

1 *Review*

2 **Removal of Indoor Volatile Organic Compounds and**
3 **Formaldehyde via Photocatalytic Oxidation: A Short Review and**
4 **Prospect**

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16 **Abstract:** Volatile organic compounds (VOCs) and formaldehyde are ubiquitous in indoor environment.
17 Inhalation of VOCs can cause irritations, difficulty breathing, and nausea, and damage the central
18 nervous system as well as other organs. Formaldehyde is even a carcinogen. Removal of VOCs and
19 formaldehyde is thus critical to control indoor air quality (IAQ). Photocatalytic oxidation has been
20 demonstrated its feasibility to remove toxic VOCs and formaldehyde from indoor environment. The
21 technique is highly-chemical stable, inexpensive, poisonless, and capable of removing a wide variety
22 of organics under light irradiation. In this paper, we review and summarize the traditional air cleaning
23 methods and current photocatalytic oxidation approaches in both of VOCs and formaldehyde
24 degradation in indoor environment. Influencing factors such as temperature, relative humidity,
25 deactivation and reactivations of the photocatalyst are discussed. Aspects on the application of the
26 photocatalytic technique to improve the IAQ are suggested.

27 **Keywords:** VOCs, formaldehyde, photocatalysis, review, influencing factors.

28 1. Introduction

29 As more illnesses being attributed by indoor air pollution, indoor air quality (IAQ) of residential
30 units and workplaces is a serious concern. Human beings spend >80% of lifetime indoors, including
31 any of living and working places such as dwellings, offices, and workshops [1,2]. Typical indoor air
32 pollutants are particulate matters (PM), nitrogen oxides (NO_x), carbon monoxide (CO), volatile organic
33 compounds (VOCs) and formaldehyde. Among those, VOCs [3] and formaldehyde [4] are classes of
34 prominent and representative indoor pollutants. The United States Environmental Protection Agency
35 (U.S.EPA) estimated that the VOCs levels in indoor air is typically 5-10 times higher than that of
36 outdoor air [5]. Currently, over 50% of the precedence-controlled pollutants proposed by U.S.EPA are
37 VOCs [6]. Formaldehyde is differentiated from VOCs because of its ubiquitous presence and various
38 adverse effects on human health. In further, it is a challenge to collect and quantify formaldehyde in the
39 airs owing to its higher polarity and reactivity compared with VOCs. Distinct monitoring and
40 measurement methods are thus required. The most commonly used offline method of simultaneous
41 determination of formaldehyde is to collect the carbonyls on solid sorbents coated with a suitable
42 derivatization agent (e.g., 2,4-dinitrophenylhydrazine (DNPH)), followed by solvent desorption and
43 liquid injection for analytical analysis (e.g., high-pressure liquid chromatography (HPLC)) [7,8].

44 Many VOCs and formaldehyde are ubiquitous in indoor environment in view of the presence of
45 typical indoor emission sources [2,9,10]. Indoor VOCs are produced from a variety of sources,
46 including the utilization of consumer household products, emissions from adhesives and building
47 materials, and combustion processes [10-12]. VOCs are easily absorbed by skin and mucous
48 membranes, causing consequences of damage to human organs and metabolic systems. Few of VOCs
49 are also linked with sick building syndrome (SBS) [13,14]. Formaldehyde is one of the representative
50 oxygenated-VOCs. More than 65% of global formaldehyde is used to synthesize resins such as urea-
51 formaldehyde (UF), phenol-formaldehyde (PF), and melamine-formaldehyde (MF) which are widely
52 used in construction materials, wood processing, furniture, textiles, carpeting, and chemical industries
53 [15]. In addition, it is strong persistent and thus can slowly release from the materials in an extensive
54 period [4]. Formaldehyde is classified as a human carcinogen and it has been given more attention
55 because of its adverse health effect [16]. Therefore, the removal of indoor VOCs and formaldehyde is
56 of widespread interest in view of avoiding the potential imposed adverse effects on human health.

57 Emission source control, ventilation, and air cleaning are the three important approaches to
58 improve indoor air quality [17]. Among these air pollution control strategies, air cleaning with
59 Advanced Oxidation Processes (AOPs) has been drawn more and more attention because of the restraint
60 in the production of secondary pollution. Photocatalysis, as a promising technology developed since
61 1972 [18], is defined as the process by which various environmental pollutants are degraded on the
62 surface of a semiconductor photocatalyst when exposed to sufficiently energetic irradiation source, and
63 is an important group of AOPs [19]. The merit of photocatalysis is that it can be operated at room
64 temperature and is capable of degrading many organics under light irradiation. In the past two decades,
65 a lot of studies have been conducted for the photocatalytic oxidation of VOCs and formaldehyde which

66 are beneficial to solve the indoor pollution issues [20]. TiO₂ has been the dominant photocatalyst
67 because of its superior photocatalytic oxidation ability, high photocorrosion resistance, and nontoxic
68 properties [21]. TiO₂ immobilized on different substrates can photocatalytically degrade indoor air
69 pollutants in a flow system under UV light irradiation [20,22]. However, TiO₂ can only be activated by
70 Ultraviolet (UV) light because of its large band gap (3.2 eV). UV light accounts for only 5% of solar
71 energy [23]. Although dye-sensitized and transition metal-doped or nonmetal-doped TiO₂ can extend
72 its optical absorption to visible light range, many researchers focus their efforts on the development of
73 novel non-TiO₂ catalysts with low band gaps [24-28]. This interest is due to the fact that stable and
74 efficient dyes are usually rare, whereas dopants can serve as recombination centers for the
75 photogenerated electrons and holes [21]. An alternative method is to combine photocatalysis with other
76 processes that enhances the degradation efficiency. For example, Tokumura and coworkers developed
77 the photo-Fenton reaction for the removal of VOCs which can efficiently prevent emission of any by-
78 products [29]. A compact scrubber and AOP process were combined to enhance the VOC oxidation
79 [30]. The combination of AOPs and gas absorption is able to transform chlorine into chloride ions
80 effectively at ambient temperature conditions [31].

81 A number of reviews about photocatalytic oxidation of VOCs and formaldehyde from different
82 aspects have been published in recent years. For example, Kabir et al. reviewed some representative
83 techniques for controlling the indoor VOCs [32]. Peral et al. discussed the basic phenomena like oxygen
84 and water vapor adsorption during gas-solid heterogeneous photocatalysis, and special interest was
85 taken in describing the different photo-reactor configurations [33]. Lim et al. reviewed the development
86 of photocatalytic materials and photoreactors which significantly affect the degradation efficiency of
87 various major air pollutants [19]. Zaleska et al. reviewed the air pollutants removal mechanisms, key
88 influencing factors on the reaction rate as well as photocatalysts preparation and immobilization
89 techniques [34]. The review conducted by Mo et al. concentrates on the preparation and coating of
90 various photocatalytic catalysts, different kinetic experiments and models, novel methods for measuring
91 kinetic parameters, reaction pathways, intermediates generated, and an overview of various
92 photocatalytic reactors and their models described in the literature [20]. Wang et al reviewed the current
93 exposure level of VOCs in various indoor environment and state of art technology for photocatalytic
94 oxidation of VOCs from indoor air [35]. Zhong and Haghghat carried out a critical review with aims
95 to examine the state-of-the-art of photocatalysis technologies in the field of air purification and their
96 application prospects [36]. Most recently, Hay et al. reviewed the viability of photocatalysis for air
97 purification, especially the catalyst lifetime and intermediates formation [37].

98 In this review, we aim to summarize and review the current progress of photocatalytic removal
99 of VOCs and formaldehyde in indoor environment. Firstly, emission sources of indoor VOCs and
100 formaldehyde and the traditional indoor air pollution control strategies are discussed. Secondly,
101 influencing factors such as temperature, relative humidity, deactivation and reactivations of the
102 photocatalyst are discussed and special interests are paid for the production of intermediates. Further
103 applications of the photocatalytic technique to improve the indoor air quality are suggested.

104 **2. VOCs and Formaldehyde in Indoor Environment**

105 2.1 Sources of VOCs and formaldehyde indoors

106 VOCs is defined as organic compounds with the boiling point in the range of 50-260 °C at room
 107 temperature and atmospheric pressure [38]. This group is composed by a large amount of low molecular
 108 weight (MW) pollutants (such as aromatic-, fatty-, halogenated-, and oxygenated-hydrocarbon, terpene,
 109 aldehydes, ketones, and esters). Table 1 lists the typical VOCs presented in indoor air and their potential
 110 sources [17]. Formaldehyde is colorless, flammable and highly reactive at room temperature.

111 **Table1. Potential sources of indoor VOCs and formaldehyde**

VOCs and formaldehyde	Possible Sources
<u>VOCs</u>	
benzene	furniture, wood-based materials, smoking
toluene	pesticide, flooring materials, insulating materials, wood-based materials, paints, adhesives, gasoline, combustion sources
acetaldehyde	wood-based materials, flooring materials, HVAC system
paradichlorobenzene	ceiling materials, wood-based materials, pesticide
ethylbenzene	furniture, paints, adhesives, gasoline, combustion sources
methylene chloride	flooring materials, furniture, HVAC system, coating and painting
chloroethylene	flooring materials, coating and painting, dry-cleaned clothes
carbon tetrachloride	coating and painting, industrial strength cleaners
chloroform	pesticide, glue
naphthalene	insulating materials, mixed materials, wall painting
Other VOCs (e.g., esters and ketones)	plastics, resins, plasticizers, solvents usage, flavors, perfumes, paints, disinfectant, adhesives
<u>formaldehyde</u>	pesticide, flooring materials, insulating materials, wood-based materials, machine, coating and painting

112
 113 The concentrations of common VOCs in a given indoor environment strongly related to the
 114 existences of emission sources and efficiencies of ventilations. In some cases, indoor VOCs levels are
 115 extremely high owing to low air exchange rates (AER) and poor ventilations [39]. For formaldehyde,
 116 the atmospheric background mixing ratio is generally in ppbv to sub-ppbv level, which is much lower
 117 than that indoors (e.g., ppmv level) such as workspaces and residential units [40]. VOCs and
 118 formaldehyde can be generated from indoor sources and can also penetrate from outdoors via air
 119 exchange.

120 *2.1.1 Indoor sources*

121 Building and decoration materials are the direct emission sources for many common VOCs. In

122 addition, the additives in solvent paint, wood preservative, plywood can release different degrees of
123 VOCs at room temperature. Flooring can emit volatile aromatic such as toluene, benzene, and xylene
124 [41]. Acetaldehyde is used as preservatives and food seasoning for fish products, which can be released
125 from aniline, cosmetics, and plastic products as well. Newspapers, magazines, and prints that people
126 regularly expose to are the source of C₈ aromatic [42]. Furthermore, dry-cleaned clothes, chlorinated
127 water, industrial-strength cleaners and room deodorants are the main source of chlorinated
128 hydrocarbons. Environmental tobacco smoke is an important source for indoor VOCs quantified a total
129 of 78 low MW chemical species, including aromatics, polycyclic aromatic hydrocarbons (PAHs),
130 carbonyls, and quinones in the cigarette gas [43]. Human metabolism is also a source of indoor VOCs.
131 Acetone, acetaldehyde, methanol and other aldehydes were detectable in the respiratory airs [44].

132 Formaldehyde is a good solvent with strong adhesive properties, thus is used to strengthen the
133 plate hardness. In addition, its insect-resistance and anticorrosive ability allow it to be applied in
134 production of urea formaldehyde (UF) resins, paint and other materials. Primary non-industrial indoor
135 sources of formaldehyde include decorative building materials and furniture bonded with UF resins, UF
136 acid-cured finishes, and UF foam insulation (UFFI) such as wood-based materials, flooring and coatings
137 [12,45]. The interior decorations of furniture and building materials (e.g., floor glue, plywood, emulsion
138 paint, synthetic fiber, and adhesives) can emit a large quantity of formaldehyde. The emission from UF-
139 bonded materials has universality, potentiality and durability [46]. The volatiles mostly locate in the
140 deep of the plank rather than on the surface, resulting in slow, continuous, and uninterrupted physically
141 releasing. However, such potential would decrease over time.

142 Heat treatment and combustion are also important sources of indoor formaldehyde. Traditional
143 fuels such as biomass, coal, kerosene and liquid petroleum are used as an energy source for in-house
144 warming, especially in most developing countries [47,48]. The heating has no doubt to emit a certain
145 amount of formaldehyde and other air pollutants that elevate the toxic levels and create a polluted indoor
146 environment. Residential cooking is considered as an anthropogenic source of indoor formaldehyde
147 [49-51]. Daily necessities and customer products such as cosmetics, cleaning detergents, pesticides,
148 chemical fiber textiles, books, and printing ink can release airborne formaldehyde.

149 *2.1.2 Outdoor sources*

150 Outdoor VOCs can be originated from anthropogenic or natural sources [52-56]. Incomplete
151 combustion processes can generate volatile dissipative of any substances with low boiling point.
152 Automobile exhaust, industrial discharges, and fuel combustion products contain many VOCs
153 represented by alkanes, olefins, aromatic hydrocarbons. The pollutants from oil-fueled automotive
154 include trace amount of rubber matrix, which consist of high numbers of alkanes and alkyl benzene.
155 For the natural sources, biological VOCs (BVOCs) can be formed from secondary metabolic reactions
156 of vegetation [57,58].

157 Formaldehyde is an intermediate of atmospheric photochemical oxidation and emission product
158 from fossil fuel combustion. The primary sources of formaldehyde include both anthropogenic and
159 natural sources as well. Natural formaldehyde can release from solid wood, forest fire and excretion of
160 animals; however, their contributions to the atmospheric level are relatively small [59]. Anthropogenic

161 emissions include motor vehicles, chemical plants, industries, coal processing, artificial biomass
162 combustion, and food barbecue. Among those, vehicle exhaust (VE) is the most critical pollution in
163 urban areas. Even though alternative fuels and additives (i.e., green energies) and more advanced
164 emission control technology have been discovered to reduce pollutant generation, the raise in amount
165 of oxygenated VOCs from VE is still found with an increases of number of vehicles [60,61].
166 Formaldehyde can be formed secondarily from oxidation of many VOCs. Alkanes, alkenes and
167 aromatics (e.g., benzene and toluene) are precursors for the photochemical processes [59] which react
168 with atmospheric ozone (O₃), NO_x, hydroxyl radical (•OH) resulting in the formation of photochemical
169 smog and production of formaldehyde or other reactive compounds.

170 **3. Traditional Removal Approaches**

171 The traditional technologies for VOCs removal include adsorption, membrane separation, liquid
172 absorption, and catalytic combustion [62]. Many of these techniques have been widely applied in
173 industries or commercial sectors, but few are being further developed or optimized [24,63-65]. Table 2
174 summarizes details of current control techniques for VOCs removal. Newly-developed technologies
175 have been demonstrated their removal efficiencies in particular testing airs or controlled environmental
176 chambers. However, many are still limited on theoretical researches without practical applications. In
177 addition, single-based removal system may not offer satisfactory purification results due to the
178 complexity of VOCs and variations on their characteristics in real world. Combinations of the
179 technologies are thus required to achieve the final goal, but both high costs and harsh conditions are
180 limitations for their practical applications. It is a need to develop more economic, effective and
181 environmental-friendly treatment methods.

Table 2. Summary on current control techniques for VOCs removal

Techniques	Principle	By-product	Advantage	Disadvantage	
Botanical purification	Air is passed through a planted soil or directly on the plants. The contaminants are then degraded by microorganisms and/or plants, the precise	CO ₂ , organic and amino acids	low cost, no secondary pollution, beautifying the indoor environment	The purification effect is bad for high concentration pollutants	[28,66]
catalytic combustion	Combustion of VOCs at low temperature with the help of a catalyst.	CO ₂ , H ₂ O	Wide range of application coverage, high efficiency, no secondary pollution	Not suitable for gas containing dust particles and droplets	[63,67]
Bio-filtration	Bio-filtration is a process in which contaminated airs passed through a biological stuffing medium that supports many kinds microorganism that	Biomass	Little or no energy needs to be added in the form of heat or radiation to support this process	The equipment is big, long residence time, easy to jam	[68,69]
Absorption	Absorption is used to remove VOCs from gas streams by contacting the contaminated air with a liquid solvent.	wastewater	Product recovery can offset annual operating costs	High demands on absorbent, complex process, high cost	[24]
Zeolite based adsorption	Air pollutants are adsorbed onto zeolites, often as filtration post-treatment	Spent zeolite and collected organics	Effective in more than 90% RH as the adsorbent might be too specific	pollutant reemission	[64]
Activated carbon based adsorption	VOCs are removed from the inlet air by physical adsorption onto the surface of the carbon	Spent carbon and collected organics	Recovery of compounds, which may offset annual operating costs	they are flammable, difficult to regenerate for high boiling solvents, promote polymerization or oxidation of some solvents to toxic or insoluble compounds,	[70]
Membrane Separation	Pollutants are passed through a membrane into another fluid by affinity separation.	Exhausted membrane	No further treatment, simple process, small energy consumption, no secondary pollution	The stability of the membrane was poor	[65]

183

184 Adsorption is the most traditional method for removal of airborne formaldehyde. Activated carbon,
185 molecular sieve and silica gel are porous materials serving a large surface area media for physical and
186 chemical adsorption. The common absorbents contain inorganic salts (e.g., ammonium and sulfurous)
187 and are composed with amine groups such as urea and its derivatives, hydrazine, and amino-containing
188 polymers [71-73]. Physical adsorption represents formaldehyde is trapped onto the materials such as
189 zeolite, activated carbon, activated alumina and molecular sieve and porous clay ore without changing
190 its original form. [Chemical absorption works with high water solubility of formaldehyde, which is then](#)
191 [reduced or decomposed by any oxidizing or completing agents in the collection solutions \[16\].](#)
192 Persistence and stability are two concerns for the absorber of aldehyde material (ACM) (Shimizu [74]).
193 The absorbed gases should be re-released subject to any change of indoor conditions such as
194 temperature and RH.

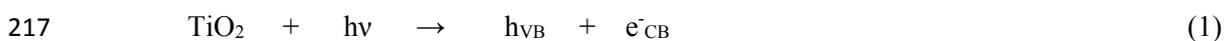
195 Catalytic oxidation technology with thermal treatment is another effective method for the VOCs
196 and formaldehyde removal. Formaldehyde reacts with oxygen (O₂) over the noble metals that produces
197 CO₂ and H₂O vapor [40]. The cost on energy consumption is a critical concern as this has to be operated
198 at high operation temperatures. For the plasma catalytic method, the molecules, particles, atoms and
199 free radicals are excited to have high chemical activities for the decomposition of VOCs, but the
200 reactions are difficult to be controlled in normal conditions and the reaction rates are usually slow [75].
201 Microbial degradation has been demonstrated its feasibility in removal of formaldehyde in both
202 wastewater and exhaust gas from industries and laboratories. Currently this technique has not been
203 widely applied for the indoor air cleaning. The composite of biological enzyme/activated carbon fiber
204 were synthesized and loaded on an AC surface [76]. Acidity is the most important factor in selecting
205 proper biological enzyme for the degradation. The experimental results showed that the removal rate of
206 formaldehyde reached 80% when the loading time was 8 h.

207 **4. Removal of indoor VOCs and formaldehyde via photocatalytic oxidation**

208 *4.1 Removal of VOCs by photocatalytic oxidation*

209 Photocatalytic oxidation (PCO) has been attracted more attention because of its unique
210 characteristics on the removal of chemicals. In recent years, PCO is perceived as a technology to remove
211 indoor VOCs. Titanium dioxide (TiO₂) is known as the most extensive studied photocatalyst due to its
212 excellent stability, high photo-activity, and suitable band gap structure. Low cost and non-toxicity are
213 also the main advantages for its application.

214 [The basic mechanism of photocatalytic degradation is that organic would be oxidized to H₂O, CO₂](#)
215 [or any inorganic harmless substances with •OH or superoxide \(•O₂⁻\) radicals, which are generated on](#)
216 [the surface of photocatalyst \(e.g., TiO₂\) under ultra-violet \(UV\) light irradiation \[77\]:](#)



220 In the heterogeneous reaction system, TiO₂ is excited by the absorption of a photon with energy
 221 greater than or equivalent to the band gap energy of the semiconductor, resulting in the electron
 222 transition from the valence band to the conduction band. The radiation could consequently produce
 223 electrons and holes (e⁻/h⁺) in conduction band and valence band, respectively. Following by the
 224 irradiation, the electrons and holes can undergo redox reaction with the adsorbed reactants on the
 225 photocatalyst's surface that leads to the formation of intermediates and products. The reaction series are
 226 co-called complete mineralization. Besides VOCs degradation, the reactions can be used as a function
 227 of disinfection and sterilization [78,79].

228 PCO of VOCs consists of a chain of stepwise reactions; that is, they take more than one elementary
 229 step to complete. Figure 1 shows a series of PCO reaction mechanism for *o*-xylene. Besides the final
 230 oxidized products, the steps also yield different oxidation states of intermediates such as aldehydes,
 231 ketones or organic acids [80]. These compounds can be qualified by real-time or offline monitoring and
 232 analytical methods such as gas chromatography/flame ionization detection (GC/FID), GC/mass
 233 spectrometry (GC/MS), high pressure liquid chromatography (HPLC), and Fourier-transform infrared
 234 spectroscopy (FTIR) [81,82]. Table 3 lists the intermediates formed in the PCO of VOCs (e.g., benzene,
 235 toluene and xylene) shown in the literature. For instance, the highly stable aromatic ring of toluene is
 236 usually intact while its active methyl group can be oxidized step-by-step to benzoic acid. The formation
 237 of the carbonyl group even causes the benzyl ring more inert because the conjugation effect reduces its
 238 electron density. The complete oxidation products such as CO₂ and H₂O would be generated from any
 239 of the intermediates until the benzyl ring is broken. However, if POC are conducted at room temperature,
 240 the active sites on the photocatalyst's surface could be gradually occupied by irreversibly chemisorbed
 241 intermediates, which retard the reactions. [For example, during the photocatalytic oxidation processes
 242 for toluene over TiO₂ catalysts, it was found that the toluene photooxidation behavior was strongly
 243 affected by the formation and oxidation behavior of intermediate compounds\[83\]. The study carried out
 244 by Nakajima et al. showed that H₂SO₄ treatment of TiO₂ surface provides higher photocatalytic removal
 245 efficiency on toluene which can be ascribed to the fast decomposition of intermediates by surface strong
 246 acid itself \[84\]. Moreover, the progresses of the research carried out into TiO₂-based photocatalysts
 247 were summarized by several recent reviews \[21,85\].](#)

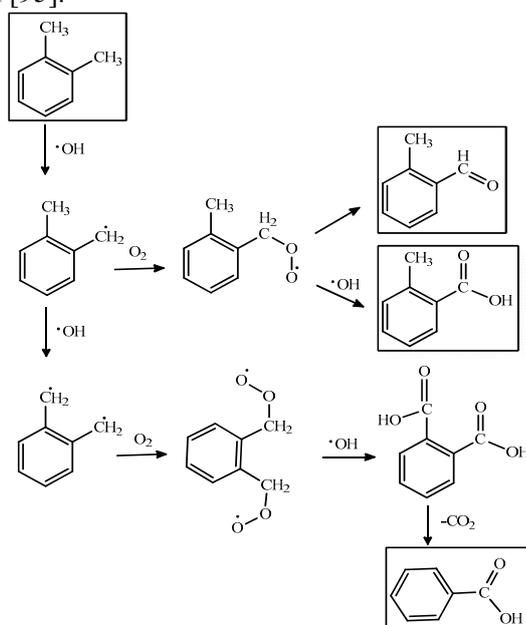
248 **Table 3. Summary on the intermediates formed in photocatalytic oxidation of typical**
 249 **indoor VOCs**

Target VOC	Concentration (ppm)	Light source	Main intermediates	Chemical formula	Analytical method	Reference
Benzene	3000-6000	4000W Xe lamp	Benzaldehyde, benzoic acid	-	GC/MS	[86]
	614	White fluorescent lamp	Phenol	Hydroquinone, 1,4-benzoquinone	GC/MS	[87]
	-	-	Phenol, hydro-	Malonic acid,	GC/MS/FTIR	[88]

			quinone, benzoic acid	benzoquinone		
Toluene	10	Black light lamp	Benzaldehyde, benzoic acid	benzyl alcohol	FTIR	[89]
	50-800	365nm UV	Acetone, acetaldehyde, formaldehyde	Acrolein, butanone	TDS-GC/MS/FID, HPLC/UV/FTIR	[90]
	370	>400nm	Benzaldehyde, benzoic acid	-	DRIFTS	[91]
Xylene	3000-6000	4000W Xe lamp	Benzaldehyde, Methyl-benzaldehydes	2,5-Furandione, 1,3-isobenzofurandione	GC/MS	[86]
	25-75	UV	o-Tolualdehyde, o-toluic acid, benzoate ion	-	FTIR	[92]

250

251 Anatase and rutile, two crystalline phases of TiO₂, have been shown their feasibilities on PCO of
 252 indoor air pollutants under UV light irradiation. The band-gap energy of anatase and rutile are 3.23 and
 253 3.02 eV, respectively. Anatase has shown better performance in PCO processes than that of rutile
 254 because of its more favorable conduction band configuration and stable surface peroxide groups. In
 255 general, TiO₂ is fixed on some substrate, such as hollow tubes, silica gel, beads, and woven fabric.
 256 These catalysts can be obtained using the methods such as electrochemical [93], plasma deposited [94],
 257 dip coating and sol-gel method [95].



258

259

Figure 1. The PCO reaction mechanism for *o*-xylene.

260

Table 4 summarizes potential photocatalysts used for removal of indoor VOCs. Different single or

261 combined photocatalysts have particular removal rates and efficiencies in PCO. Most TiO₂-based
262 catalysts have optimized performance on near-UV light region because of its large energy band gap
263 between electron-hole pairs of ~3.2 eV. A light source at a wavelength (λ) of <387 nm is required to
264 triumph over the gap, representing that the PCO can uptake *ca.* 3% from the sunlight only [96].
265 Therefore, a limited number of TiO₂ catalysts can exhibit high degradation activity under a visible light.
266 A lot of works have been thus done on the improvement of TiO₂ photocatalytic efficiency, such as
267 doping with nonmetals and metals and coupling with other supports. TiO₂ doping with a nonmetal atom
268 can enhance the photo-response in a practical application [97]. The nonmetal can substitute the oxygen
269 on TiO₂ lattice and lead to a band gap narrowing, resulting in activation at far-visible light region. The
270 common photocatalysts are primarily metal oxides, which can be doped with elements such as carbon
271 (C), nitrogen (N) or transition metal ions. For instance, the nitrogen-doped catalysts can be activated
272 more efficiently because of higher energy level of the valence band of N2p than O2p. The fluorescence-
273 assisted TiO_{2-x}N_y can decompose pollutants such as acetaldehyde through gaseous phase photocatalytic
274 reaction [98]. CaAl₂O₄: (Eu, Nd)/TiO_{2-x}N_y composite is able to store and release energies to
275 continuously inspire the visible-light responsive to TiO_{2-x}N_y even in the darkness. Such property allows
276 the fluorescence-assisted photocatalysts to function at night without supply of extra light sources.
277

Table 4. Summary on potential photocatalysts applied for indoor VOCs removal.

Photocatalyst	Preparation/coating method	Configuration	Compounds	Light source	$\eta_{\text{removal}}(\%)$	Reference
TiO ₂	Sol-gel	F	Acetone, toluene <i>p</i> -xylene	UV lamp, 254nm	77-62 (3 L/min)	[95]
TiO ₂	Electrochemical	F	Acetaldehyde	UV	99+ (110 min)	[93]
TiO ₂	Sol-gel	F	Toluene	Black light	52 (3.6 L/min)	[89]
TiO ₂	Plasma deposited	F	<i>m</i> -xylene	UV lamp	99+ (30 min)	[94]
TiO _{2-x} N _x	Calcination	P	Toluene	Visible light	99+ (3000 min)	[82]
TiO _{2-x} N _x	Hydrothermal	P	Acetaldehyde	Fluorescence	-	[98]
C-TiO ₂	Hydrothermal	P	Toluene	Visible light	60+ (120 min)	[99]
C-TiO ₂	Hydrothermal	P	Toluene	Visible light	20 (120 min)	[100]
CNT-TiO ₂	Hydrothermal	P	Styrene	UV-LED, 365nm	50 (20 mL/min)	[101]
Pt/TiO ₂	Photo-deposition	P	Benzene	Black light, 300-420	100 (100 mL/min)	[102]
Ln ³⁺ -TiO ₂	Sol-gel	P	Benzene, toluene, ethylbenzene, <i>o</i> -xylene	UV, 365nm	22-79	[103]
Ce-TiO ₂	Sol-gel	F	Toluene	Visible light	90	[104]
Fe-TiO ₂	Sol-gel	P	<i>p</i> -xylene	Visible light- LED	22 (5 min)	[105]
Fe-TiO ₂	Sol-gel	P	Toluene	Visible light	99+ (120 min)	[91]
In(OH) ₃	Ultrasound radiation	P	Acetone, Benzene, Toluene	UV lamp, 254nm	99+ (5 h)	[106]
β -Ga ₂ O ₃	Chemical deposition	P	Benzene	UV-lamp, 254nm	60 (20 mL/min)	[107]
Ag ₄ V ₂ O ₇ /Ag ₃ VO ₄	Hydrothermal	P	Benzene	White fluorescent lamp	99+ (120 min)	[87]
Pt/WO ₃	Photo-deposition	P	DCA, 4-CP, TMA	Visible light, >420 nm	99+ (3 h)	[108]
Pd/WO ₃	Calcination	P	Acetaldehyde, toluene	Fluorescent/visible light	99+ (3 h)	[26]

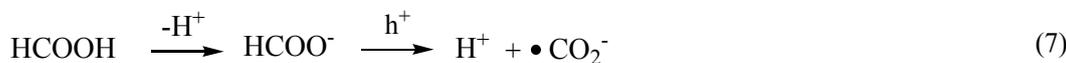
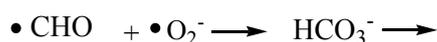
Remarks: DCA: dichloroacetate; 4-CP: 4-chlorophenol; TMA: tetramethylammonium; P: powder; F: film

255 TiO₂-Pt/TiO₂ hybrid catalyst system serves a complete oxidation of benzene to CO₂ at ambient
 256 temperature [102]. TiO₂ after doped with Pt has an increase number of active sites, which convert
 257 intermediate form of carbon monoxide (CO) to be CO₂. Pt/TiO₂ is thus the most useful catalyst for the
 258 purification of VE gases containing benzene. Doping with lanthanide ions can promote the formation
 259 of oxygen vacancies which have relatively high liquidity comparing with other oxygen species [109].
 260 In particular, cerium (Ce) is a low cost photocatalyst that has the ability to migrate between Ce⁴⁺ and
 261 Ce³⁺ through oxidization and reduction reactions. Ce doped with TiO₂ can decompose toluene under a
 262 visible light source.

263 Rather than TiO₂-based photocatalysts, other semiconductor can be also applied in the VOCs
 264 removal such as ZnO [110], ZnS [111], SnO₂[112], In(OH)₃ [106], and β-Ga₂O₃ [107]. Nano-sized
 265 porous In(OH)₃ and porous Ga₂O₃ have high activity and long-term durability for photocatalytic
 266 decomposition of acetone, benzene, toluene and other aromatic derivatives under ambient conditions.

267 4.2 Removal of Formaldehyde by photocatalytic oxidation

268 Similar to the PCO for VOCs, formaldehyde priorly reacts with •OH, which are generated on the
 269 excited photocatalyst's surface. They would form an intermediate of HCOOH which eventually is
 270 oxidized to CO₂ and H₂O vapor. The reaction mechanism is as following [113]:



280 TiO₂ and TiO₂-based (i.e., metal-doped, nonmetal-doped and composites), other metal oxides (e.g.,
 281 MnO_x, Bi₂O₃, ZnO, PdO, and composites), and new-type photocatalysts are widely used for PCO of
 282 formaldehyde. Table 5 shows a summary of the common photocatalysts and their applications and
 283 efficiency in the formaldehyde decomposition.

284

285

Table 5. Summary on the PCO used for formaldehyde degradation.

Catalyst	Preparation method	HCHO Concentration	Light source	Conversion efficiency	References
Mesoporous TiO ₂	Evaporation-induced self-assembly	30ppm	UV light	95.8%	[114]
Amorphous TiO ₂ film	CVD method	50-55ppm	UV light	80%	[115]
PEG modified TiO ₂ film	Sol-gel method	20ppm	UV light	95%	[116]
TiO ₂ coating on polyester fiber	Spray coating	24.6±2.8ppm	UV light	90%	[117]
UV/ TiO ₂ /O ₃	Sol-gel	18ppm	UV light	79.4%	[118]
Ag/TiO ₂	Incipient wet impregnation	500ppm	UV light	Above 95%	[119]
Pt@TiO ₂	Reverse micelle sol-gel	10ppm	Visible light	98.3%	[120]
Ce/TiO ₂	Sol-gel	1ppb	UV light	Above 70%	[121]
Pd-TiO ₂ film	Sol-gel dip coating	500ppb	UV light	Above 95%	[122]
Acrylic-silicon/nano-TiO ₂	Emulsion blend	0.8ppm	Vis light	83.4%	[123]
N-doped TiO ₂ film	Precipitation-peptization	0.24ppm	Vis light	90%	[124]
AC loading TiO ₂	Microwave-assisted synthetic	30ppm	UV light	58.68%	[22]
Pt@SnO ₂	Sol-gel method	—	Vis light	93.2%	[125]
α-Bi ₂ O ₃	Calcination of hydrothermally prepared (BiO) ₂ CO ₃	100ppm	Vis light	62.5%	[25]
Nano-ZnO	Mixing-calcination	2.5-25ppb	UV light	73%	[126]
Zr _{0.08} Ti _{0.92} O ₂	Sol-gel method	0.08ppb	UV-vis light	92%	[127]
Zn ₂ SnO ₄	Hydrothermal method	2ppm	UV-vis light	70%	[128]

279 4.3 Influence factors

280 Photocatalytic reaction rate, additional with the reaction kinetic and adsorption coefficients, are
 281 direct tools to evaluate the efficiency of a photocatalyst in removal of VOCs. Table 6 shows kinetic
 282 parameters and PCO conversion efficiency for the common VOCs. There are critical factors such as
 283 light source and intensity, pollutant concentration, RH, temperature, and deactivation and reactivation
 284 can control the photocatalytic reaction rate. In order to study the PCO processes, many kinetic

285 experiments for removal of common pollutants (e.g., benzene, toluene, xylene, and formaldehyde) have
 286 been thus conducted in optimal reactors. Here we summarize and review these factors.

287 **Table 6. Kinetic parameters and PCO conversion efficiency (%) for the common VOCs**
 288

Pollutants	Reactor design		Initial reaction conditions				Deactivation	Reference	
			[VOC]gas	PW(nm)/I	RH	T			
	RT	Photocatalyst	(ppm)	(mW·cm ⁻²)	(%)	(°C)			
Styrene	CR	CNT-TiO ₂	25±1.5	365/70	-	-	Y	[101]	
Benzene	CR	Pt/TiO ₂	80	300-420/-	65	Ambient	n.r.	[102]	
	CR	In(OH) ₃	920	245/-	-	25	n.r.	[106]	
Acetone	CR	In(OH) ₃	420	245/-	-	30±1	n.r.	[106]	
Toluene	CR	TiO ₂	10	>300/0.7	0-40	Ambient	Y		
	CR	TiO ₂	17-35	365/2.34	47	25	n.r.	[106]	
	CR	P25	50-800	365/10±1	0-50	25	n.r.	[90,106]	
	CR	Ce-TiO ₂	0.15-0.6	Visible/-	<3-75	42	n.r.	[104]	
	CR	Fe-TiO ₂	370	>400/-	60	25	Y+N	[91]	
	CR	Ln ³⁺ -TiO ₂	23±2	365/0.75	-	-	n.r.	[103]	
	CR	In(OH) ₃	1220	245/-	-	25	n.r.	[106]	
	CR	TiO ₂ fibers	200	365/9	20-60	-	n.r.	[129]	
	Xylene	CR	P25	25-75	UV/1.5	30-90	-	Y	[92]

289 **Remarks:**

290 CR: continuous reactor; BR: batch reactor.

291 [VOC] gas= VOC gas-phase concentration; *I* = light intensity; RH= relative humidity; *T* = temperature.

292 Y: catalyst deactivation observed; N: catalyst deactivation not observed; Y+N: catalyst partial deactivation and
 293 can be regenerated completely; n.r.: reference includes no data on catalyst deactivation,

294 -: reference includes no data on light intensity.

295 *Light source and intensity.* The electron-hole pairs of a photocatalyst must be firstly excited for
 296 the following VOCs degradation. The common catalysts (e.g., TiO₂) usually require an UV wavelength
 297 equivalent energy source for the excitation. Medium pressure mercury lamp, Xenon lamp, and UV light
 298 are common light sources for PCO. The light intensity is usually represented by units of light-irradiation
 299 (energy per unit area) or photon flux on the catalyst's surface. Theoretically, the reaction rate of PCO
 300 is proportional with the light intensity supply. The reaction rate of PCO is regulated by the first order
 301 of consumption rate of electron hole pairs and a half order of their recombination rate [130]. Thus there
 302 is no doubt that the light intensity can directly control the first-order of reaction [95]. In addition, the
 303 internal structures of photocatalysts can affect the adsorption rate of the photons and consequently
 304 impact on the conversion rate [131]. Bahnemann and Okamoto [132] investigated the relationships
 305 between UV light intensity and photocatalytic reaction rate with TiO₂. A linear correlation was found
 306 in the low intensity range whereas the degradation rate is proportional to square root of the light intensity

307 under the moderate intensities. When light intensity is greater than 6×10^{-5} Einstein $L^{-1} S^{-1}$, the
308 VOC degradation rate is not further enhanced subject to any changes.

309 As UV light is harmful to human and potentially leads to produce secondary pollutants
310 (e.g., more strong oxidizing substances) in indoor air, more attentions are drawn to applying
311 visible light stimulating catalytic reaction for the removal of VOCs. However, the influences
312 of light intensity are seldom studied with visible light sources. The formaldehyde removal rates
313 with N-doped TiO_2 photocatalyst were enhanced linearly from 25.5% to 59.6%, and stabilized
314 thereafter, when the intensity increased to 30,000 lux with an initial concentration of 0.98
315 mg/m^3 [133].

316 *Pollutant concentration.* The concentration levels of pollutants can influence the
317 photocatalytic performance in terms of the reaction rate. In the PCO process, the mass flux
318 between the surface of photocatalyst and inlet can be accounted by the convective mass transfer
319 [134]:

$$320 \quad N_A = k_A \Delta C_A \quad (9)$$

321 Where N_A is mass flux, k_A is convective mass transfer coefficient and ΔC_A is the concentration
322 difference of transfer substance between the interface and the inlet. Eventually, the pollutant
323 concentration over the photocatalyst's surface varies from that in the inlet; however, it is
324 difficult to accurately monitor the surface concentration by any means of measurement
325 techniques. As a result, the use of inlet concentration for the computation of kinetic parameters
326 may contain different degrees of errors. In order to decrease the concentration disparities, it is
327 necessary to increase the airflow rate for improving the convective mass transfer [135].

328 Pollutant concentration (C) and photocatalytic reaction rate (r) are the two kinetic
329 parameters for reaction model computation. The Langmuir-Hinshelwood (L-H) model has been
330 widely applied to establish pertinence between C and r in the PCO process for many VOCs
331 such as acetone, benzene, toluene, and xylene [136,137]. *In general, the degradation rate
332 decreases while the pollutant concentration increases* [90,95]. However, only few investigations
333 on the photocatalytic kinetics for indoor VOCs are reported. Among those, most have
334 conducted the tests at an extremely high concentration (e.g., ppmv level). The demonstration
335 concentration for a VOC would even cause instant headache, irritation, and discomfort to
336 human [138]. The results could not reflect the realistic situations in most indoor environments
337 (i.e., pptv to sub-ppbv level). Ce-doped TiO_2 had a decrease in degradation efficiency while the
338 formaldehyde levels increased from 0.1 to 0.5 mg/m^3 [121]. In addition, in a concentration range
339 of 0.1-1.0 mg/m^3 , the degradation efficiency of formaldehyde was up to 80.8% with
340 photocatalyst from 3M company, but shapely reduced to 52.9% when the concentration raised
341 to 2.0 mg/m^3 [139].

342 *Relative Humidity.* Hydroxyl groups can be generated while water molecules adsorbed on
343 the photocatalyst during the PCO processes, which can be captured by photo-generated charge
344 carriers to produce reactive radicals (e.g., $\bullet OH$) to further oxidize the indoor organic pollutants.
345 Therefore, water vapor either from indoor air or generated from the mixed reactions plays a

346 significant role in the photo-degradation [102]. In the absence of water vapor, the photo-
347 degradation of VOCs (e.g., toluene) is seriously retarded since the mineralization could not be
348 completely occurred. At the initial stage of photocatalytic reaction, hydroxyl groups were
349 expended due to the reactions between water vapor and organics on the photocatalyst's surfaces.
350 However, the presence of water vapor would lead to electron-hole recombination [140]. There
351 is also an adsorption competition between water and organics when RH is excessive. The water
352 molecules can occupy the active sites of the photocatalyst surfaces for reduction of the VOCs
353 degradation rate and suppression of photocatalytic activity. A typical breakthrough curve was
354 obtained to demonstrate the competitive adsorption of water and toluene in the TiO₂
355 photocatalytic reactions [89]. The result indicated that the photocatalyst is more sensitive to RH
356 at a low hydrophobic condition. The indoor RH is usually regulated by ventilations (e.g., air-
357 conditioning) or humidifiers, thus the competitive adsorption between water and trace
358 contaminants has strong impact on the oxidation rates [135].

359 RH is also the key factor for formaldehyde degradation, which has been demonstrated with
360 the photocatalytic performances of Zr_xTi_{1-x}O₂ at different RHs of 50±5%, 65±5%, 85±5%,
361 respectively [127]. The work reported that the activity is the highest at RH of 50±5%,
362 representing that the photocatalytic reaction can be suppressed at humid environments. Similar
363 observations were found for TiO₂-C coated and TiO₂-CN coated photocatalysts at a RH range
364 of 20-90% [141]. The effect of RH on the degradation is negligible at a formaldehyde
365 concentration of 3.3±0.3 ppmv; while at a higher concentration level (8.6±0.5 ppmv), the
366 degradation efficiency significantly dropped at a RH of 90%. It is necessary to note that the
367 impacts of water vapor on the removal efficiency for VOCs and formaldehyde were
368 inconsistent for different photocatalysts. For this reason, an optimized working RH must be
369 investigated when different systems are applied.

370 *Deactivation and reactivation.* Lifetime of a photocatalyst is an important parameter for
371 the real application in removal of indoor pollutants. This should include the consideration of
372 deactivation, regeneration, reactivation, or replacement. The gas-solid photocatalytic activity
373 decreases with time while the number of effective active sites on the catalyst surface decreases
374 at the same time. Deactivation thus occurs due to the accumulation of such partially-oxidized
375 intermediates which occupied the active sites on the photocatalyst's surface. Many kinetic
376 studies indicate that the adsorption of poisonous intermediates during the initial stage of the
377 photocatalytic reactions is almost irreversible. The initial oxidation rate is proportional to the
378 effective surface area of catalyst. For instance, acetic acid and formic acid are the two main
379 detectable intermediates formed in the photocatalytic degradation of acetaldehyde by TiO₂.
380 Even though trace amounts of these intermediates could possibly discharge into the air, these
381 polar organic compounds have a stronger affinity to be accumulated on the photocatalyst's
382 surface until they can be decomposed by further steps of PCO. In some extent, a complete
383 deactivation of the photocatalyst occurred after 20 consecutive PCO reactions due to the fully
384 occupation of the active sites by the intermediates [92]. [Mendez-Roman investigated the](#)

385 relationship between the formation of surface species and catalyst deactivation during the
386 photocatalytic oxidation of toluene, and their results showed that the accumulation of benzoic
387 acid on the surface resulting in the catalyst deactivation[142]. Recovery of photocatalytic
388 activity requires a regeneration technique. The adsorbed polar intermediates such as
389 benzaldehyde and benzoic acid can be removed completely with a heat treatment at 653 K for
390 3 h [100]. However, such reactivation of the photocatalysts is a practically difficult since it
391 consumes high energy or requires work with a furnace.

392 *Other potential factors.* Rather than the above, the loading amount of noble metal, content
393 of the photocatalyst, and gas flow rate can also affect the photocatalytic activity. These multiple
394 parameters can either advance or suppress the PCO subject to the kind of photocatalysts applied
395 for the VOCs removal.

396 **5. Summary and Outlook**

397 VOCs are omnipresent but can greatly aggravate indoor air quality. Formaldehyde is
398 highly concerned due to its carcinogenicity and universality. There is a variety of indoor
399 pollution sources such as wood-based furniture and flooring material for both of VOCs and
400 formaldehyde. A long time exposure to indoor toxics can lead to health impacts such as SBS
401 and cancer.

402 Photocatalysis is considered as one of the most promising technologies for eliminating
403 VOCs due to its high efficiency and stability. However, traditional photocatalytic materials such
404 as TiO₂ can only response to UV irradiation, limiting the light utilization efficiency.
405 Development of new single or photocatalytic composite materials which can be irrigated with
406 conventional visible or solar light is thus a need. Currently most studies demonstrated their
407 VOCs removal efficiency in a high concentration level (e.g., ppmv). More on-site
408 demonstration should be conducted in order to prove the efficiency in removal of indoor VOCs
409 and formaldehyde in realistic environments (e.g., residential and workspaces).

410 Different oxidation states of intermediates can be produced in the PCO reaction
411 mechanism. These organics can temporarily or permanently occupy the active sites on the
412 photocatalyst's surface, leading to suppression or termination of the reaction kinetics. Efficient
413 removal of these intermediates is necessary as they are even more toxic than the parent VOCs
414 and harmful to health. It is especially critical if they can be discharged into the indoor airs in
415 practical application. More advanced approaches for re-activation and regeneration of
416 photocatalyst are also essential to extend its lifetime for serving a long-term VOCs degradation.

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