Photocatalytic reactors: design for effective air purification

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Abstract This report is intended to review existing photocatalytic reactors as used in research and commercial applications. The guiding principles of effective reactor design are discussed, stressing the challenges faced by design teams. An account of specific reactor designs is given, highlighting innovative work and novel concepts. A concerted effort has been made to draw information from a wide variety of international sources, including scientific literature and web based sources.

Keywords photocatalytic oxidation; air purification; TiO_2; reactor; immobilisation

1. Introduction

The indoor air environment is currently considered to be one of the most important health concerns for industrialized nations. The U.S. Environmental Protection Agency (EPA), in 1995 stated that "Indoor air pollution is now our nation's number one environmental health concern" [1]. In developed nations increasing energy costs, coupled with a drive for environmental sustainability, has resulted in homes and buildings being sealed ever tighter with better windows, insulation and moulding, to reduce energy consumption. This sealing has resulted in a dramatic reduction in natural ventilation, drafts and leaks, which would circulate indoor air and replenish it with air from outdoors. Air pollutants are therefore able to accumulate within buildings, and it has been estimated that indoor air pollution can be 2–5 times higher than outdoor air pollution.

Since an average person may spend up to 80% of their time indoors, a polluted environment could pose a significant health risk. The House of Commons Select Committee Enquiry on Indoor Air Pollution (1991) commented that "Overall there appears to be a worryingly large number of health problems which could be connected with indoor pollution and which affect very large numbers of the population" [2].

Methods for the reduction of indoor air pollution can be divided into three categories. In order of effectiveness these methods are [3]: (1) the removal of source or control of its emissions, (2) ventilation, (3) air purification. Though air purification is not capable of replacing methods (1) and (2) it is a valuable tool for tackling the problem.

Commonly air purifiers are installed in the ducts, which are part of central heating or air-conditioning systems in buildings, or as portable units that stand alone within a room. Systems currently in commercial use include: filtration, electrostatic precipitation, ozone generators, Ion generators, and germicidal UV. Photocatalysis is now becoming a commercially viable product and has seen a major rise in global interest over the last few decades.

2. Photocatalysis

With the discovery of photoinduced water cleavage on titanium dioxide (TiO_2) semiconductor electrodes by Fujishima and Honda in the early 1970's [4], it was soon realised that this phenomenon could be applied for environmental remediation.

Photocatalytic oxidation (PCO) first saw use as a technique for water purification, following from Frank and Bard's investigation into the decomposition of cyanide using an aqueous TiO_2 suspension in 1977. Aqueous suspensions of catalysts, such as TiO_2, were found to be effective at breaking down organic pollutants. However, due to the inherent inefficiency of the process (the need to filter out the TiO_2 after purification), techniques had to be developed to immobilise TiO_2 onto support surfaces. This has lead to a technology that lends itself to air purification.

2.1 The Photocatalytic reaction

Photocatalytic Oxidation (PCO) can be defined as a chemical reaction influenced or initiated by light that removes electrons from a catalyst and adds those electrons to a compound [5]. This definition highlights the main ingredients that make photocatalytic air purification possible: a light source, a catalyst, and reactants.

Crucially PCO requires the formation of an interface between, in general, a solid photocatalyst and a liquid or gas phase containing the reactants and/or products of the photoreaction.

The series of events following the illumination of a photocatalyst-gas interface may be initiated by either (A) light absorption by the catalyst, which leads to the activation of the reactant, or by (B) direct excitation of the reactant, which is then quenched by the catalyst. Mechanisms (A) and (B) may operate simultaneously at a semiconductor-gas interface, though (A) is generally considered to be the primary step for photocatalytic oxidation.

An important step of the photoreaction is the formation of electron-hole pairs. These are created when energy is provided to overcome the atomic band gap, between valence band (VB) and conduction band (CB). When a photon, with excitation energy ($h\upsilon$) greater than the band gap energy (E_{BG}), is absorbed an electron-hole pair is created [6].

$$photocatalyst + h\upsilon \rightarrow \mathrm{e}^- + h^+ \tag{1}$$

The charge is transferred between the electron-hole pairs and an adsorbed, ground state, reactant on the photocatalyst surface. Resulting in the photo-oxidation and photo-reduction of reactants, see Fig. 1.

To what extent electron-hole pairs play a part in the destruction of pollutants is still debated. They may be too short lived to be able to react directly, unless the concentration of the pollutant is high or is strongly adsorbed.

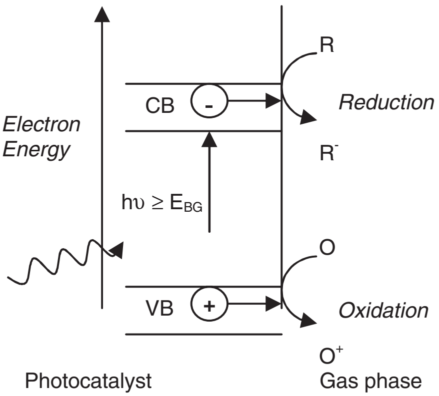

Figure 1. *Electron-hole pair generation.*

With the presence of water, as vapour form in air, the oxidizing agents known as reactive oxygen species (ROS) can be formed. These include oxygen (O_2), superoxide (O_2^-), peroxide (O_2^{-2}), and hydroxide (OH). These species can participate in a host of oxidation-reduction (redox) reactions, which are highly effective at the chemical destruction of VOC's, particulate matter, microbes, ozone, NO_x, and SO_x.

The creation of hydroxyl radicals from water and subsequent destruction of an organic compound are shown in equations 2 and 3. It is shown that, as with e-h pair oxidation, the final products of organic compound oxidation are water and carbon dioxide.

$$H_2O + h^+ \rightarrow {}^{\bullet}OH + H^+ \tag{2}$$

$$^{\bullet}OH + O_2 + C_nO_mH_{(2m-4n+5)} \rightarrow nCO_2 + (m-2n+3)H_2O \tag{3}$$

2.2 Photocatalysts

Semiconducting materials (photocatalysts) are key to the photocatalytic process. Many have been studied in either pure or doped form. The most common semiconductors researched for PCO applications have been: TiO_2, ZnO, CdS, with Fe(III) and precious metals being the most common dopants.

TiO_2 has proved to be the most suitable candidate, and is the most widely used. It is considered almost ideal for PCO applications. Firstly TiO_2 is relatively inexpensive. It is easy to produce, in large supply and is used throughout the world in a wide range of applications (e.g. as a colorant for paint, paper, and plastics, even food; and for UV protection). TiO_2 is highly stable chemically, so is unlikely to participate in unwanted reactions. Importantly, the photogenerated holes are highly oxidising (+2.53 V vs SHE), and the photogenerated electrons are reducing enough (–0.52 V vs SHE) to produce reactive oxygen species [7]. The down side to TiO_2 is that it cannot be activated by visible light.

TiO_2 has a large band gap, $E_{BG} \approx 3.2–3.0\,eV$. It is therefore limited to activation by radiation wavelengths equal to or below UV light. UV light makes up only $\sim 5\%$ of the solar spectrum.

Three are three crystalline forms of TiO_2: anatase, rutile, and brookite. The anatase form has been found to have the most favourable characteristics for PCO, as it appears to be the most active and easiest to produce of the three. Irradiation with light of 385 nm or less will generate electron-hole pairs in anatase TiO_2. The anatase form is predominantly used in most commercial PCO processes.

Commercially available Degussa P25 is a powder form of TiO_2 produced in Germany. It is a finely divided material, $50\,m^2g^{-1}$, with a 70 : 30 ratio of anatase to rutile [8]. This material is cheap, and research of this composition has shown that PCO activity is high [9]. As a result it is one of the most widely used forms of TiO_2 for both research and commercial applications.

3. Immobilisation of TiO_2

Many techniques have been devised to fix TiO_2 to a substrate, and research is ongoing. Essentially the coating needs to be mechanically robust, and must maintain a high level of photoactivity. The research drive is to create a technique that is simple, cheap, and capable of coating complex geometry's, so therefore innovative photoreactor designs, whilst maintaining the photoactivity of the semiconductor. Immobilisation techniques have been used successfully to fix TiO_2 to many supports, such as glasses, ceramics, fabrics, polymers, and metals.

Table 1 lists several common immobilisation techniques, as well as the support substrates, that have been investigated to date. Of these the Sol-gel technique appears to be the most widely used with glass plate being the most common form of substrate employed.

The sol-gel coating technique involves the transition of a system from a liquid sol (colloidal suspension of minute solid particles in a liquid) to a viscous gel in which the suspended particles are organized in a loose, but definite three-dimensional arrangement. The thin film gel is then dried (this can be repeated several times to achieve the required film thickness) and finally sintered. [10]

The sol-gel technique is popular because it is relatively simple. It can be used for a large range of substrate materials, and importantly, the formation of the thin films can be controlled in such a way to create a high proportion of anatase form TiO_2, so retaining a high level of photoactivity. Depending on the specific technique and the substrate material used the coating can maintain a good level of adhesion.

A great deal of research is still ongoing, aimed at obtaining immobilized films of TiO_2 that maintain a high level of photoactivity whilst being hardwearing and suitable for commercial application. In general it has been found that the better the film adheres to a substrate the worse is its photoactivity. However, viable commercial products have already been produced, and successful coating techniques are being employed and tested further by research and commercial activity centred on photoreactors.

Table 1. *Methods of immobilising TiO_2 and the support substrates used*

Immobilisation Technique	Substrate coated	Ref.
Dip Coating	Glass	[11]
	Quartz	[11]
	Pumice stone	[12]
	Optical fibres	[13]
Sol gel	Glass	[14]
	Red brick	[14]
	White cement	[14]
	Quartz	[9]
	Fibreglass cloth	[15]
	Polymer	[16]
	Optical Fibres	[17]
Chemical vapour deposition, CVD	Glass	[18] [19]
	Activated carbon	[20]
	γ-alumina	[20]
	Silica gel	[20]
Electrophoretic	Stainless steal	[21] [22] [11]
	Ti-4V-6Al Alloy	[21]
	Titanium Foil	[21]
	Tin oxide coated glass	[21]
	Aluminium	[23]
Sputtering	Glass	[14] [24]
	Red brick	[14]
	White cement	[14]
	Ceramic tiles	[14]

4. Photocatalytic reactors

The versatility of PCO technology, has seen many commercial applications such as in glazing [25], paving stones [26], wall paper [27], and paint [28] to name but a few, where the PCO effect is secondary to their main function. These products are typically activated solely by sunlight with photocatalytic air purification tending to be given less significance than the “self cleaning” aspect of these products. Non-the-less this marks a major shift from conventional air purifying systems. An air purifying capability can be incorporated into construction materials, surface finishes, even clothes.

Devices solely intended for purification purposes are still an important technology required to meet the need for clean air. Immobilized TiO_2 is being employed in place of conventional purifying units, or incorporated to form hybrid devices. These are typically not activated by sunlight but by UV lamps, so achieving greater efficiencies, and can be located in areas where natural light is minimal if not non-existent.

4.1 Principles of reactor design

In a photocatalytic reactor the major elements required for the PCO process are combined to form a unit, within which the pollutants are neutralized.

The aim of reactor design is to obtain the greatest reaction yield, i.e. neutralize as much pollutants as possible, whilst expending the least amount of energy. To achieve this the reactor should provide effective contact among catalyst, reactants, and photons.

For the catalyst to be effective it must have a high surface area, to allow contact with as large a volume of reactants as possible. Extremely large surface areas are possible when using powder form TiO_2. However, the need to immobilize TiO_2 on to a substrate results in a significant reduction of surface area.

Unless using sunlight, due to electricity charges and bulb replacement, the light source will tend to be the most costly component of any photoreactor. So, photons are expensive, it is essential to utilize them effectively and ensure that few are emitted that do not contact the catalyst and initiate oxidation. As well as this, efforts must be made to ensure that all reactor surfaces receive adequate irradiation from the light source, so that no flow paths through the reactor exist where the catalyst is not illuminated.

Effective design is therefore essential to maximize surface area when using immobilized TiO_2, and to ensure that it is properly irradiated. The method of immobilization also demands careful consideration to ensure an adequate coating on the substrate surface. Factors to consider include: thickness, coverage, and robustness, as well as simplicity, cost, and repeatability for commercial application.

4.2 Reactor types

Several existing reactor designs as used for research and some commercial applications are described below.

4.2.1 Flat plate reactors

The flat plate reactor in Fig. 2, designed and tested by Brandi et al. [29], is composed of two flat glass plates with a certain gap between them, through which the fluid to be cleaned is passed. The catalyst is coated on the interior surface of each plate, with an external light source irradiating the catalyst. The thickness of the catalyst layer is thin enough to allow the entire catalyst surface to be illuminated by the light. The light sources have a reflector behind them to help utilize all the available radiation by directing it onto the catalyst.

Flat plate reactors are the most basic reactor type offering low surface area, and generally, poor utilisation of available light.

4.2.2 Honeycomb monolith reactors

The honeycomb monolith configuration is the type commonly found in automobile exhaust systems for emission control. They contain a number of channels with typical internal dimensions of around 1 mm. A very thin layer of catalyst is coated onto the walls of the channels. The benefits of this type of design are that there is a low-pressure-drop and high surface area to volume ratio [31]. Raupp has investi-

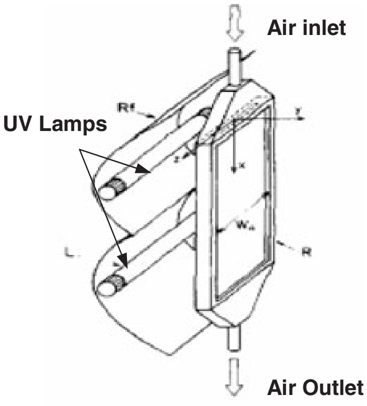

Figure 2. *Flat plate, Brandi et al. [29].*

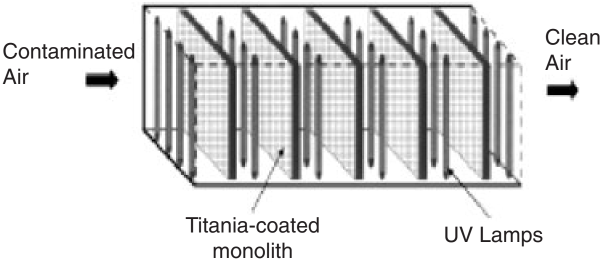

Figure 3. *Honeycomb monolith, Raupp et al. [30].*

gated reactors using various monolith formations; square channelled monoliths of various dimensions as well as porous cylindrical ceramic monoliths.

4.2.3 Fluidised bed reactors

The advantages of a fluidised bed reactor, as claimed by Dibble and Raupp, include a low pressure drop, high throughput, and very high catalyst surface area, thus efficient reactant-catalyst contact. In Fig. 4 the catalyst bed consists of silica gel impregnated with the catalyst, using a sol gel technique. The silica gel particle size is 250–450 nm in diameter, which resulted in the particles exhibiting smooth and smooth/bubbling fluidisation.

Fluidised bed reactors have seen further developments made by researchers such as Nam et al. who placed the light source at the centre of the catalyst bed, and six inlet nozzles at the base of the reactor to provide a uniform air distribution in the catalyst bed. Lim et al. combined features of a tubular and a fluidised bed reactor,

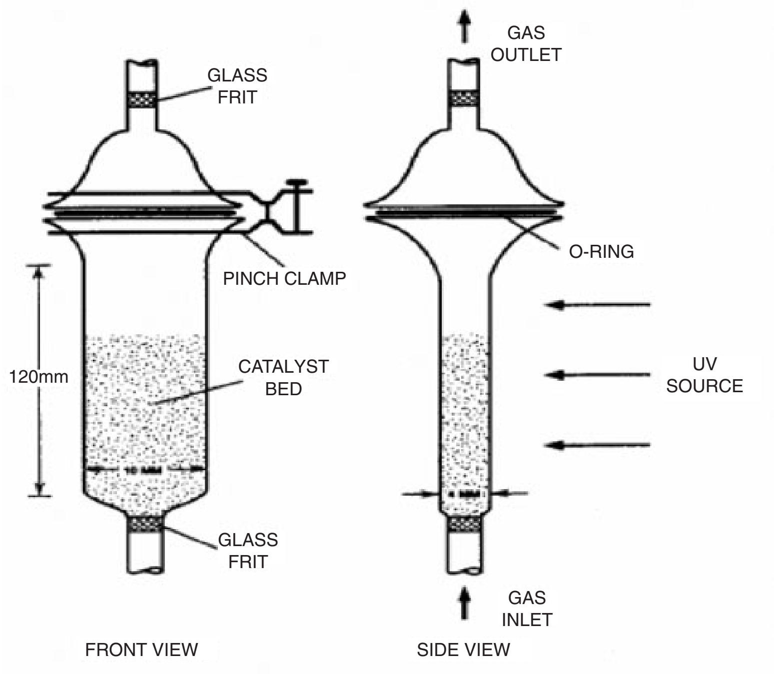

Figure 4. *Fluidised bed, Dibble et al. [32].*

forming and tubular bed with the light source in the centre. A quartz filter was used to distribute light evenly and a mirror box surrounded the reactor to minimize loss of light [31].

4.2.4 Fibre/Membrane Reactors

The reactor show in Fig. 5 was constructed and tested by Pichat et al. [33] it consists of an UV emitting lamp surrounded by a number of layers of matrix material (glass fibre) that has been coated in TiO_2. The total geometric surface area of the substrate material was calculated to be $1300\,cm^2$. The glass fibre is permeable to UV light, and if the TiO_2 layer is thin enough, effective catalyst/photon contact can be made. Other research groups working on this reactor type have been primarily concerned with water purification. They include Hidaka et al. [34] who used a reactor consisting of fibreglass cloth forming two concentric layers around a central light source. Molinari et al. [35] used a polymer membrane reactor; studying membranes of different polymer material, pore sizes and distribution, as well as thickness. Ohitani et al. [36] investigated the use of fibreglass cloth and also stainless steal mesh whose photoactivity was found to be comparable, yet the TiO_2 immobilised onto fibreglass cloth was found to be more stable.

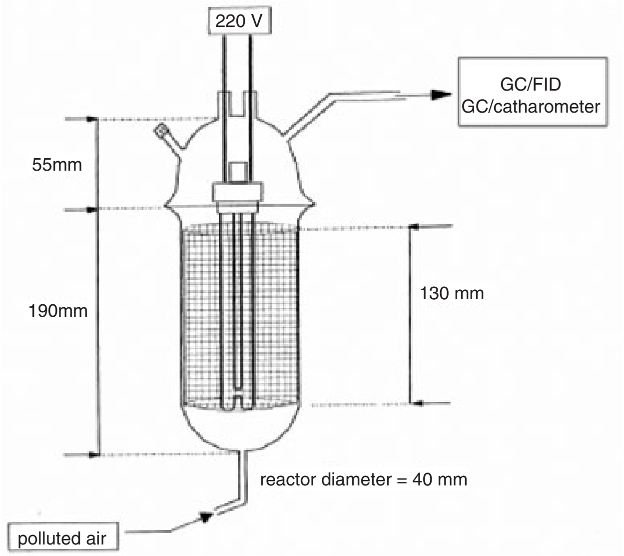

Figure 5. *Glass Fibre, Pichat et al. [33].*

4.2.5 Optical fibre reactor

The optical fibre reactor in Fig. 6 has been designed and tested by Yang and Ku. The basic design incorporates optical fibres for remote light transmission and for the solid support of the photocatalyst. The light delivery mechanism is quite different from that employed by other photocatalytic reactors. It is claimed to reduce the losses from absorption and scattering associated with the use of external light sources, so making more economic use of photons, whilst the geometry and configuration provide a high surface area.

4.3 Commercial applications

Of the reactor types described in section 4.2 the two most commonly found in commercial applications are the flat plate and fibre/membrane configurations. These are occasionally combined with other air purifying technologies, typically HEPA filtration and/or air ionisers, to form a final product. Table 2 lists a selection of commercially available products that are being marketed for domestic and light industrial use.

There is an ever-increasing volume of PCO products becoming available from a wide range of suppliers, mostly based in Japan and the USA. Not only are these product being seen as a benefit to the indoor environment of homes, offices, and industrial complexes, but PCO products are being used to prolong the life of agricultural produce, as with the Nippon Muki Co.'s Freshlong® [42] that eliminates the decay-promoting gas ethylene, and even as a potential resource against bioterrorism, as with the KES AiroCide [43], which has been proven to neutralize anthrax spores.

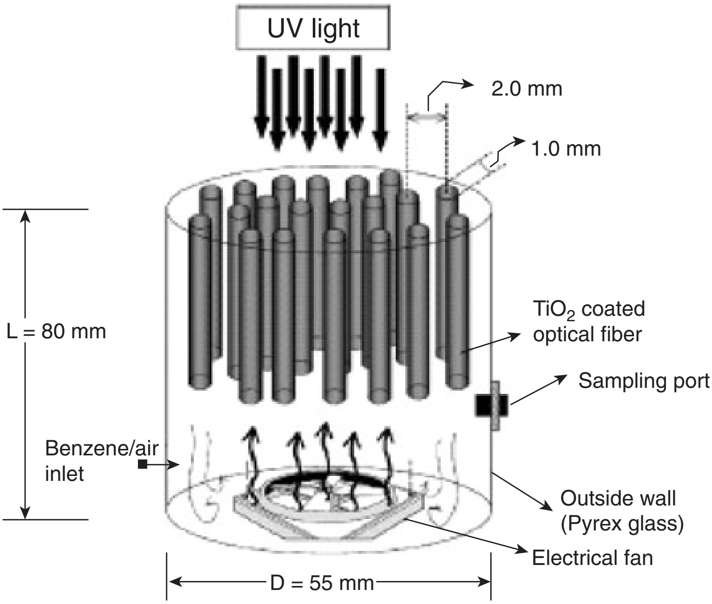

Figure 6. *Optical Fibre, Yang and Ku [37].*

Table 2. *PCO products and manufacturers*

Company	Product	Description	Ref.
Daikin	MC704VM	Ioniser plus photocatalytic filter elements	[38]
Genesis Air	GAP duct filter	Photocatalytic filter elements	[39]
Taiwan Fluorescent Lamp Co.	Air cleaning fan	Axial fan incorporating photocatalytic filter elements	[40]
Airtech international	Airsopure SP-20	HEPA filter plus photocatalytic filter elements	[41]
Airsource	Airsource 3000	Ioniser plus photocatalytic plates	[5]

5. Conclusion

The ability to coat almost any surface with photoactive material is one of the fundamental reasons why photocatalysis has been received with such enthusiasm. An extraordinary range of applications have already been commercialised and more still are in the research phase.

At the University of Nottingham researchers are working to incorporate the photocatalysis process in natural and mechanical HVAC systems as well as natural lighting schemes [44]. Fundamental research is being conducted in collaboration between the School of the Built Environment and departments of Chemistry and Materials engineering. Commercial products are being designed and tested in conjunction with industrial partners such as Pilkington and Baxi.

The future prospects of PCO look promising. The photocatalytic ability to eliminate pollutants, rather than change their phase, using non-hazardous and environmentally safe materials, while not requiring an electrical supply if using natural light, is a major advantage over competitive technologies. Of great significance is research currently underway to find a photocatalyst that can be activated by visible as well as UV light. Should this be achieved commercial activity in this area would no doubt increase significantly.

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