



Gas-phase advanced oxidation (GPAO) for benzene-containing gas by an ultraviolet irradiation/hydrogen peroxide vapour (UV/[H₂O₂]_g) process

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Abstract

Hydrogen peroxide (H₂O₂) is a remarkably strong oxidant, and its vapour ([H₂O₂]_g) has further advantages, such as a low cost and good light transmission. However, there has been very little research on its removal through gas-phase advanced oxidation (GPAO). In the present study, the photochemical oxidation of a gas that contains a series of benzene derivatives using ultraviolet (UV) irradiation and [H₂O₂]_g was investigated in a transparent bag made of fluorinated ethylene propylene (FEP). UV and [H₂O₂]_g barely reduced the pollutant within 5 h when used alone, and the reactant was also stable. When the pollutant concentration was high (248 to 756 mg/m³) and the residence time was short (3 s) compared with related studies on the removal of benzene, toluene and xylene, the apparent removal rate by UV/[H₂O₂]_g/(powder active carbon, PAC) was higher than when other methods (UV/[H₂O₂]_g, UV/[H₂O₂]_g/TiO₂ and UV/[H₂O₂]_g/ZnO), were used. However, it was found that the mineralization by UV/[H₂O₂]_g significantly decreased, which in turn decreased the conductivity after the reaction. Increasing the pollutant concentration and the pH of the H₂O₂ had a negative effect on the treatment, but the UV radiation had a positive effect at powers of up to 40 W. In addition, the characteristic absorbance of three benzene derivatives showed that the key structure of the pollutant molecules was damaged during GPAO.

Keywords Hydrogen peroxide vapour · Benzene series · Removal · Conductivity · Characteristic absorbance

Introduction

People need to breathe constantly, so nobody can avoid the influence of pollution. However, it is a widely recognized reality that air quality is not always good, especially in developing countries. Thus, pollution removal methods are increasingly studied. Such air pollution removal methods include adsorption, absorption, advanced oxidation (AO) and so on. Compared with other treatment methods, AO results in more complete and quicker removal of pollutants. Photochemical methods that do not require catalysts have received more attention in recent years (Alapi et al. 2007; Mo et al. 2009; Zhang and Anderson 2013) because there are

some distinctive advantages, such as energy efficiency and running costs, that must be evaluated for real-world applications (Matthew et al. 2014).

Both hydrogen peroxide and ozone are well-known oxidants that are usually used in aqueous and gaseous systems, respectively (Wang and Ray 2000). Although ozone is probably the leading oxidant species because its 2.07 V oxidation–reduction potential (ORP) is higher than that of hydrogen peroxide 1.8 V, both substances can produce the •OH radical, which is regarded as the key oxidant in the AO process. Furthermore, H₂O₂ has multiple benefits. First, it is very easy to generate as long as electron donors are present in water, and chromophoric substances can produce hydrated electrons by illumination and combine with dissolved oxygen, which leads to a series of reactions for H₂O₂ outcome (Anastasio et al. 1997; Wohlgemuth et al. 2001). Second, artificial production of hydrogen peroxide is very convenient, even in situ production (Choudhary and Jana 2007; Fang et al. 2013). Third, H₂O₂ can be used as an indicator in the AO process because the representative oxidant, the hydroxyl radical (•OH), which has an (oxidation reduction potential)

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ORP of 2.8 V, can lead to H_2O_2 formation by reacting with volatile organic compounds (VOCs), CO, SO_2 , and other pollutants. It is evident that in AO involving multiple radical chain reactions, hydrogen peroxide plays a central role (Möeller 2009).

Due to its unique properties, H_2O_2 is usually used as an oxidative solution in tandem with ultraviolet (UV) radiation in AO processes to remove liquid pollutants (Xu et al. 2007; Muruganandham and Swaminathan 2004). Because the reactants are in solution during this process, the waste has a long residence time. For gas pollutant treatment, some studies have shown that UV/ H_2O_2 can oxidize SO_2 , NO_x and Hg^0 at low concentrations in flue gas (Yangxian et al. 2011; Yangxian et al. 2014). However, research focusing on GPAO by $[\text{H}_2\text{O}_2]_g$ is very rare, especially that which involves a high pollutant concentration and short residence time, even if the vapour is only used for sterilization (French et al. 2004). In reality, the vapour form of H_2O_2 not only maintains the properties of the solution phase but also provides some advantages, such as a lesser oxidant consumption and decreased loss of UV light that passes through the reaction system.

In this work, the GPAO by H_2O_2 vapour was studied to understand the effect of harsh conditions, such as high concentration pollution and short residence time compared with those of related studies. The experiment was conducted to investigate the decomposition of benzene, toluene and xylene by UV/ $[\text{H}_2\text{O}_2]_g$. The reactions took place in a transparent bag made of fluorinated ethylene propylene (FEP, Teflon) due to its high reactive inertia and UV transmittance (Du et al. 2007).

Benzene derivatives were chosen as the target compounds because such organic pollutants, including benzene, toluene and xylene, are frequently encountered, for example, benzene and toluene are frequently encountered in indoor and outdoor environments (Wallace et al. 1993; Bernstein et al. 2008), are representative organic contaminants (Liu et al.

2016) and can cause multiple diseases in humans. Research on pollutant removal methods includes adsorption, evaporation, membrane separation, microbial degradation and catalysis (Liu et al. 2018; Miaralipour et al. 2017; Varjani et al. 2017). However, GPAO is rarely used (Zhang et al. 2003; Johnson et al. 2014), and studies using H_2O_2 as an oxidant have not been reported.

Experimental section

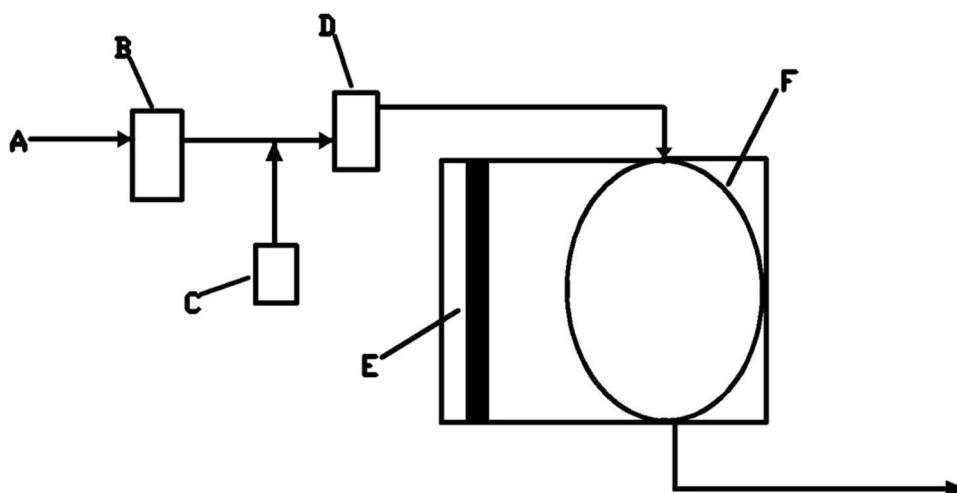
Materials and analysis

The reagents were used as received without further treatment. Benzene, toluene, xylene, NaOH, HCl and 30% commercial H_2O_2 solution (Guoyao Chemical Reagent Co., China) were analytical-grade reagents (AR). The powdered active carbon (PAC, < 75 μm diameter, 0.45 g/cm^3), ZnO (5.6 g/cm^3) and TiO_2 (4.28 g/cm^3 , anatase) powder with a diameter of 20 nm and purity of 99.9% were separately purchased from Liyang Nanfang Activated Carbon Factory, Brofos Nanotechnology Ningbo Co., China.

Experimental procedure

The experimental system used in this study is illustrated in Fig. 1. All gas flow channels in the experimental system were FEP pipes. In this procedure, equal quantities of PAC, TiO_2 and ZnO (2 g) were added into compartment D, and air was pumped by a micro air pump (ACO-001, Guangzhou Weierma Instrument Co., Ltd. China) into the FEP pipes, regulated by rotameters to transfer the vapour of benzene and H_2O_2 to D. Here, the gas was thoroughly mixed with the powder due to the small particle size and low density of the powder. Then, the mixture was transferred to the FEP bag and irradiated under a UV lamp to undergo a

Fig. 1 Schematic of experimental system. Legend: A, air; B, liquid pollutant vessel; C, H_2O_2 solution vessel; D, mixing tank; E, UV lamp (253.7 nm); F, FEP bag



photochemical reaction. The inlet and outlet concentrations of the reactants were measured through the gas bypass line. To determine the apparent removal rate (dependent on the measurement for the pollutants), the pollutant concentrations were monitored via gas chromatography (GC) with FID and an AB-INOWAX fused silica capillary column (sampling time of 50–90 min). The retention time was 1.2–1.6 min and was calculated by Eq. (1); the mineralization rate, which was dependent on the measurement of the terminal degradation product, was calculated with measurements from a CO₂ detector (GT-1000-CO₂, KORNO Co. Ltd. China). The characteristic absorbance of the pollutants and [H₂O₂]_g was determined by UV-vis spectroscopy and H₂O₂ detector (GT-1000-H₂O₂, KORNO Co. Ltd. China) separately. The apparent and mineralization efficiencies were calculated using Eqs. (2) and (3), respectively.

$$C(\text{mg}/\text{m}^3) = (A - 318.5)/2.39 \quad (1)$$

$$\text{Apparent removal efficiency (\%)} = (C_{\text{in},p} - C_{\text{out},p}) * 100\% / C_{\text{in},p} \quad (2)$$

$$\text{Mineralization efficiency (\%)} = (C_{\text{out},c} - C_{\text{in},c}) * 100\% / C_{T,c} \quad (3)$$

where A is the peak area of the waste concentration determined by GC, and the correlation coefficient for (1) is 0.9997; C is the concentration of the benzene derivative in mg/m³; $C_{\text{in},p}$ and $C_{\text{out},p}$ are the inlet and outlet concentrations of pollutants, respectively; $C_{\text{in},c}$ and $C_{\text{out},c}$ are the CO₂ concentrations; and $C_{T,c}$ is the total CO₂ concentration upon complete oxidation of the pollutant.

To investigate the effect of pH and conductivity, the pH of the H₂O₂ solution was adjusted by adding HCl and NaOH and was determined using an acidimeter (Model Starter 2100/3C Pro, Shanghai Hongji Instrument Co., Ltd. China). After 1 h of absorption in a 25-mL opaque sampling tube with pure water, the conductivity was measured using a Conductivity Meter (DDSJ-319L, INESA Scientific Instrument Co., Ltd. China).

Results and discussion

Effect of UV or H₂O₂ alone

Apparent removal by UV radiation

Benzene was used as the target compound in these experiments. Figure 2 shows that with a maximum UV radiation power of 30 W, benzene is resistant to removal under different reaction times, and the apparent removal rate was in the range of 0.5–1%. There was little direct UV photo-oxidation and that which occurred was indirectly caused by substances

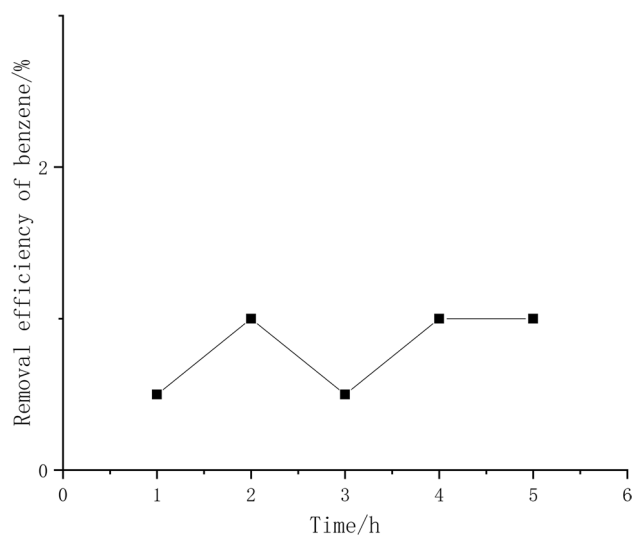


Fig. 2 Apparent removal of benzene when only UV radiation was applied

in the ambient air, such as O₂ and H₂O. This observation is in accordance with studies showing that toluene and benzene in air could hardly be degraded under > 200 nm UV light (Zhang et al. 2003; Zhuangzhuang et al. 2019) because 253.7 nm is too long of a wavelength to break the benzene molecular bond because the photons do not have enough energy. Although dimethylbenzene could be reduced under 254 nm light after a long time (> 3 h) (Ma et al. 2007), the substance is different from the benzene derivatives used in this study. In addition, it is critical to understand the impact of the reactor walls on gas-phase reactivity; due to the background NO_x off-gas and other reactive species, heterogeneous reactions can generate “chamber radical sources” upon irradiation (Carter et al. 1982). However, Fig. 2 shows that the effects did not effectively influence pollutant reduction under certain conditions.

Conditions The experimental conditions were as follows: benzene concentration: 455 mg/m³; gas flow: 9 L/min; reactant temperature (room temperature): 25 °C; UV energy: 30 W; and residence time: 3 s.

Apparent removal by [H₂O₂]_g

The performance of benzene oxidation with only [H₂O₂]_g was investigated without UV radiation. Figure 3 shows that the pollutant quantity decreased very slightly (≤ 2%) over 5 h of reaction time; this result implies that although ambient light with very weak ultraviolet radiation could be transmitted into the reactor to cause [H₂O₂]_g to release ·OH, the production of free radicals was too rare to enhance the reaction. Thus, the benzene was extremely inert and was not easily degraded by [H₂O₂]_g oxidation. This phenomenon also suggested that

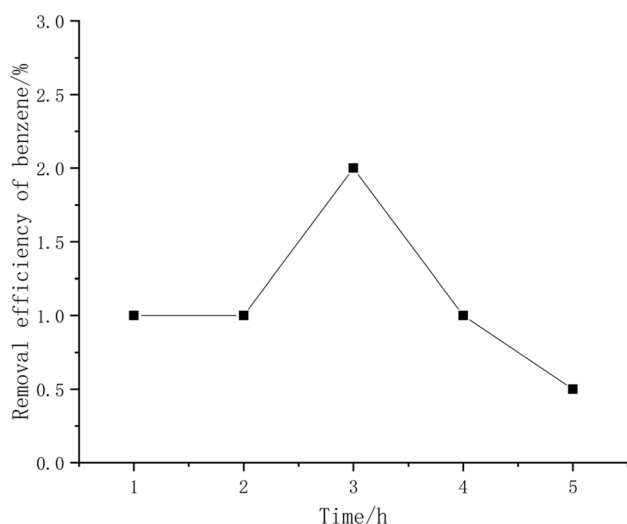


Fig. 3 Apparent removal of benzene with only [H₂O₂]_g

generating hydroxyl radicals is necessary for GPAO. The results also show that the reactor was stable under [H₂O₂]_g oxidation. Other studies have proven that FEP Teflon bags meet the requirements of accuracy and small loss of gas due to photochemical reactions under different humidity, illumination, and substance conditions (Jia et al. 2011; Jia and Xu. 2014; Li et al. 2021). Therefore, based on the results of the test mentioned above, the reactor satisfactorily met the requirements of the study. However, Fig. 3 showed that after 3 h of treatment, the removal of benzene was the highest (2%), while 5 h of treatment resulted in a lower inefficiency (0.5%). According to the observations in this work, [H₂O₂]_g formed by 30% solution could slowly accumulate in the FEP pipe (not transparent) and bag (transparent). Therefore, when accumulation initially occurred in the pipe, the rate remained stable due to a lack of additional ·OH production in the reactor; then, when the accumulative H₂O₂ reached the bag, radicals could be generated to slightly enhance the reaction. However, with the increase in the substances accumulated on the bag wall, the transmission of the weak light was easily hindered, resulting in the subsequent decrease in the efficiency. After the reaction, the accumulation measurement for H₂O₂ was also carried out (Supplementary Information in Fig. S5).

Conditions The experimental conditions were similar to those in the “Apparent removal by UV radiation” section, in which 4.6 mol/m³ [H₂O₂]_g was used to replace the UV light.

Apparent removal and mineralization

Apparent removal using different methods

All four treatment methods could reduce the contaminants, as shown in Fig. 4. Although the maximal efficiency was

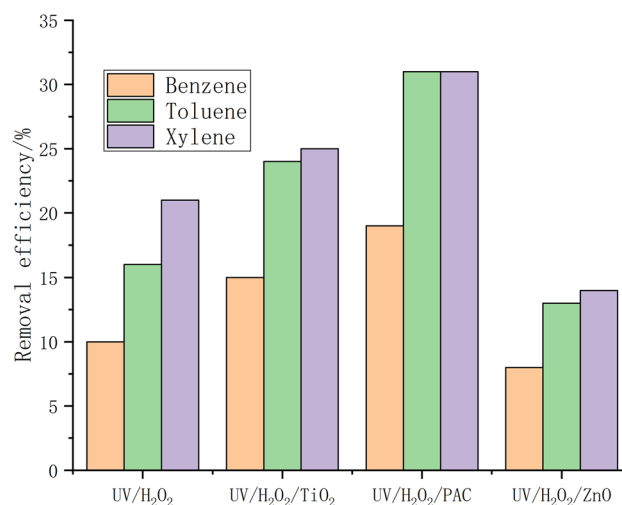
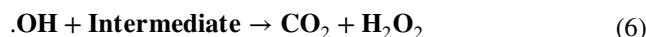


Fig. 4 Apparent removal rate by different pathways with H₂O₂

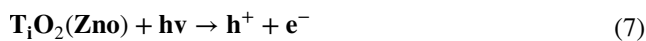
31% for toluene, which is lower than that reported in related research (Yangxian et al. 2014; Xu et al. 2007), the toluene was obtained with a very short residence time and high concentration of pollutants. Thus, the result was still very meaningful, as [H₂O₂]_g played an important role as an oxidant. The fundamental role of H₂O₂ lies in its decomposition into ·OH free radicals under UV radiation at a certain wavelength according to the following reaction (Lee et al. 2004).



Then, the removal of pollutants can be expressed by the following equations (Xu et al. 2007):



The catalyst and PAC completely left in the process, the determination of departure time and the subsequent removal after that time were also carried out (Supplementary Information in Table S1 and Fig. S4). In these three pathways, the potential problem was that H₂O in the H₂O₂ vapour could hinder the removal because of its competitive adsorption onto the reaction surface of the contaminants (Herrmann et al. 1999; Nazir et al. 2003). However, some promotion of removal occurred in the process. In addition to the ·OH radical produced by H₂O₂, it was also produced by the two catalysts under 253.7 nm UV light. The electrons (e⁻) in the electron-filled valence band could be excited by photoirradiation (hν > band gap energy) to the vacant conduction band, leaving a positive hole (h⁺) in the valence band; this h⁺ caused the OH⁻ from H₂O oxidation to produce ·OH, as shown in Eqs. (7) and (8).



Then, $\cdot\text{OH}$ could oxidize the compounds adsorbed on the surface of a photocatalyst. In addition, owing to the higher electron affinity of H_2O_2 (1.8 eV) than that of O_2 (0.44 eV), the excited electrons in the conduction band could be captured by H_2O_2 more easily than by O_2 , which increased the production of $\cdot\text{OH}$. Although the particle size of the PAC was the largest, its lowest density ensured ideal uniformity of mixing, and the adsorption performance could be helpful for removing pollutants, although the benzene derivatives were not completely degraded. There were fewer negative impacts of TiO_2 and PAC than positive impacts, and the latter treatment is better for situations in which the adsorption surface area is very great. For the three pollutants, the desired enhanced effect of ZnO did not occur, but rather, the removal was less than when the other treatments were used. It was concluded that ZnO was prone to photocorrosion (Hoffmann et al. 1995; Han and Zhao 1999), and its density was highest, which caused deposition, leading to the shortcomings caused instability and heterogeneity during the reaction.

Conditions The experimental conditions were similar to those in the “Apparent removal by UV radiation” section, in which $4.6 \text{ mol/m}^3 [\text{H}_2\text{O}_2]_g$ was also applied.

Mineralization by UV/ $[\text{H}_2\text{O}_2]_g$

In Fig. 5, it is shown that xylene had the highest apparent removal and mineralization rates, and benzene had the lowest. Toluene and xylene have one and two methyl groups substituting the hydrogen on the benzene ring, respectively; this substitution weakens the stability compared with that of benzene. These exocyclic groups are points that are easily attacked by oxidative species, which leads to the destruction of the whole molecular structure. Due to the greater number of methyl substitutions, the removal rate of xylene was most remarkable.

The results also showed that although the apparent removal rate was more than 10%, the mineralization was very low; the maximum was less than 5%. In general, the advanced oxidation by $\cdot\text{OH}$ can be expressed as Eq. (9).



To remove the series of benzene derivatives, C=C or C-H were first cracked to form active organic radicals, and then the radicals continued to produce multiple fragments (intermediates), of which only some were oxidized to CO_2 and H_2O ; these observations were in accordance with those reported in multiple studies (Kuo-Pin and Grace 2007;

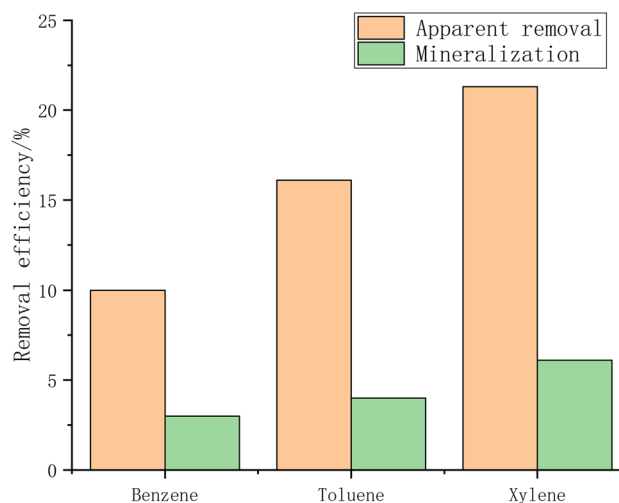


Fig. 5 Comparison between mineralization and apparent removal

Johnson et al. 2014). Due to the differences between the two measurement methods in this work, it was noted that most of the degraded pollutants did not mineralize, and the corresponding products were intermediates (not terminal degradation products), which caused a remarkable gap between the apparent removal and mineralization rates. For the other treatments with PAC and catalysts, similar results were also obtained (Supplementary Information in Fig. S1-PAC, S2- TiO_2 and S3-ZnO). These phenomena also revealed the limitations of GPAO, which cannot completely remove contaminants, hence limiting its applications.

Conditions The experimental conditions were in accordance with those described in the “Apparent removal using different methods” section.

Characteristic absorbance

Due to the multiple intermediate GPAO products, it is meaningful and necessary to focus on the characteristics of the molecules to determine whether pollutant structures are damaged. For the benzene derivatives, the conjugated ethylene in the benzene ring plays a directive role, and UV-Vis was used to investigate the inlet and outlet differences of the indicator. Although the benzene derivatives had a B-absorption band (230–270 nm) and E2 band (200–210 nm), the latter intensity was always higher than the former. Thus, E2 was used as the maximum absorbance in the experiment, and only the results of UV/ $[\text{H}_2\text{O}_2]_g$ degradation are mentioned in Table 1. These results show that the wavelength and absorption intensity of toluene and xylene are higher than those of benzene because there were redshifts and hyperchromic effects. It is believed that the methyl groups in the two pollutants are auxochromes, and their electrons in the n orbital

can produce a conjugation effect with π electrons from the chromophores. Then, the electrons in the electron cloud can influence each other, which changes the absorption intensity and peak position (Li et al. 2010). Table 1 also shows that there was a higher absorbance for inlet pollutants than for outlet pollutants. This phenomenon proved that it is also a feasible way to determine the removal effect. In addition, although this method involves a clear response to the conjugated ethylene in the benzene ring, it is rarely used in GPAO because the low concentrations of pollutants in the sample solution lead to very weak absorption peaks; in this work, this problem was solved by increasing the initial pollutant concentration and ending collection after longer sampling times.

Conditions The experimental conditions were in accordance with those in the “Apparent removal using different methods” section.

Variation of conductivity

In similar works, conductivity was not studied as the measurement index, although it can indicate changes in the substances after reaction. Figure 6 shows the conductivities of the inlet and outlet for removal of different pollutants. The data slightly decrease in the blank test (absorption by pure water), but opposite effects are observed with the pollutants. The result in the blank test could be explained by the fact that the UV decomposition of H_2O_2 caused its ionization reduction. For the latter measurement, Martina et al. found that the increase in polar products had a positive correlation with conductivity (Hein et al. 1998); therefore, it was believed that intermediates such as aldehydes, ketones and alcohols were polar products accelerated by the reaction and could not be completely eliminated due to the limited retention time. In addition, the increased values for benzene, xylene and toluene were 3.6, 1.2 and 0.2, respectively. This result also indicated the difference in the quantity of polar intermediates, which is in accordance with some other studies (Kislov et al. 2004; Hamilton et al. 2005; Yu et al. 1997).

Conditions The experimental conditions were in accordance with those described in the “Apparent removal using different methods” section.

Table 1 characteristic absorbance by UV–Vis

Pollutants	Benzene	Toluene	Xylene
Characteristic Wavelength (nm)	200	210.4	207.2
Absorbance of inlet pollutant	0.06813	0.0944	0.08744
Absorbance of outlet pollutant	0.02290	0.03012	0.05159

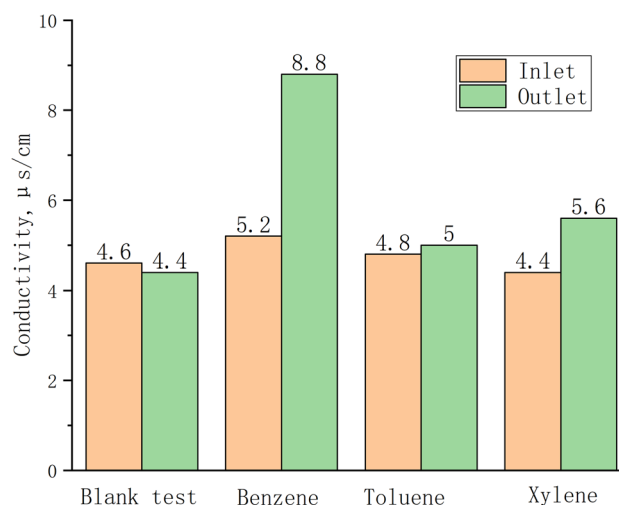


Fig. 6 Variation in conductivity for removal of different pollutants

Effect by pH of H_2O_2

Benzene vapour was selected to investigate the influence of pH. The effect on the waste removal efficiency is shown in Fig. 7, in which the rate decreased from 28.6 to 1.7 as the H_2O_2 solution pH increased from 4 to 9. It is widely recognized that H_2O_2 has stronger oxidizing properties under acidic conditions; therefore, the degradation was reduced with increasing solution pH. For these reasons, some research has proven that H_2O_2 in solutions can produce HO_2^- species, which scavenge $\cdot OH$ radicals via a hydrolysis reaction (Meng-Wen and Chia-Chun 2010; Liu et al. 2011). When the solution is alkaline, OH^- can consume H^+ and accelerate HO_2^- formation.

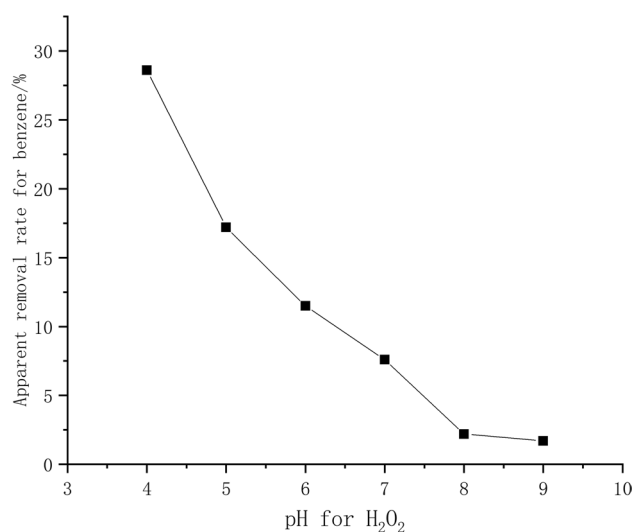
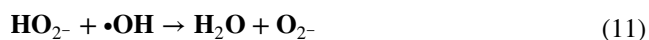


Fig. 7 Effect of H_2O_2 pH for benzene removal



In addition, OH^- can also consume $\cdot\text{OH}$ radicals through the following reaction (Zehavi and Rabani 1971), thereby reducing the removal efficiency of benzene:



Conditions Aside from the pH, the experimental conditions were in accordance with those described in the “Apparent removal using different methods” section.

Apparent removal by different UV power levels

The influence of UV radiation was investigated at different UV powers, as shown in Fig. 8. The results show that the removal increases with increasing UV power at values from 10 to 40 W. The improvement between 30 and 40 W was less than at lower power levels, and no improvement was observed from 40 to 50 W. It could be deduced that the $\cdot\text{OH}$ concentration was increased when the UV intensity was increased; thus, the chain reactions were strengthened to obtain a higher reactant consumption rate (Anglada et al. 2002). However, when the UV radiation power exceeded a certain value, several side reactions (reactions 13–16) also simultaneously occurred in the system, resulting in the loss of $\cdot\text{OH}$. Thus, with further increases in the light power to 40 W, the pollutant removal efficiency only increased slightly and then did not change at 50 W, which was the equilibrium state of the opposite action.

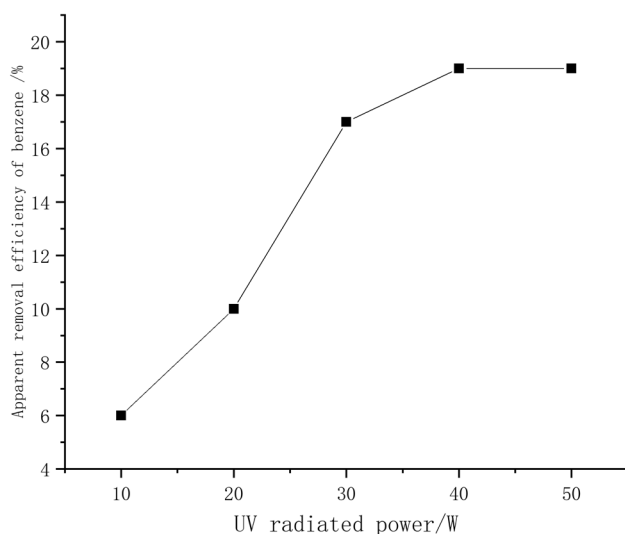
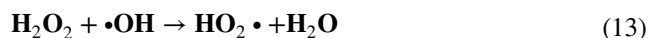


Fig. 8 Influence of UV power for benzene removal



Conditions Aside from the UV light intensity, the other experimental conditions were in accordance with those described in the “Apparent removal using different methods” section.

Effect of initial benzene concentration

Under the same reaction conditions, benzene was used to study the effect of pollutant concentrations from 248 to 756 mg/m^3 in the experiments. As shown in Fig. 9, the removal efficiency decreased when the initial concentration of benzene was increased. For a single chemical reaction system, according to the general theory, increasing the reactant concentration can raise the rate. However, in the photochemical reaction, improving the removal required more oxidants for degradation, and the higher oxidant concentrations were increasingly obstructive to UV radiation, causing the OH to only be irradiated by a smaller amount of UV light, which led to the formation of fewer free radicals (Aleboye et al. 2005). Thus, the rate decreased.

Conditions Aside from the pollutant concentration, the other experimental conditions were in accordance with those

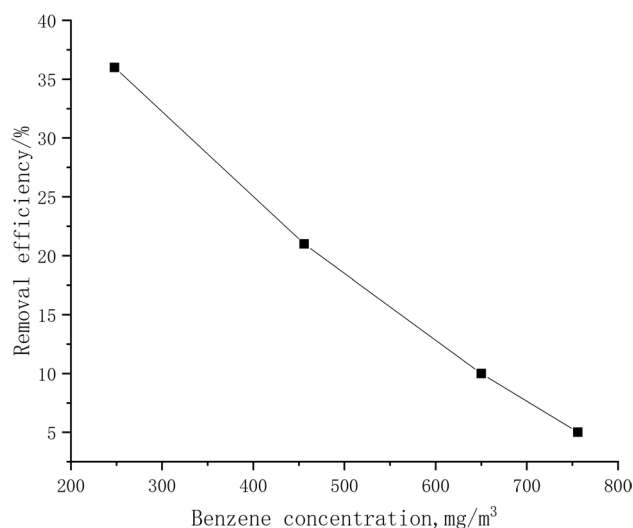


Fig. 9 Influence of concentration on benzene removal

described in the “[Apparent removal using different methods](#)” section.

Conclusion

The reaction system, which included a reactor made of FEP film, $[H_2O_2]_g$ as an oxidant, and UV light, was stable in the study and could also remarkably remove the benzene derivatives. In the present work, the reaction conditions were unfavourable compared with the conditions used in related studies. In particular, this study used shorter residence times and higher pollutant concentrations; thus, it was expected that with increasing reaction time, satisfactory efficiency would be obtained. In addition, it was very important to ensure the stability of the H_2O_2 , pollution load and light transmittance; therefore, enhancing the power of the UV light and reducing the pH of the H_2O_2 solution and the pollutant concentration could improve the treatment effect in the experiment. In the GPAO experiments, although the characteristic structures of the benzene derivatives were destroyed, there were multiple intermediate polar products that caused the apparent removal rate to be much higher than the mineralization rate. Thus, it is necessary to further research the complete removal of waste.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11356-021-16920-w>.

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Author contribution Yuping Jiang analysed and interpreted the experiment and data of present study and was a major contributor in writing the manuscript; Juanjuan Song review the content of the article; Andong ZHU prepared the instrument for the experiment. All authors read and approved the final manuscript.

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Availability of data and materials All data generated or analysed during this study are included in this published article.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent to publication Not applicable.

Competing interests The authors declare no competing interests.

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