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Electrochemical Advanced Treatment of Organics from Coking Wastewater Using Three-Dimensional Electrodes

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Saturated coke, as filler materials, was applied to advanced treat coking wastewater by three-dimensional fixed bed electrode reactor. It was indicated that coke powder could be used as catalytic electrode, and optimized conditions were of electrolysis time 1 h, current density 8A, particle size 10-20 mesh number, dosing quantity 400 mL, plate spacing 1 cm, pH 3, the removal efficiency of COD was up to 70 %. Meanwhile, the degradation mechanism of COD in the experiment was also carried out and obeyed the first order dynamics.

Key Words: Coking wastewater, Coke, Three-dimensional electrode, Advanced treatment.

INTRODUCTION

Coking wastewater is a high concentration industrial wastewater with complicated composition, high toxicity and decomposition bio-refractory, which is generated by coking industry[1-3]. It contains large amounts of organic pollutants, such as phenols, polycyclic aromatic hydrocarbons, chlorinated organic compounds and heterocyclic compounds and cause great harm to the environment[4].

As a new type of electrochemical processing, three-dimensional electrode technology has been paid more and more attention in the wastewater treatment field for the advantages of larger specific surface area, better transfer effect, higher current efficiency and space time yield[5-7]. Most academic studies have focused on reaction mechanism and determination of reaction intermediate, rather studies on the influence of various factors on experimental effect are relatively seldom[8-10]. Therefore by taking coking wastewater as the research object and using single factor experiment, the author analysis COD treatment efficiency in coking wastewater and the effects of various factors on treatment, providing the basis for the popularization and application of this technology[11]. Therefore, this experiment use coke as three-dimensional electrode filler material, so that the coke recycling can be reached and discharge to reached standard of coking wastewater can be realized for circular economy[12].

EXPERIMENTAL

Coke and coking wastewater: Water for this study was fed from secondary sendimentation tank in a coking plant in Zhangjiakou, Hebei, China. The composition of the coking wastewater used in all experiments and recycling targets are shown in Table-1. The coke was fine coke, taken from a coking plant coke bin in Shanxi.

TABLE-1
RAW WATER QUALITY AND RECYCLED WATER INDEXES

Characteristics	COD (mg/L)	Ammonium nitrogen (mg/L)	Chroma (mg/L)	pH
Raw water quality	260.98	132.246	200	6.3

Electrolytic system: The electrolytic cell was made of synthetic glass with a dimension of 140 mm × 90 mm × 130 mm and an effective volume of 1.3 L (Fig. 1). Stainless steel plates (130 mm × 65 mm × 1 mm) and Ti/RuO_2-IrO_2 plates (130 mm × 65 mm × 1 mm) were used as cathodes and anodes, respectively. A pump offered air at 0.36 m^3/h and a DC power supply (MPS702, Beijing, China) provided constant currents and the corresponding voltages. During the experiments, samples were drawn with apinoid needle tube from the cell at every ten minutes and were analyzed.

Methods: Before the test, with a focus in acid-base solution for degreasing and washed with distilled water, place the coke in drying oven at 105 ºC for 12 h, cooled and then focus powder to screening. In the experiment, put the coke powder into the raw water for a long term immersion firstly and periodic determinate COD of coking wastewater after soaking, then add the raw water after removing the filtrate until variation of COD content in the coking wastewater before and after soaking

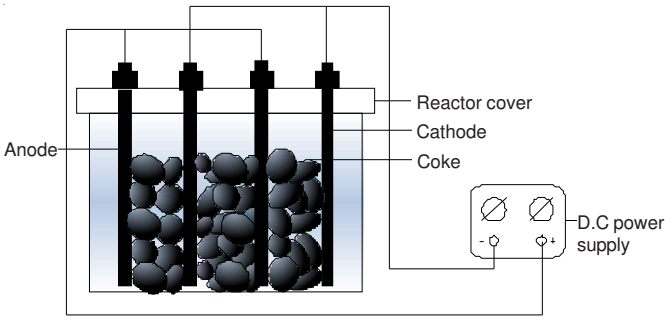

Fig. 1. Schematic diagram of the experimental apparatus

is small. So the adsorption of coke powder could be saturated and its adsorption properties are removed. Take 1 L coking wastewater into the reactor with coke powder between electrodes, collect sample at different time points into a dry beaker, with the organic filter membrane removing insoluble matter, then detect the COD. Detect COD by potassium dichromate method.

RESULTS AND DISCUSSION

Influence of electrolytic current: As shown in Fig. 2, the removal rate of COD could reach to 73 % by the electrolysis when the current was 8 A and increased quickly with increasing of current density when the current was less than 8 A, but it was decreased if the current exceeds 8 A with the increase of temperature. This is because along with the enhancement of the current, the reaction driving-power and the electric energy consumption increased, as well as the removal rate of COD. But there exist removal and energy inputs requirement extreme for pollutants in fixed bed. For the input energy is under the extreme circumstance, there is a higher removal efficiency and better redox reaction, with the input energy increases. On the contrary, with the increasing of input energy, instead of removal efficiency increasing, adverse reactions happened and the current efficiency decreased. Thus the proper current of the experiment is 8 A.

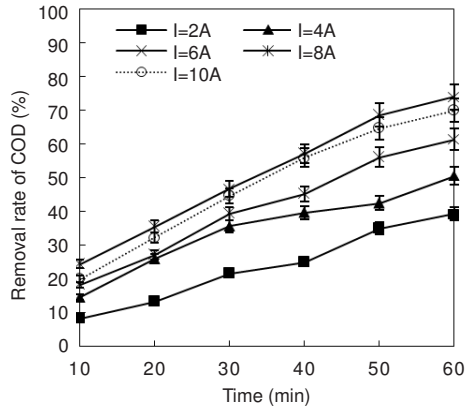

Fig. 2. Influence of electrolytic current on COD removal rate (electrolysis time = 1 h, plate spacing = 1 cm, particle size = 10-20 mesh number, pH = 3, dosing quantity = 400 mL)

Influence of dosing quantity: As shown in Fig. 3, the removal rate of COD increased firstly and then decreased, while the dosing quantity of coke powder increased and reached a maximum when the dosing quantity was 400 mL. As the dosing

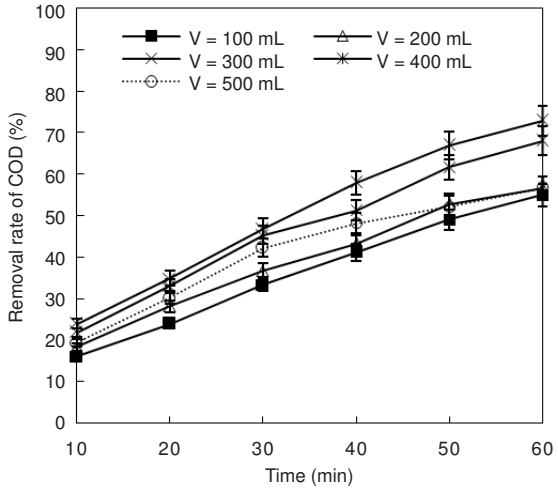

Fig. 3. Influence of dosing quantity on COD removal rate (electrolysis time = 1 h, current density = 8 A, plate spacing = 1 cm, particle size = 10-20 mesh number, pH = 3)

quantity was more than 400 mL, the removal of COD decreased. It is because the more the coke powder, the larger the electrode surface area. And the more the organic molecules on electrode, the speed of degradation increased. However, excessive coke powder could lead to worse treatment effect as worked coke remained but the short circuit current increased.

Influence of pH: As shown in Fig.4, the removal rate of COD reached a maximum *ca.* 73 % when the pH was 3. The cathode generated the most $^{\bullet}OH$ and H_2O_2 at pH 3. Lower pH leads to higher concentration of H^+ and more production of H^+ with the decrease of $^{\bullet}OH$ and H_2O_2. But higher pH with lower concentration of H^+ could not support enough H_2O_2. The reaction could be very active in alkaline solution. Therefore, the pH should be controlled around 3 to reduce the side reactions in the electrolysis process.

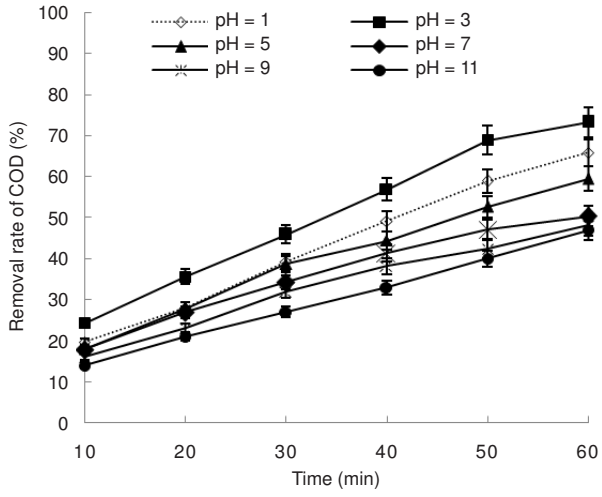

Fig. 4. Influence of pH on COD removal rate (electrolysis time = 1 h, current density = 8 A, plate spacing = 1 cm, particle size = 10-20 mesh number, dosing quantity = 400 mL)

Dynamic characteristic of coke in three-dimensional electrode: According to above method,sample at different time point and detect and ln C and time 't' curve and their linear regression equations are shown in Fig. 5,[10,11].

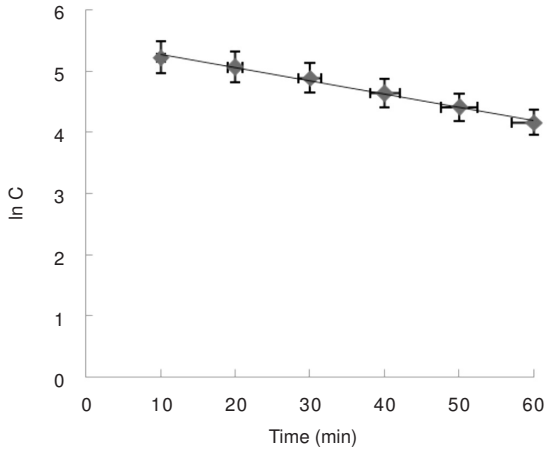

Fig. 5. Logarithm residual concentration-time linear graph

As shown in Fig. 5, the correlation coefficienct between degradation time and In C was 0.992. The degradation of COD in three-dimensional electrode obeyed the first order dynamics under the condition of the experiment.

Conclusion

The optimum technological conditions were determined by single factor test, the optimal parameters are s current 8 A, particle size 10-20 mesh, dosing quantity 400 mL, pH 3, plate spacing 1 cm and reaction time 1 h, this system could remove more than 70 % of organic matter under the optimum conditions. The degration of COD in three-dimensional electrode obeyed the first order dynamics under the condition of the experiment.

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