STUDY ON FENTON OXIDATION PROMOTED BY ULTRASOUND IRRADIATION FOR PULPING AND PAPERMAKING WASTEWATER

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In order to improve the degradation efficiency of pulping and papermaking wastewater, the combination oxidation process of Fenton with ultrasonic irradiation was carried out. The processes of ultrasonic irradiation alone and Fenton oxidation alone on pulping and papermaking wastewater treatment were optimized respectively. Basing on the optimum process of Fenton oxidation, the synergistic effect of ultrasonic treatment was also confirmed. Through the optimum process treatment, chroma removal rate of wastewater reached above 90%, turbidity removal rate over 80%, and COD reduced to 60 mg/L. The FTIR (ATR) spectra of wastewaters, treated by three oxidation systems above, were tested respectively. The results showed that Fenton treatment and ultrasound irradiation both caused the damage of ester linkage of carboxylic group of lignin and the degradation of aromatic structure. Comparably, US-Fenton (ultrasound and Fenton treatment) process had better oxidation ability than that of Fenton oxidation alone.

Keywords: Fenton oxidation, advanced oxidation processes, ultrasonic irradiation, papermaking wastewater, FTIR (ATR)

INTRODUCTION

Pulping and papermaking wastewater has high organic content and complex composition, which makes it difficult to biodegrade. To make wastewater reach the national discharge standard, advanced oxidation processes (AOPs) are applied in this research. AOPs include many oxidation techniques, which have proven their efficiency in the treatment of refractory organic pollutants. They focus on the production of hydroxyl radicals, highly reactive species, which promote the degradation and mineralization of organic pollutants. The AOPs involve Fenton oxidation,[1] photo-Fenton process,[2] high voltage electrochemical discharge, heterogeneous photocatalysis,[3] ultrasonic irradiation and electro-Fenton process.[4,5,6]

In order to improve the degradation efficiency, the AOPs are combined with other methods, such as ultrasonic treatment. This research aims to present the optimum process of ultrasonic irradiation alone and a combination of ultrasound and Fenton's reagent for the treatment of papermaking wastewater. The results demonstrate that ultrasonic irradiation has potential as a method for the removal of organic pollutants, as shown in Table 1.

Table 1
Effects of different processing methods on the papermaking wastewater indexes

Processing method	COD removal rate	Turbidity removal rate
Ultrasonic treatment	< 10%	< 10%
Fenton's reagent	< 80%	< 70%
Ultrasound +Fenton's reagent	> 85%	> 80%

In order to further improve the removal efficiency, Fenton's reagent was added to this treatment method, which strengthened the oxidation ability of the whole reaction system.

Table 2
Indexes of original papermaking wastewater

pH	COD_{Cr} (mg/L)	Turbidity (NTU)	Chroma
8	441.4	37	75

EXPERIMENTAL

Chemicals

NaOH (10%) (CAS NO: 8012-01-9), H_2SO_4 (10%) (CAS NO: 7664-93-9), H_2O_2 (30%) (CAS NO: 7722-84-1), $FeSO_4 \cdot 7H_2O$ (purity 99%) (CAS NO: 7720-78-7), diethyl ether (60-29-7), and other chemicals were of laboratory reagent grade and applied without further purification. All chemicals were purchased from Beijing Chemical Works and Beijing Yili Fine Chemicals Co., Inc (China). All the solutions were prepared by using high purity deionized water.

Characteristics of papermaking wastewater

The papermaking wastewater sample was obtained from the secondary sedimentation tank in a papermaking corporation, located in Dongying City, China, which mainly produced wastewater containing acids, alcohols, benzene series and esters, for example, acetic acid, ethanol, cis-propanediol, 2,4-di-t-butylphenol, pentacosane, et al. The indexes of the original wastewater sample are shown in Table 2.

Wastewater analysis

CODcr of the wastewater was measured by the potassium dichromate method according to GB11914-87. The instrument used was a HATO @CTL-12 COD tester. The chroma of the wastewater was tested by the dilution multiple method according to GB11903-89. The turbidity of paper was tested by a high turbidimeter (WZS-185).

FTIR-ATR analysis

A Fourier transform infrared system (FTIR), Tensor 27 spectrometer was used. The infrared absorption spectra were recorded in the region of 4000-400 cm^{-1}.

Experimental methods

Treatment by Fenton process alone

The optimum Fenton treatment conditions were determined. The wastewater samples were degraded in a 250 mL flask. Before treatment, a 100 mL wastewater sample was added in. The initial pH of the wastewater sample was adjusted with diluted NaOH or H_2SO_4 solution to the target pH. Samples were taken at different time intervals, and the pH was adjusted to 8 to remove the irons. The CODcr, chroma, and turbidity of the wastewater were determined after 30 min standing.

Treatment by ultrasound alone

The ultrasound process was carried out for comparing the treatment efficiency with that of the Fenton process.

Treatment by combination of ultrasonic irradiation and Fenton's reagent

Fenton oxidation promoted by the ultrasonic irradiation process was carried out by ultrasonic generator (power: 750 W, freq.: 20 kHz, Sonics Co. LT. type: VCX750) at different treatment time.

RESULTS AND DISCUSSION

Fenton oxidation process optimization

Effect of initial pH value on Fenton oxidation system

The pH value is one of the most important factors in Fenton oxidation, which directly influences the existence form of ferrous ions. It has been proven that the most suitable pH value is 2-4. On the one hand, when the pH further increases, Fe (III) is formed from Fe (II) and exists as deposit. Therefore, the catalytic ability of Fenton's reagent decreases, which makes the oxidation efficiency decrease. On the other hand, when the pH value is less than 2, H_2O_2 cannot be effectively decomposed to OH· by Fe^{2+}. Under these conditions, H_2O_2 converts into $H_3O_2^+$ by capturing one proton. $H_3O_2^+$ is electrophilic, leading to the decrease of the reaction rate between H_2O_2 and Fe^{2+}.[7]

The effects of initial pH on CODcr removal rate of wastewater are shown in Fig. 1a. The other reaction conditions were as follows: the dosage of hydrogen peroxide was 0.1 mL (0.0098 mol/L), the molar ratio of H_2O_2 to Fe^{2+} was 3, and reaction time was 20 min in 100 mL raw wastewater. From Fig. 1a, it can be easily noted that about 43% of COD content was removed at the initial pH of 2 after 20 min reaction.

When the initial pH increased to 4, the average COD removal efficiency was increased to 75%. The further increase to 7 resulted in a negative decline of COD removal efficiency. This is because Fenton's reagent is a mixture of hydrogen peroxide

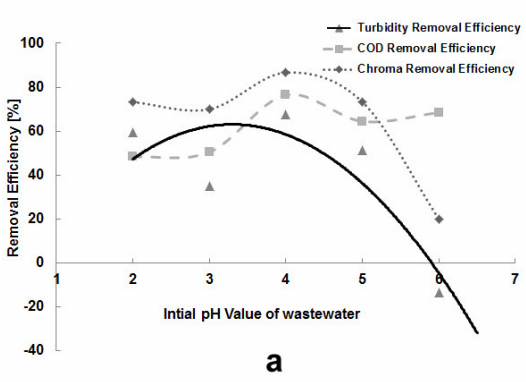
and ferrous ions that produce OH radicals, which is decided by the original pH value of the wastewater to a great extent. In a strong acidic medium, the high hydrogen ions accelerate the following reaction (1), which results in a large amount of Fe (III) and OH·. The amount of Fe (II) reduces and inhibits the dissociation of H_2O_2, as follows from Eq. 2.

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

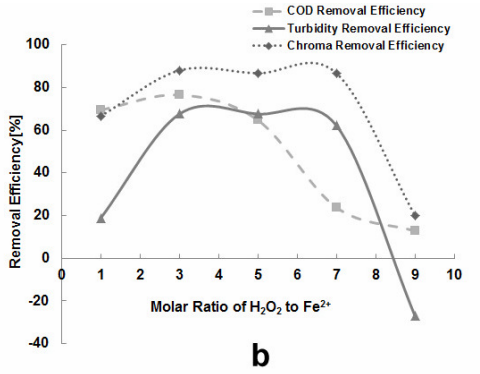
$$OH\cdot + Fe^{2+} \rightarrow OH^- + Fe^{3+} \quad (2)$$

As shown in Fig. 1a, the CODcr removal rate had the best performance when the pH value was 4. When the pH value increases continuously, Fe^{3+} will react with hydrogen peroxide to produce hydroxyl complexes and the excess of ferric ions will catalyze hydrogen peroxide and cause it to convert into water and oxygen.[8] Fe^{2+} and Fe^{3+} are also precipitated by OH^- under higher pH value, which decreases the catalytic ability of ferrous ions.

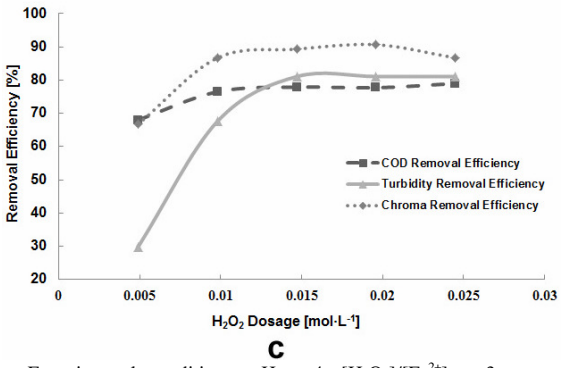
Just as the variation trend of COD removal rate, in Fig. 1a, the removal efficiency of chroma and turbidity had an obvious escalating trend when the initial pH value ranged from 2 to 4. When the pH value increased to 4, they decreased obviously. Fenton's reagent can be employed for the degradation of organic pollutants and the damage of chromophores by its oxidation and coagulation in the solution. Because many organic compounds in wastewater were oxidized at a lower pH value (pH = 2~4), the chroma and turbidity of wastewater further reduced. As the pH value gradually increased, the coagulation effect of ferric ions played a dominant role in treating effluents and formed precipitates as $Fe(OH)_3$, which hindered the catalytic ability of ferrous ions and resulted in the relative decrease of chroma and turbidity removal efficiency.



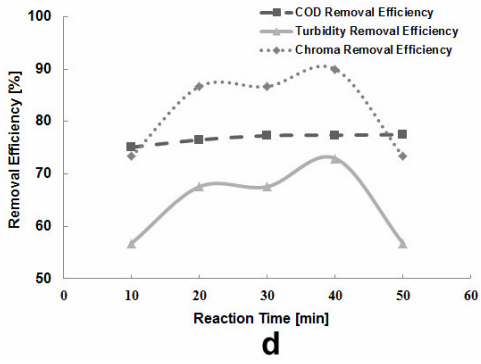
Experimental conditions: dosage of H_2O_2 = 0.1 mL, $[H_2O_2]/[Fe^{2+}]$ = 3, reaction time = 20 min



Experimental conditions: pH = 4, dosage of H_2O_2 = 0.1 mL, reaction time = 20 min



Experimental conditions: pH = 4, $[H_2O_2]/[Fe^{2+}]$ = 3, reaction time = 20 min



Experimental conditions: pH = 4, dosage of H_2O_2 = 0.15 mL, $[H_2O_2]/[Fe^{2+}]$ = 3

Figure 1: Effects of different factors on the removal rate of wastewater indexes

Effect of molar ratio of H_2O_2 to Fe^{2+} on removal rate of wastewater indexes

The molar ratio of H_2O_2 to Fe^{2+} is another important factor in the Fenton oxidation system.[9] When $[H_2O_2]/[Fe^{2+}]$ is less than 1, the coagulation of Fe (II) plays a major role in the wastewater treatment. When $[H_2O_2]/[Fe^{2+}]$ is more than 1, Fe (II) can catalyze H_2O_2 to produce hydroxyl radicals

for the oxidation effect.[10] Although Fe^{2+} can react with H_2O_2 to generate OH·, Fe^{2+} and H_2O_2 cannot be excessive unilaterally because of the undesired side reactions as follows:[11]

$$Fe^{2+} + OH\cdot \rightarrow Fe^{3+} + OH^- \quad (2)$$

$$H_2O_2 + OH\cdot \rightarrow HO_2\cdot + H_2O \quad (3)$$

$HO_2\cdot$ is an oxidant, but its potential oxidation is much weaker than that of OH·. The influence of molar ratio of H_2O_2 to Fe^{2+} is illustrated in Fig. 1b, the other reaction conditions being as follows: initial pH = 4, dosage of hydrogen peroxide = 0.1 mL, and reaction time = 20 min, in 100 mL papermaking wastewater. In Fig. 1b, the maximum degradation could be obtained by the Fenton process with a molar ratio of 3. Under this ratio, the COD, chroma and turbidity removal efficiency of wastewater reached the highest value. As the molar ratio continued to increase, the concentration of Fe^{2+} became lower, and could not provide enough OH· in the reaction with H_2O_2. It will further weaken the oxidation of Fenton's reagent to the organics, especially some effluents containing chromophoric groups. Therefore, it will cause a worse catalytic ability and reduce the removal rate of COD, chroma and turbidity.

Effect of H_2O_2 dosages on removal rate of wastewater indexes

Fenton's reagent is an aqueous solution of hydrogen peroxide and ferrous ions. The reaction between H_2O_2 and Fe^{2+} can yield hydroxyl radicals that degrade many organic compounds of papermaking wastewater. The running cost of the Fenton oxidation system depends on the dosage of H_2O_2 to a great extent. The appropriate dosages of H_2O_2 make a balance between cost and reaction efficiency. According to the reaction equation (1) listed above, if the dosage of H_2O_2 is not enough, the production of OH· radicals decreases and the oxidation ability becomes weak. Hydrogen peroxide, one capture agent of OH·, also consumes OH· radicals, according the reaction equation (3), making the oxidation ability decrease.

Fig. 1c illustrates that the CODcr removal rate had no obvious increase when the dosage of hydrogen peroxide was more than 0.15 mL (0.0147 mol/L). The production of many small bubbles, one special experimental phenomenon, was observed when the dosage increased to 0.15 mL. This is caused by the invalid decomposition of the excessive hydrogen peroxide. Taking running cost and treatment effect into consideration, the dosage of 0.15 mL was the optimum point.

Chroma removal rate presented a slight decrease, as may be noted in Fig. 1c, which was caused by the increase of Fe (II) concentration. The excessive ferrous ions also increased the chroma of the wastewater and decreased the reaction efficiency. CODcr, chroma and turbidity removal efficiency had different variation trends, which suggests that different organic compounds influence CODcr and chroma indexes of wastewater. Above all, Fenton oxidation system has different oxidation ability to different kinds of pollutants.[12]

Effect of reaction time on removal rate of wastewater indexes

The results in Fig. 1d show that the CODcr removal rate was of about 74% when the reaction time was 10 min, and without any significant improvement as the reaction time was extended to 50 min, with the other reaction conditions as follows: initial pH = 4, dosage of H_2O_2 = 0.15 mL, molar ratio of H_2O_2 to Fe^{2+} = 3. The chroma and turbidity removal rate increased as the reaction time increased from 10 min to 40 min gradually. A turning point was observed at the time of 40 min. When the time was too short, such as 10 min, the reaction between H_2O_2 and Fe^{2+} may not have been completed, which led to lower production of OH· radicals and made the reaction efficiency decrease. When the time was too long (more than 40 min), the chroma and turbidity of the water sample were on a clear rise, which resulted in resource waste.[13] The data in Fig. 1d suggest that the optimal reaction time was 40 min, considering the removal rate of CODcr, chroma, and turbidity.

Ultrasonic-Fenton process optimization

Effect of different methods of Fenton and ultrasonic treatment on wastewater indexes

Ultrasound is defined as a sound wave with a frequency greater than the upper limit of human hearing, approximately in the range of 20 kHz to 10 MHz. The ultrasound employed in this research was 20 kHz.

Both ultrasound and Fenton's reagent have effects on the decomposition of organic compounds, some researchers have tried to use combined methods. These methods utilize the advantages of ultrasound and Fenton's reagent, allowing better degradation of organic pollutants. These findings suggest that a combination of ultrasound and Fenton's reagent is more attractive for practical application.[11]

The ultrasound wave consists of expansion and compression cycles. The expansion cycle can result in acoustic cavitation. The cavitation mechanism is

very complicated. When the cavitation bubbles collapse, the temperature and pressure in the bubbles can reach up to several thousand Kelvin and several hundred atmospheres, respectively.[14,15] Under these conditions, organic compounds are decomposed directly by pyrolysis inside the bubbles. The cavitation effect hits the surface with a tremendous force, which can activate surface catalysis, force the impregnation of the catalytic material into the fibers in wastewater, and improve mass and heat transfer at the surface by decreasing the width of interfacial boundary layers. Another degradation mechanism is via the pyrolytic formation of hydroxyl radicals in the bubbles. The hydroxyl radicals can be transferred to the bubble interface, promoting the degradation of organic compounds outside the bubbles.[16]

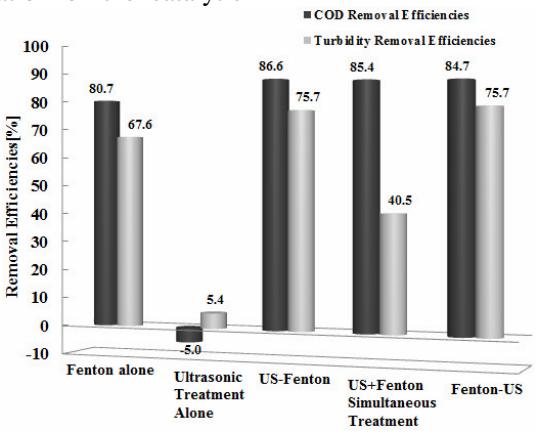


Figure 2: Effects of different methods of Fenton and ultrasonic treatment on wastewater indexes
Experimental conditions: ultrasound treatment: irradiation time = 40 min, treatment temperature = 20 °C, ultrasound pulse on(30 s)-off(30 s), amplitude = 19%; Fenton treatment: initial pH = 4, dosage of H_2O_2 = 0.15 mL, $[H_2O_2]/n[Fe^{2+}]$ = 3, reaction time = 40 min; US-Fenton: ultrasonic pretreatment, Fenton's reagent follow-up treatment; Fenton-US: Fenton's reagent pretreatment, ultrasonic follow-up treatment

Fig. 2 illustrates that ultrasonic treatment alone had little influence on the CODcr and turbidity of the wastewater. The ultrasound treatment can degrade some chromogenic macromolecular organics into some colorless small molecular substances in the wastewater, for example, damage to the aromatic or heterocyclic rings and change to their structures.[17] However, large amounts of organic pollutants have low volatility and good solubility in the papermaking wastewater. During the ultrasonic degradation, it is difficult for those organics to reach the bubble-liquid interface region, which reduces the occurrence of oxidation reactions and virtually has no influence on the COD removal. The removal rate of wastewater indexes had a significant increase through the combination of ultrasonic and Fenton treatment. The US-Fenton (ultrasonic-Fenton) and Fenton-US (Fenton-ultrasonic) had better treatment effects than Fenton alone and US+Fenton simultaneous treatment. It appeared that the US-Fenton procedure led to the best treatment effects according to the removal rate of wastewater CODcr and turbidity. In comparison, different combination methods of ultrasound and Fenton led to similar CODcr removal efficiency, while the US-Fenton treatment produced better a turbidity removal rate. This phenomenon is caused by the synergistic effect in US+Fenton process, in which the flocculation and oxidation effects decrease dramatically by the ultrasonic treatment. The pyrolytic reaction resulted from ultrasound can increase the temperature in the solution system. In a complex reaction system, such as Fenton's agent, the increase of temperature accelerates the positive reaction of the system, as well as the side reactions. The appropriate temperature can stimulate the production of hydroxyl radicals, but higher temperature instead results in the invalid decomposition of hydrogen peroxide into water and oxygen.[18] Above all, although the synergistic effect of the combination of ultrasound and Fenton's reagent can oxidize the refractory pollutants, the increase of temperature in the aqueous solution activates the ineffective degradation of H_2O_2 and further reduces the turbidity removal rate.

Effect of ultrasonic treatment time on removal rate of wastewater indexes

From Fig. 3a, it may be observed that the CODcr, chroma and turbidity removal efficiency of the wastewater decreased from 10 min to 50 min,

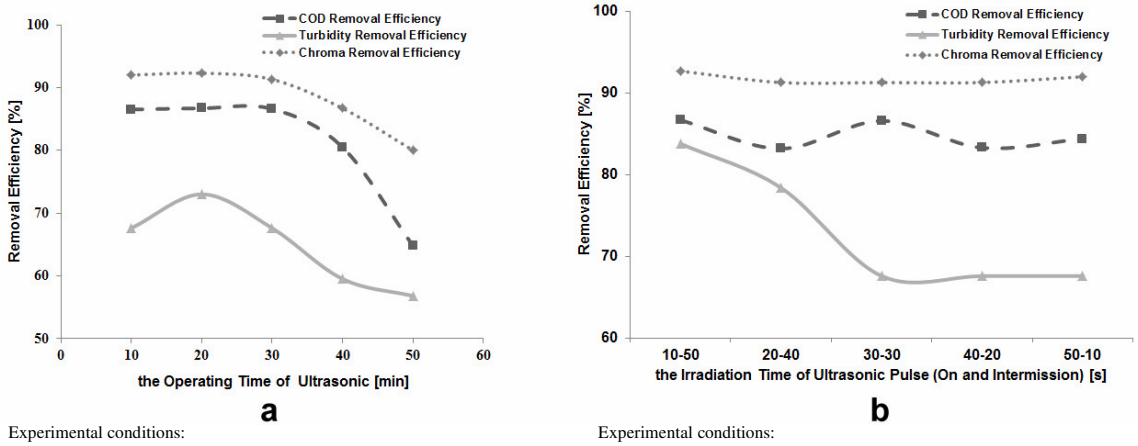
especially after 30 min, when Fenton oxidation was applied as a subsequent process, following ultrasound irradiation.

The terminal oxidation and flocculation effects were relatively poor when ultrasound irradiation time was less than 10 min, which was because the refractory organic compounds were still not degraded in wastewater. Fig. 3a illustrates that the removal efficiency decreased dramatically when ultrasound irradiation time was over 30 min. The excessive processing time had a little contribution to the removal of COD and turbidity, because the majority of refractory organic compounds were degraded.[19]

Longer ultrasound irradiation time produces more energy consumption and higher temperature in the wastewater, which weakens Fenton oxidation. Considering the costs and the reaction efficiency, the ultrasound irradiation time of 20 min was accepted as the most suitable. The easily degradable organic compounds can be oxidized in a short time by ultrasound irradiation, leading to the decrease of CODcr and turbidity. The refractory compounds, such as benzene ring, can be degraded into small molecular compounds as ultrasound irradiation time increases, which makes the COD removal efficiency decrease.

Effect of operating time of ultrasonic pulse on removal rate of wastewater indexes

Taking 1 min as a running cycle, Fig. 3b illustrates that the pulse of ultrasound irradiation had an influence on the removal rate of CODcr and turbidity. The results show that the ultrasonic pulse of 10 s-50 s (ultrasound on 10 s and intermission 50 s) had better effects than that of other ultrasonic pulse treatment time. Compared with CODcr and chroma removal rate, the turbidity removal rate changed relatively significantly. This is because different pulse had different ultrasound running time for the wastewater treatment. Although ultrasound irradiation has good degradation ability to papermaking wastewater, longer irradiation time will make refractory compounds degrade, which causes the increase of CODcr value.



Experimental conditions:
Ultrasonic pretreatment, treatment temperature = 20 °C, ultrasound pulse on (30 s)-off (30 s), amplitude = 19%; Fenton follow-up treatment, pH = 4, dosage of H_2O_2 = 0.15 mL, $[H_2O_2]/[Fe^{2+}]$ = 3, reaction time = 40 min

Experimental conditions:
Ultrasonic pretreatment, operating time 40 min, treatment temperature 20 °C, amplitude 19%; Fenton follow-up treatment, pH = 4, dosage of H_2O_2 = 0.15 mL, $[H_2O_2]/[Fe^{2+}]$ = 3, Reaction time = 40 min

Figure 3: Effects of the operating time and ultrasonic pulse on removal rate of wastewater indexes

FTIR/ATR analysis

Fig. 4 illustrates the infrared spectra of the raw sample of wastewater and of the samples treated by different oxidation methods, including Fenton oxidation alone, ultrasound irradiation alone, and US-Fenton treatment. Fenton treatment conditions were the following: initial pH = 4, dosage of H_2O_2 = 0.15 mL (0.0147 mol/L), molar ratio of H_2O_2 to Fe^{2+} = 3, and reaction time = 40 min. Ultrasound irradiation was carried out with 20 min treatment time, 20 °C temperature, pulse 10 s-50 s and 19% amplitude.

The treatment results were investigated by FTIR-ATR analysis. The spectra of the treated and untreated samples are presented in Fig. 4, and some differences may be observed therein.

The absorbance at 1595 cm^{-1} is associated with the aromatic skeleton vibration. The data in Fig. 4

indicate that the wastewater processed by Fenton's reagent presented no significant variation in comparison with the raw sample, while the water sample treated by ultrasound or US-Fenton had obvious absorbance. It may be concluded that the Fenton oxidation system has little influence on the aromatic skeleton structure, while the ultrasound oxidation and US-Fenton oxidation system can make the aromatic structure degrade.

In the spectra of the treated wastewater samples, an increased absorbance of distinct bands can be observed between 1430 and 1373 cm^{-1} corresponding to –CH– stretching, bending deformations, and rocking vibrations of methylene groups, according to Kamel *et al.*,[20] which suggest the decomposition of long-chain organic compounds. The decomposition occurred much more obviously under ultrasound irradiation and US-Fenton treatment.

The absorbance band at 1123-1033 cm^{-1} is assigned to the ether bonds. The absorbance of Fenton-treated and ultrasound-treated water decreased, which suggests the degradation of refractory organic compounds in wastewater.

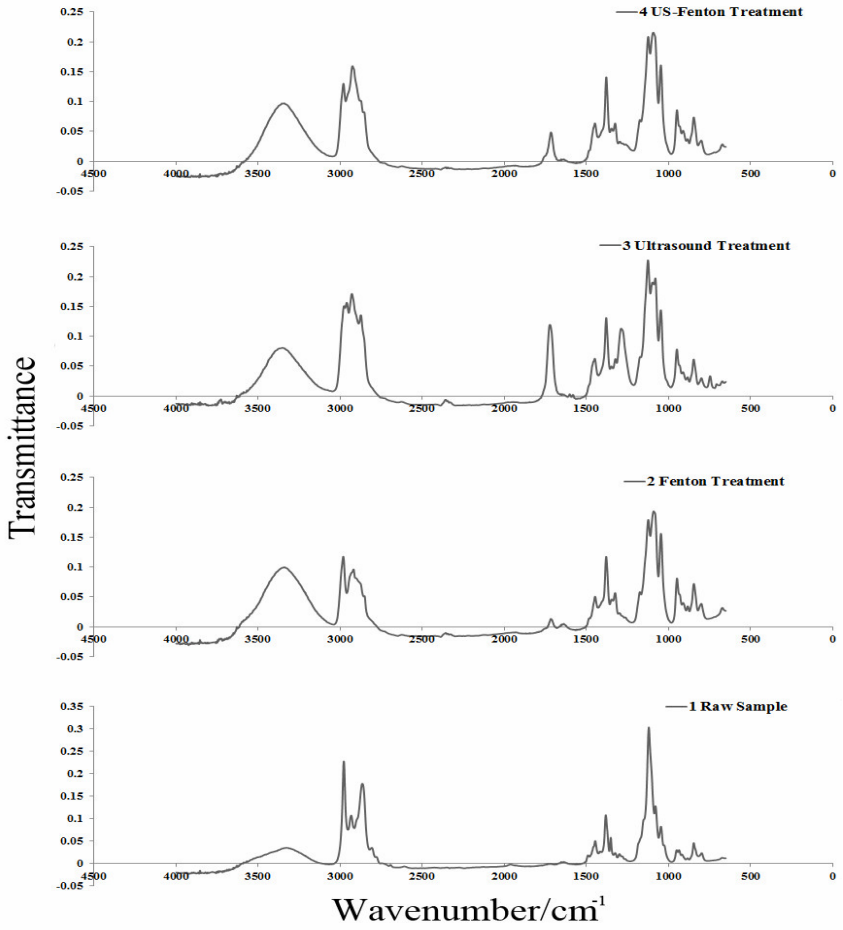


Figure 4: FTIR spectra of raw and treated wastewater

CONCLUSION

The present investigation indicates that the combination of ultrasonic pretreatment and subsequent treatment with Fenton's reagent is more attractive for practical application. The most suitable ultrasound irradiation conditions are listed as follows: ultrasound irradiation time = 20 min, treatment temperature = 20 °C, pulse 10 s (on) - 50 s (off), and amplitude = 19%. The optimum Fenton oxidation conditions were confirmed to be the following: initial pH = 4, dosage of H_2O_2 = 0.15 mL (0.0147 mol/L), molar ratio of H_2O_2 to Fe^{2+} = 3, and treatment time = 40 min. By the above optimum processing treatment, the wastewater COD can be decreased to less than 60 mg/L. Chroma removal rate exceeds 90% and turbidity removal rate is over 80% – results that are in agreement with the national standards. In the experiment for testing the effect of different H_2O_2 dosages in Fenton's system, it was observed that

the variation of COD and chroma in wastewater had different trends, which proves that the organic composition influencing the COD value is different from those influencing the chroma in aqueous solution. The FTIR-ATR analysis suggested that Fenton treatment and ultrasound irradiation both caused damage to the ester linkage of the carboxylic group of lignin and even the degradation of the aromatic structure. Comparably, the US-Fenton (ultrasound and Fenton treatment) process had better oxidation ability than that of Fenton oxidation alone.

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