

Treatment of waste cutting fluid by coagulation-anaerobic hydrolysis (with or without cosubstrate glucose)-aeration process

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ABSTRACT

A coagulation-anaerobic hydrolysis-aeration process was used to treat the waste cutting fluid, which was discharged from a metalworking plant, and the operating conditions of each unit were optimized in this paper. The results showed that 9 g/L poly-aluminum chloride and 0.3 g/L cationic polyacrylamide were added in the coagulation stage, the TOC was removed by 78.94% and the BOD₅/COD of the waste cutting fluid increased from 0.046 to 0.312 before and after coagulation. The coagulation effluent was further treated by anaerobic hydrolysis-aeration, and the TOC removal efficiencies of the biological process and the whole chemical-biological process were 92.77% and 98.48% respectively. Adding glucose as a cosubstrate into the anaerobic hydrolysis can improve the TOC removal efficiency; when the TOC content ratio of coagulation effluent to glucose solution was 7:3, the TOC removal efficiencies of the biological process and the chemical-biological process reached 97.16% and 99.40%, and the total oil removal efficiency of the whole process reached 99.99%. The effluent quality parameters of the coagulation-anaerobic hydrolysis (with cosubstrate glucose)-aeration process met the Class C limits specified in the *Wastewater Quality Standards for Discharge to Municipal Sewers* (GB/T 31962-2015); that is, the effluent COD, TN, TP and total oil were below 300 mg/L, 25 mg/L, 5 mg/L and 110 mg/L, respectively, and the effluent pH was between 6.5–9.5.

Key words: aeration, anaerobic hydrolysis, coagulation, cosubstrate, waste cutting fluid

HIGHLIGHTS

- The waste cutting fluid could be well treated by the coagulation-anaerobic hydrolysis-aeration process.
- The influence of coagulant and coagulant aid on the biodegradability of waste cutting fluid was analyzed.
- The removal efficiency of the treatment system was further improved by adding some glucose into the anaerobic hydrolysis unit.
- The traditional method had low cost and high treatment efficiency.

INTRODUCTION

Cutting fluid is a kind of machining fluid used to clean, cool and lubricate tools during the metal cutting and grinding process, it helps to improve the life cycle and function of machining equipment (Yan *et al.* 2016). After use, the cutting fluid will lose the lubrication and cooling performance, and then need to be replaced periodically, which will eventually generate a large amount of cutting fluid wastewater. Waste cutting fluid has a high organic and oil content and poor biodegradability, which is extremely harmful to the environment and human health when it is discharged directly (Srikant *et al.* 2009; Feng *et al.* 2017). In recent years, the treatment methods for waste cutting fluid have been reported, mainly including electrochemical methods (Kobyas *et al.* 2008; Chachou *et al.* 2015; Chawaloeshosiya *et al.* 2015; Xiong *et al.* 2020), advanced oxidation methods (Sánchez-Oneto *et al.* 2007; Seo *et al.* 2007; Liu *et al.* 2017; Zhang *et al.* 2017; Su *et al.* 2018) and membrane separation methods (Chen *et al.* 2019; Popović *et al.* 2019). However, these methods often require high-cost drugs and expensive equipment, beyond the economic capacity of the ordinary factory, so it is difficult to apply in practical engineering (Chiang *et al.* 2001; Emami-Meibodi *et al.* 2016; Intini & Liberti 2016; Boczkaj & Fernandes 2017).

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Anaerobic biological treatment has low energy consumption and sludge production, and can undertake high organic loads. In addition, aerobic organisms have a fast multiplication rate and short acclimation time. Therefore, some researchers have found that a combined anaerobic-aerobic process is a feasible way to treat industrial wastewater (Alayu & Yirgu 2018; Sun *et al.* 2018; Dhaouefi *et al.* 2019). However, the organics and oil in waste cutting fluid, which can reach 10^4 – 10^5 mg/L, are much higher than those of ordinary industrial wastewater. So, in order to remove a large amount of emulsified oil and some organic matter, the coagulation method was planned to pretreat the waste cutting fluid and then the biological treatment process was considered. Compared with methanogenic bacteria, hydrolytic acidifying bacteria are less sensitive to temperature, pH and toxic substances. Hence, the anaerobic metabolism of microorganisms is controlled in the hydrolysis acidification stage, which will not only shorten the operation time of the treatment process, but also be more suitable for the treatment of refractory organic wastewater such as waste cutting fluid (Perez *et al.* 2006; Shi 2011). Therefore, the waste cutting fluid was treated by the coagulation-anaerobic hydrolysis-aeration process, and the operating conditions of each treatment unit were optimized, and meanwhile the role of glucose as a cosubstrate of the waste cutting fluid in anaerobic hydrolysis unit was investigated.

MATERIALS AND METHODS

Wastewater characteristics and standard limits

The waste cutting fluid was taken from an aluminum metalworking plant in Tianjin, and its color was opaque milky white. The main characteristics of the waste cutting fluid and corresponding limits specified in the *Wastewater Quality Standards for Discharge to Municipal Sewers (GB/T 31962-2015)* are shown in Table 1.

Table 1 | Main characteristics of the waste cutting fluid and corresponding limits specified in the *Wastewater Quality Standards for Discharge to Municipal Sewers (GB/T 31962-2015)*

Parameter	Raw waste cutting fluid	Limits specified in GB/T 31962-2015		
		Class A	Class B	Class C
TOC (mg/L)	15,915–17,078	/	/	/
COD (mg/L)	49,726–50,541	500	500	300
BOD ₅ (mg/L)	2,275–2,483	350	350	150
BOD ₅ /COD (B/C, mg/L)	0.046–0.049	/	/	/
Total nitrogen (TN, mg/L)	115.51–121.43	45	45	25
Total phosphorus (TP, mg/L)	17.55–18.42	8	8	5
Total oil (mg/L)	12,951–13,053	115	115	110
pH	9.4–9.7	6.5–9.5	6.5–9.5	6.5–9.5
Total dissolved solids (mg/L)	/	1,500	2,000	2,000

"/" indicated that the limit of this parameter was not specified in *GB/T 31962-2015* or the unmeasured parameter in raw waste cutting fluid.

Analytical methods

The dissolved oxygen (DO) and pH were measured *in situ* by the portable DO meter (WTW FDO925) and pH meter (WTW SenTix940-3). The TOC, COD, BOD₅, TN, TP, total oil and total dissolved solid were analyzed according to Chinese standard methods (CEPB 2002).

Experimental procedure

Screening coagulant

Six coagulants including calcium chloride (CaCl₂), polymerized ferric sulfate (PFS), polymerized aluminum chloride (PAC), ferric chloride (FeCl₃), iron sulfate (Fe₂(SO₄)₃) and aluminum sulfate (Al₂(SO₄)₃) were selected for coagulation pretreatment with different dosages (3 g/L, 6 g/L, 9 g/L, 12 g/L, 15 g/L, 18 g/L).

350 ml waste cutting fluid and the coagulant were put into a 500 ml beaker successively, and then the beaker was placed on the ER4-6 coagulation experiment mixer with quick stirring at 200 rpm for 4 minutes, then slow stirring at 50 rpm for 5 minutes. Finally, the beaker with the mixture was settled for 2 hours. The supernatant

sample was taken from the beaker and used to analyze TOC and Zeta potential. The optimum dosages of the six coagulants and the best coagulant were determined according to Zeta potential, TOC removal efficiency, pH and B/C.

Determination of the best coagulation conditions

Four 500 ml beakers were taken and 350 ml waste cutting fluid was put into each beaker, and then the four beakers were added with the best coagulant of 3 g/L, 6 g/L, 9 g/L and 12 g/L respectively. The four beakers were placed on the ER4-6 coagulation experiment mixer. After rapid stirring at 200 rpm for 4 minutes, 0.3 g/L cationic polyacrylamide (CPAM) were added into each beaker, and then slowly stirred at 50 rpm for 5 minutes with static settling for 2 hours. The supernatant sample was taken from the beaker and used to analyze TOC, pH and B/C, and then the optimal dosage of the best coagulant with 0.3 g/L coagulant aid was determined.

The optimal dosage of the best coagulant was fixed, and the CPAM dosage (g/L), the rapid stirring speed (r/min), and rapid stirring time (min) were selected for three factors and three levels $L_9(3^4)$ orthogonal design to determine the optimal coagulation conditions. Under the optimal coagulation conditions, the TOC, COD, B/C TN, TP, total oil and pH of coagulation effluent were measured.

Acclimation of the biological reactors

Both anaerobic hydrolysis and aerobic reactors were cylindrical and made of plexiglass. The total volume and working volume of each reactor were 5 L and 3.5 L respectively. Both anaerobic hydrolysis and aerobic reactors were equipped with a stirrer to mix the activated sludge and waste cutting fluid. Moreover, an aerator was set in the aerobic reactor to oxygenate and keep DO not less than 3 mg/L. The schematic and physical diagrams of the biological treatment apparatuses are shown in Figure 1(a) and 1(b) respectively. The influent compositions of anaerobic hydrolysis reactor during each biological treatment stage are listed in Table 2.

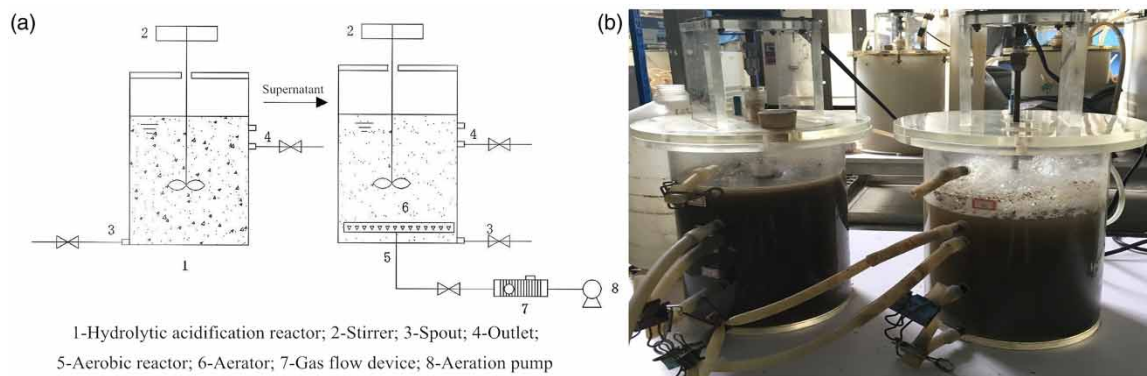


Figure 1 | Schematic and physical diagrams of the biological treatment apparatuses: (a) Schematic diagram, (b) Physical diagram.

Table 2 | Anaerobic hydrolysis reactor influent compositions in each phase

Phases	Days (d)	Volume ratio of anaerobic hydrolysis reactor influent (%)		
		Glucose solution	Coagulation effluent	
Domestication phases	Phase I	1-7 (7d)	100	0
	Phase II	8-14 (7d)	70	30
	Phase III	15-28 (14d)	50	50
	Phase IV	29-42 (14d)	30	70
	Phase V	43-56 (14d)	0	100
Stable operation phase	57-119 (63d)	0	100	
Cometabolism phase	120-147 (28d)	30	70	

The TOC concentration of glucose solution was similar to that of coagulation effluent, about 3,600 mg/L. In each phase, the anaerobic hydrolysis reactor effluent of the day was injected into the aerobic reactor as the aerobic reactor influent.

The seed sludge was acquired from the secondary sedimentation tank of a municipal WWTP in Tianjin. The anaerobic hydrolysis and aerobic reactors were operated intermittently (namely daily running 22 h, static settling 1.5 h, drainage and inflow 0.5 h).

500 ml supernatant was discharged from the anaerobic hydrolysis reactor, and then 500 ml synthetic wastewater or coagulation effluent were added into the anaerobic hydrolysis reactor, and the ratio of coagulation effluent to glucose solution in synthetic wastewater gradually increased during acclimation, which is shown in Table 2. 500 ml supernatant was discharged from the aerobic reactor too, and then the 500 ml anaerobic hydrolysis effluent of the day was added into the aerobic reactor.

Ammonium chloride and potassium dihydrogen phosphate were added as the nitrogen and phosphorus sources to keep BOD₅:N:P at 200: 5: 1 in the anaerobic hydrolysis reactor and 100: 5: 1 in the aerobic reactor (Liu 2004; Fang *et al.* 2006). The trace element solution (per L: 1.5 gFeCl₃·6H₂O, 0.15 g H₃BO₃, 0.03 gCuSO₄·5H₂O, 0.03 gKI, 0.06 g (NH₄)₆Mo₇O₂₄·7H₂O, 0.03 gMnCl₂·4H₂O, 0.12 g ZnSO₄·7H₂O and 0.15 gCoCl₂·7H₂O) was added into the anaerobic hydrolysis reactor and aerobic reactor respectively for 1 ml/L (Zhou & Peng 2015). Meanwhile NaHCO₃ was added into anaerobic hydrolysis reactor to maintain the pH between 6.5 and 7.5.

At the end of stable operation phase, the anaerobic hydrolysis and aerobic reactors were continuously operated for 24 hours. 50 ml of mixture was extracted from the anaerobic hydrolysis reactor and aerobic reactor respectively every 3 hours, and after static settling the supernatant TOC of each 50 ml mixture was measured, and then the optimum running times of biological reactors were determined by the TOC variations with time.

Cometabolism experiment

After the stable operation phase, glucose was studied as the cosubstrate. The influent of the cometabolism phase is listed in Table 2. Except that the running time was adjusted to the optimum conditions within the last 7 days of the cometabolism phase, other experimental conditions were the same as above.

RESULTS AND DISCUSSIONS

Screening coagulant

Since the chloride and sulfate ions in some coagulants may affect the accuracy of COD measurement, TOC was used as the main parameter to characterize the organic matter treatment results of the waste cutting fluid. As shown in Figure 2(a), six coagulants had different treatment effects on waste cutting fluid. In the beginning, the coagulation effluent TOC decreased with the increase of coagulant dosages. When the coagulants were overdosed, the removal effect of TOC became worse, which may be due to the restabilization of the coagulant (Zhang *et al.* 2007).

The stability of colloidal particles is related to their Zeta potential. The particles with higher absolute value of zeta potential have larger repulsive force, are not easy to settle and are relatively stable; the particles with lower absolute value of zeta potential have smaller repulsive force, and are more likely to settle. When the Zeta

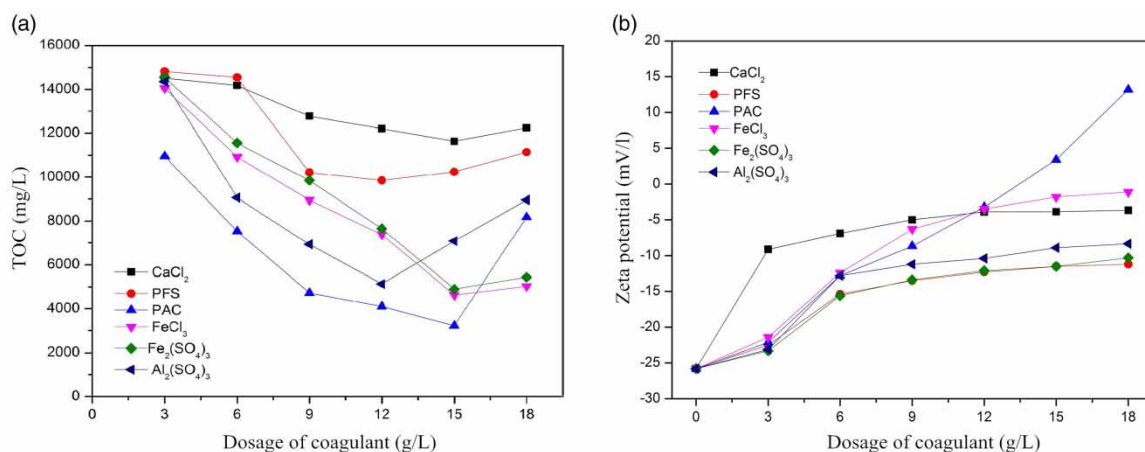


Figure 2 | TOC and Zeta potential variations under different dosages of six coagulants: (a) TOC variations; (b) Zeta potential variations.

potential is 0, the particles are the most unstable and easiest to settle (Cheng *et al.* 2012). Therefore, Zeta potential variations of six coagulants at different dosages were also measured to determine the best coagulant. As shown in Figure 2(b), the original Zeta potential value of the waste cutting fluid was -25.8 mV, which was mainly caused by a large amount of anionic surfactant in the waste cutting fluid. During coagulation, the positively charged coagulant hydrolysate gradually neutralized the negatively charged particles, and then the repulsive force between particles was reduced. Therefore, the absolute values of Zeta potential decreased with the increase of the coagulant dosages. When the coagulant dosages increased to a certain value, the change of the Zeta potential values tended to be slow, which was consistent with the TOC variations.

According to the variations of TOC and Zeta potential, the optimal dosage of each coagulant was determined. Meanwhile the TOC removal efficiency, pH, and B/C of the coagulation effluent at the optimal dosage of each coagulant are listed in Table 3.

Table 3 | Characteristics of the coagulation effluent at the optimal dosage of six coagulants

Coagulants	Optimal dosage (g/L)	pH	B/C	TOC removal efficiency (%)
CaCl ₂	15	9.102	0.102	27.3
PFS	12	6.825	0.089	38.5
PAC	15	6.301	0.491	79.8
Fe ₂ (SO ₄) ₃	15	2.657	0.114	69.3
FeCl ₃	15	2.425	0.365	71.1
Al ₂ (SO ₄) ₃	12	2.745	0.252	67.9

As shown in Table 3, coagulation pretreatment changed the pH value and biodegradability of waste cutting fluid. However, CaCl₂ is a strong acid-strong base salt, and it is ionized after being dissolved in wastewater, so it had the least effect on the coagulation effluent pH among the six coagulants.

However, Fe₂(SO₄)₃, FeCl₃ and Al₂(SO₄)₃ coagulants are strong acid-weak base salts, which perform acidity after being dissolved in wastewater, and their dosages were large, so they had a great impact on the coagulation effluent pH. In addition, the proton with positive charge neutralized the negative charge on the particle, which lowered the absolute value of the Zeta potential, promoted particle destabilization and enhanced coagulation. Therefore, the demulsification performances of the above three coagulants were better than that of CaCl₂.

During the hydrolysis of PAC and PFS, a large number of hydroxide polymers or complex ions are generated, thus PAC and PFS not only have the same salting out effect as CaCl₂ but also the adsorption effect of polymer coagulant (Tang & Luan 1997; Chawaloesphonsiya *et al.* 2018). However, the waste cutting fluid contains a lot of polycyclic aromatic hydrocarbons and heterocyclic compounds, and most of these organics have molecular weights greater than 100 ku. Under the same dosage, PAC has a better effect than PFS on the removal of macromolecular organics (Han *et al.* 2018). Therefore, B/C of PAC coagulation effluent was higher than PFS coagulation effluent, and PAC had the best effect on the removal of organics and improvement of biodegradability.

As shown in Figure 2(a), when the dosage of each coagulant was not more than 15 g/L, PAC had the best demulsification performance under the same dosage. As shown in Figure 2(a) and Table 3, when the dosage of PAC was 15 g/L, the TOC removal efficiency reached 79.8% and the B/C of the waste cutting fluid increased to 0.491, but the coagulation effluent pH was 6.301, which was lower than the optimal pH range (i.e., 6.5–8.5) for microbial growth and not conducive to subsequent biological treatment. In summary, PAC was selected as the best coagulant for subsequent experiments, and the dosage of PAC was preferably less than 15 g/L.

Optimization experiment of coagulation conditions

In order to reduce the cost of coagulant, a small amount of coagulant aid was used to reduce the PAC dosage.

As shown in Figure 3(a), under different PAC dosages, the removal efficiencies of TOC with CPAM were improved by 3% ~ 6% compared with that without CPAM. When 9 g/L PAC and 0.3 g/L CPAM were added, the TOC removal efficiency was 78.3%. When only 12 g/L PAC was added, namely without CPAM, the TOC removal efficiency was 76.0%. Therefore, adding 0.3 g/L CPAM could reduce the dosage of PAC by 3 g/L and the cost of wastewater treatment by 3.9 CNY/m³ (0.6 USD/m³; the China Yuan (CNY) cost was converted to

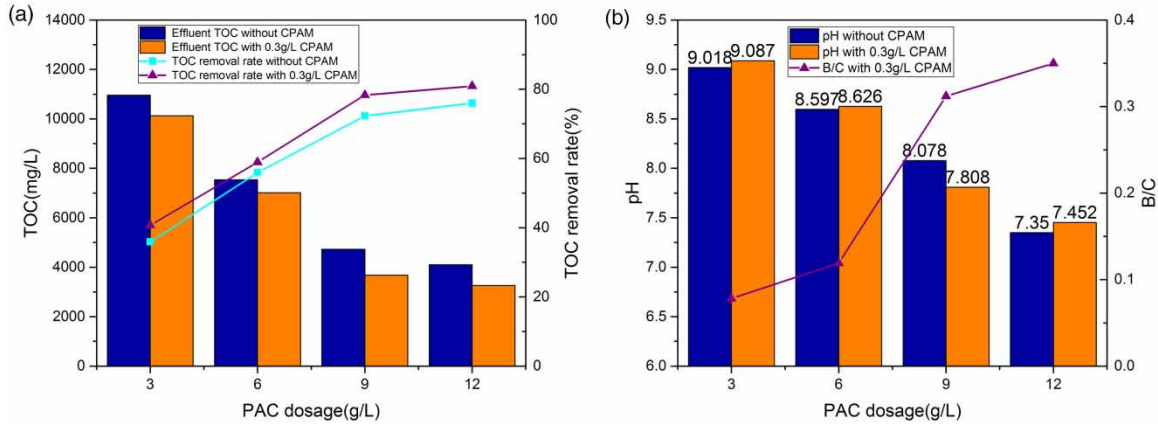


Figure 3 | The influence of CPAM on the coagulation performance: (a) coagulation effluent TOC and TOC removal efficiencies with different PAC dosages, and with or without CPAM; (b) coagulation effluent pH and B/C with different PAC dosages, and with or without CPAM.

US Dollars (USD) at the rate of 1 USD = 6.5 CNY, and the reagents prices are listed in the section Estimation of reagent cost), and meanwhile the removal efficiency of TOC would not be decreased. As shown in Figure 3(b), when 9 g/L PAC and 0.3 g/L CPAM were added, the coagulation effluent pH was 7.808 and B/C was 0.312, both of which met the growth conditions of microorganisms. Hence the dosage of PAC was selected as 9 g/L when CPAM was used as the coagulant aid.

PAC dosage was fixed at 9 g/L. The CPAM dosage, rapid stirring speed, and rapid stirring time were selected for three factors and three levels $L_9(3^4)$ orthogonal design, as shown in Table 4. The results of orthogonal design are shown in Table 5.

Table 4 | Factors and levels of orthogonal experiment

Level	Factors		
	CPAM dosage (g/L) A	Rapid stirring speed (r/min) B	Rapid stirring time (min) C
1	0.2	250	3.5
2	0.3	350	5
3	0.4	450	6.5

According to the orthogonal experiment results, the factors affecting the coagulation performance were ranked as $B > A > C$, namely rapid stirring speed > CPAM dosage > rapid stirring time. The optimal coagulation conditions were B3A2C2, namely 0.3 g/L CPAM, and quick stirring for 5 minutes at 450 r/min. The verification experiment was carried out under the optimal coagulation conditions, namely adding 9 g/L PAC and 0.3 g/L CPAM, quick stirring for 5 min at 450 r/min, slow stirring for 5 min at 50 r/min, and then settling for 2 h. The verification experiment results are shown in Table 6.

Acclimation of the biological reactors

The influent and effluent TOC and TOC removal efficiencies of the anaerobic hydrolysis and aerobic reactors are shown in Figure 4.

The domestication process was implemented for five phases (I ~ V), a total of 56 days, and each phase ran for 7 or 14 days. The proportion of coagulation effluent in the influent of anaerobic hydrolysis reactor gradually increased at each phase.

As shown in Figure 4(a), in the phase of domestication I, 500 ml glucose solution, seed sludge and trace element solution were fed into the anaerobic hydrolysis reactor on the first day, and tap water was used to make up the working volume until the total volume of mixed liquid reached 3.5 L. Therefore, because of dilution effect, the effluent TOC was low and the TOC removal efficiency was high on the first day. With the continuous

Table 5 | Results of orthogonal experiment

Experiment number	Factors			TOC removal efficiency (%)
	A	B	C	
1	Level 1	Level 1	Level 1	78.11
2	Level 1	Level 2	Level 2	79.10
3	Level 1	Level 3	Level 3	78.48
4	Level 2	Level 1	Level 2	78.78
5	Level 2	Level 2	Level 3	78.47
6	Level 2	Level 3	Level 1	77.38
7	Level 3	Level 1	Level 3	78.39
8	Level 3	Level 2	Level 1	79.67
9	Level 3	Level 3	Level 2	78.10
K1	78.56	78.23	78.39	
K2	78.72	78.29	78.66	
K3	78.21	78.78	78.45	
R	0.51	0.75	0.27	
Influencing order	B > A > C			
Optimal coagulation conditions	A2	B3	C2	

Table 6 | Coagulation pretreatment performance under the optimal conditions

Parameter	Measurement result	TOC removal efficiency (%)
TOC (mg/L)	3,595	78.94
COD (mg/L)	9,876	80.46
B/C	0.312	–
TN (mg/L)	11.17	90.8
TP (mg/L)	2.79	84.85
Total oil (mg/L)	22	99.83
pH	7.8	–

inflow of the influent every day, organic matter was continuously accumulated in reactor, and the dilution effect was weakened day by day, and then the effluent TOC gradually increased and TOC removal efficiency decreased. In the phase of domestication II, when the coagulation effluent was added as a part of the anaerobic hydrolysis reactor influent, because microorganisms did not adapt to the coagulation effluent of waste cutting fluid, the effluent TOC increased rapidly and TOC removal efficiency dropped sharply. During domestication phases III and IV, because microorganisms gradually adapted to the coagulation effluent of waste cutting fluid, the increase of effluent TOC and the decrease of TOC removal efficiency slowed down, even though the proportion of coagulation effluent in the influent increased. At the end of domestication phase II, the anaerobic hydrolysis effluent became clear. After domestication phase IV, the activated sludge in the anaerobic hydrolysis reactor became flocculent from the initial paste, and settled better. During domestication phase V, the effluent TOC of anaerobic hydrolysis reactor no longer increased, even though all anaerobic hydrolysis influent was the coagulation effluent. At the end of domestication phase V, the effluent TOC and TOC removal efficiency of the anaerobic hydrolysis reactor were about 2,700 mg/L and 24.19% respectively.

The aerobic reactor influent was the anaerobic hydrolysis effluent of the day. As shown in Figure 4(b), in the phase of domestication I, the anaerobic hydrolysis effluent TOC was increased, so the effluent TOC and TOC removal efficiency of the aerobic reactor slightly increased and decreased, respectively. As the microorganisms gradually adapt to the waste cutting fluid from domestication phase II to domestication phase IV, the aerobic

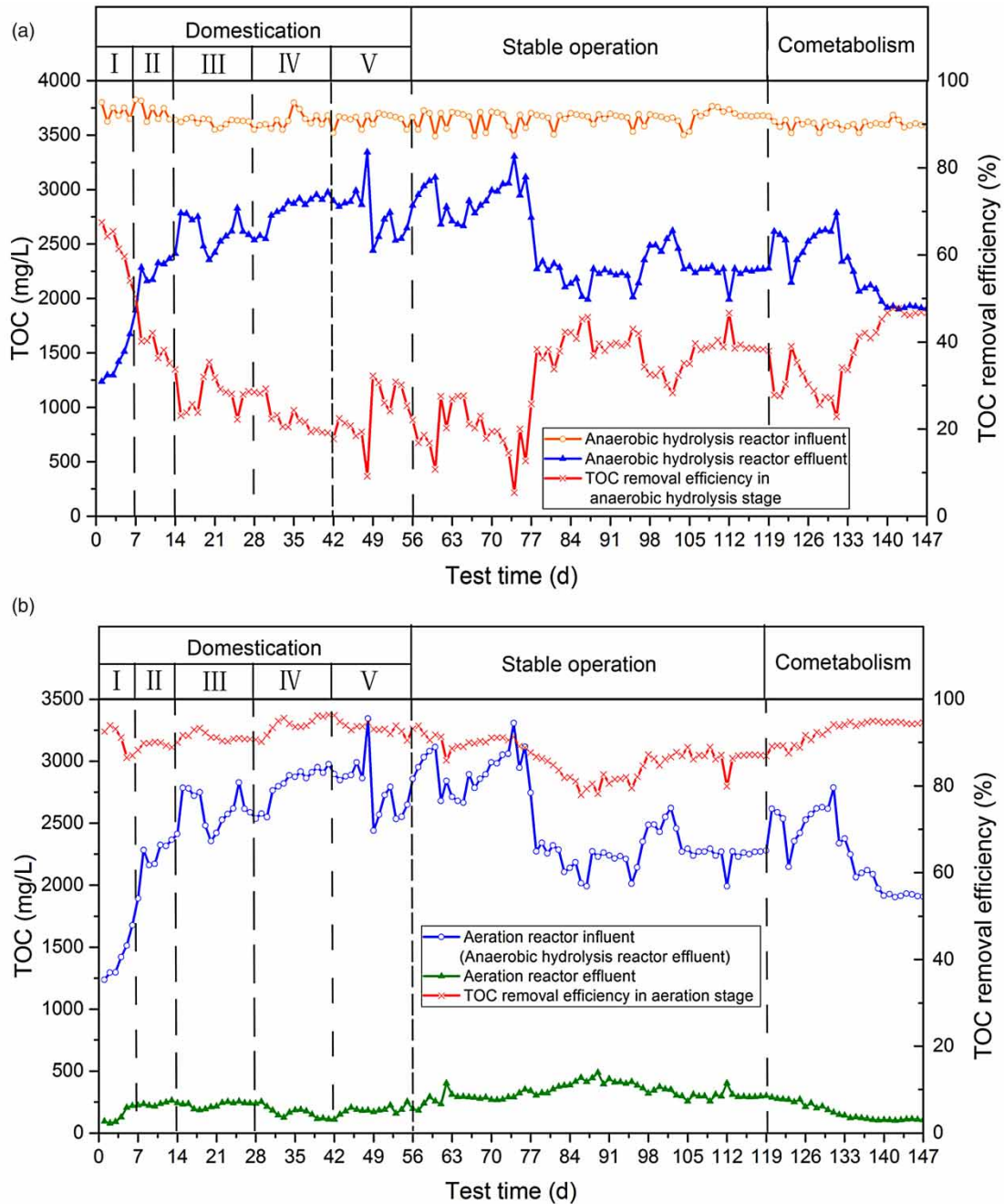


Figure 4 | TOC and TOC removal efficiencies of the biological reactors: (a) Anaerobic hydrolysis reactor; (b) aerobic reactor.

reactor effluent TOC dropped gradually and TOC removal efficiency rose step by step, even though the proportion of coagulation effluent in the anaerobic hydrolysis influent increased. However, in the phase of domestication V, due to the influent of anaerobic hydrolysis reactor being 100% coagulation effluent of waste cutting fluid, the effluent TOC and TOC removal efficiency of the aerobic reactor were greater and lower than those of domestication IV respectively. At the end of domestication phase V, the aerobic reactor effluent TOC was about 200 mg/L.

In the stable operation phase, as in domestication phase V, all influent of the anaerobic hydrolysis reactor was coagulation effluent. As shown in Figure 4(a), in the first 21 days of the stable operation phase, the anaerobic hydrolysis effluent TOC was similar to that in domestication phase V. In the next 42 days of stable operation phase, because microorganisms adapt to the coagulation effluent better, the anaerobic hydrolysis effluent TOC dropped to about 2,200 mg/L, and the TOC removal efficiency rose to about 38.8%, and the B/C of the anaerobic hydrolysis effluent was increased to 0.47. Therefore, in the anaerobic hydrolysis reactor, the refractory organics

could be hydrolyzed into small molecule biodegradable organics and part of the readily biodegradable organics was likely to be used for anaerobic methane production.

As shown in Figure 4(b), the effluent TOC and TOC removal efficiency of the aerobic reactor fluctuated less although the influent of the aerobic reactor changed greatly, which indicated that the aerobic reactor removed the organic matters effectively and had a good resistance to shock load. The aerobic reactor effluent TOC in the stable operation phase was about 260 mg/L, and the TOC removal efficiency of the aerobic reactor was about 88.18%.

The TOC removal efficiency of the biological treatment process, namely the anaerobic hydrolysis-aeration process, was 92.77%, and the TOC removal efficiency of the entire treatment process, namely the coagulation-anaerobic hydrolysis-aeration process, reached 98.48%.

At the end of the stable operation phase, within 24 h, the effluent TOC of the anaerobic hydrolysis and aerobic reactors were measured respectively every 3 h to investigate the effect of the running time on the organics removal in the biological treatment reactors.

As shown in Figure 5, the effluent TOC of the anaerobic hydrolysis and aerobic reactors had an obvious decrease in the first 12 hours, which indicated that the biodegradation was almost complete when the reactors ran for 12 hours. In the full-scale treatment plant, if the biological treatment reactor runs longer than its needs, it will not only increase the reactor volume but also increase the operation and investment costs. Therefore, the optimal running time for both anaerobic hydrolysis and aerobic reactors was 12 hours.

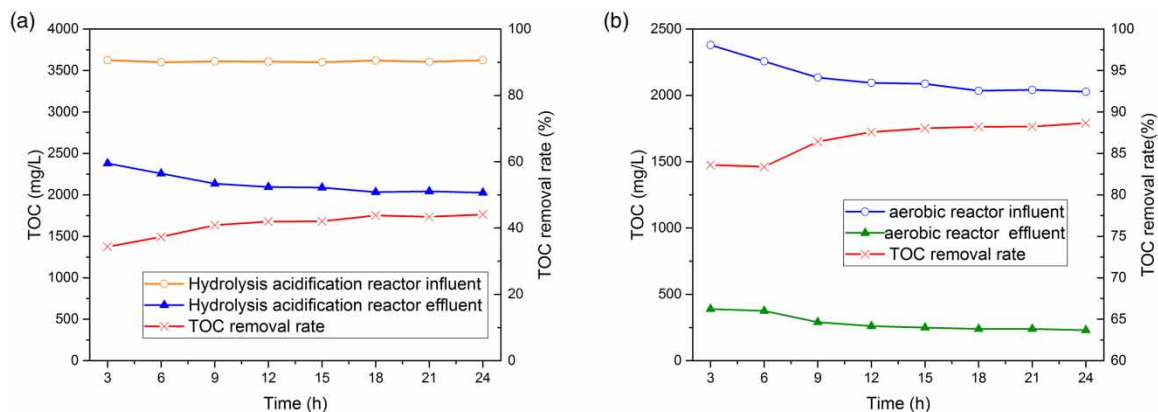


Figure 5 | Variations of effluent TOC and TOC removal efficiency with the running time in each reactor: (a) Anaerobic hydrolysis reactor; (b) aerobic reactor.

Cometabolism experiment

When the waste cutting fluid was treated by the coagulation-anaerobic hydrolysis-aeration process, in the stable operation phase, the effluent TOC of the above process was about 260 mg/L, which failed to meet the organics limit listed in Table 1. However, it was found that the effluent TOC in the domestication phase IV was lower than that in domestication phase II, III, V and stable operation phase. In the domestication phase IV, the TOC content ratio of coagulant effluent to glucose solution in the anaerobic hydrolysis influent was 7:3, which indicated that under the same influent organic load, adding some readily biodegradable organics was conducive to the refractory organic's removal. So, after the stable operation phase, glucose solution was added into the anaerobic hydrolysis influent again to verify the effect of cometabolism.

As shown in Figure 4, at the end of cometabolism phase, compared with the stable operation phase, the effluent TOC of the anaerobic hydrolysis reactor decreased from about 2,200 mg/L to about 2,000 mg/L, and the effluent TOC of the aerobic reactor decreased from about 260 mg/L to about 100 mg/L. If there was no cometabolism of glucose, and all the glucose was degraded in biological treatment reactors, the effluent TOC of aerobic reactor should be about 180 mg/L (namely, $260 \times 0.7 = 182$ mg/L) due to the dilution of coagulation effluent. Therefore, that indicated the glucose could be used as a cosubstrate in the biological treatment process of waste cutting fluid.

The waste cutting fluid was treated by the coagulation-anaerobic hydrolysis (with cosubstrate glucose) -aeration process, and both anaerobic hydrolysis and aerobic reactors were operated for 14 hours in one operational period

(namely running 12 h, static settling 1.5 h, drainage and inflow 0.5 h), and then the effluent characteristics at the end of cometabolism phase were measured and shown in Table 7.

Table 7 | Effluent characteristics by the coagulation-anaerobic hydrolysis (with cosubstrate glucose) -aeration process

Parameter	Measurement result	Total removal efficiency (%)
TOC (mg/L)	102	99.40
COD (mg/L)	271	99.46
TN (mg/L)	10.83	81.08
TP (mg/L)	1.89	89.74
Total oil (mg/L)	-	99.99
pH	7.8	-
Total dissolved solid (mg/L)	1,375	-

'-' indicated that the total oil was too small to be detected. '-' indicated that was not calculated.

As shown in Table 7, when the waste cutting fluid was treated by the coagulation-anaerobic hydrolysis -aeration process and the glucose as a cosubstrate was added into the anaerobic hydrolysis reactor with the TOC content ratio of coagulation effluent to glucose solution was 7:3, the main pollutants in the aerobic reactor effluent met the Class C limits specified in the *Wastewater Quality Standards for Discharge to Municipal Sewers (GB/T 31962-2015)*, and the removal efficiencies of TOC, COD and total oil were all over 99%.

Estimation of reagent cost

According to the dosage of PAC 9 g/L, CPAM 0.3 g/L, NaHCO₃ 10 g/L, glucose 3.86 g/L, and the unit price of PAC 2,500 CNY/tonne (384.62 USD/tonne), CPAM 12,000 CNY/tonne (1,846.16 USD/tonne), NaHCO₃ 2,500 CNY/tonne (384.62 USD/tonne), glucose 5,000 CNY/tonne (769.23 USD/tonne), the treatment reagent cost of waste cutting fluid was 70.4 CNY/m³ (10.83 USD/m³).

CONCLUSION

Compared with CaCl₂, Fe₂(SO₄)₃, FeCl₃, PFS and Al₂(SO₄)₃, PAC had the best effect on the removal of organics and could improve the biodegradability of the waste cutting fluid. The CPAM was used as coagulant aid to reduce the dosage of PAC. When the dosages of PAC and CPAM were 9 g/L and 0.3 g/L, after the coagulation pretreatment, the TOC removal efficiency was 78.94%, the total oil removal efficiency reached 99.83%, and the B/C value of the coagulation effluent increased to above 0.3.

The organic matters in the coagulation effluent could be removed by the anaerobic hydrolysis-aeration treatment process, and the biodegradability of wastewater was improved by anaerobic hydrolysis reactor, and the B/C of anaerobic hydrolysis effluent was increased to 0.47. The TOC removal efficiencies of the biological process and the whole chemical-biological process were 92.77% and 98.48% respectively.

Adding glucose as a cosubstrate into the anaerobic hydrolysis unit promoted the degradation of the refractory organics. When the TOC content ratio of coagulation effluent to glucose solution was 7:3, the effluent TOC of aerobic reactor was reduced to about 100 mg/L. Nevertheless, the average effluent TOC of aerobic reactor was about 260 mg/L without glucose as a cosubstrate. The effluent COD, TN, TP, total oil and pH of the coagulation-anaerobic hydrolysis (with cosubstrate glucose)-aeration process met the Class C limits specified in the *Wastewater Quality Standards for Discharge to Municipal Sewers (GB/T 31962-2015)*.

As the quality of waste cutting fluid produced by different metalworking plants would be different, the above treatment process could be used for the treatment of different waste cutting fluids, but the dosages of PAC and CPAM, and coagulation conditions need to optimized again.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

- Alayu, E. & Yirgu, Z. 2018 Advanced technologies for the treatment of wastewaters from agro-processing industries and cogeneration of by-products: a case of slaughterhouse, dairy and beverage industries. *International Journal of Environmental Science and Technology* **15**(7), 1581–1596.
- Boczkaj, G. & Fernandes, A. 2017 Wastewater treatment by means of advanced oxidation processes at basic pH conditions: a review. *Chemical Engineering Journal* **320**, 608–633.
- CEPB 2002 *Analysis Methods for Water and Wastewater 4th edn (Supplement Edition)*. China Environmental Science Press, Beijing, China (in Chinese).
- Chachou, L., Gueraini, Y., Bouhalouane, Y., Poncin, S., Li, H. Z. & Bensadok, K. 2015 Application of electro-Fenton process for cutting fluids mineralization. *Environmental Technology* **36**(15), 1–32.
- Chawaloeshosiya, N., Mongkolnauwarat, J., Prommajun, C., Wongwailikhit, K. & Painmanakul, P. 2015 Treatment of cutting-oily wastewater by electrocoagulation-flotation (ECF) process: modeling approach. *Environmental Engineering Research* **20**(4), 392–396.
- Chawaloeshosiya, N., Guiraud, P. & Painmanakul, P. 2018 Analysis of cutting-oil emulsion destabilization by aluminum sulfate. *Environmental Technology* **39**(11), 1450–1460.
- Chen, P., Yin, D., Song, P., Liu, Y., Cai, L., Wang, H. & Zhang, L. 2019 Demulsification and oil recovery from oil-in-water cutting fluid wastewater using electrochemical micromembrane technology. *Journal of Cleaner Production* **244**, 118698.
- Cheng, F., Qin, T., Zhao, X. Y., Sun, L. P. & Wang, X. D. 2012 Effect of coagulant dosage and hydraulic stirring speed on rainwater coagulation. *Chinese Journal of Environmental Engineering* **6**(11), 3905–3909 (in Chinese).
- Chiang, L. C., Chang, J. E. & Chung, C. T. 2001 Electrochemical oxidation combined with physical-chemical pretreatment processes for the treatment of refractory landfill leachate. *Environmental Engineering Science* **18**(6), 369–379.
- Dhaouefi, Z., Toledo-Cervantes, A., Ghedira, K., Chekir-Ghedira, L. & Muñoz, R. 2019 Decolorization and phytotoxicity reduction in an innovative anaerobic/aerobic photobioreactor treating textile wastewater. *Chemosphere* **234**, 356–364.
- Emami-Meibodi, M., Parsaeian, M. R., Amraei, R., Banaei, M., Anvari, F., Tahami, S. M. R., Vakhshoor, B., Mehdizadeh, A., Fallah Nejad, N., Shirmardi, S. P., Mostafavi, S. J. & Mousavi, S. M. J. 2016 An experimental investigation of wastewater treatment using electron beam irradiation. *Radiation Physics and Chemistry* **125**, 82–87.
- Fang, Y. F., Lin, F. K. & Lu, Z. 2006 Nutrient supplements of optimize treatment of industrial wastewater by activated sludge system. *Technology of Water Treatment* **32**(9), 15–18 (in Chinese).
- Feng, W., Yin, Y., Mendoza, M. D. L., Wang, L., Chen, X., Liu, Y., Cai, L. & Zhang, L. 2017 Freeze-thaw method for oil recovery from waste cutting fluid without chemical additions. *Journal of Cleaner Production* **148**, 84–89.
- Han, Z. R., Yu, J. J., Wang, S. P., Chen, Z. B. & Sun, L. P. 2018 Characteristics of organic substances removal from wastewater containing cutting fluid by aluminium salt and ferric salt coagulation. *Industrial Water Treatment* **38**(3), 81–85 (in Chinese).
- Intini, G. & Liberti, L. 2016 UF and RO membrane fouling during wastewater desalination in the petrochemical industry. *Desalination and Water Treatment* **57**(2), 479–486.
- Kobyas, M., Ciftci, C., Bayramoglu, M. & Sensoy, M. T. 2008 Study on the treatment of waste metal cutting fluids using electrocoagulation. *Separation and Purification Technology* **60**(3), 285–291.
- Liu, W. 2004 *Study on the Efficiency and Mechanism of High Efficient Hydrolysis-Acidogenesis Wastewater Process*. Master thesis, Jilin University, Changchun, China (in Chinese).
- Liu, Y., Wang, X. & Yu, L. 2017 Treatment of waste cutting fluid enhanced by combining emulsion-breaking coagulation with sponge iron/Fenton oxidation processes. *Desalination and Water Treatment* **67**, 140–144.
- Perez, M., Rodriguez-Cano, R., Romero, L. I. & Sales, D. 2006 Anaerobic thermophilic digestion of cutting oil wastewater: effect of co-substrate. *Biochemical Engineering Journal* **29**(3), 250–257.
- Popović, S., Karadžić, M. & Cakl, J. 2019 Optimization of ultrafiltration of cutting oil wastewater enhanced by application of twisted tapes: response surface methodology approach. *Journal of Cleaner Production* **231**, 320–330.
- Sánchez-Oneto, J., Portela, J. R., Nebot, E. & de la Ossa, E. M. 2007 Hydrothermal oxidation: application to the treatment of different cutting fluid wastes. *Journal of Hazardous Materials* **144**(3), 639–644.
- Seo, D. C., Lee, H. J., Hwang, H. N., Park, M. R., Kwak, N. W., Cho, I. J., Cho, J. S., Seo, J. Y., Joo, W. H., Park, K. H. & Heo, J. S. 2007 Treatment of non-biodegradable cutting oil wastewater by ultrasonication-Fenton oxidation process. *Water Science and Technology* **55**(1–2), 251–259.
- Shi, C. 2011 Research progress and application prospect of hydrolysis acidification. *Environmental Science and Technology* **24**(S2), 105–107 (in Chinese).
- Srikant, R. R., Nageswara Rao, D. & Rao, P. N. 2009 Experimental investigations on the influence of emulsifier content in cutting fluids on machined surface. *Proceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology* **223**(2), 195–201.
- Su, C., Cao, G., Lou, S., Wang, R., Yuan, F., Yang, L. & Wang, Q. 2018 Treatment of cutting fluid waste using activated carbon fiber supported nanometer iron as a heterogeneous Fenton catalyst. *Scientific Reports* **8**, 10650.

- Sun, F., Hu, J., Zhou, Y., Mei, R., Wang, C., He, Y. & Wu, W. 2018 High efficient alternating anaerobic/aerobic process for polyester resin wastewater treatment: performance and microbial community structure. *Biochemical Engineering Journal* **138**, 121–130.
- Tang, H. X. & Luan, Z. K. 1997 The differences of coagulating behavior and mechanism between preproduced inorganic polymeric and traditional coagulants. *Environmental Chemistry* **16**(6), 497–505 (in Chinese).
- Xiong, Y., Huang, X., Lu, B., Wu, B., Lu, L., Liu, J. & Peng, K. 2020 Acceleration of floc-water separation and floc reduction with magnetic nanoparticles during demulsification of complex waste cutting emulsions. *Journal of Environmental Sciences-China* **89**, 80–89.
- Yan, P., Rong, Y. & Wang, G. 2016 The effect of cutting fluids applied in metal cutting process. *Proceedings of the Institution of Mechanical Engineers, Part B: Journal of Engineering Manufacture* **230**(1), 19–37.
- Zhang, Z. G., Luan, Z. K., Zhao, Y., Cui, J. H., Chen, Z. Y. & Li, Y. Z. 2007 Breakage and regrowth of flocs coagulation with polyaluminum chloride (PACl). *Environmental Sciences* **28**(2), 346–351 (in Chinese).
- Zhang, Q., Yu, C. J., Fang, J., Xu, H. Y., Jiang, Q. L., Yang, S. K. & Wang, W. K. 2017 Using the combined fenton-MBR process to treat cutting fluid wastewater. *Polish Journal of Environmental Studies* **26**(3), 1375–1383.
- Zhou, L. & Peng, B. 2015 Effect of trace elements for improving the aerobic biological treatment of industrial wastewater. *Journal of Tsinghua University* **55**(6), 653–659 (in Chinese).

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