



Preparation of ceramic-corrosion-cell fillers and application for cyclohexanone industry wastewater treatment in electrobath reactor

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ABSTRACT

As new media, ceramic-corrosion-cell fillers (Cathode Ceramic-corrosion-cell Fillers – CCF, and Anode Ceramic-corrosion-cell Fillers – ACF) employed in electrobath were investigated for cyclohexanone industry wastewater treatment. 60.0 wt% of dried sewage sludge and 40.0 wt% of clay, 40.0 wt% of scrap iron and 60.0 wt% of clay were utilized as raw materials for the preparation of raw CCF and ACF, respectively. The raw CCF and ACF were respectively sintered at 400 °C for 20 min in anoxic conditions. The physical properties (bulk density, grain density and water absorption), structural and morphological characters and toxic metal leaching contents were tested. The influences of pH, hydraulic retention time (HRT) and the media height on removal of COD_{Cr} and cyclohexanone were studied. The results showed that the bulk density and grain density of CCF and ACF were 869.0 kg m⁻³ and 936.3 kg m⁻³, 1245.0 kg m⁻³ and 1420.0 kg m⁻³, respectively. The contents of toxic metal (Cu, Zn, Cd, Pb, Cr, Ba, Ni and As) were all below the detection limit. When pH of 3–4, HRT of 6 h and the media height of 60 cm were applied, about 90% of COD_{Cr} and cyclohexanone were removed.

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1. Introduction

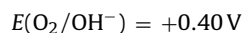
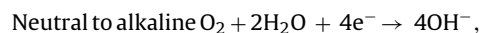
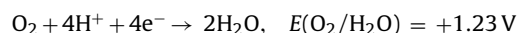
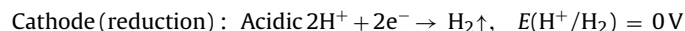
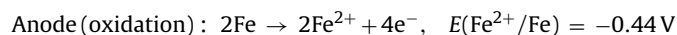
Sewage sludge, as a by-product of wastewater treatment, contains pollutants and unstable pathogen substance such as abundant organics, heavy metals and pathogenic bacteria [1–3], thus leading to potential hazards to human health and environment. Traditional treatments such as incineration, landfilling or composting were not economical, safe or sanitary [4,5]. The most widely used final disposal method of sludge is landfilling, with all the risk of soil contamination and degradation of the urban landscape [6,7]. Therefore, in order to prevent secondary pollution and convert sludge into useful resources, an effective and suitable method to treat large amounts of sludge should be found [8].

Scrap iron is a kind of solid waste from production process of machinery plants. The large quantity of scrap iron not only occupies a lot of land, but also causes environmental pollution. How to reduce environmental pollution induced by scrap iron and convert it into useful resources has been widely investigated [9–11].

Ceramics are widely used in construction industry, chemical industry, metallurgy, agriculture and environmental protection [12,13]. Sludge and scrap iron are utilized as additives in ceram-

ics production, which can reduce the usage of clay. Thus wastes can be turned into valuable substances [14].

Micro-electrolysis technology was developed on the basis of electrochemistry in Europe during the 1960s [15]. It was believed to operate on the principles very similar to electrochemical methods, and the electrons were supplied from the galvanic corrosion of many micro-scale sacrificial anodes instead of external power supply. Numerous microscopic galvanic cells were formed between the particles of iron and carbon when they were in contact with wastewater (electrolyte solution). The half-cell reactions can be represented as follows [16]:



As a kind of physical-chemical methods, micro-electrolysis technology is widely used to treat refractory wastewater [17], including pesticide wastewater [18], pharmacy wastewater [19], and dye wastewater [20,21]. Micro-electrolysis technology can be used to break the construction of organic pollutants [22] and increase the ratio of BOD₅ to COD [23] which can facilitate biological treatment, remove color of wastewater, and reduce energy

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consumption and infrastructure investment. At present, the service life of the electrobath reactor of micro-electrolysis is too short and failure of the electrobath reactor is frequent. CCF and ACF have lower bulk density and grain density, which is beneficial for the backwash process. Therefore, they are suitable to be used as fillers of electrobath reactor for micro-electrolysis.

The cyclohexanone wastewater contains hazardous substance such as cyclohexanone and cyclohexane which is harmful to the blood vessel of human body and results in coagulation necrosis of viscera and brain. Therefore, it is significant to find an effective treatment method. At present, the treatment of cyclohexanone mainly includes extraction and electrochemistry. There are few studies on the application of micro-electrolysis for cyclohexanone wastewater treatment.

This research investigated the three aspects as follows:

- (1) The possibility of novel media-ceramic-corrosion-cell fillers (CCF and ACF) used as fillers in electrobath of micro-electrolysis.
- (2) The electrobath employed in cyclohexanone industry wastewater treatment.
- (3) The optimum conditions (including pH, HRT and the height of fillers) for the wastewater treatment in terms of the removal efficiency of COD_{Cr} and cyclohexanone.

2. Materials and methods

2.1. Raw materials

Clay and dried sewage sludge (DSS) were utilized as raw materials to prepare CCF, and clay and scrap iron were used as raw materials for ACF production. Clay was obtained from a brickfield (in Zibo, Shandong Province), and dried sewage sludge and scrap iron were obtained from Jinan Wastewater Treatment Plant and Jinan Machinery Plant. Clay, DSS and scrap iron were dried at 105°C for 4 h, crushed to pass sieve No. 100 (the diameter of mesh was 0.154 mm), and stored until being used in polyethylene vessels to avoid humidification.

2.2. Preparation of CCF and ACF

The raw pellets were thermally treated according to the following three steps as shown in Fig. 1:

Step 1: Dosage, mixing and drying. Raw pellets were prepared with clay and DSS or clay and scrap iron, which were completely mixed, respectively. Then, the mixture was poured into a pelletizer (DZ-20 equipment) to produce pellets (about 7.00 wt% of water was added). Two sieves (the diameters of meshes were 5.00 mm and 6.00 mm, respectively) were used to sift the pellets, which were selected for the following treatment. Before thermal treatment,

raw pellets were stored in draught cupboard at room temperature (22°C) for 24 h.

Step 2: Sintering treatment. The dried raw pellets were transferred into electric tube furnace (KSY-4D-16, made in China) and sintered at 400°C for 20 min in anoxic conditions. The pellets were placed in the center of the heated zone.

Step 3: Cooling treatment. After the sintering process, the pellets were kept in draught cupboard until they cooled down to room temperature (22°C).

2.3. Characterization of CCF and ACF

Water absorption and bulk density were determined according to GB/T 17431.2-1998 [24]. Before the tests, sintered pellets were kept in an exsiccator (105°C) for 4 h. Then 100 g of dried pellets were put in a measuring cylinder (500 mL) and leveled completely. The bulk volume of dried pellets was determined. 200 mL of water was added into the measuring cylinder to cover the pellets completely. A dry towel was used to dry the surface of the wet pellets after 1 h, and 1 h saturated wet pellets were weighed. Water absorption and bulk density were calculated by Eqs. (1) and (2), respectively:

Water absorption

$$= \frac{\text{mass of 1 h saturated wet pellets} - \text{mass of dry pellets}}{\text{mass of dry pellets}} \times 100\% \quad (1)$$

$$\text{Bulk density (kg m}^{-3}\text{)} = \frac{\text{mass of dry pellets}}{\text{bulk volume of dry pellets}} \quad (2)$$

Grain density was determined by the dry mass (M_{dry}) and the volume of the sintered pellets (V_{grain}). Individual grain density was calculated according to the Archimedes' principle [25].

Structural and morphological analysis was conducted by scanning electron microscopy (Hitachi S-520) both in the surface and in the cross-section (Au coated).

1000.00 g of CCF (or ACF) was soaked into 1.00 L of hydrochloric acid (0.20 mol L^{-1} ; HCl: $\rho = 1.19 \text{ g mL}^{-1}$ Guaranteed Reagent (GR)) for 24 h. 1.00 mL of leach solution obtained from the supernatant was collected for leaching test of the toxic metal elements. Toxic metal concentrations (Cu, Zn, Pb, Cr, Cd, Hg, Ba, Ni, and As) of 1000.00 g of CCF (or ACF) were determined by ICP-AES (IRIS Intrepid II XSP equipment) and were compared with GB 5085.3-2007 [26].

2.4. Electrolysis reactor

An electrobath reactor was set up as shown in Fig. 2. The cylindrical reactor made from polymethyl methacrylate had a diameter of 200 mm and an effective volume of 12.5 L with a height of 1.15 m.

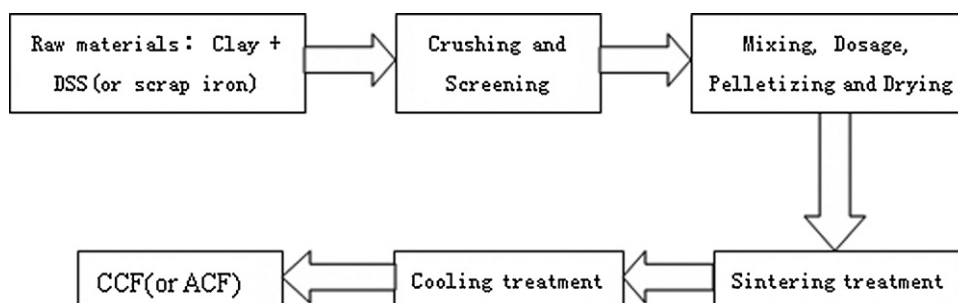


Fig. 1. Flow chart for preparation of CCF and ACF.

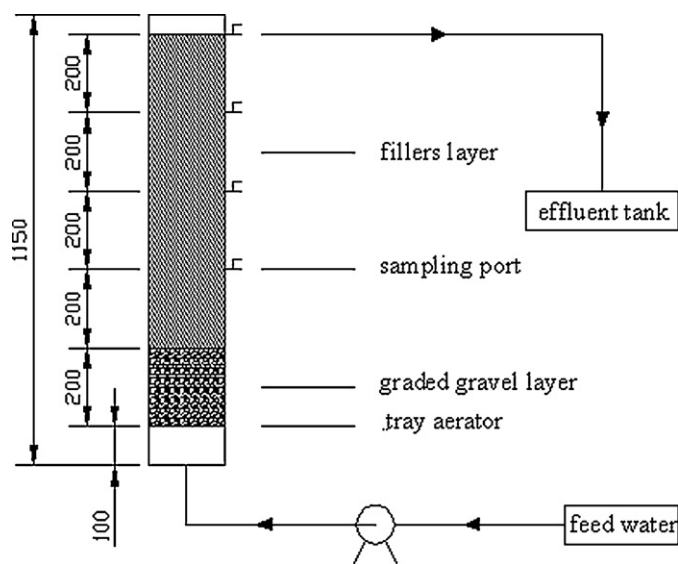


Fig. 2. Schematic diagram of experiment (dimensioning unit: mm).

CCF (12.5 L) and ACF (12.5 L) were filled in the column and the height of fillers was 80 cm, and the height of graded gravel layer was 20 cm. Tray aerator was installed at the bottom of reactor, aerated sufficiently. The raw wastewater was fed into the column with a feed pump and the treated effluent was collected by an effluent tank.

2.5. Starting and running

During the whole experiment period, the electrobath reactor was fed with cyclohexanone industry wastewater which was obtained from a chemical plant (in Dongying, Shandong Province). COD_{Cr} of the influent was about $64,000 \text{ mg L}^{-1}$, and the percentage of cyclohexanone in the influent was about 4.5 wt%.

At the start-up, the electrobath was inoculated with the cyclohexanone industry wastewater, the basic operation parameters were as follows: pH of 3, HRT of 12, and the height of fillers of 80 cm. Then pH was varied, HRT was reduced, and the height of fillers was changed, respectively. The optimum conditions were determined in terms of the removal of COD_{Cr} and cyclohexanone.

2.6. Analytical methods

The concentrations of COD_{Cr} in influent and effluent were measured by standard methods [27]. The concentrations of cyclohexanone in influent and effluent were measured by GC–MS (GC/MS-2010QP). Other parameters such as pH, temperature and dissolved oxygen, were monitored regularly. All measurements of each sample were determined in three replicates. The system was operated at room temperature ranging from 25.2 to 30.5 °C.

Table 1
Properties of CCF and ACF.

Ceramics	Bulk density (kg m^{-3})	Grain density (kg m^{-3})	Water absorption (wt%)
CCF	869.0	1245.0	12.5
ACF	936.3	1420.0	14.4

3. Results and discussion

3.1. Properties of CCF and ACF

The physical properties of CCF and ACF were shown in Table 1. It indicated that CCF and ACF had low bulk density and water absorption, and grain density was a little higher than density of water which was beneficial for backwashing and could prevent failure of the electrobath reactor.

Table 2 showed the results of the toxic metal leaching test of CCF and ACF. It revealed that all the nine metal (Cu, Zn, Pb, Cr, Cd, Hg, Ba, Ni, and As) concentrations in lixivium were much lower than the limits of the national standards specified by GB 5085.3–2007, China (Hazardous Wastes Distinction Standard–Leaching Toxicity Distinction). It can be seen that CCF and ACF used as fillers for wastewater treatment will not cause secondary pollution.

The appearance and microstructure of CCF and ACF ((A) surface of CCF, (B) fracture surface of CCF, (C) surface of ACF, (D) fracture surface of ACF) were shown in Fig. 3. It can be seen from Fig. 3A and C that the surfaces of CCF and ACF were rough and some small apertures contributed to the rough surface, which could increase their specific surface area, meanwhile increase the probability of fillers contacting with wastewater. Fig. 3B and D revealed that there were some large apertures among the frameworks of CCF and ACF, wastewater could easily flow into the fillers, which increased the time for wastewater contacting with fillers. The analysis above indicated that CCF and ACF utilized as fillers in this electrobath for wastewater treatment could increase pollutant removal efficiency.

3.2. Influence of pH on removal of COD_{Cr} and cyclohexanone

pH was a crucial parameter in micro-electrolysis process. Ten pHs (1–10) were selected and other conditions were as follows: HRT of 12 h and air–liquid ratio (A/L) of 1:1.

Fig. 4 showed the effect of pH on removal of COD_{Cr} and cyclohexanone. It can be seen that both the removal efficiency of COD_{Cr} and cyclohexanone decreased gradually as pH increased. When pH increased from 1 to 4, the removal efficiency of COD_{Cr} and cyclohexanone was almost constant, which reached about 90%. When pH increased from 4 to 10, the removal efficiency of COD_{Cr} and cyclohexanone decreased rapidly, which was lower than 10% as pH was 10. It can be deduced that acidic condition was beneficial for micro-electrolysis process, and that the removal efficiency was higher when pH was lower. It may be due to that the electrode voltage in an acidic condition (+1.23 V) was higher than that in a neutral or alkaline condition (+0.40 V), which enhanced micro electrolysis

Table 2
Toxic metal leaching tests of CCF and ACF.

Toxic metal	Contents (mg kg^{-1} of CCF)	Contents (mg kg^{-1} of ACF)	Threshold (mg kg^{-1} of hazardous waste)	Toxic metal	Contents (mg kg^{-1} of CCF)	Contents (mg kg^{-1} of ACF)	Threshold (mg kg^{-1} of hazardous waste)
Total Cu	0.05	0.07	100.00	Total Hg	–	–	0.10
Total Zn	0.03	0.01	100.00	Total Ba	0.03	0.05	100.00
Total Cd	0.01	–	1.00	Total Ni	–	–	5.00
Total Pb	0.07	0.03	5.00	Total As	–	0.01	5.00
Total Cr	–	0.02	15.00				

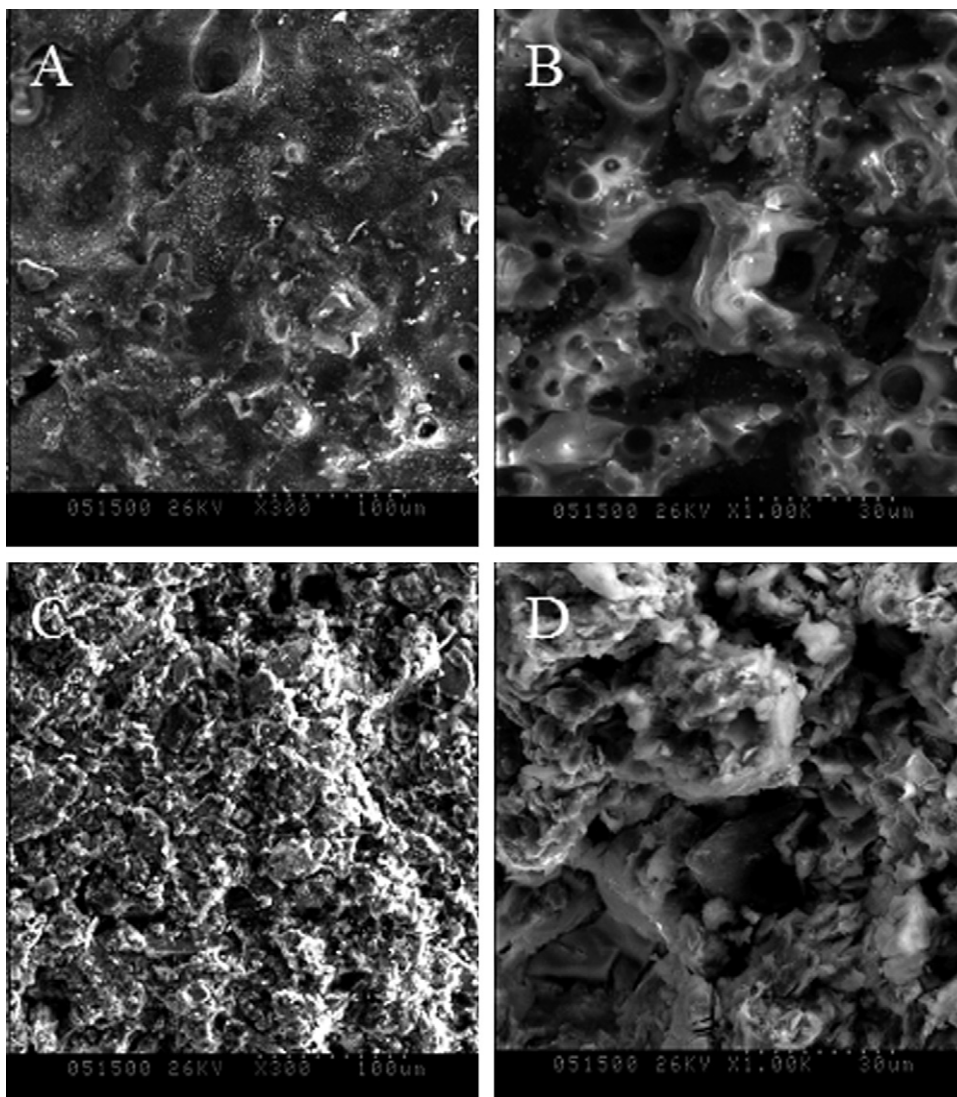


Fig. 3. The appearance and microstructure of CCF and ACF (SEM): (A) surface of CCF, (B) fracture surface of CCF, (C) surface of ACF, (D) fracture surface of ACF.

reactions and accelerated the dissolving of iron. As a result, more energy was provided which was beneficial for micro-electrolysis process [28]. However, the speed of micro electrolysis reactions and the dissolving of iron decreased rapidly as pH increased, which

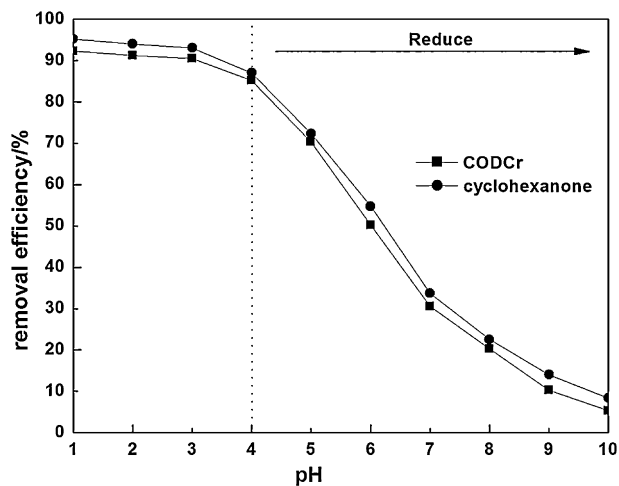


Fig. 4. Influence of pH on removal of COD_{Cr} and cyclohexanone.

led to the decrease of the removal efficiency of COD_{Cr} and cyclohexanone. Especially, when pH of wastewater was higher than 7, the removal efficiency of COD_{Cr} and cyclohexanone was much lower than that in lower pH, it may be that micro electrolysis reactions were greatly inhibited in basic solution [29].

When pH of wastewater was too low, there would be some disadvantages such as erosion. Overall, in order to ensure the removal efficiency of COD_{Cr} and cyclohexanone and to reduce cost of wastewater treatment, pH of 3–4 was selected as the optimum pH.

3.3. Influence of HRT on removal of COD_{Cr} and cyclohexanone

HRT was a key influence factor in micro-electrolysis process. According to the effective volume of the column, ten HRTs (0.5, 1, 2, 3, 4, 5, 6, 8, 10 and 12) were selected and determined. Other conditions were as follows: pH of 3–4 and A/L of 1:1. The flow rate of raw wastewater ranged from 1.0 to 25.0 L h⁻¹.

Fig. 5 showed the influence of HRT on the removal of COD_{Cr} and cyclohexanone. It was shown that the removal efficiency of COD_{Cr} and cyclohexanone increased gradually as HRT increased. When HRT increased from 0.5 h to 6 h, the removal efficiency increased rapidly, which reached about 90% as HRT was 6 h. When HRT increased from 6 h to 12 h, the removal efficiency was almost

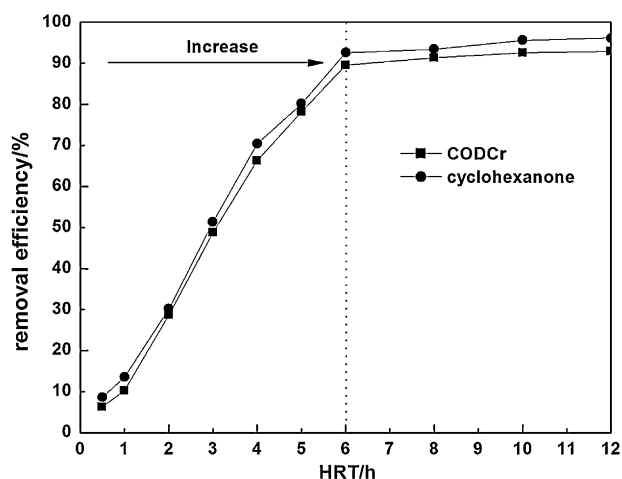


Fig. 5. Influence of HRT on removal of COD_{Cr} and cyclohexanone.

constant. It can be deduced that the wastewater was still in acidic condition, which could still enhance micro electrolysis reactions and accelerate the dissolving of iron. Moreover, when pH of wastewater was higher than 4, Fe³⁺ mainly precipitated as Fe(OH)₃, the flocculation precipitation process from ferric hydroxides would result in further removal of organic contaminants [30]. Therefore, micro-electrolysis process was more sufficient as HRT increased, which led to the increase of removal efficiency of COD_{Cr} and cyclohexanone as HRT increased from 0.5 h to 6 h. However, the influence of HRT on the removal of COD_{Cr} and cyclohexanone was insignificant when HRT was too long. It may be due to that when HRT was longer than 6 h, the wastewater has reached the alkaline condition, where micro electrolysis reactions and the dissolving of iron were greatly restrained, and flocculation precipitation process from ferric hydroxides was also greatly inhibited. Therefore, the removal efficiency of COD_{Cr} and cyclohexanone was almost constant as HRT was longer than 6 h.

Overall, considering the removal efficiency of COD_{Cr} and cyclohexanone and investment in wastewater treatment, HRT of 6 h was a better choice than other HRTs.

3.4. Influence of height of fillers on removal of COD_{Cr} and cyclohexanone

The effect of height of fillers on removal of COD_{Cr} and cyclohexanone was investigated at pH of 3–4, HRT of 8 h and A/L of 1:1. Four heights of fillers (20 cm, 40 cm, 60 cm, and 80 cm) were selected.

The results of the effect of height of fillers on the removal of COD_{Cr} and cyclohexanone were shown in Fig. 6. It revealed that the removal efficiency of COD_{Cr} and cyclohexanone increased rapidly as the height of fillers increased, which reached about 90% when the height was 60 cm. The removal efficiency was almost constant when the height increased from 60 cm to 80 cm. It can be inferred that increasing the height of fillers could increase HRT, which was beneficial for micro-electrolysis process. When media height was low, the contact time between wastewater and fillers was short, and the wastewater was still in acidic condition, where micro electrolysis reactions and flocculation precipitation process from ferric hydroxides still went on, therefore the removal efficiency of COD_{Cr} and cyclohexanone increased as media height increased from 0 to 60 cm. However, when media height reached 60 cm, the contact time between wastewater and fillers was too long, and the wastewater was already in alkaline condition, where micro electrolysis reactions and flocculation precipitation process from ferric hydroxides were inhibited greatly, therefore, the removal efficiency of COD_{Cr} and cyclohexanone almost did not increase as media height

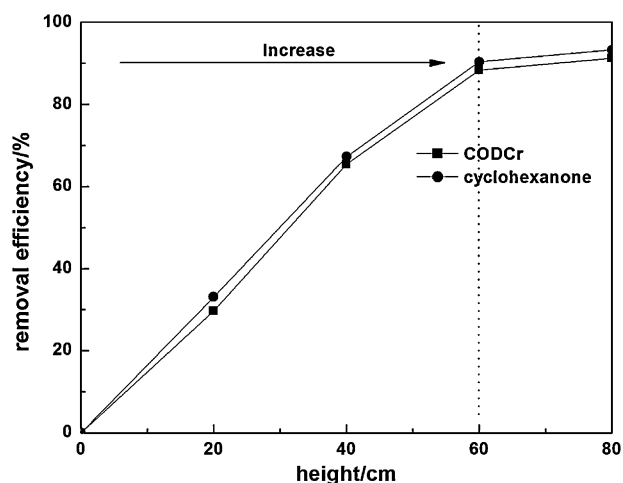


Fig. 6. Influence of height of fillers on removal of COD_{Cr} and cyclohexanone.

was higher than 60 cm. Overall, 60 cm should be selected as the optimum height of fillers.

During the whole experiment, attention should be paid to the fact that the removal of COD_{Cr} was almost the same as that of cyclohexanone. It can be deduced that cyclohexanone in the wastewater was almost degraded entirely in the electrobath. The results indicated that the removal efficiency of COD_{Cr} and cyclohexanone was satisfactory for the wastewater treatment when CCF and ACF were utilized as fillers in the electrobath.

3.5. Influence of aeration on removal of COD_{Cr} and cyclohexanone

Aeration played a key role in micro-electrolysis process. The experiment was performed at pH of 3–4, HRT of 6 h and the height of fillers of 60 cm. The result revealed that the removal efficiency of COD_{Cr} and cyclohexanone in aerobic condition reached about 90%, while in the anaerobic condition it was less than 30%. It may be due to that the electrode voltage in aerobic condition (+0.4–1.23 V) was much higher than that in anaerobic condition (0 V), and more energy was provided when oxygen participated in the cathodic reaction, which was beneficial for micro-electrolysis process [31]. Overall, aeration was selected as the experimental condition.

3.6. Backwashing

The backwashing operation for the electrobath was water backwashing, and this step was continued for 30 min. According to the effective volume of the column, the superficial velocities of backwash water (40.5 m h⁻¹) were determined. And the electrobath was backwashed every three days. As bulk density, grain density and water absorption of CCF and ACF were low, hydraulic resistance of backwash water in backwashing operation was small. It can be deduced that fillers with low bulk density, grain density and water absorption were easily moved, which was beneficial for backwashing. And failure of the electrobath reactor did not occur during the whole experiment.

4. Conclusions

As new media, ceramic-corrosion-cell fillers (CCF and ACF) employed in electrobath were investigated for cyclohexanone industry wastewater treatment. The results were as follows:

- (1) The feasibility of DDS and scrap iron as raw materials of ceramics was verified according to the properties of CCF and ACF.

- (2) CCF and ACF used as fillers in micro-electrolysis process for cyclohexanone industry wastewater treatment were feasible, which could prevent failure of the electrobath reactor.
- (3) The optimum conditions for wastewater treatment were pH of 3–4, HRT of 6 h, height of fillers of 60 cm and in aerobic condition.

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