Journal of Hazardous Materials 169 (2009) 480–485

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Interior microelectrolysis oxidation of polyester wastewater and its treatment technology

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ARTICLE INFO

Article history:
Received 17 October 2008
Received in revised form 26 March 2009
Accepted 26 March 2009
Available online 2 April 2009

Keywords:
Polyester wastewater
Interior microelectrolysis
Anaerobic
Aerobic
Surfactant

ABSTRACT

This paper has investigated the effects of interior microelectrolysis pretreatment on polyester wastewater treatment and analyzed its mechanism on COD and surfactant removal. The efficiency of interior microelectrolysis is mainly influenced by solution pH, aeration and reaction time. Contaminants can be removed not only by redox reaction and flocculation in the result of ferrous and ferric hydroxides but also by electrophoresis under electric fields created by electron flow. pH confirms the chemical states of surfactants, Fe(II)/Fe(III) ratio and the redox potential, and thus influences the effects of electrophoresis, flocculation and redox action on contaminant removal. Anaerobic and aerobic batch tests were performed to study the degradation of polyester wastewater. The results imply that interior microelectrolysis and anaerobic pretreatment are lacking of effectiveness if applied individually in treating polyester wastewater in spite of their individual advantages. The interior microelectrolysis–anaerobic–aerobic process was investigated to treat polyester wastewater with comparison with interior microelectrolysis–aerobic process and anaerobic–aerobic process. High COD removal efficiencies have been gotten by the combination of interior microelectrolysis with anaerobic technology and aerobic technology. The results also imply that only biological treatment was less effective in polyester wastewater treatment.

1. Introduction

Polyester is a material produced on a large scale as a component of textile fiber, which results in a great deal of discharge wastewater with various additives and detergents, including wetting agents, softening agents, antioxidant, surfactant, detergent and antiseptic. Surfactant induces foaming and consequently cut down on the oxygen concentration in water. Antioxidants used in textile industry to inhibit the oxidation of the fiber could resist the oxidation of contaminations in wastewater treatment and antiseptic take negative effect on growth of bacteria. Therefore, these pollutants discharged from various stages of the polyester manufacturing process are characterized by hard oxidation, toxicity and poor biodegradation. Consequently, most of the traditional methods are becoming inadequate, and unsatisfied to the requirement of environment.

Most of the studies on decolourization of textile wastewater [1–3] have been carried out, but few researchers focus on the surfactant removal in textile wastewater. Indeed, one of the main aspects of the treatment of polyester wastewater is the removal of surfactant and the poor biodegradable organics. The presence of surfactants in water is highly visible which seriously results in foaming [4], and surfactant can be biodegraded but at very low rate [5–7]. In addition, various types of surfactant are gradually invented and utilized in the polyester process to improve the characteristics of fabrics and subsequently increase the difficulties of the polyester wastewater treatment.

Traditional methods cannot be employed for polyester effluent treatment due to time consuming and lack of effectiveness if applied apart [8–11]. Moreover, toxicants inhibit the biological activity of biomass and even cause process upset [12]. It is necessary to develop pretreatment technologies available to eliminate the hazardous pollutant before biological treatment. Therefore, different technologies developed so far have been investigated for possible application in the pretreatment of poor biodegradable and even toxic polyester wastewater [13–15]. Hongjun et al. [16] have studied the UASB-AF process to treat polyester wastewater, and hydrolytic acidification hybrid membrane bioreactor [17] also has been studied to treat polyester wastewater. Besides, ozone and hydrogen peroxide oxidation technologies and photochemistry technology [18–19] have been employed to enhance polyester wastewater treatment. However, many of these technologies suffer the limitation of either removal efficiency or treatment cost. For toxic and poor biodegradable substances in polyester wastewater, an ideal process is to pretreat wastewater to removal toxics or change to biodegradable substances.

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doi:10.1016/j.jhazmat.2009.03.123

Table 1
Characteristics of polyester wastewater.

	COD (mg/L)	BOD_5 (mg/L)	TS (mg/L)	NH_3–N (mg/L)	TN (mg/L)	TP (mg/L)	pH	Surfactant (mg/L)
Maximum	4298	1071	210	28	34	5.4	5.5	158
Minimum	1100	262	55	9	14	0.6	4.2	90
Average	3365	970	73	17	26	2.1	4.5	102

Several researches have shown that interior microelectrolysis can be regarded as effective pretreatment of poor biodegradable wastewater [20–22]. Waste iron chips form numerous galvanic cells between iron and carbon in wastewater, which results in the galvanic cell reaction. Products released from the galvanic cell reaction include hydroxyl, atomic hydrogen and Fe(II) which have high activities to decompose contaminants. Furthermore, interior microelectrolysis pretreatment is low-cost and effective because it does not require chemical coagulant and external power as in the cases of coagulation and electrolysis. In addition, anaerobic processes have been shown as effective methods in removing organic loads in textile wastewater [23–26].

In this paper, the effects of interior microelectrolysis pretreatment were performed by interior microelectrolysis batch tests, and degradable mechanism and its influence factors of interior microelectrolysis have also been investigated. Anaerobic and aerobic batch tests have been used to evaluate the biodegradabilities of polyester wastewater. Furthermore, another objective of the present work was to evaluate the efficiency of interior microelectrolysis–anaerobic–aerobic process on COD and surfactant removal in the treatment of real polyester wastewater.

2. Materials and methods

2.1. Materials and analysis methods

Polyester wastewater was obtained from a real polyester factory (Tianjin, China). The composition of wastewater varied widely due to the variety of raw material and agents used in the production process. The characteristics of wastewater sampled from the polyester finishing mill are given in Table 1. Wastewater shows very low pH values and associated with low nitrogen and phosphorous content, and wastewater contains 90–158 mg/L surfactant. Moreover, High fluctuation in COD, BOD and pH are found.

Waste iron chips (3.5% carbon) were collected from a metal machining mill and 5–10 mm in length and 3.0 mm in width. Iron chips were first degreased in a 10% NaOH solution, and then soaked in a diluted (5%) hydrochloride acid solution, and finally cleaned by deionized water.

COD, BOD_5, NH_3–N, mixed liquor suspended solids (MLSS), surfactant concentration, total nitrogen (TN) and total phosphorus (TP) were analyzed referring to the standard methods for Examination of Water and Wastewater [27]. COD was determined by potassium dichromate oxidation method, NH_3–N by distillation and titrimetric method, TN by potassium persulfate oxidation and ultraviolet–visible spectrophotometer (UV-1000, Beifenruili) determination, TP by sulfuric acid and nitric acid digestion method, MLSS by gravimetric method, surfactant by extraction and ultraviolet–visible spectrophotometric (UV-1000, Beifenruili) determination.

Oxygen concentration was measured by dissolved oxygen concentration meter (ORION, 805A) and was adjusted by air flow.

2.2. Batch experiments

Batch tests were performed to study the degradation of polyester wastewater. In interior microelectrolysis batch tests, iron chips, 10% (v/v) working volume of reactor, were first spread at the bottom of the reactor (1 L) with a diffuser on the bottom and aeration can be adjusted and controlled by a flowmeter. The extent of interior microelectrolysis degradation of polyester wastewater was detected as compared with a blank without the addition of the iron chips. COD, surfactant and pH values in supernate were detected regularly by an interval of time.

In anaerobic batch tests [28] anaerobic sludge collected from sludge anaerobic digester tanks in a domestic wastewater plant was first involved in a sealed container to reduce the original organic substrate at 35 °C for 7 days without any nutrition addition, and then anaerobic sludge was inoculated to polyester wastewater for 30 days before tests. In anaerobic batch experiment, 50 mL anaerobic sludge with 20 g MLSS/L and 50 mL polyester wastewater were first involved into a sealed triangle flask. pH value of mixture was adjusted to 7.0 ± 0.2 by sodium bicarbonate. After deaired by nitrogen, flask was put into water boiler at 35 ± 1 °C. To study the effect of interior microelectrolysis pretreatment on COD removal in anaerobic unit, COD removal of polyester wastewater after interior microelectrolysis pretreatment was also investigated in anaerobic batch test.

In aerobic batch test, aerobic sludge from a real polyester wastewater treatment plant (Tianjin, China) was inoculated to polyester wastewater at 25 °C with air aeration for 2 days. 50 mL aerobic sludge 10 MLSS g/L and 50 mL polyester wastewater were mixed into a flask with a diffuser at the bottom of the flask, and oxygen concentration was kept at 2.5 g/L. COD in the supernate was carried out to determine the extent of aerobic biodegradation.

2.3. Pilot-scale experiment setup and procedure

Interior microelectrolysis–anaerobic–aerobic process (IM-A/A) was applied to treat polyester wastewater in this study. Interior microelectrolysis reactor with 8 m^3 working volume was filled with helix iron chips 2 m^3 and was aerated through a tube diffuser on the bottom of reactor. A pressure reducing valve and a flow meter were installed to adjust and check the air flow rate. UASB was chose as the anaerobic reactor. The effective volume of the reactor was 8 m^3 and the flow distributor was set at the bottom of the reactor to distribute the influent evenly from the bottom. The solid–gas–liquid separator was put in the upper part of the reactor to prevent the loss of sludge from the reactor and release the biogas produced by anaerobic digestion. UASB was seeded up to 1/3 of its height with anaerobic sludge 50 g MLSS/L and was fed with polyester wastewater discharged from interior microelectrolysis reactor. A plug flow tank was used as aerobic reactor with three diffusers on the bottom of reactor. Anaerobic and aerobic process (A/A) and interior microelectrolysis–aerobic process (IM/A) were also investigated to treat polyester wastewater in order to compare with IM-A/A process. COD, surfactant and pH in the influent and the effluent were detected. In Table 2, the main operational parameters applied to the three processes are summarized.

3. Results and discussion

3.1. Interior microelectrolysis batch tests

In the unit of interior microelectrolysis, iron and carbon of iron chips are applied as anode and cathode respectively during

Table 2
Operation conditions in the different processes.

Phase	Total HRT (h)	IM-A/A (HRT, h)			IM-A (HRT, h)		A/A (HRT, h)	
		IM	Anaerobic	Aerobic	IM	Aerobic	Anaerobic	Aerobic
1	20	6	6	8	8	12	8	12
2	26	9	9	8	12	14	12	14
3	32	12	12	8	16	16	16	16
4	38	15	15	8	20	18	20	18

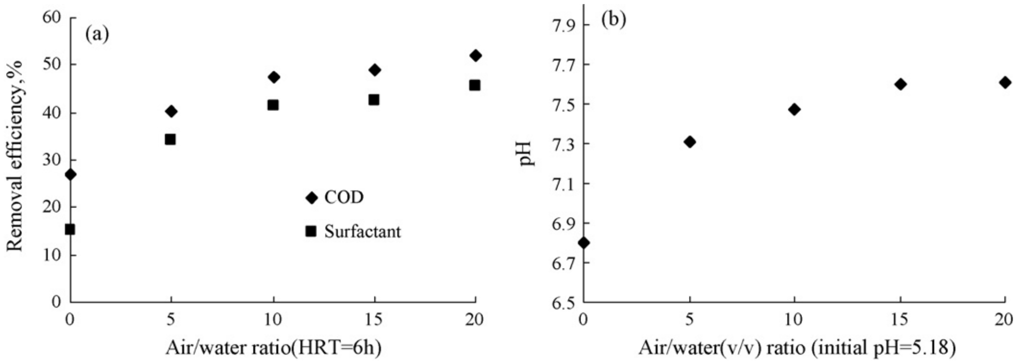

Fig. 1. Effects of air/water ratios on COD, surfactant and pH in interior microelectrolysis.

the corrosion of iron chips, but mechanisms are quite different to decompose contaminants with and without aeration. Without aeration, Fe was oxidized into Fe(II) ($E^0_{\mathrm{Fe(II)+/Fe}} = -0.44\,\mathrm{V}$) and H^+ was reduced into H_2 ($E^0_{\mathrm{H+/H2}} = 0.00\,\mathrm{V}$). Consequently, contaminants mainly decreased through reduction reaction with fresh hydrogen. If provided oxygen, Fe was first oxidized into Fe(II) and then oxidized to Fe(III) [29] along with hydroxyl radical formation [30,31]. Consequently, organic contaminants are broken into small molecule substances due to the strong oxidization property of hydroxyl radical. Although contaminants can both be removed by ferrous and ferric hydroxide flocculation, Fe(III) show a better flocculating effect with less solubility. Accordingly, the Fe(III)/Fe(II) ratio should influence the COD removal, which is changed with the air/water ratio.

The effects of air/water ratio on COD and surfactant removal in interior microelectrolysis unit are given in Fig. 1(a). COD removal was only 27% without aeration but the total COD removal could get 47.3% at air/water ratio 10 and was 52% at air/water ratio 20. Surfactant was removed 15.3% without aeration and the surfactant removal can get 41.5% at air/water ratio 10. The results indicate that aeration enhances COD removal and surfactant removal in interior microelectrolysis unit. That can be explained that the increase of air/water ratio results in the high oxygen concentration in water and consequently improves the total COD removal. However, further increase in air/water ratio did not increase the total COD removal and surfactant removal significantly, indicating that oxygen concentration tends to saturation in water.

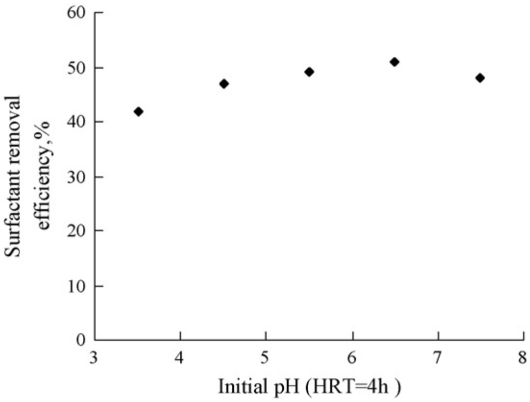

Fig. 2. Effects of initial pH on surfactant removal.

Surfactant agents are usually poor biodegradable, and only a small amount of surfactant can induce foaming and wrap bacteria in biotreatment unit and subsequently the growth of microbe was restrained due to nutrient substance scarcity. Effective surfactant removal in pretreatment could be benefit for the following biotreatment. The results from interior microelectrolysis batch tests suggest that interior microelectrolysis is an effective and economic pretreatment method for removing surfactant agents.

pH values increase after interior microelectrolysis pretreatment, given in Fig. 1(b). Indeed, the redox of galvanic cell reaction consumes acidity of water. Furthermore, the increases in air/water ratio enhance the oxidation of Fe(II) to Fe(III), which accompany the production of basicity ($4\mathrm{Fe(II)} + O_2 + 2H_2O \rightarrow 4\mathrm{Fe(III)} + 4OH^-$). Accordingly, the pH of the treated effluent should increase with the increase of air/water ratio. However, the increase of pH enhances the production of ferrous hydroxide and ferric hydroxide at the same time, which could restrain the increase of pH. After the reactions balance, the pH of the treated effluent was near neutral and was in the range of 6.80–7.61.

In interior microelectrolysis unit, contaminants can be removed not only by redox reaction and flocculation in the result of ferrous and ferric hydroxides but also by electrophoresis under electric fields created by electron flow. pH confirms the chemical states of surfactants, Fe(II)/Fe(III) ratio and the redox potential, and thus influences the effects of electrophoresis, flocculation and redox action on contaminant removal. From Fig. 2, initial pH influences the surfactant removal, and the surfactant removal efficiency increased from 47% to 51% when initial pH was changed from 4.5 to 7.5. The results showed that pH values of the treated effluent were adjusted to near neutral (6.8–7.5) when initial pH was in the range of 4.5–7.5.

COD and BOD were also measured in the interior microelectrolysis batch tests at air/water ratio 10. Interior microelectrolysis reduced COD of polyester wastewater from 3356.6 to 1679 mg/L after 12 h, which corresponds to removal efficiency 50.0%, given in Fig. 3. COD decreased quickly at the beginning of the test and

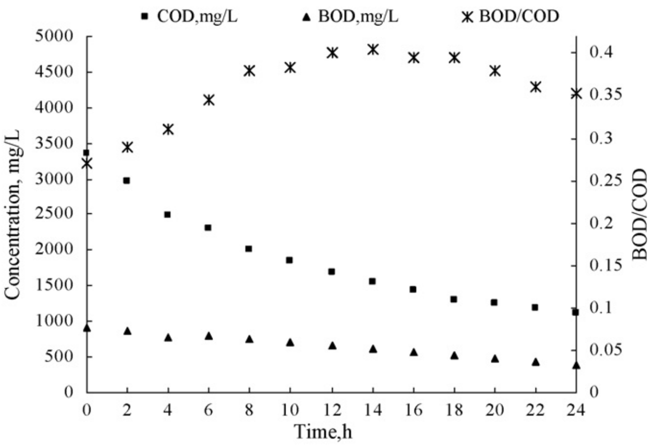

Fig. 3. COD, BOD changes in interior microelectrolysis.

then reduced slowly after 16 h. COD and BOD both decreased with extending retention time, but BOD decreased more slowly compared with the decrease of COD. Considering BOD/COD ratios in Fig. 3, BOD/COD ratios increased as HRT was in the range of 0–14 h, which showed that the biodegradability of polyester wastewater was improved after interior microelectrolysis pretreatment. In interior microelectrolysis unit, poor biodegradable substances can be broken into biodegradable substances by the oxidation of hydroxyl radical [30], and electrophoresis results in the removal of colloid and surfactant. Those could be the reasons about the increase of BOD/COD after interior microelectrolysis pretreatment.

3.2. *Anaerobic biodegradable batch tests*

Anaerobic biodegradabilities of polyester wastewater with and without interior microelectrolysis pretreatment were both measured, given in Fig. 4. COD of polyester wastewater without interior microelectrolysis pretreatment decreased quickly at the first beginning 6 days, and COD removal was 42.9% 6 days later. After that, COD reduced slowly and COD removal was 56.7% 16 days later, and it seems that COD was almost constant after 16 days. However, COD of polyester wastewater decreased more quickly after interior microelectrolysis pretreatment. COD removal was 68.5% 6 days later and 78.2% 16 days later. These results indicate that polyester wastewater after interior microelectrolysis pretreatment showed advantages in anaerobic biodegradabilities in comparison with polyester wastewater without any pretreatment.

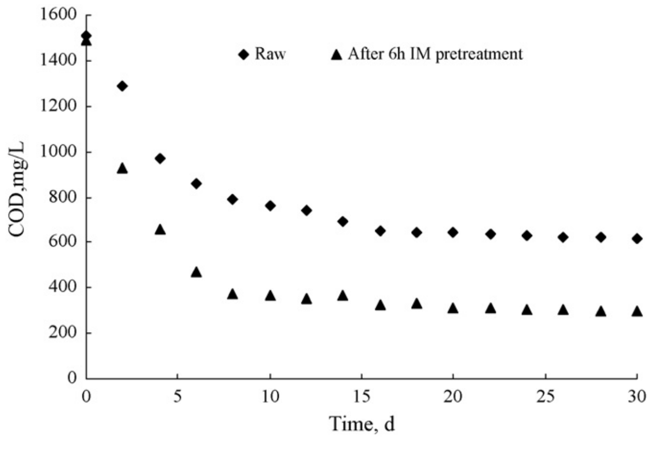

Fig. 4. COD changes in anaerobic batch tests.

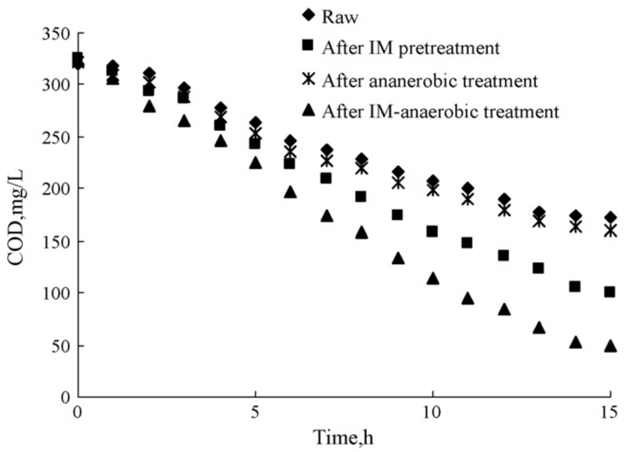

Fig. 5. COD changes in aerobic batch tests.

3.3. *Aerobic biodegradable batch tests*

Initial COD of samples were adjusted to almost the same level in aerobic batch tests for facilitating the comparison of aerobic biodegradability of polyester wastewater with and without pretreatment, given in Fig. 5.

COD removal of polyester wastewater without any pretreatment was 46.3% 15 h later, which showed poor aerobic biodegradability. Under the same operating condition, COD removal efficiency gets to 69.3% by interior microelectrolysis pretreatment, whereas is only 50.2% by anaerobic pretreatment. In contrast, COD removal can get to 84.5% through interior microelectrolysis–anaerobic pretreatment. The results imply that interior microelectrolysis–anaerobic pretreatment shows the highest COD removal efficiencies than only interior microelectrolysis pretreatment or only anaerobic pretreatment. Although interior microelectrolysis and anaerobic pretreatment have individual advantages, they are lacking of effectiveness if applied individually in treating polyester wastewater.

3.4. *IM-A/A process*

The IM-A/A process reduced COD of polyester wastewater from 3345.7–3476.9 mg/L to 76.2–257.9 mg/L, which corresponds to removal efficiencies between 92.5% and 97.7%, given in Table 3. COD in effluent decreased with the increase of the applied HRT, and COD of treated polyester wastewater could be below 150 mg/L at HRT 26 h and below 100 mg/L at HRT 32 h.

Especially, when influent COD values were at its maximum level (3800–4300 mg/L), COD in the effluent increased with most of values in the range of 102.3–147.6 mg/L at HRT 32 h. The results show that even at high loading rates, COD removal efficiency can get 96% in IM-A/A process.

From Table 4, BOD decreased after interior microelectrolysis pretreatment, but BOD/COD ratio increased from 0.27 to 0.38–0.44, which is in accord with the results from interior microelectrolysis batch tests. Moreover, when HRT of interior microelectrolysis was extended from 9 to 15 h, COD removal increased from 45.6% to 58.5% and was relatively sensitive to reaction time, but BOD/COD ratio was relatively constant (0.42–0.44). In anaerobic unit, COD removal efficiencies increased with the extension of HRT and were in the range of 51.3–66.2%. BOD/COD ratios decreased somewhat and were in the range of 0.31–0.41. Although the same HRT was kept in aerobic unit, high COD removal of aerobic unit was observed when the high COD removal was gotten in interior microelectrolysis and anaerobic unit, which imply the effects of interior microelectrolysis and anaerobic unit on total COD removal in IM-A/A process.

Table 3
COD and COD removal in IM-A/A process.

HRT (h)	COD[a] (mg/L)				COD removal (%)			Total COD removal (%)
	Influent	IM	Anaerobic	Aerobic	IM	Anaerobic	Aerobic	
20	3443.2	2207.1	1074.9	257.9	35.9	51.3	76.0	92.5
26	3345.7	1820.1	797.2	142.7	45.6	56.2	82.1	95.7
32	3476.9	1536.8	579.4	89.2	55.8	62.3	84.6	97.2
38	3353.2	1391.6	470.4	76.2	58.5	66.2	83.8	97.7

[a] 10 days average at every operating condition.

Table 4
BOD and BOD/COD ratio in IM-A/A process.

HRT (h)	BOD[a] (mg/L)				BOD/COD			
	Influent	IM	Anaerobic	Aerobic	Influent	IM	Anaerobic	Aerobic
20	929.7	849	413.2	37.3	0.27	0.38	0.38	0.14
26	903.3	800.8	326.8	15.7	0.27	0.44	0.41	0.11
32	938.8	668.4	208.6	8.0	0.27	0.43	0.36	0.09
38	905.4	590.5	144.2	6.4	0.27	0.42	0.31	0.08

[a] 10 days average at every operating condition.

Surfactant removal efficiency was 98.5% at total HRT 32 h and was 98.7% at total HRT 38 h and its concentration in treated effluent was in the range of 0.2–1.3 mg/L. The alkalinity required for buffering of polyester wastewater affects the economical feasibility of the anaerobic treatment. pH values changed near neutrality (6.25–7.04) after interior microelectrolysis pretreatment and were quite close to those optional for the adequate metabolism of bacteria. After anaerobic treatment, pH values decreased somewhat and was 5.23 at 6 h anaerobic HRT, which showed a little acidic. However, when anaerobic HRT was extended above 12 h, pH value increased and was 6.81. The effluent discharged from aerobic unit was near neutrality, in the range of 6.43–6.96.

Effect of the nutrient addition was detected according to COD removal efficiency. The initial COD:N:P ratio of polyester wastewater was 100:0.77:0.06, but after interior microelectrolysis pretreatment, COD:N:P ratio showed 100:2:0.2 due to the reduction of COD in interior microelectrolysis unit. The COD:N:P of the effluent discharged from interior microelectrolysis was modified to 100:5:1 through supplementation with $NaNO_3$ and an equimolar mixture of KH_2PO_4 and Na_2HPO_4 [11]. COD in the effluent showed a slightly lower value, but only decreased 5–10 mg/L.

The effects of IM/A process and A/A process on polyester wastewater treatment were also discussed. From Fig. 6, IM-A/A process showed the best COD removal efficiencies in comparison with IM-A process and A/A process under the same HRT. COD was below 100 mg/L in effluent under 32 h HRT by IM-A/A process, whereas in IM/A process, HRT should be 38 h due to lack of anaerobic unit. It is quite different between anaerobic bacteria and aerobic bacteria to decompose the organics in mechanisms. Contaminants are mainly removed by hydrogenation in anaerobic process, whereas by oxidation in aerobic process. The results showed that some organic substances in polyester wastewater degraded easily under anaerobic condition. In A/A process, COD in the effluent was 244.5 mg/L at HRT 32 h and 223.6 mg/L at HRT 38 h. COD in effluent decreased only 25.3 mg/L even if extended HRT 6 h, which indicated that only biological treatment was less effective in treatment of polyester wastewater and the effluents contained non-biodegradable organics. The results also implied that some non-biodegradable organics was removed by interior microelectrolysis.

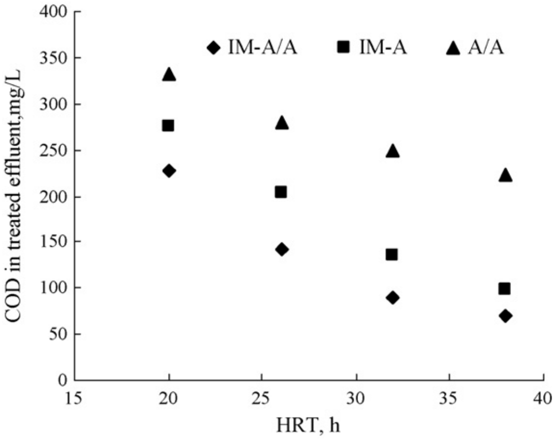

Fig. 6. COD values in final effluents in three different processes.

4. Conclusion

Interior microelectrolysis is an effective and economic pretreatment method for removing COD and surfactant agents. BOD/COD ratios increased after interior microelectrolysis pretreatment, and the biodegradability of polyester wastewater was improved. The organic contaminants were removed by the combination effects of redox reaction, flocculation and electrophoresis.

Polyester wastewater showed advantages in anaerobic biodegradabilities after interior microelectrolysis pretreatment in comparison with polyester wastewater without any pretreatment. In aerobic batch tests, interior microelectrolysis–anaerobic pretreatment showed highest COD removal efficiencies than only interior microelectrolysis pretreatment or only anaerobic pretreatment.

IM-A/A process showed the best COD removal efficiencies in comparison with IM-A process and A/A process under the same operating conditions. HRT was only 32 h by interior microelectrolysis–anaerobic–aerobic process as COD in effluent was below 100 mg/L, but HRT had to be 38 h by interior microelectrolysis–aerobic process and HRT should be above 38 h by anaerobic–aerobic process.

Acknowledgements

This work was supported by research foundation of SINOPEC. The authors acknowledge supports of Professor Penglin Jia and Professor Hong Gong.

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