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Coking wastewater treatment by coagulation coupled with Fenton and hydrodynamic cavitation/Fenton

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ABSTRACT

In the present study, chemical oxygen demand (COD) was taken as the primary aspect of investigation in order to evaluate the efficiency of advanced treatment of coking wastewater by coagulation–flocculation coupled with Fenton and hydrodynamic cavitation (HC)/Fenton. The coagulation process had a COD removal rate of 71.49% with the $FeCl_3$ = 800 mg/L, pH of raw water, and no flocculant polyacrylamide (PAM) added. Further treatment of Fenton and HC/Fenton was carried out on the coagulated effluent. Using Fenton processes, it was investigated the dosage, reaction time and the influence of mole ratio of H_2O_2:Fe^{2+}. It was proven, that the molar ratio of H_2O_2:F_e^{2+} = 4:1, H_2O_2 = 50 mM, reaction for 80min, the total removal rate was 88.83%, met emission standards. HC/Fenton process was applied at the same Fenton ratio and dosage, and the total removal rate was 98.51% at 40 min, with a remaining COD = 3.4 mg/L, being almost completely removed. Fenton dosage was 30 mM for a 50 min reaction in HC/Fenton process, the obtained total removal rate being 87.91%, with a remaining of COD = 27.66 mg/L, which meets the 40 mg/L stipulated in the Emission standard of pollutants for coking chemical industry (GB16171-2012). HC/Fenton increased the reaction rate, which was three times higher than that of Fenton process, and reducing in this way the amount of applied Fenton reagent. Meanwhile, the cost-efficiency of each process is calculated, the coagulation-Fenton process cost reaching 4.205 CNY/m^3, and the coagulation-HC/Fenton cost of 7.75 CNY/m^3.

Keywords: Coking wastewater; Coagulation; Fenton; Hydrodynamic cavitation; Chemical oxygen demand

1. Introduction

Coking wastewater is toxic and harmful wastewater produced in the process of high-temperature coalification, gas purification, and chemical product refining in coking plants [1]. The coking wastewater mainly comes from the gas condensate in the initial cooling section, at the separation of water in the crude benzene section, the washing water of the used gas in the final cooling section and at the gas generating section. In the process of coke production, a large number of pollutants such as cyanides, phenol, quinolines, thiocyanate, indoles, and pyridines as well as a variety of refractory organic compounds, polycyclic aromatic hydrocarbons (PAHs), persistent organic pollutants (POPs), chemical oxygen demand (COD), etc. [1–5] would cause high toxicity of coking wastewater. These components in coking wastewater have various pollution effects on the environment, especially the phenolic compounds, which are highly toxic substances according to

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Regulation (EC) No 1272/2008 [EU-GHS-CLP] [6]. Phenols have a direct toxic effect on various biological cells/systems, leading to dizziness and insomnia, and having a corrosive effect on the mucosal epidermis, being able to cause significant harm to human beings, aquatic products, and crops [7,8]. Therefore, it is essential to treat coking wastewater.

Nowadays, there are many investigations on coking wastewater remediation. The literature showed that microorganisms can have a remarkable effect on the treatment of coking wastewater [9–12]. The most common physicochemical processes used for coking wastewater treatment are based on coagulation/flocculation, reverse osmosis [13], adsorption [14–16], and advanced oxidation processes (AOPs).

Coagulation is an easy to operate, cost-efficient, and effective method which has been successfully applied to the remediation of coking wastewater [17,18]. Selection of the suitable coagulant, determination of best operational/experimental conditions, assessments of pH effect, and investigation of optimal dosage are necessary for the optimization of the performance.

AOPs over the past years have made these processes more cost-competitive compared to the traditional water treatment processes such as activated carbon. Studies revealed that the advanced oxidation methods used in coking wastewater treatment at present mainly could include catalytic oxidation [17,18], electrochemical oxidation [19], Fenton-based and, micro-electrolysis process [20]. Among them, Fenton is widely used because of its simple operation, economic efficiency, and no secondary pollution [21]. The Fenton oxidation process refers to the reaction of H_2O_2 and Fe^{2+}(1) in an acidic solution to produce hydroxyl radicals ($^{\bullet}OH$). This is a highly oxidizing substance that is responsible for degrading pollutants [22]:

$$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^- \quad (1)$$

$k = 63\ M^{-1}\,s^{-1}$.

Chu et al. [5] used a separate Fenton process to treat coking wastewater, reacted for 1 h at pH < 6.5, and the COD removal rate was 44%–50%, which improved the biodegradability of wastewater. Jiang et al. [16] used Fenton combined with biological activated carbon process for treating coking plant effluent showing that the combined method consistently produced a final effluent of COD < 50 mg/L and total cyanide <0.5 mg/L. Thus, the combined approach of Fenton can be considered an effective method.

To reduce the use of harmful chemicals and shorten the reaction time, further improvement of treatment progress is necessary, enabling the development of a cost-efficient and viable technology from engineering for coking wastewater treatment. Co-conditioning the coking wastewater with Fenton and HC was a competitive pathway to reduce the reaction time and process costs. HC can degrade organic matter in coking wastewater in three ways: using mechanical [23], pyrolysis [24], and supercritical oxidation [25–28]. There are already some reports of the usage of HC combined with other AOPs, because when is combined with other AOPs, it has a significantly enhanced number of produced radicals, exhibiting a synergic-based performance [29]. At present, the HC/Fenton system has been demonstrated in several publications to be efficient for the elimination of organic contaminants [30–33].

Taking into account the aspects mentioned above, the purpose of this work is the investigation of a hybrid method for advanced remediation of coking wastewater to reach the legal discharge limits in China in natural waters (COD < 40 mg/L). The coking wastewater was pretreatment by a coagulation/flocculation process and then the efficiency of Fenton and HC/Fenton techniques were evaluated. The research results provided a solid foundation for the application of HC and Fenton in coking wastewater treatment.

2. Material and methods

2.1. Material

Coking wastewater after biochemical treatment was collected with a polyethylene bucket from a coking plant in Shanxi, China. The samples were stored at 4°C without any pre-treatment. Table 1 indicates the main characteristics of the wastewater used in the experiments.

Ferric chloride ($FeCl_3$), polyaluminium chloride (PAC), polyferric sulphate (PFS), polyacrylamide (PAM) are all analytically pure purchased from Sinopharm Chemical Reagent Co., Ltd., (Shanghai, China). Sodium hydroxide, sulfuric acid, ferrous sulfate, and hydrogen peroxide were all analytically pure purchased from Tianjin Kemiou Chemical Reagent Co., Ltd., (Tianjin, China).

2.2. Experimental set-up

In this work, HC equipment is shown in Fig. 1. The equipment is composed of a cistern, a booster pump (rated power 2.2 kW, rated flow 2 m^3/h, head 81 m), and regulating valves (V_1, V_2, and V_3). The cavitation generator was fitted in the main line and the flow rate of main line was controlled through the bypass line. The cistern was designed with a cooling shroud to adjust the temperature of the liquid. Pressure gauges were set to measure the inlet pressure and fully recovered pressure downstream. A flowmeter was installed on the main line to measure the flow rate. The perforated plate was used as a cavitation generator with a plate thickness of 5 mm, outside diameter of 45 mm. The plate had 22 holes with a diameter of each hole being 1.5 mm. The dimensions of the orifice plate were shown in Fig. 2, Table 2 shows the components and functions of the device.

2.3. Analytical method

In this study, pH was determined by a pH meter (ZD-2, Shanghai INESA Scientific Instrument Co., Ltd., Shanghai,

Table 1
Coking wastewater characteristics

COD (mg/L)	228.76
NH_3–N (mg/L)	1~2
pH	7.5
Total suspended solids (mg/L)	61.4

China). The COD was determined by the potassium dichromate method with a procedure from the National Standard of the People's Republic of China (GB/T 11914-1989). The COD removal rate is calculated by the following equation:

$$\mathrm{COD_{Cr}} = \left(V_0 - V_1\right) \times C \times 8 \times \frac{1{,}000}{V} \tag{2}$$

where C is the concentration of ammonium iron(II) sulfate (FAS) standard solution (mol/L); V_0 is the volume of FAS standard solution when titrating blank (mL); V_1 is the volume of FAS standard solution when titrating water sample (mL); V is the water sample volume (mL); 8 is the Oxygen (1/2O) molar mass (g/mol).

2.4. Experimental method

2.4.1. Coagulation/flocculation

Coagulant was added to the coking wastewater at room temperature. The mixture was stirred at 200 rpm for 2 min followed by 50 rpm for 30 min using a Coagulation Test Stirrer (model MY3000-6B, Wuhan Meiyu Instrument Co., Ltd., China). After stirring, the floc was allowed to settle for 30 min for sedimentation, and then the COD content was determined by sampling from 2 cm below the liquid surface. For this experiment, the effect of pH, coagulant type, and dosage was investigated on COD removal. Afterward, the coagulation effluent is prepared for the next steps of Fenton oxidation and HC/Fenton.

2.4.2. Fenton's reagent

After coagulation, coking wastewater was transferred into the Fenton oxidation reactor for further advanced treatment. The stock solution of Fe^{2+} was prepared by dissolving $FeSO_4·7H_2O$ in distilled water. In addition to the iron sulfate reagent, 30% H_2O_2 was used in the oxidation process. All Fenton experiments were carried out at room temperature. First, 500 mL coagulation treated coking wastewater was put into a beaker, and placed on the coagulation test stirrer. pH was adjusted to the ideal value using H_2SO_4 and NaOH before the start of oxidation process. $FeSO_4·7H_2O$ and H_2O_2 solutions were added into the beaker, vigorously (200 rpm) and gently (50 rpm) stirred for 1 and 60 min, respectively static precipitated, then 100 mL supernatant was taken out, and NaOH was used to adjust pH to 10. The supernatant was heated with stirring to 80°C rapidly to terminate this reaction [34]. The values of COD also were measured by the potassium dichromate method.

2.4.3. HC/Fenton

A self-designed cavitation instrument was used in this experiment. The schematic diagram of the experimental

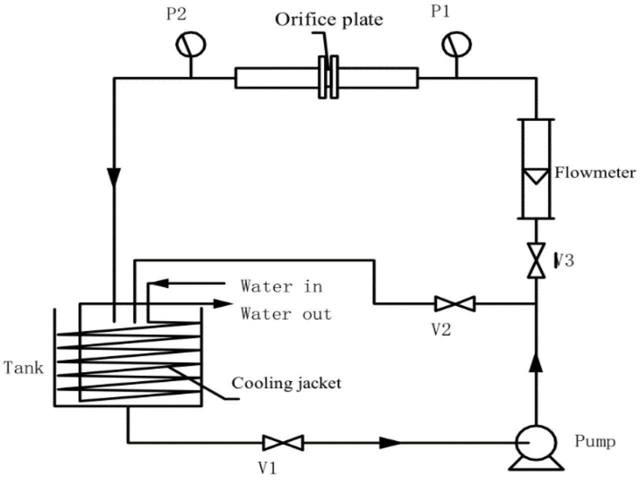

Fig. 1. Schematic diagram of HC experimental device.

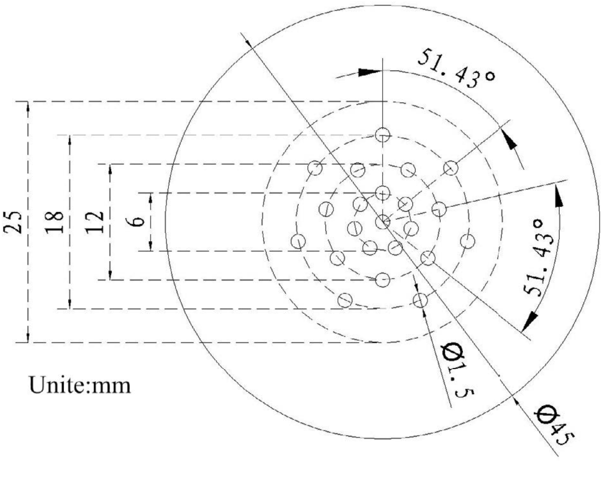

Fig. 2. Dimensions of the orifice plate.

Table 2
The components and functions of the HC device

Device composition	Model and manufacturer
Booster pump	Model: CDLF-100 (Shanghai Yuquan Pump Co., Ltd., Shanghai, China)
Water tank	Made of PVC material, dimensions 500 mm × 500 mm × 600 mm, thickness: 5 mm, capacity: 150 L
Condensing tube	Made with rubber hose
Valve	Stainless steel, DN40 ball valve
Pipeline	The main pipeline is made of stainless steel. The side piping is made of PVC and the pipe diameter is DN40.
Flow meter	Model: LWSY-25, measuring range 1–10 $m^3 h^{-1}$ (Dongtai Dongxing Instrument Factory, China)
Pressure gauge	Model: BD-801K, Range 0–1.0 Mpa (Shanghai Kaixun Technology Co., Ltd., China)

device and dimensions of the orifice plate is shown in Figs. 1 and 2, respectively. A 50 L coke plant wastewater after coagulation/flocculation was added to the cavitation water tank. The initial pH was adjusted to the desired values by H_2SO_4 and NaOH. Turned the cavitation equipment on and rotated valves to adjust the inlet pressure to optimum condition (0.4 MPa, from pre-experiment result [35]). Fenton reagents were added into the tank when the inlet pressure was stable. A condenser was used to maintain the optimum cavitation temperature at 35°C–38°C during the reaction stage. Every 10 min, 200 mL of solutions were sampled for analysis. Each treatment was repeated for three times.

3. Results and discussion

3.1. Coagulation/flocculation stage

The results of coagulant selection and optimization are shown in Fig. 3.

3.1.1. Coagulant selection and pH influence for COD reduction

The effects of COD removal by PAC, PFS, $FeCl_3$, PAC+PFS, and PAC+$FeCl_3$ are compared in Fig. 3A. At the same dosage (600 mg/L) at initial pH = 7.5, $FeCl_3$ was the most effective coagulant which the COD removal rate was 57.11%. Therefore, $FeCl_3$ was selected for coagulation in this study.

The effectiveness of COD removal with pH was evaluated at $FeCl_3$ = 600 mg/L. As shown in Fig. 3B, pH in the range of 4–9, the COD reduced with the pH increased, the maximum removal rate of 69.88% when pH = 7 and 8. When pH continued to increase, the removal rate of COD showed a downward trend. With a low pH value, organic acids are produced by a large amount of H^+ react with organic ligands in the solution, which reduced the colloidal particles adsorbed around the coagulant, and the charge neutralization and adsorption bridging mechanisms were weakened, thus, the coagulation effect is poor. Under basic conditions, the Fe^{3+} reacts with the OH^- to form $Fe(OH)_3$ or $Fe(OH)_4^-$. Fe^{3+} in the forms of $Fe(OH)_3$ and $Fe(OH)_4^-$ with neutral and negative charge loss its ability to neutralize the negatively charged colloidal system of coking wastewater [36]. At the same time, $Fe(OH)_3$ that formed by Fe^{3+} react with OH^- can promote the flocculation as a flocculant and promoted the best destabilization and aggregation of colloids. Meanwhile, in this study, in order to save the agent, pH 7.5 (original value) was selected for coagulation.

3.1.2. Effect of coagulant dosage

The COD removal efficiency was evaluated by using $FeCl_3$ with different dosages (400–1,600 mg/L). Fig. 3C shows that the COD removal was found to increase from

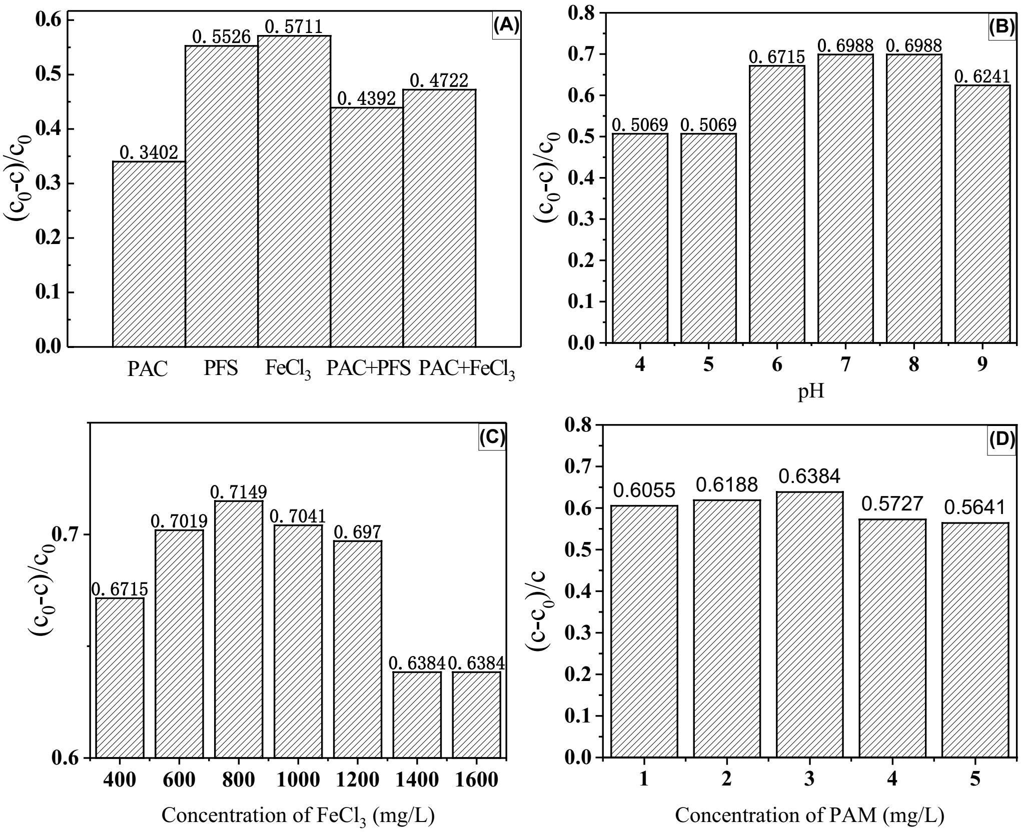

Fig. 3. (A) The effect of coagulant types on COD removal at pH = 7.5 and coagulant 600 mg/L, (B) The effect of pH on COD removal with $FeCl_3$ Conc. 600 mg/L, (C) The effect dosage of $FeCl_3$ on COD removal at pH = 7.5 and (D) The effect dosage of PAM on COD removal while pH = 7.5, $FeCl_3$ = 800 mg/L.

67.15% to 71.49% when the dosage of $FeCl_3$ was increased from 400 to 800 mg/L. The reduction COD was found to decrease to 70.41% and 63.84% when the dosage of $FeCl_3$ was further increased to 1,600 mg/L. Generally, there is an optimal dosage range in coagulation systems with compression double layer and charge neutralization as the main mechanism. Beyond this range, the removal rate decreases gradually due to the charge reversal. When $FeCl_3$ is added to the wastewater, floc was formed by the reaction of Fe^{3+} and colloidal particles with compression double layer, charge neutralization and absorption bridge, which deceased COD. When the amount of $FeCl_3$ was in excess, the colloid was stabilized again as a result of charge repulsion due to the presence of excess positively charged ions. When $FeCl_3$ was 800 mg/L, the COD removal rate was the highest. So $FeCl_3$ dosage was selected as 800 mg/L.

3.1.3. Influence of PAM dosage

PAM dosage (1–5 mg/L) was investigated at pH = 7.5 and $FeCl_3$ injection amount was 800 mg/L. From Fig. 3D, the removal effect of COD was poor after adding PAM. This is because the flocs generated by the hydrolysis of $FeCl_3$ are large and easy to settle, while the polymer chains generated by the addition of PAM will cover the colloidal particles, making the colloidal particles unable to condense and settle. Therefore, PAM was not used. In conclusion, the best coagulation condition was that $FeCl_3$ was selected as a coagulant, with pH = 7.5, $FeCl_3$ = 800 mg/L, PAM = 0 mg/L, and COD removal rate was 70.27%. After 20 repeated experiments, the final remaining COD stabilized at about 68 mg/L. Table 3 shows the water quality after coagulation.

3.2. Chemical oxidation by Fenton reagent

From Table 3, the addition of $FeCl_3$ would reduce the pH of wastewater. The pH of the coagulation effluent was 3. Numerous studies have shown that the pH of the Fenton reaction is 2–4.5 [20,37], so the pH of the wastewater is not adjusted for the oxidation experiment. The water samples used in the following experiments are all coagulation effluent (COD = 68 mg/L).

3.2.1. Influence of mole ratio of H_2O_2:Fe^{2+}

The relationship between COD removal effect and molar of H_2O_2 to Fe^{2+} ratio is shown in Fig. 4A. When $n(H_2O_2)$:$n(Fe^{2+})$ = 2:1 and 4:1, the removal rate was the best, 65.14% and 63.15%, respectively. Both of the removal rate of COD was similar. The reduction of COD decreased as Fe^{2+} was decreased [38]. The main reason is that when Fe^{2+} concentration is too low, Fe^{2+} reacts slowly with H_2O_2 and little •OH is produced. $n(H_2O_2)$:$n(Fe^{2+})$ = 4:1 is used in order to avoid excessive production of iron sludge and waste of agents.

Table 3
Comparison of water samples before coagulation and after coagulation

Scheme	COD (mg/L)	pH
Raw water	228.76	7.5
After coagulation	68	3

3.2.2. Effect of Fenton dosage and oxidation time

In Fig. 4B, with the increase of Fenton reagent concentration, the COD removal rate was increased. When the dosage of H_2O_2 was 50 mM/L (H_2O_2/COD = 0.74), the COD reduction reached 38.6%, and the COD in the water was 41.75 mg/L, close to the discharge standard of 40 mg/L. The COD content was further reduced with reaction time extended appropriately, as shown in Fig. 4C.

The COD content increased slightly before 20 min, decreased to the initial value at 40 min, then continued to decrease. Most organic compounds can be oxidized based on the detection method of COD, straight-chain aliphatic compounds can be oxidized entirely by acid potassium dichromate when Ag_2SO_4 was existed as a catalyst, while other organic compounds were more difficult to be oxidized, for example, aromatic compounds, pyridine and volatile straight-chain aliphatic compounds [39]. The reduced oxidation effects were due to the lower contact area with the vapor phase and oxidant solution. The reason for COD content increased in this experiment can be explained as undetectable COD in the raw water can be detected after oxidation treatment due to insufficient •OH production at the beginning of Fenton reaction. Large-molecule organics and polycyclic aromatic hydrocarbons in the raw water can be transformed into small molecules and chain-like organics under weak oxidizing conditions, respectively. They were oxidized by acidic potassium dichromate, and leaded to an increased COD content. •OH radicals were generated in large quantities during the stable period of the reaction, and further oxidized the small-molecule organics. Thus, the content of COD was decreased in reaction solutions. When the reaction time reached 80 min, the COD reduction was 62.41% and the content was 25.56 mg/L, which met the emission standard (40 mg/L). This is because with the extension of reaction time, the amount of •OH produced increases so that the removal rate of COD increases.

3.3. HC/Fenton process

3.3.1. HC alone process

In order to compare the effects of the single method and the combined method, we conducted a separate HC experiment without the addition of an oxidant. Interestingly, we found results that are contrary to other experiments. As shown in Fig. 5, with the increase of HC time, COD increased gradually. Wang et al. [40] used HC alone treatment of landfill leachate, COD has also seen the rise of the phenomenon.

The phenomenon can be explained as follows: the efficiency of HC is low, which produced fewer and less active free radicals. The macromolecular organics substances in coking wastewater are decomposed into lower molecule weight organics matters and the organic ring compounds are opened into chain compounds through high mechanical shear force, pyrolysis, and minimal free radical action [41]. Coking wastewater contains a variety of complex organic compounds, especially aromatic substances

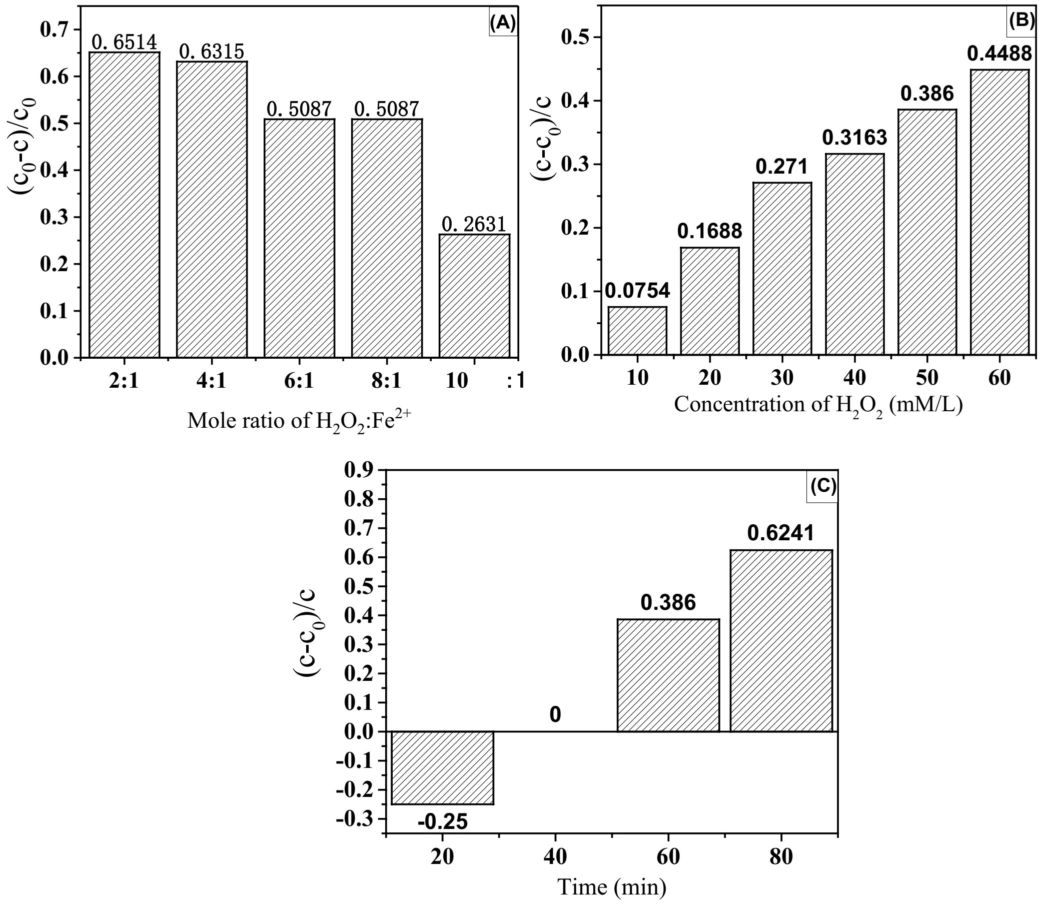

Fig. 4. (A) The effect of mole ratio of H_2O_2:Fe^{2+} on COD removal with H_2O_2 = 100 mM/L for 60 min reaction, (B) The effect of concentration of H_2O_2 on COD removal with H_2O_2:Fe^{2+} = 4:1 for 60 min reaction and (C) The effect of time on COD removal with H_2O_2 = 50 mM/L.

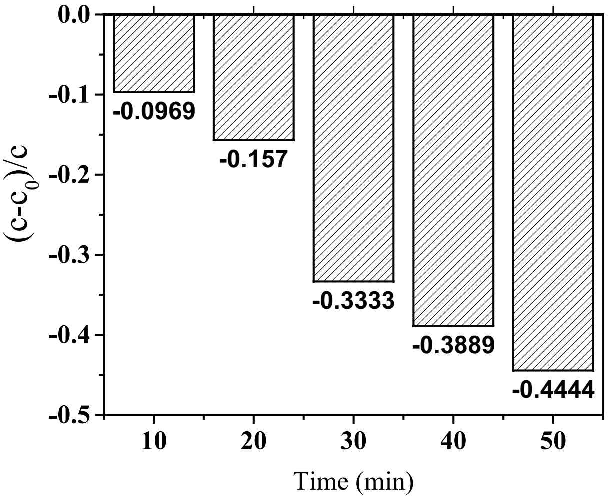

Fig. 5. The change of COD removal rate with HC alone.

with a benzene ring. These complex organic compounds were oxidized to chain hydrocarbon secondary molecules after HC treatment, which were easier to be oxidized by potassium dichromate, and increased COD level [40].

3.3.2. Effect of Fenton dosage and oxidation time in HC/Fenton process

The removal of COD by different concentrations of the Fenton reagent combined with HC was investigated. The results are shown in Fig. 6. Almost all oxidation was completed with 50 mM/L H_2O_2 at 40 min, COD reduction was 95%, and the remaining COD was 3.4 mg/L. The reduction of COD was 29.04% and 59.34% after 50 min treatment at the H_2O_2 dosage of 10 and 30 mM/L, respectively. When the adding amount was 30 mM/L, the residual COD was 27.66 mg/L, which met the emission standards. COD removal rate rose slowly after 50 min reaction, which may be due to that a small amount of Fenton was consumed completely.

The COD removal rate was significantly improved after the combination of the HC and Fenton methods, so there was a synergistic effect between the HC and Fenton process. The combination of the two methods makes up for the limitations of the single method: the micro-jet produced by HC increased the contact area between pollutants and Fenton reagent, improved the utilization rate of the Fenton reagent, and the reaction rate was accelerated. Besides, the HC itself also produced a part of •OH that could react with organic matter. Meanwhile, •OH produced by the Fenton reaction can make up for the shortage of free radicals generated by

HC. The two methods have synergistic effect and reinforce each other, so that the COD removal rate can be improved significantly.

The average reaction rate of the Fenton process alone for 80 min and the HC/Fenton process for 40 min were 0.5305 and 1.615 mg/min, separately. The reaction rate of the HC/Fenton process is about three times as fast as that of Fenton alone.

4. Cavitational yield and total treatment cost

According to the method from literatures [25,42,43], we calculated the cavitation yield. It was meaningless to calculate the cavitation yield of HC alone since the HC alone rose the COD. Therefore, we only calculated the cavitation yield of HC/Fenton. From Table 4, on the premise of ensuring high COD removal rate, the maximum cavitation yield of HC loadings of H_2O_2 50 mM/L is 5.83 mg/J × 10^{-4} mg/J, when energy utilization was highest and COD was almost completely removed.

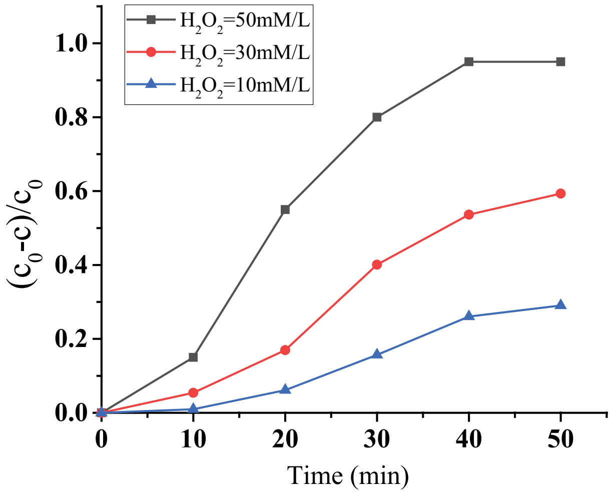

Fig. 6. The effects of different concentrations of H_2O_2 based on HC on COD removal

Table 5 shows the costs of each process. In the coagulation process, the amount of $FeCl_3$ is 800 mg/L, $FeCl_3$ (96%) cost is 3,000 CNY/t from Shandong Zhizheng International Trade Co. Ltd., (Shandong, China). The coagulation cost was 2.4 CNY/t.

Taking all these into account, the total COD reduction of coagulation-HC/Fenton process was 98.51%, and the total cost was 7.75 CNY/m^3. Although coagulation-HC/Fenton is more expensive than coagulation-Fenton, the HC/Fenton improved the reaction rate and utilization of the oxidant, thereby reducing the secondary harm to the environment caused by the excessive oxidant.

5. Conclusions

This study presents a general scheme for determining the optimal combination treatment line for coking wastewater. With COD as the evaluation index, this scheme should include coagulation studies in the first stage and then AOPs were used for chemical treatment.

In the coagulation/flocculation process, ferric chloride has the best effect on the reduction of COD from coking wastewater. The optimal conditions selected were the natural pH of 7.5 and a coagulant dosage of 800 mg/L of such reagent. The process of coagulation combined with Fenton led to higher COD reduction, but in this way, the treatment time was increased and the COD was not removed entirely.

The combination of the HC/Fenton process and coagulation pretreatment showed a COD removal of 98.51% (final COD of 3.4 mg/L) with an H_2O_2 consumption of 50 mM in 40 min. The reaction rate of the HC/Fenton process is three

Table 4
Comparison of different processes in terms of cavitational yield

Scheme	H_2O_2 Conc. (mM/L)	COD reduction (%)	Cavitational Yield × 10^{-4} (mg/J)	Energy required (kWh)
HC	–	–44.44	–	–
HC/Fenton[a]	10	29.04	1.78	0.1061
HC/Fenton[b]	30	59.34	3.64	0.0519
HC/Fenton[c]	50	95.00	5.83	0.0324

Note: [a]Electrical energy cost = 0.5 CNY/kWh; [b]H_2O_2 (30%) cost = 1,050 CNY/t from Guangzhou Jielong Chemical Co. Ltd., Guangdong, China; [c]$FeSO_4·7H_2O$ (99%) cost = 350 CNY/t from Renqiu Pengyu Chemical Co. Ltd., Hebei, China.

Table 5
Total treatment cost of each process

Scheme	COD (mg/L)	COD reduction (%)	Cost (CNY/m^3)
Raw wastewater	228.76	0	0
Coagulation	68	70.27	2.4
Coagulation-Fenton	25.56	88.83	4.205
Coagulation-HC/Fenton	3.4	98.51	7.75

times higher than that of Fenton itself. HC combined with Fenton also can significantly improve the energy efficiency. Under these conditions the coking wastewater treated can be discharged in natural water bodies.

In conclusion, the HC/Fenton process can be an economical, efficient, and low-energy consumption method, which is of great significance for practical industrial application.

Acknowledgments

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References

[1] S. Giannakis, K.A. Lin, F. Ghanbari, A review of the recent advances on the treatment of industrial wastewaters by Sulfate Radical-based Advanced Oxidation Processes (SR-AOPs), Chem. Eng. J., 406 (2021) 127083, doi: 10.1016/j.cej.2020.127083.

[2] W. Zhang, C. Wei, C. Feng, B. Yan, N. Li, P. Peng, J. Fu, Coking wastewater treatment plant as a source of polycyclic aromatic hydrocarbons (PAHs) to the atmosphere and health-risk assessment for workers, Sci. Total Environ., 432 (2012) 396–403.

[3] X. Yu, C. Wei, H. Wu, Z. Jiang, R. Xu, Improvement of biodegradability for coking wastewater by selective adsorption of hydrophobic organic pollutants, Sep. Purif. Technol., 151 (2015) 23–30.

[4] D. Guo, Q. Shi, B. He, X. Yuan, Different solvents for the regeneration of the exhausted activated carbon used in the treatment of coking wastewater, J. Hazard. Mater., 186 (2011) 1788–1793.

[5] L. Chu, J. Wang, J. Dong, H. Liu, X. Sun, Treatment of coking wastewater by an advanced Fenton oxidation process using iron powder and hydrogen peroxide, Chemosphere, 86 (2012) 409–414.

[6] S. Fodor, G. Kovács, K. Hernádi, V. Danciu, L. Baia, Z. Pap, Shape tailored Pd nanoparticles' effect on the photocatalytic activity of commercial TiO_2, Catal. Today, 284 (2017) 137–145.

[7] J.S. Melo, S. Kholi, A.W. Patwardhan, S.F. D'Souza, Effect of oxygen transfer limitations in phenol biodegradation, Process Biochem., 40 (2005) 625–628.

[8] X.-X. Wei, Z.-Y. Zhang, Q.-L. Fan, X.-Y. Yuan, D.-S. Guo, The effect of treatment stages on the coking wastewater hazardous compounds and their toxicity, J. Hazard. Mater., 239–240 (2012) 135–141.

[9] D.R. Joshi, Y. Zhang, Z. Tian, Y.X. Gao, M. Yang, Performance and microbial community composition in a long-term sequential anaerobic-aerobic bioreactor operation treating coking wastewater, Appl. Microbiol. Biotechnol., 100 (2016) 8191–8202.

[10] Y. Huang, X. Hou, S. Liu, J. Ni, Correspondence analysis of bio-refractory compounds degradation and microbiological community distribution in anaerobic filter for coking wastewater treatment, Chem. Eng. J., 304 (2016) 864–872.

[11] D. Wu, X. Yi, R. Tang, C. Feng, C. Wei, Single microbial fuel cell reactor for coking wastewater treatment: simultaneous carbon and nitrogen removal with zero alkaline consumption, Sci. Total Environ., 621 (2018) 497–506.

[12] Y. Zhao, L. Fan, D. Yang, Z. Dong, Y. Wang, X. An, Comparative study of electrochemical performance and microbial flora in microbial fuel cells by using three kinds of substrates, Chem. Res. Chin. Univ., 35 (2019) 292–298.

[13] X. Jin, E. Li, S. Lu, Z. Qiu, Q. Sui, Coking wastewater treatment for industrial reuse purpose: combining biological processes with ultrafiltration, nanofiltration and reverse osmosis, J. Environ. Sci. (China), 25 (2013) 1565–1574.

[14] I. Vázquez, J. Rodríguez-Iglesias, E. Marañón, L. Castrillón, M. Álvarez, Removal of residual phenols from coke wastewater by adsorption, J. Hazard. Mater., 147 (2007) 395–400.

[15] N. Wang, Q. Zhao, H. Xu, W. Niu, L. Ma, D. Lan, L. Hao, Adsorptive treatment of coking wastewater using raw coal fly ash: adsorption kinetic, thermodynamics and regeneration by Fenton process, Chemosphere, 210 (2018) 624–632.

[16] W.-x. Jiang, W. Zhang, B.-j. Li, J. Duan, Y. Lv, W.-d. Liu, W.-c. Ying, Combined Fenton oxidation and biological activated carbon process for recycling of coking plant effluent, J. Hazard. Mater., 189 (2011) 308–314.

[17] P. Lai, H.Z. Zhao, C. Wang, J.R. Ni, Advanced treatment of coking wastewater by coagulation and zero-valent iron processes, J. Hazard. Mater., 147 (2007) 232–239.

[18] J. Li, X. Yuan, H. Zhao, F. Li, Z. Lei, Z. Zhang, Highly efficient one-step advanced treatment of biologically pretreated coking wastewater by an integration of coagulation and adsorption process, Bioresour. Technol., 247 (2018) 1206–1209.

[19] M.S.P. Indu, K.G. Ashok, Anodic oxidation of coke oven wastewater: multiparameter optimization for simultaneous removal of cyanide, COD and phenol, J. Environ. Manage., 176 (2016) 45–53.

[20] W.-w. Liu, X.-y. Tu, X.-p. Wang, F.-q. Wang, W. Li, Pretreatment of coking wastewater by acid out, micro-electrolysis process with in situ electrochemical peroxidation reaction, Chem. Eng. J., 200–202 (2012) 720–728.

[21] P.V. Nidheesh, R. Gandhimathi, Trends in electro-Fenton process for water and wastewater treatment: an overview, Desalination, 299 (2012) 1–15.

[22] A.G. Gutierrez-Mata, S. Velazquez-Martínez, A. Álvarez-Gallegos, M. Ahmadi, J.A. Hernández-Pérez, F. Ghanbari, S. Silva-Martínez, Recent overview of solar photocatalysis and solar photo-Fenton processes for wastewater treatment, Int. J. Photoenergy, 2017 (2017) 1–27.

[23] Z. Meng-Fu, Z. Yan, D. Cheng, S. Hong-Bo, Y. Xiu-Dong, C. Ping, W. Jian-You, Application and research progress of hydrocavitation in water treatment, Environ. Sci. Technol., 33 (2010) 445–449.

[24] K.S. Suslick, S.J. Doktycz, E.B. Flint, On the origin of sonoluminescence and sonochemistry, Ultrasonics, 28 (1990) 280–290.

[25] M. Gągol, A. Przyjazny, G. Boczkaj, Wastewater treatment by means of advanced oxidation processes based on cavitation – a review, Chem. Eng. J., 338 (2018) 599–627.

[26] V.B. Manisha, R.G. Parag, Wastewater treatment using hybrid treatment schemes based on cavitation and Fenton chemistry: a review, Ultrason. Sonochem., 21 (2014) 1–14.

[27] S. Shu-Ting, Hydro-Cavitation Combined with Chlorine Dioxide to Degrade Methyl Orange, North University of China, 2016, pp. 69.

[28] K. Wei-Dian, Study on Hydro-Cavitation Enhanced Chlorine Dioxide Degradation of Phenol Wastewater, North University of China, 2017, pp. 76.

[29] Z. Mojca, K. Tina, P. Martin, D. Matevž, K. Boris, Š. Brane, B. Željko, H. Ester, Removal of pharmaceuticals from wastewater by biological processes, hydrodynamic cavitation and UV treatment, Ultrason. Sonochem., 20 (2013) 1104–1112.

[30] R.G. Parag, N.P. Pankaj, Combined treatment technology based on synergism between hydrodynamic cavitation and advanced oxidation processes, Ultrason. Sonochem., 25 (2015) 60–69.

[31] A.M. Torabi, R. Ghiaee, Decontamination of unsymmetrical dimethylhydrazine waste water by hydrodynamic cavitation-induced advanced Fenton process, Ultrason. Sonochem., 23 (2015) 257–265.

[32] G.C. Anand, R.G. Parag, E.B. Arthur, H.B. David, Industrial wastewater treatment using hydrodynamic cavitation and heterogeneous advanced Fenton processing, Chem. Eng. J., 152 (2009) 498–502.

[33] H.B. David, D.C. Stefano, G.C. Anand, C. Giancarlo, Mineralisation of 2,4-dichlorophenoxyacetic acid by acoustic or hydrodynamic cavitation in conjunction with the advanced Fenton process, Ultrason. Sonochem., 15 (2008) 416–419.

[34] S.E. Hanife, A. Omer, Final treatment of young, middle-aged, and stabilized leachates by Fenton process: optimization by

response surface methodology, Desal. Water Treat., 54 (2015) 342–357.
[35] Y. Si-jing, Hydro-Cavitation Enhanced ClO_2 to Crack the Mechanism of Methyl Orange and benzo[a]pyrene, North University of China, 2018, pp. 185.
[36] R.I. Ahmad, S.H. Fauziah, M. Sharifah, S.T. Kheng, Stabilized landfill leachate treatment by coagulation-flocculation coupled with UV-based sulfate radical oxidation process, Waste Manage., 76 (2018) 575–581.
[37] D. Yang, D.E. James, Treatment of landfill leachate by the Fenton process, Water Res., 40 (2006) 3683–3694.
[38] B. Su-Bei, Experimental Study on Advanced Treatment of Coking Wastewater by Fenton Method, Dalian Maritime University, 2020.
[39] L. Jia-Jia, Method validation of "water quality determination of chemical oxygen demand by dichromate method", Green Technol., (2019) 111–112.
[40] W. Kun, Experimental Study on Coagulation/Hydraulic Cavitation Enhanced Chlorine Dioxide Combined Treatment of Landfill Leachate COD, North University of China, 2020, pp. 92.
[41] A.P.J. Scandelai, E. Sloboda Rigobello, B.L.C.D. Oliveira, C.R.G. Tavares, Identification of organic compounds in landfill leachate treated by advanced oxidation processes, Environ. Technol., 40 (2019) 730–741.
[42] M. Gągol, A. Przyjazny, G. Boczkaj, Effective method of treatment of industrial effluents under basic pH conditions using acoustic cavitation – a comprehensive comparison with hydrodynamic cavitation processes, Chem. Eng. Process., 128 (2018) 103–113.
[43] T. Pooja, P. Mihir, R.G. Parag, Degradation of carbamazepine using hydrodynamic cavitation combined with advanced oxidation processes, Ultrason. Sonochem., 40 (2018) 567–576.