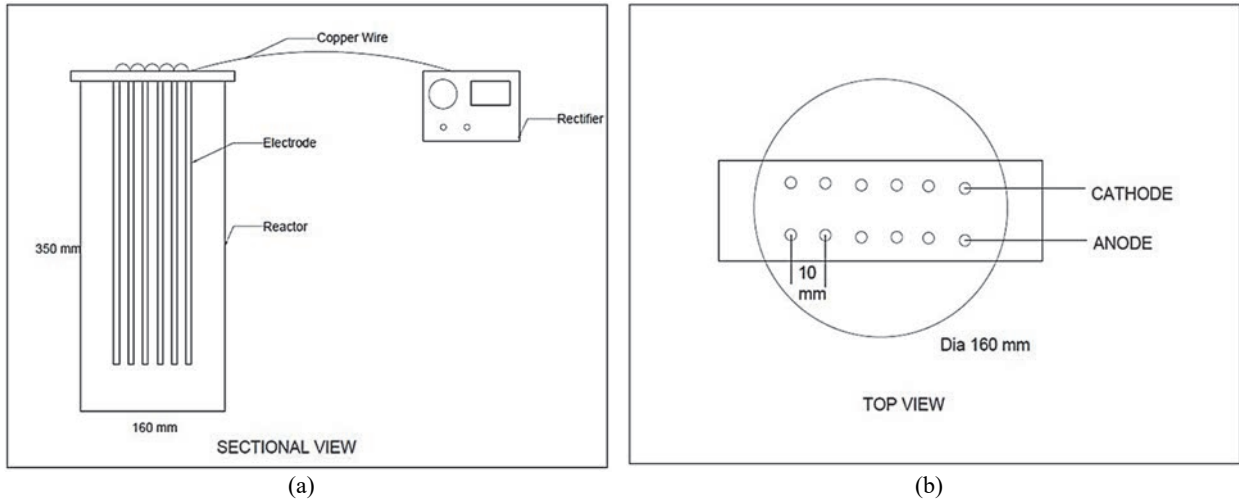


Fig. 2. (a) Sectional view of EAOP reactor; (b) top view of EAOP reactor

For COD analysis, the sample is taken in the beaker, then potassium dichromate and sulphuric acid reagent were added to it in the required proportion. After that, the prepared solution is refluxed at 150°C in the reflux tube for about two hours. Then mercuric sulfate and ferroin indicator were added and titrated it against ferrous ammonium sulfate solution.

**3. Results and discussion**

In the present study, the EAOP process has been performed without adding any external chemical to the sample. The sample present in the reactor acted as an electrolyte. The various parameters such as COD, BOD, TSS, temperature, pH have been investigated for different EAOP reaction time.

*3.1. Effect of voltage on pollutants removal*

The voltage supply in the reactor was altered as mentioned above. If the current supply in the reactor increased, the COD removal efficiency also increased. The current input directly influenced organic pollutant removal. The COD removal in the first hour seems to be high when compared to a second, third, and fourth hour for all different voltages applied. The highest COD removal was 88% which was achieved at 7.79V. Ultimately, the COD of the wastewater sample was reduced to discharge standards in a four hour of

treatment. The BOD of the wastewater was also kept on reducing as the voltage increased. For BOD also the highest removal efficiency was achieved at 7.79V. At the end of four hours of treatment 84% of BOD was removed. In the case of BOD removal, the discharge standards could not be achieved.

The effect of voltage on TSS removal was studied to check the level of organic and inorganic solids removed during the process. The TSS removal was very high compared to all other parameters. At 3.2V, there was a good removal of TSS of about 83% after four hour reaction time. However, as the voltage increased, there was a small increase in TSS removal percentage subsequently the highest TSS removal of about 87% was achieved at 7.79V. The values of TSS, BOD, COD removal at 3.2V has shown in Fig 3. The removal percentage at 5.65 has shown in Fig 4 and removal percentage at 7.79V has shown in Fig 5. The obtained values of TSS, BOD, and COD at different voltage are listed in Table 2.

To know the change in pH of the sample due to the movement of ions inside the reactor when the current is applied, the pH of the sample was tested for all applied voltage. The results showed that the pH of the solution does not change much, there was only a slight variation. The temperature of the sample before the start of the reaction was 32°C. During the process, no change in temperature was observed.

Table. 2. Results of EAOP on fish meal industry wastewater using Ti/RuO<sub>2</sub> electrode

Sample	Parameter	Values at 3.2 voltage				Values at 5.65 voltage				Values at 7.79 voltage			
		1 <sup>st</sup> hour	2 <sup>nd</sup> hour	3 <sup>rd</sup> hour	4 <sup>th</sup> hour	1 <sup>st</sup> hour	2 <sup>nd</sup> hour	3 <sup>rd</sup> hour	4 <sup>th</sup> hour	1 <sup>st</sup> hour	2 <sup>nd</sup> hour	3 <sup>rd</sup> hour	4 <sup>th</sup> hour
1	TSS (mg/L)	136	112	89	76	109	91	79	68	102	85	71	60
2	BOD (mg/L)	1002	832	703	580	955	764	564	385	818	531	379	203
3	COD (mg/L)	1699	1320	1050	828	1565	1140	783	514	1186	738	492	246
4	BOD/ COD	0.59	0.63	0.67	0.7	0.61	0.67	0.72	0.75	0.69	0.72	0.77	0.82

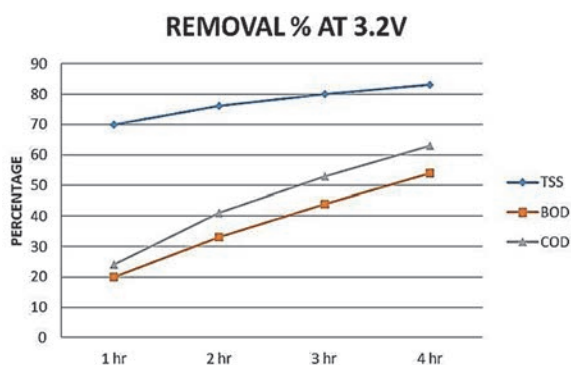


Fig. 3. Removal percentage of TSS, BOD, COD at 3.2 V

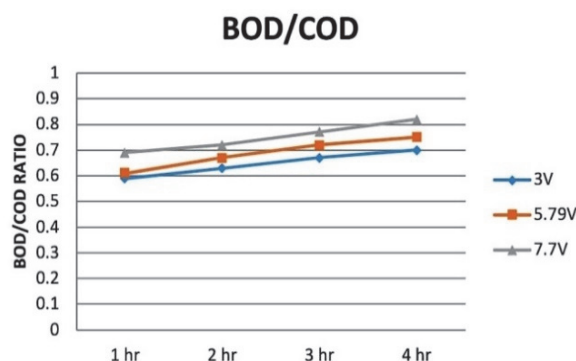


Fig. 6. Ratio of BOD to COD

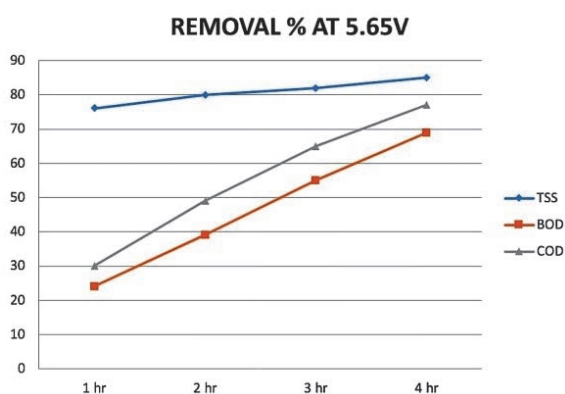


Fig. 4. Removal percentage of TSS, BOD, COD at 5.65 V

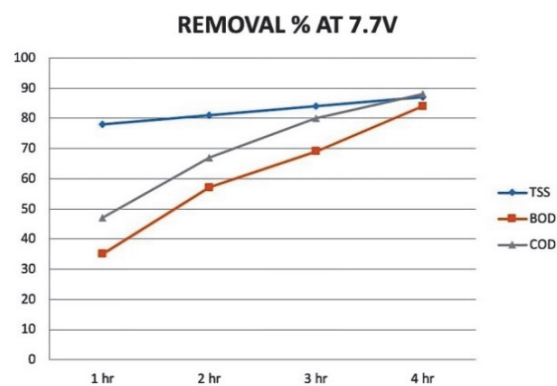


Fig. 5. Removal percentage of TSS, BOD, COD at 7.79V

The constant temperature was there in the reactor for all applied voltage until the end of the reaction time. Also during the experiment, the color change can be seen through the naked eyes. Before the start of the experiment, the color of the sample was dark gray which is then changed to white color and finally colorless.

If the BOD/COD is greater, then the effluent is easily biocompatible (GilPavas et al., 2018). The BOD/COD ratio of the raw sample was 0.56. The BOD/COD ratio of the sample was calculated to know the biodegradability of the solution. The BOD/COD of the wastewater increased as the reaction time increased. The final BOD/COD ratio was 0.82. The BOD/COD ratio for different voltages has shown in Fig. 6.

### 3.2. Effect of pH on pollutants removal

pH plays a very important role in organic pollutants removal by the electrochemical method (Brillas et al., 2005). In some literature, it is said that the pH does not influence the pollutant removal rate (Ellouze et al., 2015). In some cases, it is said removal rate of pollutants will be high if the solution is alkaline (Canizares et al., 2005; Lissens et al., 2003) and to contrast, some have said that removal rate will be high in acidic solution (Martinez et al., 2005; Scialdone et al., 2008) so to check the influence of pH in removing pollutants in fish meal wastewater and check whether initial pH change should be given to the sample for the efficient removal of COD the pH of the sample was altered.

Two different pH range, 3 and 9 were taken. For these two runs the voltage in the reactor was kept as 7.79V. The results showed that there was no major change in the COD removal percentage for both acidic and alkaline nature. The removal efficiency was 89% for COD at acidic pH and 87% at alkaline pH of the sample. This confirms that only direct oxidation takes place inside the reactor, which means the organic matters are removed only by the hydroxyl radicals produced around the electrodes. Therefore, no initial pH adjustment is required for efficient COD removal in fish meal wastewater.

### 3.3. Post-treatment after EAOP

In the EAOP process, COD of wastewater has been reduced to 246 mg/L, which meets the discharge standards in India. The BOD of the wastewater was reduced to 203 mg/L which does not meet the discharge standards. Fish processing wastewater is treated using constructed wetlands in which BOD removal was found to be 55% (Pineyro et al., 2019). Moreover, 78% to 91% of BOD can be removed in the domestic wastewaters using wetlands (Juwarkar et al., 1995). Sehar et al. (2019) have used constructed wetland to treat various industrial wastewater such as olive mill wastewater, aquaculture, and seafood processing industry wastewater, milk and cheese industry wastewater. They have concluded that constructed wetlands are an effective and sustainable



approach in treating domestic and industrial wastewater. Constructed wetlands is the most economical and environmentally friendly method to treat fish and shrimp aquaculture effluents (Yalcin and Fulya, 2018) So for further reduction in BOD in cost-effective manner constructed wetlands can be used after EAOP process.

#### 4. Conclusions

In this study, electrochemical advanced oxidation process is found to be effective in treating fish meal industry wastewater. The 88% of COD, 84% of BOD, 87% of TSS, removal was achieved by the EAOP process for a retention time of about four hours at 7.79V. For achieving the BOD discharge standards Constructed wetland process can be adopted followed by the EAOP process.

The Ti/RuO<sub>2</sub> electrode can be effectively used in the EAOP process to treat fish meal industrial wastewater. It forms no byproduct during the reaction and it the main advantage of this process. Also, there is no trace of a toxic substance at the end. Though the initial investment cost for electrodes is high, the maintenance cost will be comparatively low. However, further research needs to be conducted to know the actual efficiency of EAOP with constructed wetland process.

Constructed wetland process is considered to be economically feasible compared to other methods for BOD removal so as to meet the Indian standards.

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