

Gas-Phase Advanced Oxidation (GPAO) for Gas Containing Benzene Series by Ultraviolet Irradiation/Hydrogen Peroxide Vapour (UV/[H₂O₂]g) Process

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2 Benzene Series by Ultraviolet Irradiation/Hydrogen Peroxide Vapour
3 (UV/[H₂O₂]_g) Process

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7
8 **ABSTRACT**

9 Hydrogen peroxide (H₂O₂) has the remarkable characteristics of strong
10 oxidation, in which its vapour ([H₂O₂]_g) has further advantages, such as
11 economize and good light transmission. But there is very rare research
12 about its degradation effect in Gas-Phase Advanced Oxidation (GPAO).
13 In the present study, the photochemical oxidation for gas containing
14 benzene series, using ultraviolet (UV) irradiation and [H₂O₂]_g, was
15 investigated in a transparent bag made of fluorinated ethylene
16 propylene (FEP). Only UV or [H₂O₂]_g hardly reduced the pollutant in
17 5 hours in which the reactor was also stable. With the high pollutant
18 concentration (248 to 756 mg/m³) and short residence time (3S)
19 compared with the related studies, studying the degradation of
20 benzene, toluene and xylene, the apparent rate by
21 UV/[H₂O₂]_g/(powder active carbon, PAC) was higher compared with

22 the different way ($UV/[H_2O_2]_g$, $UV/[H_2O_2]_g/TiO_2$ and
23 $UV/[H_2O_2]_g/ZnO$), but it was found that the terminal degradation by
24 $UV/[H_2O_2]_g$ significantly decreased, in which the conductivity
25 decreased after reaction. The effects of increasing pollutant
26 concentration and H_2O_2 pH were negative for the treatment, but the UV
27 radiated power positive till 40W. In addition, the characteristic
28 absorbance for three benzene series showed that the key structure of
29 pollutant molecule were damaged in GPAO.

30 **Key word:**

31 Hydrogen Peroxide Vapour; Benzene series; Degradation;
32 Conductivity; Characteristic absorbance

33

34 **Introduction**

35 People need to breathe every time, so everyone can not avoid
36 the influence of the pollution. But as widely known in reality, air
37 quality is not too well, specially in development countries. Thus the
38 removal ways are increasingly studied, in which the removal
39 methods of air pollution include adsorption, absorption, advanced
40 oxidation (AO) and so on. Compared with other treatment ways, AO
41 shows more complete and quicker removal of pollutants, in which

42 photochemical methods (without catalyst) have obtaining more
43 attention in recent years [Alapi et al., 2007; Jinhan et al., 2009;
44 Lianfeng et al., 2013], because there are some distinctive
45 advantages, such as energy efficiency and running costs which must
46 be evaluated in reality [Matthew et al., 2014].

47 Both hydrogen peroxide and ozone are the well known oxidants,
48 are usually used in aqueous and gas systems respectively [Wang et
49 al., 2000]. Although ozone is probably the oxidant leader-species
50 because its 2.07V oxidation-reduction potential (ORP) is higher than
51 hydrogen peroxide 1.8V, the two substances have the same effect
52 that can produce the .OH radical regarded as the key oxidant in AO
53 process. Further more, H₂O₂ has multiple features. First, it is very
54 easy to generate the substance as long as electron donators exist in
55 the water, chromophoric substances can produce hydrated electrons
56 by illumination and combine with dissolved oxygen, which leads to
57 a series of reactions for H₂O₂ outcome [Anastasio et al., 1997;
58 Wohlgemuth et al., 2001]. Second, the artificial production for
59 hydrogen peroxide is very convenient, even for the in-situ
60 production [Choudhary and Jana, 2007; Fang et al., 2013]. Third,
61 H₂O₂ can be used as an indicator of AO process, because the
62 representative oxidant, hydroxyl radical (OH.) with 2.8V ORP, can
63 be led to H₂O₂ formation by reacting with VOCs, CO, SO₂ and so on.

64 It is obvious that in the AO with multiple reaction by radical chains,
65 hydrogen peroxide plays the central role [Detlev et al., 2009].

66 Due to the uniqueness for H_2O_2 , the AO by H_2O_2 was usually
67 used as oxidatant solution to remove liquid pollutants, combined
68 with ultraviolet radiation(UV) [Bin et al., 2007; Muruganandham
69 et al. 2004], in which the solution state of reactants caused the long
70 residence time for the waste. To the gas pollutants treatment, some
71 researchs displaied that the UV/ H_2O_2 could oxidize SO_2 , NO_x and
72 Hg^0 with low concentration in flue gas[Yangxian et al.,
73 2011;Yangxian et al., 2014]. However, the researchs focusing on
74 GPAO by [H_2O_2]_g were very rare, especilly with the high pollutant
75 concentration and short residence time, even if the vapour was only
76 used for sterilization [French et al., 2004;Xue et al., 2012]. Actually,
77 the vapour form for H_2O_2 not only maintain the features of
78 solution-phase, but also cause some advantage, such as saving
79 oxidant consumption and reducing the loss of UV light when passing
80 the reaction system.

81 To research the GPAO by H_2O_2 vapour, and understand the
82 effect for the harsh condition compared with the related studies, such
83 as high concentration pollution and short residence time. We
84 conducted the experiment to investigate the decomposition of
85 benzene, toluene and xylene by UV/[H_2O_2]_g, the reactions took place

86 in the transparent bag of fluorinated ethylene propylene (FEP) .

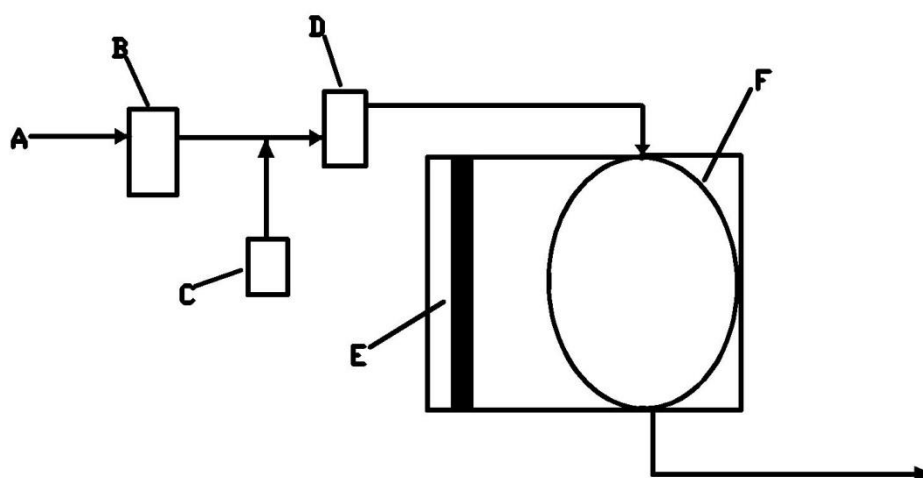
87 Benzene series were chosen as the target compounds because
88 they were the frequently encountered organic pollutants including
89 benzene, toluene and xylene, for example benzene and toluene is the
90 frequently encountered organic compounds in indoor and outdoor
91 environments [Wallace et al., 1985; Bernstein et al., 2008], serve
92 as the representative organic contaminant [Liu et al., 2016], can
93 cause the multiple diseases for people.

94 **2. Experimental section**

95 **2.1 Materials and analysis**

96 The reagents were used as received without further treatment.
97 Benzene, toluene, xylene, Na(OH), HCl and 30% commercial H₂O₂
98 solution (Guoyao chemical Reagent Co., China), were analytical
99 reagents (AR).

100 **2.2 experimental procedure**



101

102 Fig. 1. Schematic of experimental system. Legend: A, Air; B, liquid
103 pollutant vessel; C, H₂O₂ solution vessel; D, gas mixing tank; E, UV
104 lamp (253.7nm) ; F, FEP bag.

105

106 The experimental system used in this study was illustrated in
107 Fig. 1. The flows of air was blown through the FEP pipes and
108 regulated by the rotameters, to transfer and mix the vapour of
109 benzene series and H₂O₂. According to the requirement, powder
110 active carbon (PAC), TiO₂ and ZnO of the equal quality (2g) were
111 put into gas mixing tank. Then the mixed gas photo-reacted in the
112 FEP bag under the UV lamp irradiation. The inlet and outlet
113 concentrations of reactants were measured through the gas bypass
114 line. To determined the apparent degradation rate, the pollutant
115 concentrations were monitored via gas chromatography (GC) with

116 FID and AB-INOWAX fused silica capillary column in which the
117 retention time was 1.2-1.6 min and calculated by equation (1); to
118 terminal rate, vis CO₂ detector (GT-1000-CO₂, made in KORNO Co.
119 Ltd. China). The characteristic absorbance for pollutant was
120 determined by UV-vis spectra. The apparent and terminal removal
121 efficiency is calculated using the following equation (2) and (3)
122 respectively.

$$123 \quad C(\text{mg}/\text{m}^3)=(A-318.5)/2.39 \quad (1)$$

$$124 \quad \text{Apparent removal efficiency } (\%)=(C_{\text{in,p}}-C_{\text{out,p}})*100\%/C_{\text{in,p}} \quad (2)$$

$$125 \quad \text{Terminal removal efficiency } (\%)=(C_{\text{out,c}}-C_{\text{in,c}})*100\%/C_{\text{o,c}} \quad (3)$$

126

127 Where A is the peak area of waste concentration determined by GC,
128 the correlation coefficient for (1) is 0.9997; C is the concentration of
129 benzene series, mg/m³; C_{in,p} and C_{out,p} were the inlet and outlet
130 concentration of pollutant separately; C_{in,c} and C_{out,c} of CO₂; C_{o,c}
131 was the total concentration of CO₂ corresponding to the pollutant by
132 complete oxidation.

133 To investigate the effect for pH and conductivity, the H₂O₂ pH
134 value was adjusted by adding HCl and NaOH, was determined using
135 an acidimeter (Model Starter 2100/3C Pro, Shanghai Hongji
136 Instrument Co., Ltd. China), the conductivity using an
137 Conductivity Meter (DDSJ-319L, INESA SCIENTIFIC

138 INSTRUMENT CO., LTD. China).

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140 **3 Result and dicussion**

141 **3.1 Single effect with UV or H₂O₂**

142 3.1.1 Apparent degradation by UV radiation

143 Benzene was used as the target compound for this research section.

144 With the maximum power of UV radiation in our study as 30W,

145 Fig. 2 shows benzene is resistant under different time in which the

146 apparent degradation rate was in the range of 0.5%-1%, there was

147 little direct UV photo-oxidation and the indirect way caused by the

148 substance in ambient air, such as O₂ and H₂O. It is accordant with

149 the studies that toluene and benzene in air could be hardly

150 degraded under >200nm UV ligh [[Zhang et al., 2003](#); [FENG et](#)

151 [al., 2019](#)], because 253.7 nm is too high to break the benzene

152 molecular bond by the photon energy at this wavelength. Although

153 dimethylbenzene could be reduced under 254nm after taking a long

154 time (>3h) [[Ma et al., 2007](#)], the substance is different from benzene

155 series. In addition, it is critical to understand the impact of reactor

156 walls on gas-phase reactivity, due to the background offgasing of

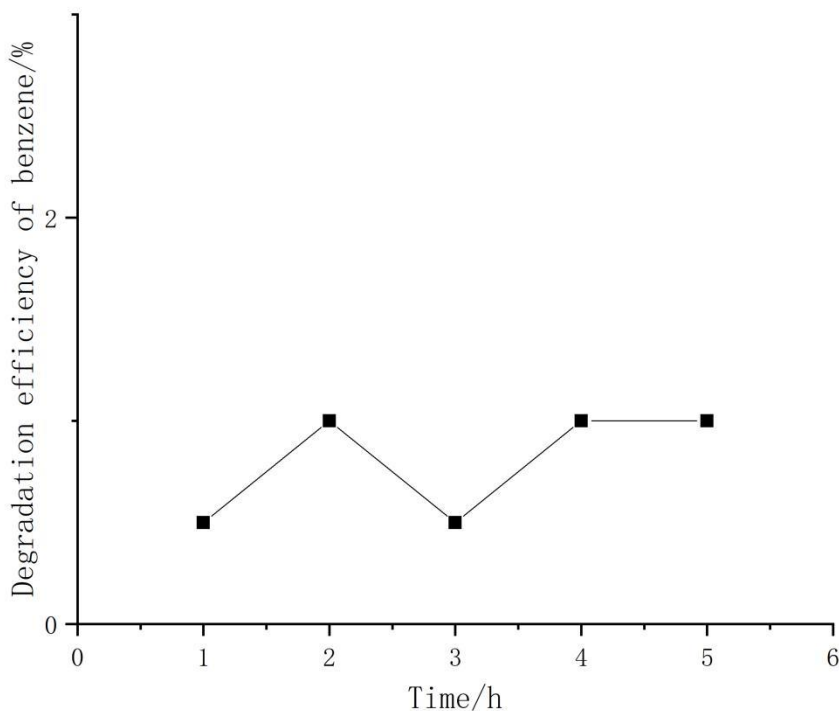
157 NO_x, other reactive species and heterogeneous reactions that cause

158 “chamber radical sources” upon irradiation [[Carter et al., 1982](#)].

159 But Fig.2 shows that the effects did not effectively influence the
160 pollutant reduction in our study.

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

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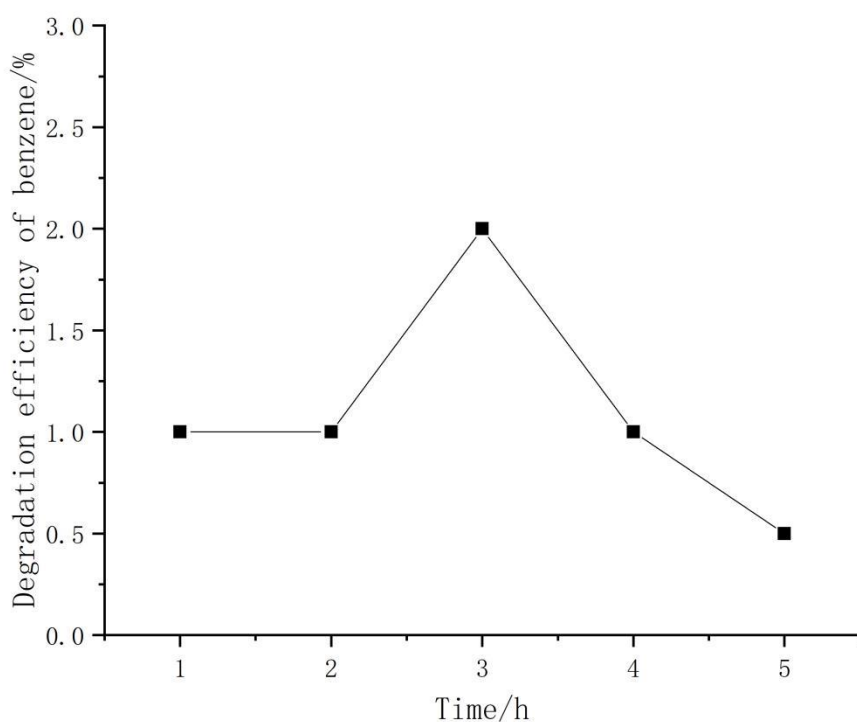
166 Fig. 2 apparent degradation of benzene only by UV radiation

167 3.1.2 Apparent degradation by [H₂O₂]_g

168 The performance of benzene oxidation only by [H₂O₂]_g was
169 investigated without UV radiation. In Fig.3, it could be found that
170 the pollutant quantity in 5 h reaction decreased very slightly (≤1%),
171 which implied that benzene is extremely inert and difficult to be

172 degraded by $[\text{H}_2\text{O}_2]_{\text{g}}$ oxidation alone. It can be deduced that the
173 oxidation capacity of H_2O_2 to benzene is very limited, and can not
174 release $\cdot\text{OH}$ free radicals for the reaction without the radiation of UV.
175 This phenomenon also proved that generating hydroxyl radicals is
176 the very necessary for GPAO. On the other hand, it was obvious that
177 the reactor was also stable under $[\text{H}_2\text{O}_2]_{\text{g}}$ oxidation.
178 *Conditions.* The experimental conditions were similar to that in 3.1.1,
179 in which 4.6mol/m^3 $[\text{H}_2\text{O}_2]_{\text{g}}$ were used to replace the UV light.

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Fig. 3 apparent degradation of benzene only by $[\text{H}_2\text{O}_2]_{\text{g}}$

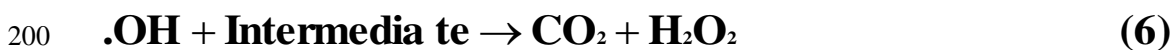
184 **3.2 Apparent and terminal degradation**

185 3.2.1 Apparent degradation by different way

186 All the three treatment ways could reduce the contaminants as
187 shown in Fig.4. Although the maximal efficiency is 31% for the
188 toluene which lower than the related researchs [Yangxian et al.,
189 2014; Bin et al., 2007], it is obtained in the case of very short
190 residence time and high concentration pollutant. Thus the result was
191 still very meaningful in which [H₂O₂]_g, TiO₂ and ZnO played the
192 important oxidant role in the course. The fundamental role for H₂O₂
193 lies in the decomposing into •OH free radicals under UV radiation
194 with a certain wavelength according to the following reaction [Lee
195 et al., 2004].



197 Then the degradation of pollutants could be expressed as the
198 following equations [Bin et al., 2007]:



201 In the other three treatment ways mixing with catalyist and PAC, the
202 potential problem was that the H₂O in H₂O₂ vapor could hinder the
203 degradation because of its competitive adsorption onto the reaction
204 surface with contaminants [Herrmann et al., 1999; Nazir et al.,
205 2003]. But there were also some promotions in the progress, because

206 the two catalysts also can produced the .OH radical under 253.7nm
207 UV light; in addition, owing to the higher electron affinity of H₂O₂
208 (1.8eV) than O₂ (0.44 eV), the excited electrons on the conduction
209 band of TiO₂ could be captured by H₂O₂ more easily than by O₂,
210 which increased the production of ·OH. For the PAC, the adsorption
211 performance could be helpful to the pollutant reduction. It is obvious
212 that the negative impacts for TiO₂ and PAC were less than the
213 positive, and the latter treatment is better in which the huge
214 adsorption surface area has made a great contribution. For the three
215 pollutant, the desired enhanced effect for ZnO did not occurred, but
216 rather weakened compared with the other treaments. It was
217 concluded that ZnO was prone to photocorrosion [HOFFMANN et
218 al., 1995; Han et al., 1999], and its density is most highest, then the
219 shortcomings caused the instability and heterogeneity in the reaction
220 course.

221 *Conditions.* The experimental conditions were similar to that in 3.1.1,
222 in which 4.6mol/m³ [H₂O₂]g was also applied.

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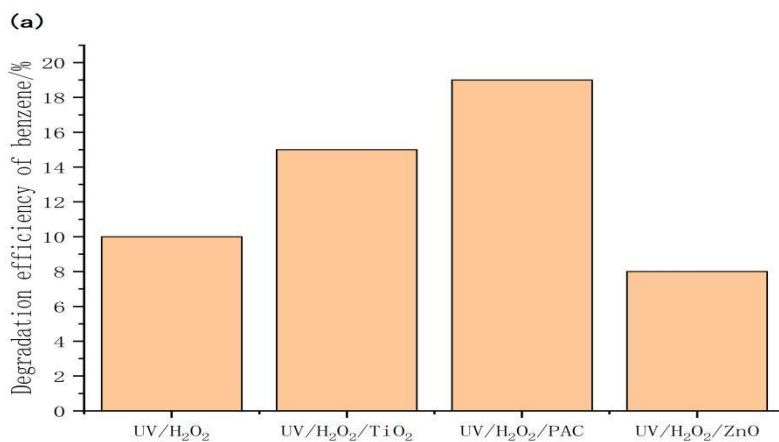
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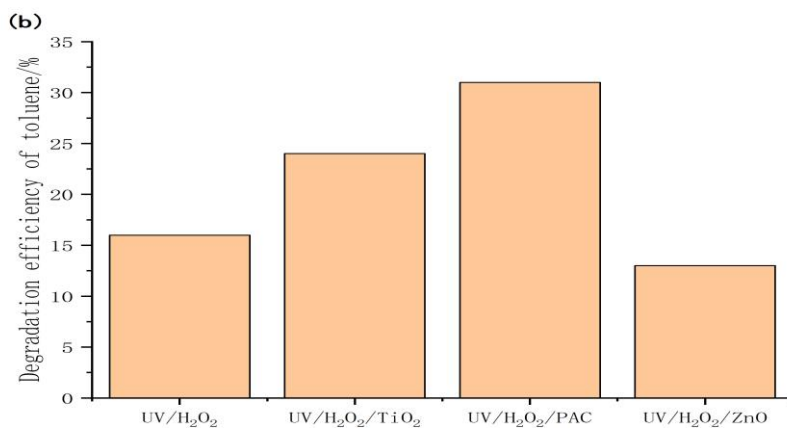
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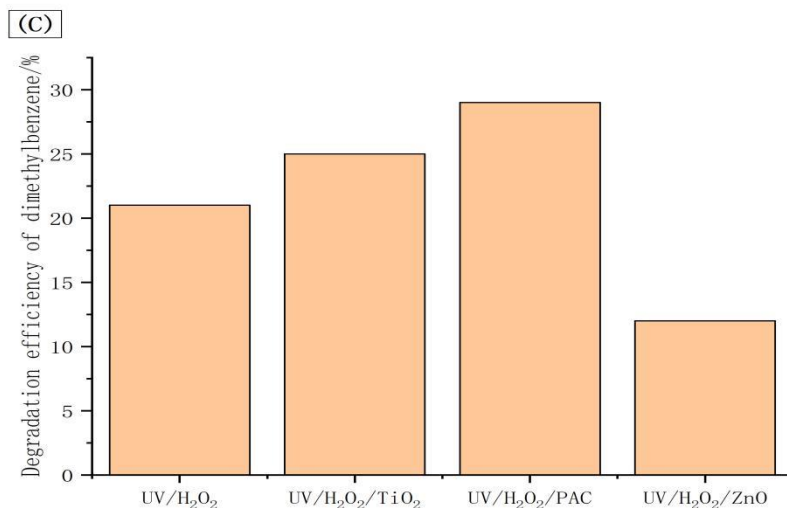
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Fig.4 Apparent degradation rate by different ways with H₂O₂

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252 3.2.2 Terminal degradation by UV/[H₂O₂]_g

253 In Fig.5, it was obvious that for both apparent and terminal rate,
254 the xylene is highest, the benzene lowest. For the toluene and xylene,
255 there are separately one and two methyl groups substituting the
256 hydrogen in benzene ring, which weaken the stability compared with
257 benzene. These exocyclic groups easily became the attacked points
258 for oxidative species, then destroyed the whole molecular structure.
259 Due to more substituting, degradation rate of xylene was most
260 remarkable.

261 The results also showed that although the apparent effects were
262 more than 10%, the terminal were very low in which the maximum
263 was less than 5%. It was denoted that most of the products were
264 intermediates, which accordant with multiful studies [[Kuo-Pin et al.,](#)
265 [2007](#); [Matthew et al., 2014](#); [Feng et al., 2019](#)]. This phenomenon
266 also revealed the uncertainty for the GPAO which can not
267 completely removed the contaminants, hence limited the
268 application.

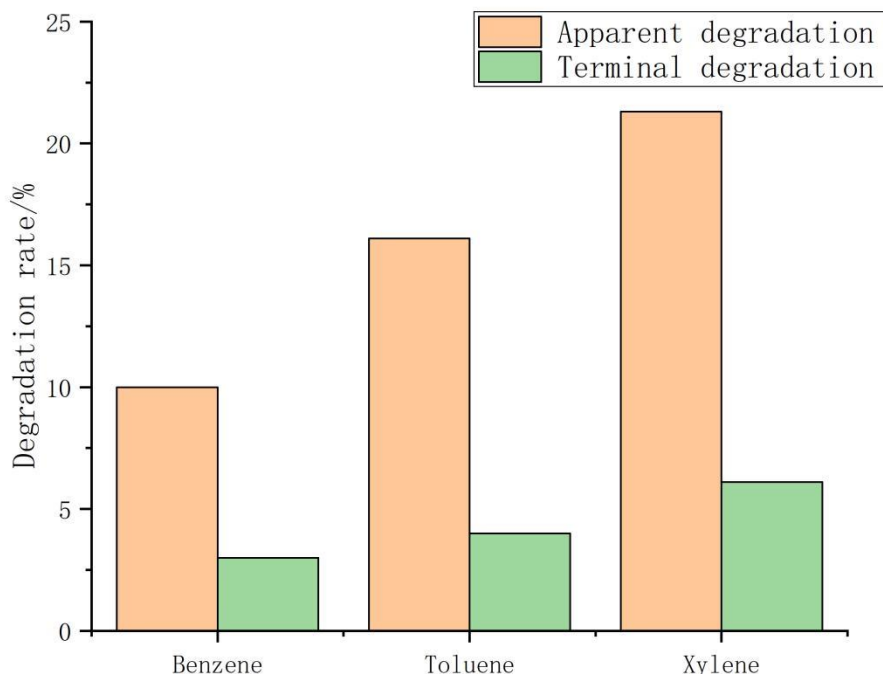
269 *Conditions.* The experimental conditions were accordant with that in
270 3.2.1.

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Fig. 5 Comparison between terminal and apparent degradation

277 3.3 characteristic absorbance

278 Due to the multiple intermediate products in GPAO, it is meaningful
279 and necessary to focus characteristic of molecule for determining
280 whether pollutant structure were damaged. For benzene series,
281 conjugated ethylene in benzene ring plays the directive role, UV-Vis
282 was used to investigate the inlet and outlet difference for the
283 indicator. Although benzene series had B-absorption band
284 (230-270nm) and E2 (200-210nm) respectively, the latter intensity

285 is always higher than the former. Thus E2 was used as the the
286 maximum absorbance in our experiment, and the results are
287 mentioned in Table 1, which shows that the wavelengths value and
288 absorption intensity for toluene and xylene are higher than benzene,
289 because there were red-shift and and hyperchromic effect. It is
290 believed that the methyl groups in the two pollutants are the
291 auxochromes, their electrons in n orbit can produce conjugation
292 effect with them π from the chromophores, then the electron cloud
293 can influence each other, which change the absorption intensity and
294 peak position [LI et al., 2010]. Table 1 also dispalys that there were
295 high absorbance for inlet pollutant than outlet, this phenomenon
296 proved that it is also the feasible way for determining the
297 degradation effect. In addition, although this method has clear
298 response to the conjugated ethylene in benzene ring, it is rarely used
299 in GPAO because the very few pollutant quantity in the sample
300 solution weaken the absorption peak very much; in our work, this
301 problem was solved by increasing pollutant initial concentration and
302 ending collection through longer time sampling over.

303 *Conditions.* The experimental conditions were accordant with that in
304 3.2.1.

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Table 1 characteristic absorbance by UV-Vis

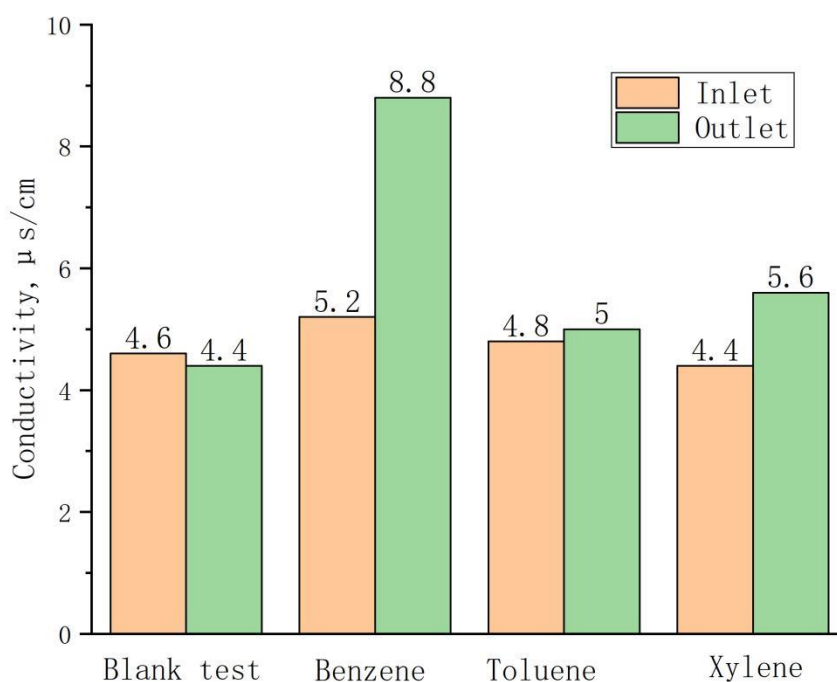
Pollutants	benzene	toluene	xylene
Characteristic Wavelength (nm)	200	210.4	207.2
Absorbance of inlet pollutant	0.06813	0.0944	0.08744
Absorbance of outlet pollutant	0.02290	0.03012	0.05159

309 **3.4 Variation of conductivity**

310 In the similar works, conductivity was not studied as the
 311 measure index, although it can indicate the special substance
 312 changes after reaction. Fig.6 shows the conductivities of inlet and
 313 outlet for different pollutant degradation, in which the data slightly
 314 decrease in blank test, but there are opposite effect on other
 315 pollutants. The result in blank test could be explained by the fact that
 316 the UV decomposition of H₂O₂ caused its ionization reduction. For
 317 the latter measurement, [Martina et al.](#) found that the improvement
 318 of polar products was a positive correlation with conductivity
 319 [\[Martina et al., 1998\]](#), therefore it was believed that the
 320 intermediates such as aldehyde, ketone and alcohol, were polar

321 products accelerated by the reaction and could not be
322 completely subtracted due to the limited retention time. In addition,
323 the increase values for benzene, xylene and toluene were 3.6, 1.2,
324 0.2 respectively. This result also indicated the difference for
325 their quantity of polar intermediates, which is accordant with some
326 studies [Kislov et al., 2004; Hamilton et al., 2005; Yu et al., 1997].
327 *Conditions.* The experimental conditions were accordant with that in
328 3.2.1.

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Fig.6 Variation of conductivity for different pollutant degradation

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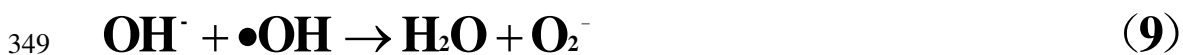
333 **3.5 Effect of H₂O₂ pH**

334 The benzene vapour was selected for investigated the pH
335 influence. The effect on the waste removal efficiency is shown in Fig.
336 **7** in which the rate decreased from 28.6 to 1.7 with the H₂O₂ solution
337 pH increase 4 to 9. It is widely recognized that H₂O₂ has stronger
338 oxidizing property under acidic conditions, therefore the degradation
339 was reduced with increasing the solution pH. For the reasons, some
340 researchs proved that H₂O₂ in solutions can produce the HO₂⁻ species,
341 which is a scavenger of ·OH radicals via the following hydrolysis
342 reaction[Chang et al., 2008; Liu et al., 2011]. When the solution is
343 alkaline, OH⁻ can consume H⁺ and accelerate HO₂⁻ formation.



345

346 Besides, the OH⁻ can also consume ·OH radicals through the
347 following reaction [Zehavi and Rabani; 1971], thereby reducing
348 the removal efficiency of benzene



350 *Conditions.* Except for pH, the other experimental conditions were
351 accordant with that in 3.2.1.

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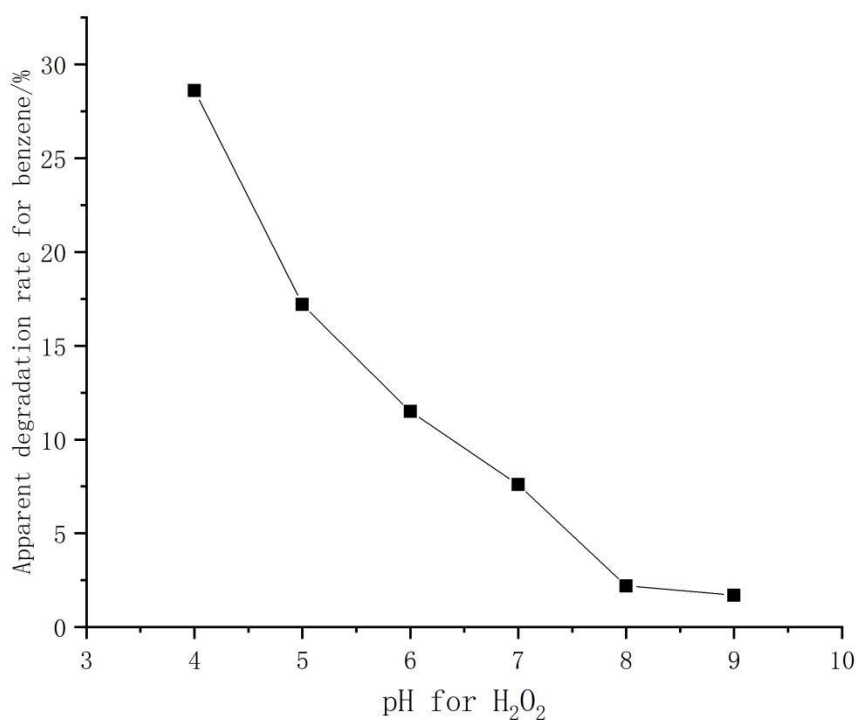


Fig.7 Effect of H₂O₂ pH for benzene degradation

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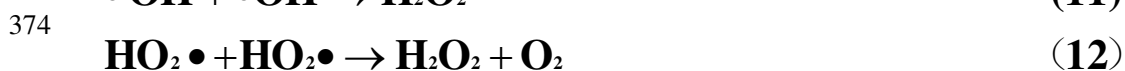
356

3.6 Apparent degradation by different UV power

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358 The influence of UV radiation has been investigated by the
 359 different UV power of 10W, 20W, 30W, 40W and 50W, in which the
 360 apparent degradation rate of benzene were 6%, 10%, 17%, 19%,
 361 19% respectively, as shown in the fig. 8. The results display that
 362 the degradation effect increase with raising the UV power at the
 363 value from 10W to 40W, in which the improvement between 30W
 364 and 40W was slighter than the former and no progress from 40W to
 365 50W. It could be deduced that the .OH concentration were increased

366 by increasing the UV intensity, thus the chain reactions were
367 strengthen to obtain a higher reactant consumption rate [Tadic et al.,
368 2002]. However, when the UV radiated power exceeds a certain
369 value, several side reactions (reaction 10-13) also simultaneously
370 occurred in the system, resulting in the self-loss of ·OH. Thus, with
371 further increases in the light power of 40W, the pollutant removal
372 efficiency only increased slightly, then not change in 50W which
373 was the equilibrium state of opposite action.



375

376 *Conditions.* Except for UV light intensity, the other experimental
377 conditions were accordant with that in 3.2.1.

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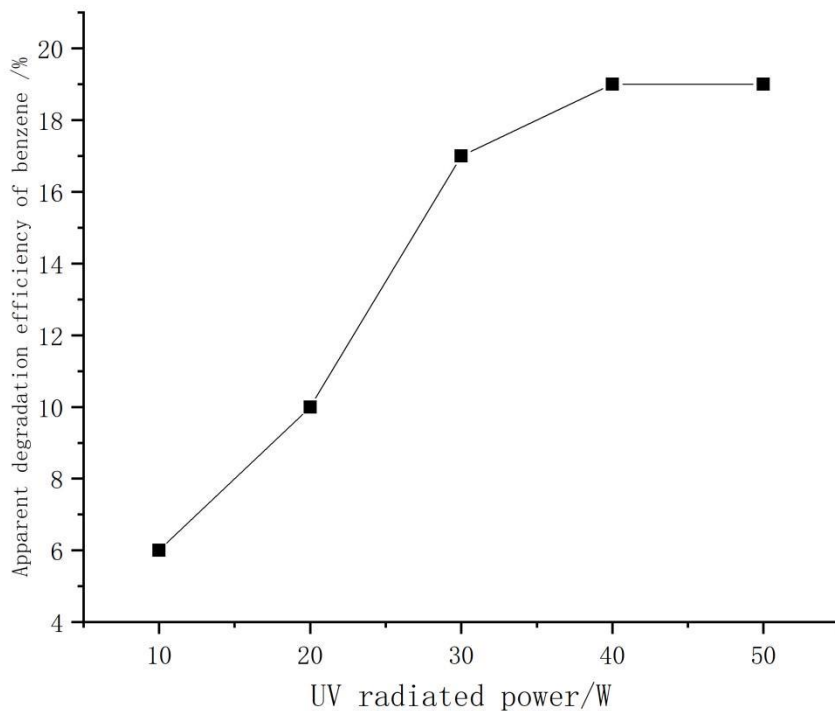


Fig. 8 Influence of UV power for benzene degradation

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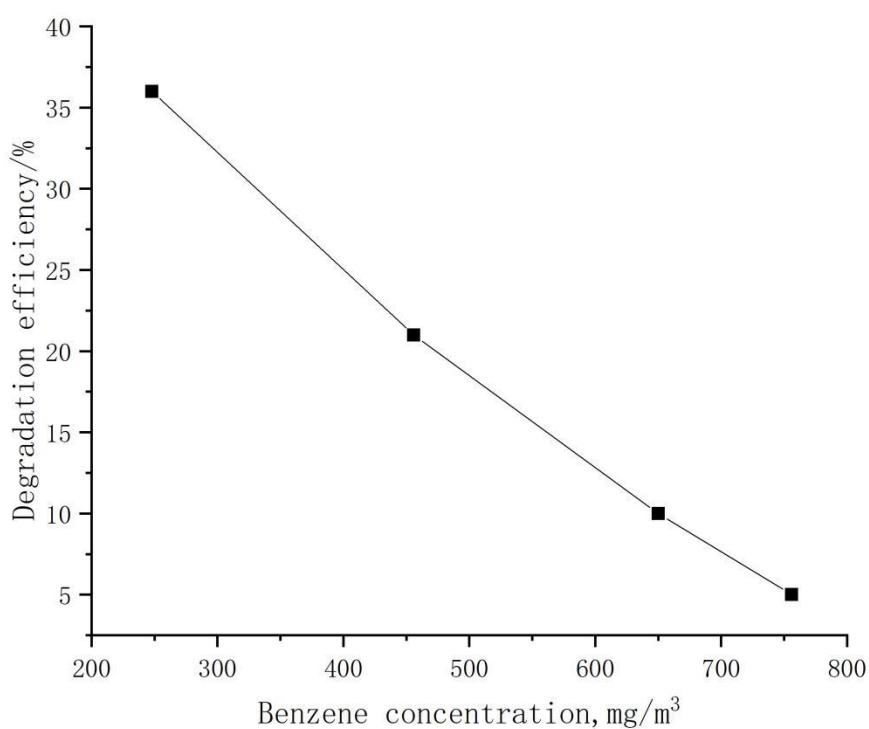
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383 **3.7 Effect of initial benzene concentration**

384 Under the same condition of reaction, benzene was used to
385 study the effect of pollutant concentration from 248 to 756 mg/m³ in
386 the experiments. The results are shown in Fig. 9, in which the
387 degradation efficiency decreased when the initial concentration of
388 benzene is increased. For a single chemical reaction system, by the
389 general theory, increasing reactant concentration is able to raise the
390 rate. But for the photochemical reaction in which improving
391 degradation required more oxidants for degradation, elevating

392 induced more and more impermeable to UV radiation which cause
393 the .OH can only be irradiated by a smaller portion of UV light to
394 form lower free radicals [Aleboyeh et al., 2005], thus the rate
395 decreased.
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Fig. 9 Influence of concentration for benzene degradation

Conditions. Except for pollutant concentration, the other experimental conditions were accordant with that in 3.2.1.

405 **Conclusion**

406 The reaction system including the reactor made of FEP film,
407 $[\text{H}_2\text{O}_2]_g$ as oxidant with UV light, was stable in the study, could also
408 remarkably removal benzene series. For the present work, the reaction
409 condition are unfavorable compared with the related, especially to shorter
410 residence time and higher pollutant concentration, thus it is expected that
411 with the increase of reaction time, satisfied efficiency can be obtained. In
412 addition, it was very important to ensure the stability of H_2O_2 , pollution
413 load and light transmittance, therefore enhancing the power of UV-light,
414 reducing the pH of H_2O_2 and pollutant concentration could improve the
415 treatment effect in our experiment. In the GPAO course, although the
416 characteristic structure of benzene series can be destroyed, there were
417 multiple intermediate polar products, which cause the apparent
418 degradation rate was much higher than the terminal. Thus it is necessary
419 to further reserach for completely removing the waste.

420 **Acknowledgments**

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423 work.

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425 **-Consent to Participate:** Not applicable.

426 **-Consent to Publish:** Not applicable.

427 **-Authors Contributions:**Yuping JIANG analyzed and interpreted the
428 experiment and data of present study, was a major contributor in writing
429 the manuscript; Juanjuan SONG review the content of the article; Andong
430 ZHU prepared the instrument for the experiment. All authors read and
431 approved the final manuscript.

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433 Technology Project (2019B2015).

434 **-Competing Interests:**The authors declare that they have no competing
435 interests.

436 **-Availability of data and materials:** All data generated or analysed
437 during this study are included in this published article.

438

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Figures

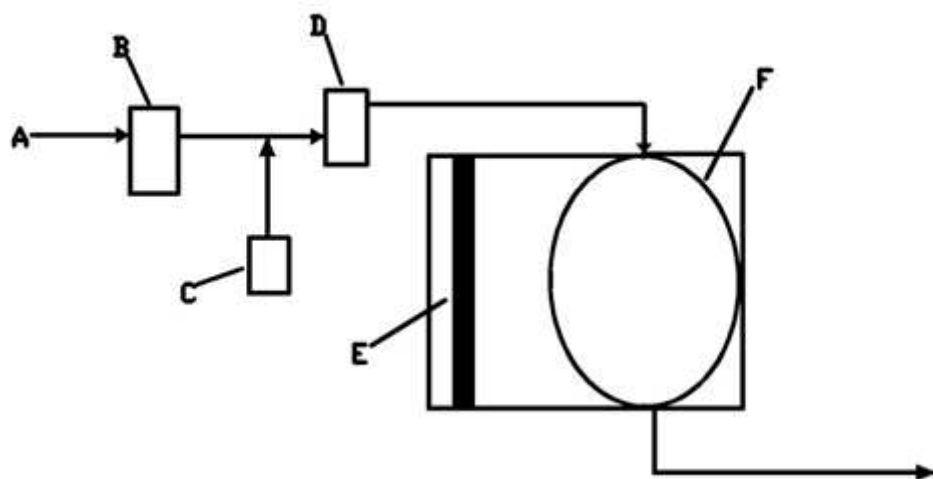


Figure 1

Schematic of experimental system. Legend: A, Air; B, liquid pollutant vessel; C, H₂O₂ solution vessel; D, gas mixing tank; E, UV lamp (253.7nm); F, FEP bag.

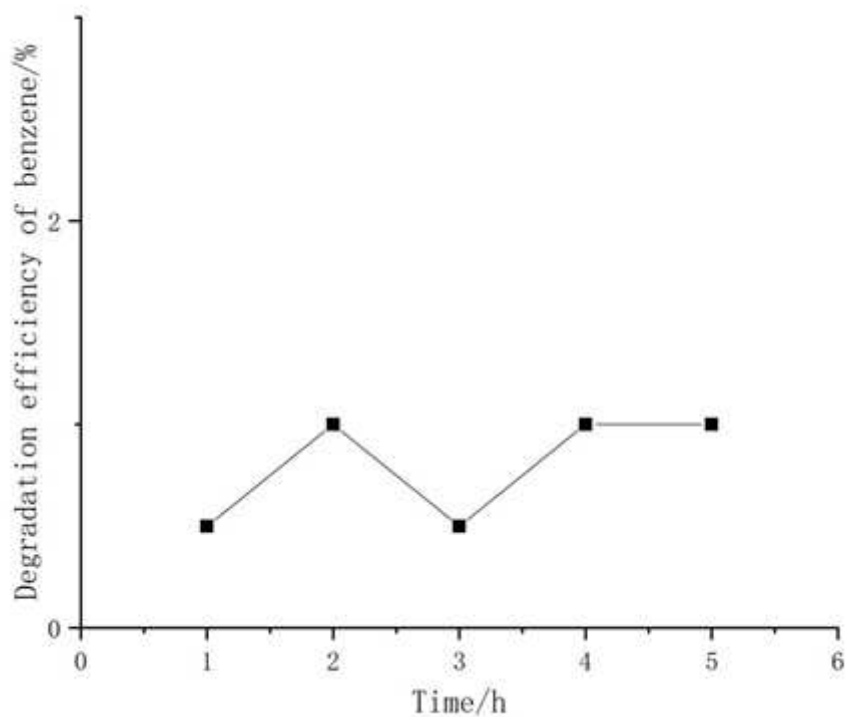


Figure 2

apparent degradation of benzene only by UV radiation

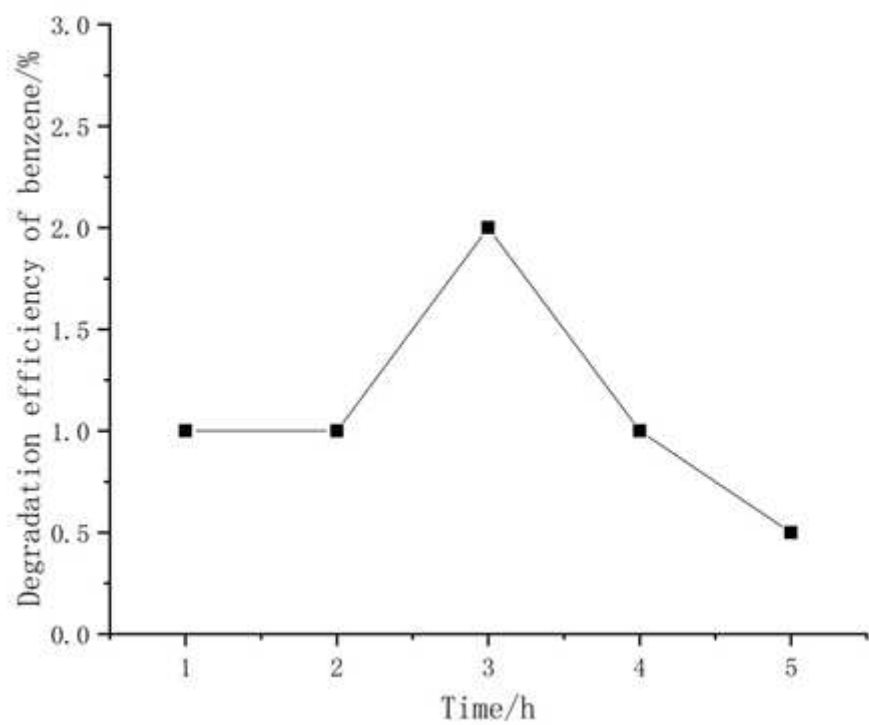


Figure 3

apparent degradation of benzene only by [H₂O₂]_g

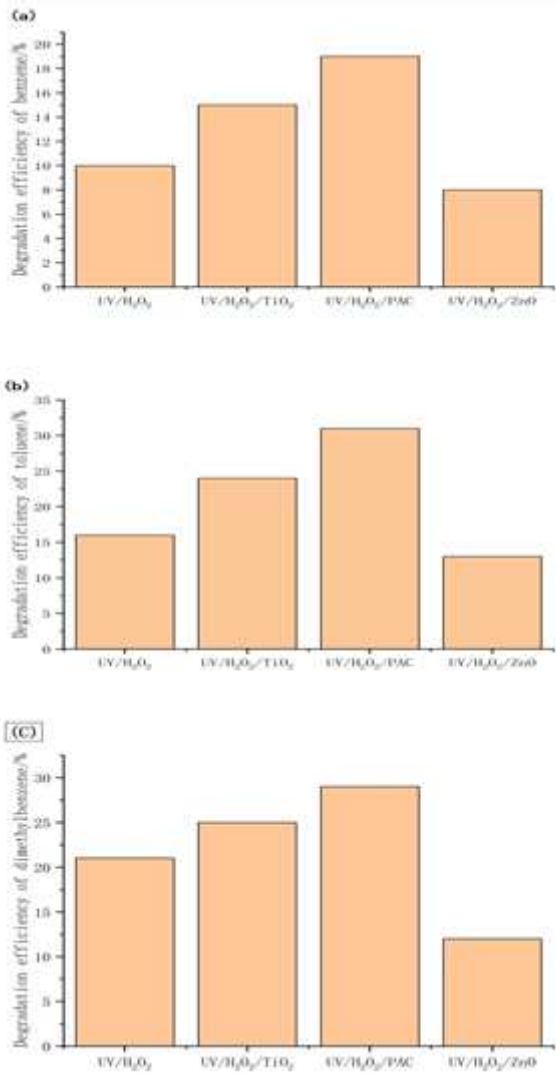


Figure 4

Apparent degradation rate by different ways with H₂O₂

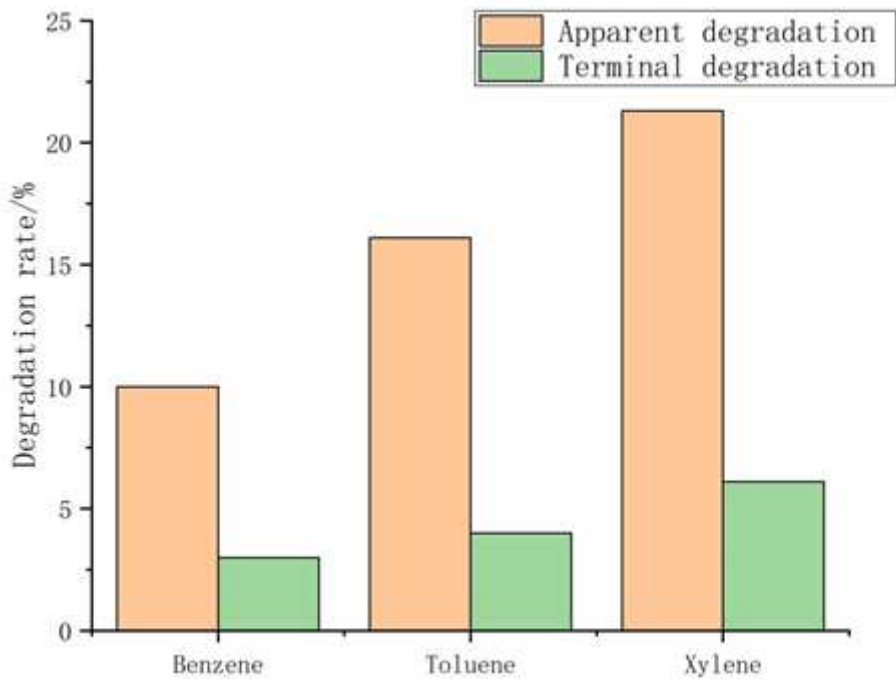


Figure 5

Comparison between terminal and apparent degradation

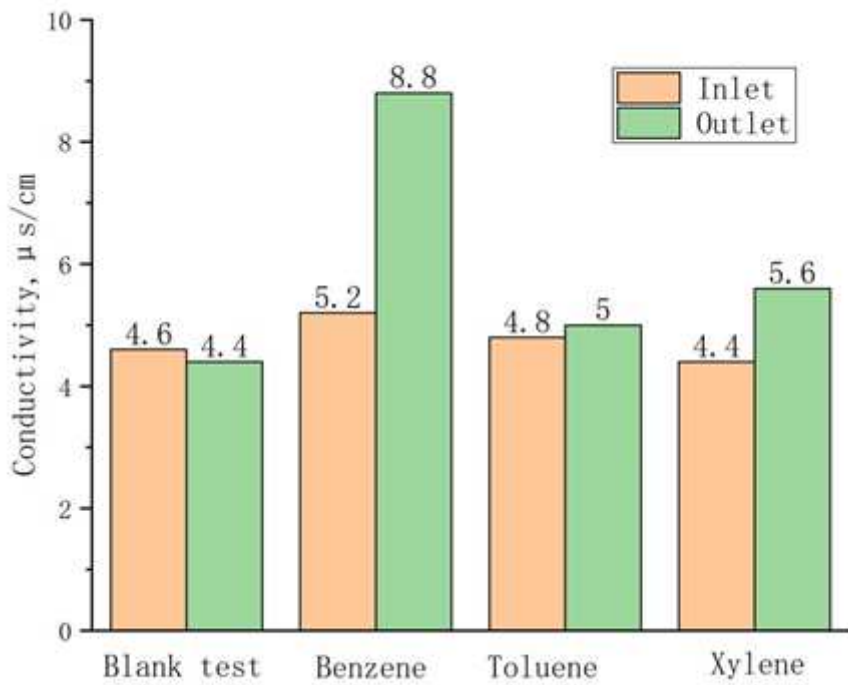


Figure 6

Variation of conductivity for different pollutant degradation

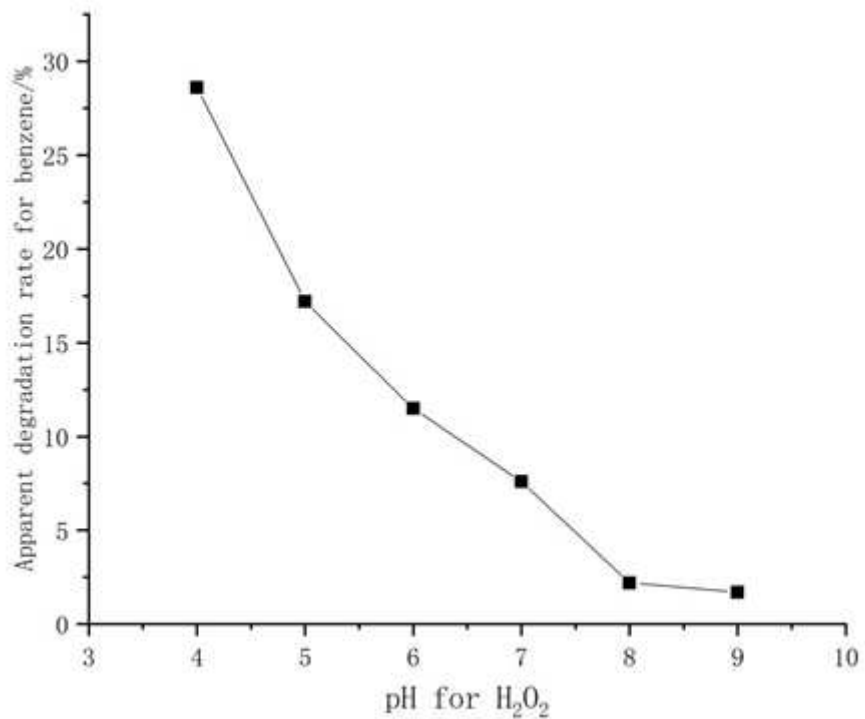


Figure 7

Effect of H₂O₂ pH for benzene degradation

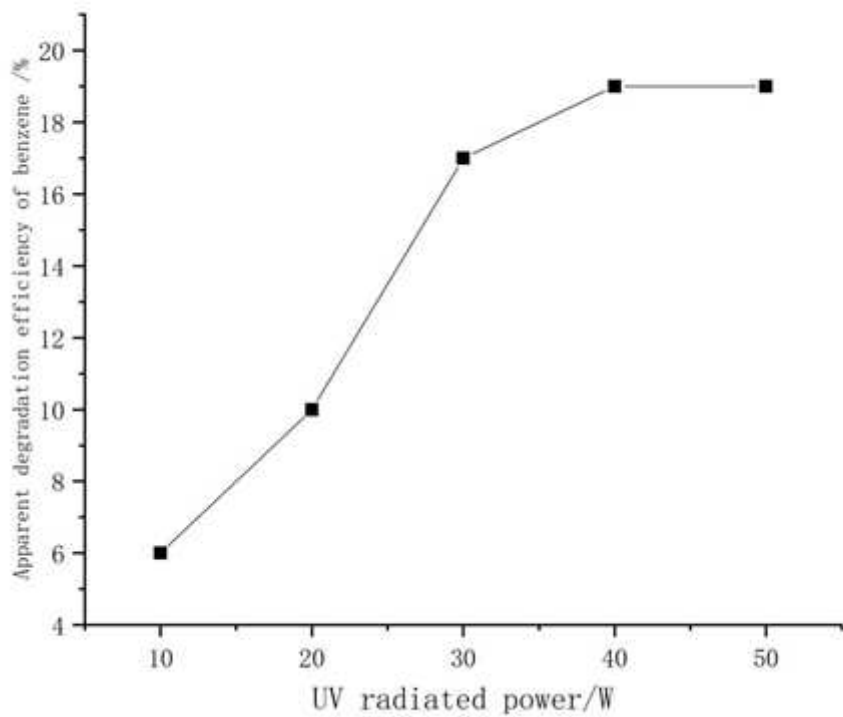


Figure 8

Influence of UV power for benzene degradation

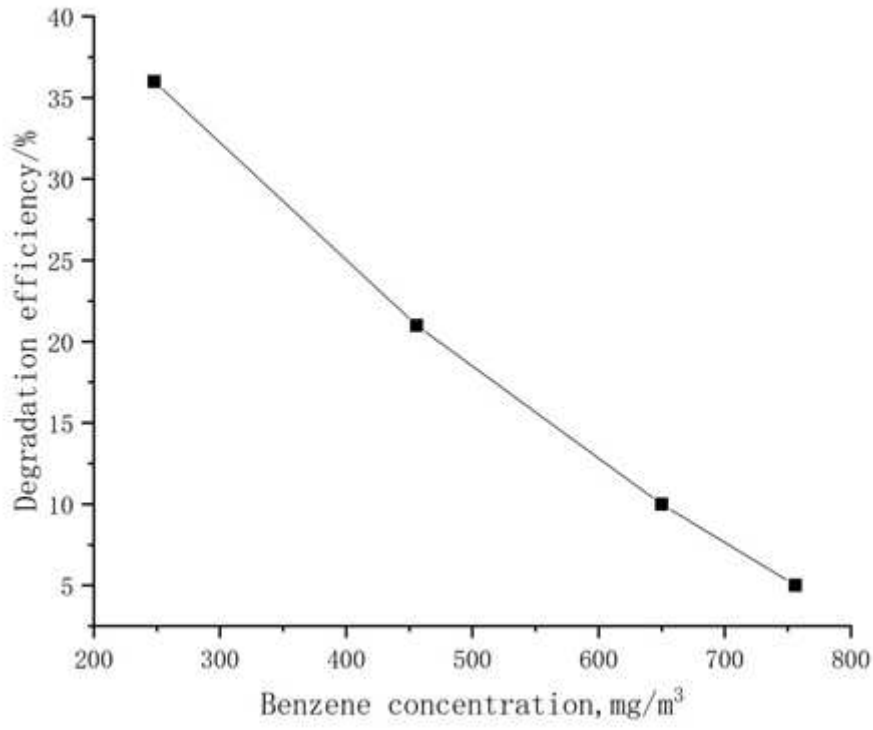


Figure 9

Influence of concentration for benzene degradation