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Pierre Pichat

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Title: A brief survey of the practicality of using photocatalysis to purify the ambient air (indoors or outdoors) or air effluents

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Corresponding Author: Dr. Pierre Pichat,

Corresponding Author's Institution:

First Author: Pierre Pichat

Order of Authors: Pierre Pichat

1. Introduction

Photocatalysis using semiconductors was proposed long ago as an air-purifying means, which was presented as being robust, versatile, non-specific and easy to operate. However, it is impaired by the inherent decreases in the photocatalytic rates of pollutants removal resulting from the recombination of the charges generated by the photo-activation. The higher the recombination rate, the higher the loss of energy, hence the higher the cost whose impact depends on the type of air purification targeted. Much research has been devoted to finding solutions to this impediment, which indeed concerns all potential applications of heterogeneous photocatalysis, not only air purification. Until now, it does not appear that photocatalysts with a sufficiently satisfying increase in efficacy have been reported, especially considering that other prerequisites would be preparation meeting the industrial-scale needs and using innocuous, low-cost chemical elements, at least for some of the envisaged applications. Heterogeneous photocatalysis also presents two other drawbacks, viz. the unavoidable formation of degradation intermediate products, and final products, issued from some heteroatoms of the pollutants, that can form solid deposits on the photocatalyst and, hence, alter the activity. The effects on air purification of these predictable, well-known drawbacks have often been underestimated. This survey will attempt to evaluate their impact on the purification of indoor air, outdoor air and air effluents. For that, a few selected examples, thought to be informative, will be presented and the consequences for the practicality of the use of heterogeneous photocatalysis will be discussed. The topic of the photocatalytic effects on microorganisms will not be considered as it would merit a lengthy survey itself. Recent general reviews [1,2] and multi-authored books [3–6] on the fundamentals and various aspects of heterogeneous photocatalysis are cited here for

background to the readers **who would like further information and the professionals of air purification who may need it.**

2. Indoor air purification

The obvious benefits expected are for the people to breathe an air that is healthier and to increase the comfort by removing irritating and malodorous compounds. In the case where the air is recirculated, another advantage would be to decrease the flow of air introduced from outdoors, which would correspond to an energy saving on heating or cooling.

With respect to other purification techniques, photocatalysis appears a priori to offer several assets as mentioned in the Introduction. Regarding indoor air, other advantages can be underlined. The pollutants are degraded instead of being transferred or chemically neutralized, or, in the case of malodorous compounds, masked by compounds having a pleasant odor. Also, apart from CO, CH₄ and NO_x, most pollutants have concentrations, comprised between 0.1 and 50 ppbv, that are compatible with the potential of the relatively low photocatalytic removal rates. Consequently, the air pollutants can be removed simultaneously because no saturation of the photocatalyst surface is expected to occur in the ppbv concentration range. This is exemplified by Figure 1 showing that no noteworthy difference was observed in the removal rate of a given pollutant whether it was alone or mixed with nine other ones at 5 to 45 ppbv levels [7].

An optimistic view has resulted from some of the aforementioned considerations: complete mineralization of the organic pollutants would be achievable, that is, the initial toxic, irritating, malodorous pollutants would be removed and replaced by CO₂, H₂O and innocuous inorganics. This view has often been highlighted.

It is challenged by the following data [8]. The curves of Figure 2 show examples recorded with a prototype purifier placed in an ordinary room which hence was non-airtight. The concentrations of toluene and m,p-xylenes decreased initially and then reached a pseudo-stationary state corresponding to a kind of equilibrium between the photocatalytic removal rates and the rates of introduction from outside the room as well as emission inside the room, for the same pollutants. For o-xylene and benzene that were at lower levels, the decreases were much smaller and even insignificant, as was expected given the anticipated coverage of the photocatalytic material surface by these molecules. Overall, the concentration decreases of some of the initial aromatic pollutants illustrated the cleaning capability of photocatalysis. Moreover, these measurements regarding the initial aromatic pollutants were completed by measurements regarding some of the intermediate products of the photocatalytic degradation. In particular, those with a low number of carbon atoms are volatile; accordingly, they can be released from the photocatalyst surface into the air. Data were obtained for formaldehyde, acetaldehyde and acetone under the same conditions as those of Figure 2. They showed that the ranges of concentration measured for these three carbonyl compounds were significantly higher when the purifier UV lamps were on than when they were off (Figure 3) [8,9]. That is understandable because these carbonyls can be formed permanently from the many organic pollutants present in the room and their formation rates can exceed their degradation rates, at least during part of the study time period. The concentration ranges observed remain low. However, recommended concentrations for chronic exposure are also extremely low,

especially for formaldehyde. Accordingly, these data clearly indicate that an individual purifier must be adapted to the volume of the room in which it will operate and even sufficiently over-sized to cope with a pollution prone to vary. A concern has sometimes been raised about CO, which obviously can be formed from the photocatalytic incomplete oxidation of organic pollutants; however, given the concentrations generally < 10 ppbv of each of these pollutants, the contribution to the 0.5 to 5 ppmv-CO concentration in homes without stoves cannot be significant, considering the permanent exchange with outdoor air, which is a source of CO.

These measurements [8,9] had the interest to report on a real case, i.e., a non-airtight, non-occupied ordinary room containing actual pollutants at genuine concentrations. Conversely, the data thus obtained could not be modeled because the exchange of air with the outside was unknown, and, obviously, they were restricted to the particular case examined at given time periods (as in Figure 2).

Airtight test chambers have mostly been utilized for the ranking of individual purifiers, e.g. [10,11]. The efficiencies were measured using a few typical air pollutants or their mixtures introduced in the chamber at selected concentrations (unfortunately, sometimes higher than those of indoor environments). The rates of initial pollutant(s) removal and CO₂ production were determined. Formation of formaldehyde, acetaldehyde, acetone and other intermediate products were found, depending on the purifier and the initial pollutant(s). Over time, with proper purifiers, the air was cleaned within the chamber operated in the batch mode, which meant that the concentrations of the intermediate products passed through maxima. Similarly, when pollutants were not introduced but emitted from wooden panels placed in the airtight chamber, the action of the purifier caused the concentrations of the emitted pollutants to decrease over days, whereas the concentrations of the low molar-mass carbonyls increased initially [11]. All these observations were those expected on the basis of the knowledge of photocatalysis. They confirm the potential health risk due to the formation of hazardous compounds. Unfortunately, the rates related to the purifiers tested in the chambers, cannot easily be used to calculate stationary-states of pollutant concentrations in ordinary rooms.

An air photocatalytic purifier could also be used in heating, ventilating and air conditioning (HVAC) systems, e.g., in large office buildings. In a particularly thorough performance evaluation [12], an optimized photocatalytic purifier was installed in a well-ventilated, single room structure. **Figure 4 shows a typical design of a purifier using multiples of the module in the drawing.** The outlet of the device was ducted to a venturi flow meter and a blower was mounted downstream. This set-up mimicked purification using air ducts. The inlet contained single organic pollutants, typical of the ambient air, or mixtures of many of them at concentrations < 10 ppbv (i.e., realistic for indoor air). Formaldehyde, acetaldehyde and acetone were detected in the outlet air, as well as, in some experiments, formic acid and acetic acid which are expected to be the products of the photocatalytic degradation of these carbonyls, inter alia. From the viewpoint of photocatalysis, these results are not surprising. Undoubtedly, they draw our attention on the risk of delivering air containing hazardous compounds at too high concentrations relative to the recommendations, if the air residence time in the purifying device is insufficient.

Another problem regarding the implementation of photocatalysis to purify indoor air is the deactivation by inorganic, non-oxidizable solids deposited on the photocatalytic material. It is

well known that nitrate, sulfate, phosphate can be formed from the oxidative photocatalytic degradation of organic compounds containing the corresponding heteroatoms. It has been much more recently discovered that compounds containing Si atoms can also be at the origin of solid deposits [13,14]. Deactivation can, not only decrease the removal rates of the initial pollutants, but also promote the release of intermediate products, which is an added disadvantage.

Deactivation has been observed in photocatalytic purifiers containing different TiO₂-based coatings, deployed in the ambient air of offices in various buildings and countries [13,14]. The examination of the photocatalyst surface showed the presence of a SiO_x deposit (Si/Ti at. % = 0.3-1.5) supposed to be due to the oxidation of ambient siloxanes (~ 2ppbv). This interpretation was supported by a laboratory experiment showing a clear relationship between the percentage of deactivation, using propanal as a test compound, and the log of the ratio % Si/% Ti, Si being issued from exposure to tetramethylsilane under UV-irradiation (Figure 5) [13]. This type of deactivation can be limited by a simple means, that is, the use of a filter for siloxanes before admitting the air in the purifier. An increase in the TiO₂ coating lifetime by a factor of 2 to 4 thus gained was, nevertheless, considered by the company to be economically insufficient. Therefore, the use of a more elaborate means was explored. It consisted in adding an overlayer of a porous, UV-transparent adsorbent to reduce the encapsulation of TiO₂ by the SiO_x deposits. Though this development has been patented, it has apparently not been implemented on an industrial scale [13,14].

To conclude, safety seems to be the main obstacle to the implementation of photocatalytic purification of indoor air. In the case of individual purifiers, whether their management (including changing the photocatalytic material over time) can securely be left to the responsibility of the users, without proper control devices, is questionable. In the case of purifiers incorporated in air-ducts in buildings, planes, submarines, etc., adequate control can possibly be achieved permanently via proper sensors automatically adjusting the air flow to deliver healthy air, taking into account the pollution variations in the air before the treatment. For extra safety, if the pressure drop enables it, an adsorbing filter can be included before air admission in the occupied spaces. The cost of the photocatalytic treatment is also an issue, according to articles by a potential manufacturer well known for producing HVAC systems worldwide [13,14].

3. Outdoor air purification

Some years ago, a manufacturer of cementitious materials, published a cartoon claiming that if most of the building facades and the street paving were covered by TiO₂-containing coatings, the air will remarkably be healthier in our cities (<http://www.fotocatalisis.org/assets/doc497.pdf>). For the scientists, this claim raised the question: is it a dream or a reasonable objective?

Indeed, some measurements of NO_x abatement seem to support the potential of photocatalytic coverings to clean air outdoors. About the choice of NO_x, note that because of their usual concentrations, their roles in ozone formation and the toxicity of NO₂, they are target compounds to evaluate outdoor air pollution. In a pilot-scale canyon street with TiO₂ (3 wt.%) coated walls, a daytime NO_x abatement of 40 % to 80 % was observed during a

summer period of high sunshine and light wind, i.e., under favorable conditions [15,16]. It was successfully modelled, according to the street dimensions and some weather parameters, by the use of a three-dimensional CFD model into which the removal rates determined with the same photocatalytic materials in the laboratory were incorporated. In real streets, a TiO₂-coated paving enabled averaged NO_x abatements of 30 % at 30 cm above the ground, decreasing to 20 % at 180 cm, in the north of Italy in winter (<http://www.fotocatalisis.org/assets/doc497.pdf>), and between 10 and 20 % at a height of 180 cm, in Denmark from October to June (<https://www.dti.dk/projects/light2cat>).

However, these data showed that the effect was limited to relatively small distances because, obviously, the NO_x, as well as other atmospheric pollutants, need to be transferred from air to the photocatalytic surfaces. Unsurprisingly, they also illustrated the influence of the season, evidently related to the availability of UV light necessary to photo-excite TiO₂. On the whole, the NO_x abatements recorded were most often low. On the other hand, the expected positive effect on the concentration of ozone which can be photocatalytically removed [17] - while it is formed via NO₂ photolysis - should be determined in real environments. Additionally, the already low photocatalytic efficacy was reported to decrease over time because of various solid deposits (not always removable by rain), especially on the paving [18]; this is another factor to consider regarding the interest of TiO₂-containing materials for outdoor air purification.

Also, the release of the intermediate products of the photocatalytic degradation of air pollutants other than the NO_x has not been investigated in relevant conditions, likely because dilution was assumed to be sufficient to avoid a significant impact of these products on the outdoor air quality.

Overall, even if the additional cost relative to ordinary coverings would be acceptable according to the manufacturers, the interest of photocatalytic materials for purifying outdoor air appears questionable, especially because of the very restricted volume where the effect would be significant. Clearly, the self-cleaning property [19] must be put forward (though there are also limitations[20]), not the purification of air [19].

4. Purification of gaseous effluents

A priori, some factors seem favorable for implementing photocatalysis in the treatment of air effluents. First, the nature and concentrations of the main pollutants to be eliminated are, in general, relatively well-known and their over-time variations are also foreseeable, at least in some cases. Second, the outlet quality requested is pre-defined. Third, the industrial environment enables well-adapted monitoring of the purification system and possibly on-site regeneration of the photocatalyst. Fourth, the implementation of this technique can be regarded as relatively simple from an industrial viewpoint. Fifth, including a post- (or pre-) treatment should be possible, if need be.

Moreover, if intermittency is acceptable, the use of sunlight could be envisaged. Apart from the saving in electrical power to supply the necessary irradiation of the photocatalytic material, photovoltaic devices could also be employed to operate side devices, e.g., pumps, for supplementing economy. A compound parabolic collector of sunlight was tested at the laboratory scale with air containing trichloroethene and a TiO₂-SiMgO_x composite whose

added adsorbent was destined to minimize the effect of the irradiation variations [21]. A pilot-scale, field demonstration using a flat solar panel (Figure 6), has shown that the efficacy was a priori satisfactory: the removal of trichloroethene (10 ppmv) was > 95 % at $17 \text{ m}^3 \text{ h}^{-1}$, and also adequate for the other chlorinated alkenes present in the air stripped stream [22]. No loss in efficacy was observed over the 3-week long operating period. In spite of these fairly good results, there was no commercial outcome, to this author's knowledge. It might have been that the market and the constraints of the regulations for these low flow rates were not appealing enough for air purification companies.

The use of flat solar panels might be inappropriate for small facilities in large cities because of the lack of space. Consequently, a cylindrical, vertical solar reactor, including a scattering mirror, was cleverly designed (Figure 7) [23]. This reactor (2.1 m height; 0.3 m diameter) was tested to remove toluene (5 ppmv). At flow rates of 90 to 600 L h^{-1} , the average removal was incomplete. The positive message was that the activity was unchanged over the 6 months of testing, at least with this single pollutant and the TiO_2 -coated materials utilized. Apparently, no commercial outcome has been reported to date.

On the other hand, cost calculations have shown that photocatalysis using UV lamps, can be more economical than some conventional techniques at flow rates below $\sim 8500 \text{ m}^3 \text{ h}^{-1}$ ($\sim 5000 \text{ cfm}$) (Figure 8) [24]. Given this limitation, the potential photocatalytic applications could be the treatment of waste streams with sufficiently low flow rates, the remediation of sites where the flow rates are adjustable, and the purification of emissions from small point-sources, such as small businesses (cf. the previous paragraph), and from facilities in which waste solids are stored.

In addition to the cost, the presence of undesirable intermediate compounds in the outlet can be an issue, though not as crucial as for indoor air because solutions can be applied and adapted, e.g., if the inlet composition/concentrations vary. For instance, in the case of a field trial of a pilot-scale TiO_2 photocatalytic reactor for treatment of off-gases from a soil vapor extraction containing chloroethenes and 1,1,1-trichloroethane, some toxic chlorinated compounds, including COCl_2 previously found in laboratory experiments [14,25], were detected in the outlet, despite the high conversion [26]. In that case, the solution was merely to adequately dilute the inlet stream with air, that is, to decrease the molar flow rate of initial pollutants. Obviously, for a given mass of photo-excited catalyst and a given air flow rate, the amount of a given pollutant that can be mineralized is limited, considering the time scales of the adsorption phenomena and surface (or near-surface) reactions; beyond this limit, the pollutant(s) will be incompletely eliminated, or volatile intermediate products will be released, or both.

Some years ago, the hope to use photocatalysis to purify gaseous effluents led to a large-scale demonstration process [27] (Figure 9). The gas to be photocatalytically treated was extracted from a soil, containing chlorinated hydrocarbons, whose remediation was requested. The maximum flow-rate was $\sim 1175 \text{ m}^3 \text{ h}^{-1}$ ($\sim 700 \text{ cfm}$), which was high, below, however, the aforementioned threshold for efficiently competing with other conventional techniques on a cost basis. The UV lamps had a total power of 19 kW. Behind the photocatalytic reactor, a caustic scrubber was used to remove HCl. Over the 4 months of operation, the removal of trichloroethene (average concentration: 1000 ppmv; 100 % relative humidity) was excellent, and all other contaminants in varying, lower concentrations were also removed. Despite these

apparently good results from the purification viewpoint (https://tools.niehs.nih.gov/srp/researchbriefs/view.cfm?Brief_ID=131), the process has not been expanded. Perhaps, the cost was estimated too high because of the needs (i) of an important electrical power to ensure the requested continuous removals starting from inlet ppmv levels, and (ii) of regenerating the photocatalytic material too frequently. Other reasons could be advanced: insufficient, appropriate market; as usual, competition with other well-established technologies and reluctance to implement a technology without sufficient background.

Another pilot-scale trial is worth being mentioned in this survey because it addressed the case of the purification of air issued from a small workshop and it used a photocatalytic unit behind biotrickling filtration [28]. The air to be treated contained more than 45 compounds, among which 6 major compounds (ethyl acetate and substituted benzenes) were selected to monitor the purification, which was found to be > 95 % with flow rates between 1333 and 3000 m³ h⁻¹ for the combined techniques in all conditions (varying humidity and concentrations of pollutants, etc.) over the 90 days of operation. These data illustrated the interest of using photocatalysis behind another more conventional treatment in order to increase the purification performance.

5. Conclusions

In this survey, the barriers for the implantation of heterogeneous photocatalysis to clean air have been presented and discussed. Their relative importance depends on the application targeted. In this author's view, the most crucial barriers are: the release of light molar-mass carbonyl compounds for indoor air; the inherent restriction to confined spaces and short distances for outdoor air; the potentially insufficient removal rates for treating effluents at a competing cost. The gradual deactivation of the photocatalytic material concerns all the application domains. Apart from the case of deposition of atmospheric dust and other types of dirt, the cause of deactivation arises from the photocatalytic process itself: formation of organic deposits, essentially in the case of the treatment of gas effluents because of the higher concentrations of pollutants; formation of inorganic, non-oxidizable deposits, such as nitrates, sulfates, and also Si oxides, resulting from the total oxidation of some pollutants, including siloxanes, particularly relevant in indoor air. In the case of outdoor air, these salts can be washed away by rain, depending on the location and climate. For the other potential applications, the periodical regeneration or renewal of the photocatalytic material will be necessary and hence will increase the cost.

Whatever the domain, despite much research, pilot-scale reactors, demonstration sites, there has been no really successful commercial application so far. Presently, it seems that the likely prospects will concern the treatment of some air effluents (maybe in combination with another technique), and possibly the purification of indoor air distributed by ducts, provided the conditions are well-controlled by responsible agencies using proper sensors. For both prospects, the cost will, obviously, remain an essential issue for the companies.

To pass the obstacles to development in both domains, improvements are needed for both the reactors and the materials, in general and also on a case-by-case basis. For the reactors, the usual goals are to efficiently supply the pollutants to the photocatalyst surface and to irradiate it as much as possible. For the materials, aside from the efficacy, the goals are the retention of

the volatile products and the resistance to deactivation according to the industrial necessities. Regarding the effluents, the goal for the companies is to seek cases where the characteristics of heterogeneous photocatalysis can help meet economically the constraints of the environmental regulations, which are expected to grow. Concomitant, concerted and significant progress to reach these practical goals is required to implement this technology.

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Captions to Figures

Fig. 1. Reacted fractions of the compounds indicated when they were introduced into the purifying device individually or as a mixture with the other 9 compounds. Error bars represent ± 1 standard deviation of the means. 2-BE: 2-Butoxyethan-1-ol. MIBK: 4-Methylpentan-2-one. Reprinted with permission from reference 7.

Fig. 2. Figures showing the concentration variations for the pollutants indicated in an 83-m³ ordinary room on two different days. t is the functioning time of the air purifier prototype. **The detection limit was ~1 ppbv.** Reprinted with permission from reference 8.

Fig. 3. Measured concentration ranges of the carbonyl pollutants indicated in an 83-m³ ordinary room with (black bars) or without (gray bars) photocatalytic air treatment over 2-h periods. Reprinted with permission from reference 8.

Fig. 4. Generic, multi-stage, coated honeycomb-monoliths photocatalytic purifier. Reprinted with permission from reference 14.

Fig. 5. The deactivation % of a TiO₂ P25 coating against the log of the ratio % Si/% Ti as determined by XPS analysis. Reprinted with permission from reference 13.

Fig. 6. Schematic of the solar reactor system used to treat an air stripped stream at a field remediation site. The dimensions of the flat panel were 1.2 m x 2.4 m. TiO₂ P25 was coated onto a structured, perforated, polypropylene tubular packing. Reprinted with permission from reference 22.

Fig. 7. Schematic and photo of a cylindrical, vertical, small footprint, solar purifier prototype. Reprinted with permission from reference 23.

Fig. 8. Cost evaluations of the technologies indicated versus flow rate. Values based on a hydrocarbon - which put photocatalysis at a relative disadvantage - as pollutant (at 500 ppmv). **For photocatalysis, they were based on laboratory/pilot tests, and engineering estimates.** Reprinted with permission from reference 24.

Fig. 9. Photo (provided by Trojan Technol.) of the installation built to purify the air stripped from contaminated groundwater at a former plant. The photocatalytic units are on the left.

Fig. 1

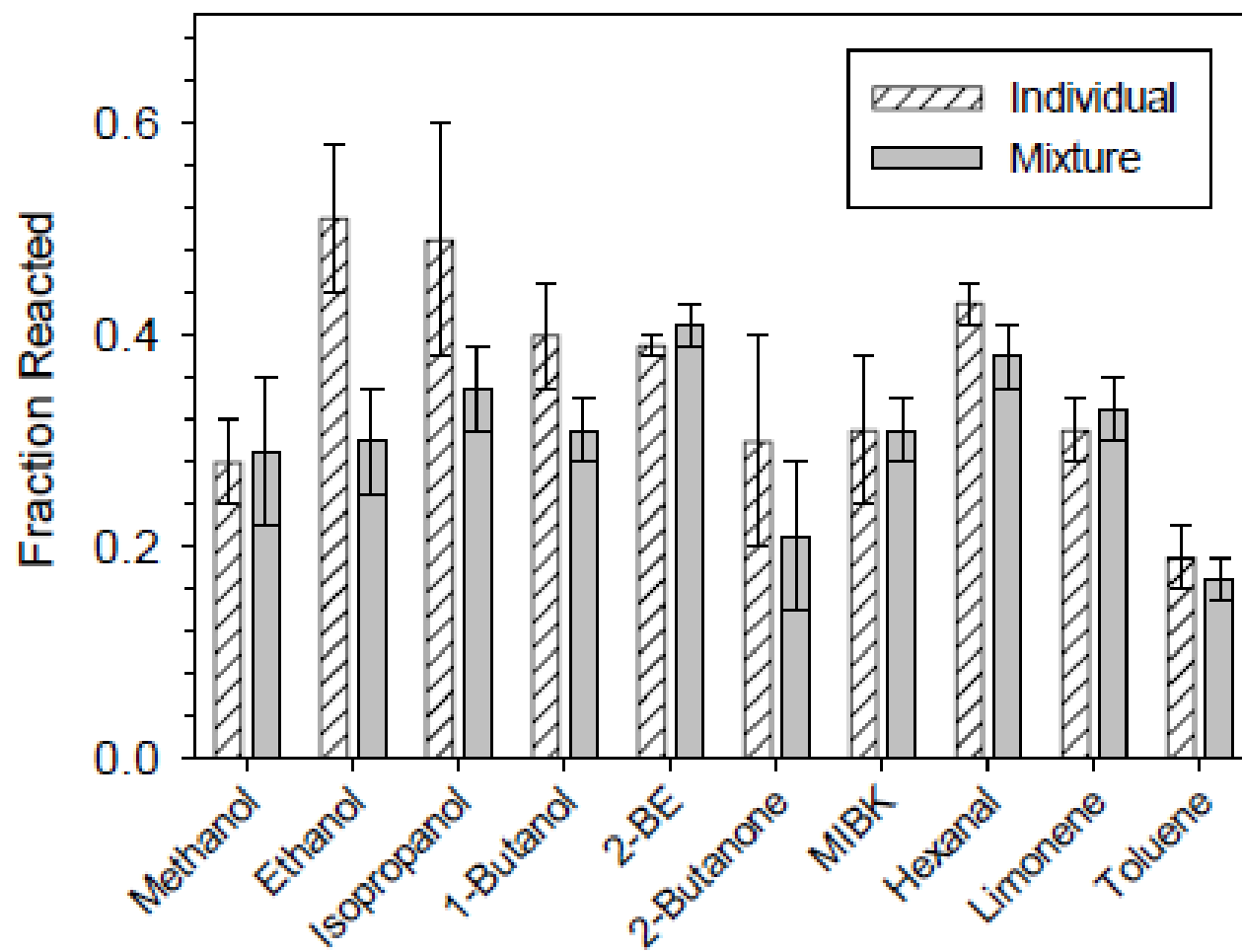


Fig. 2

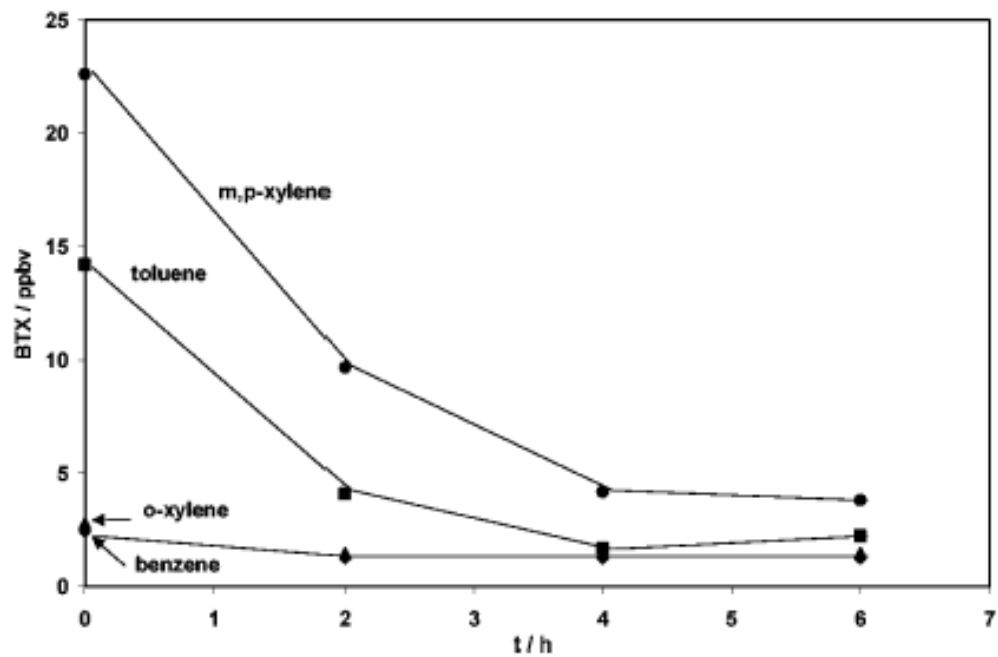
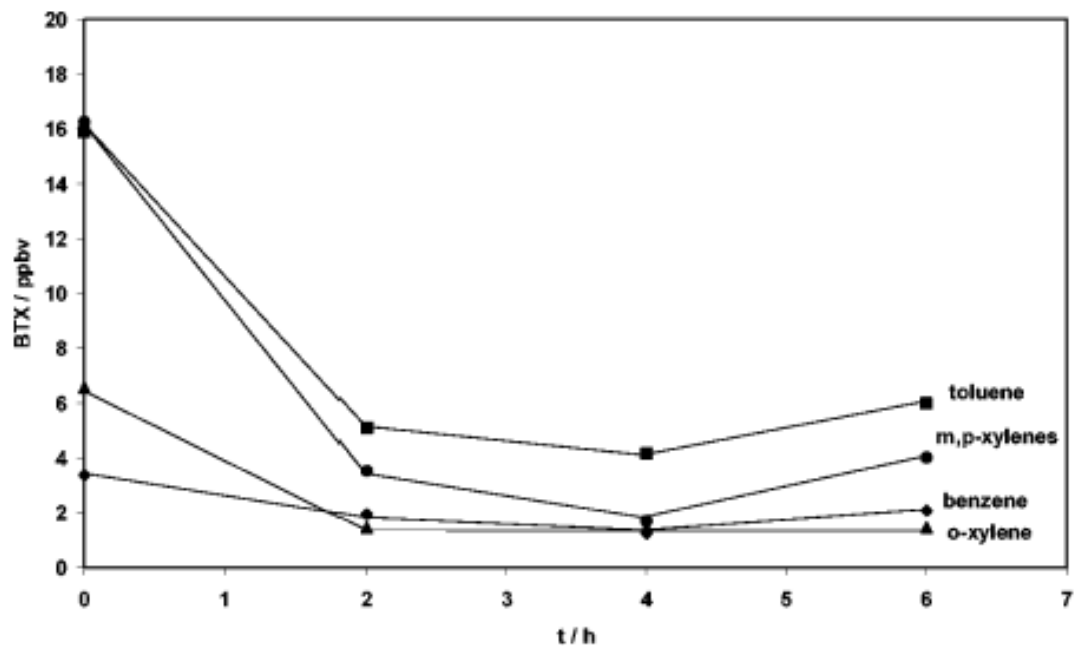


Fig. 3

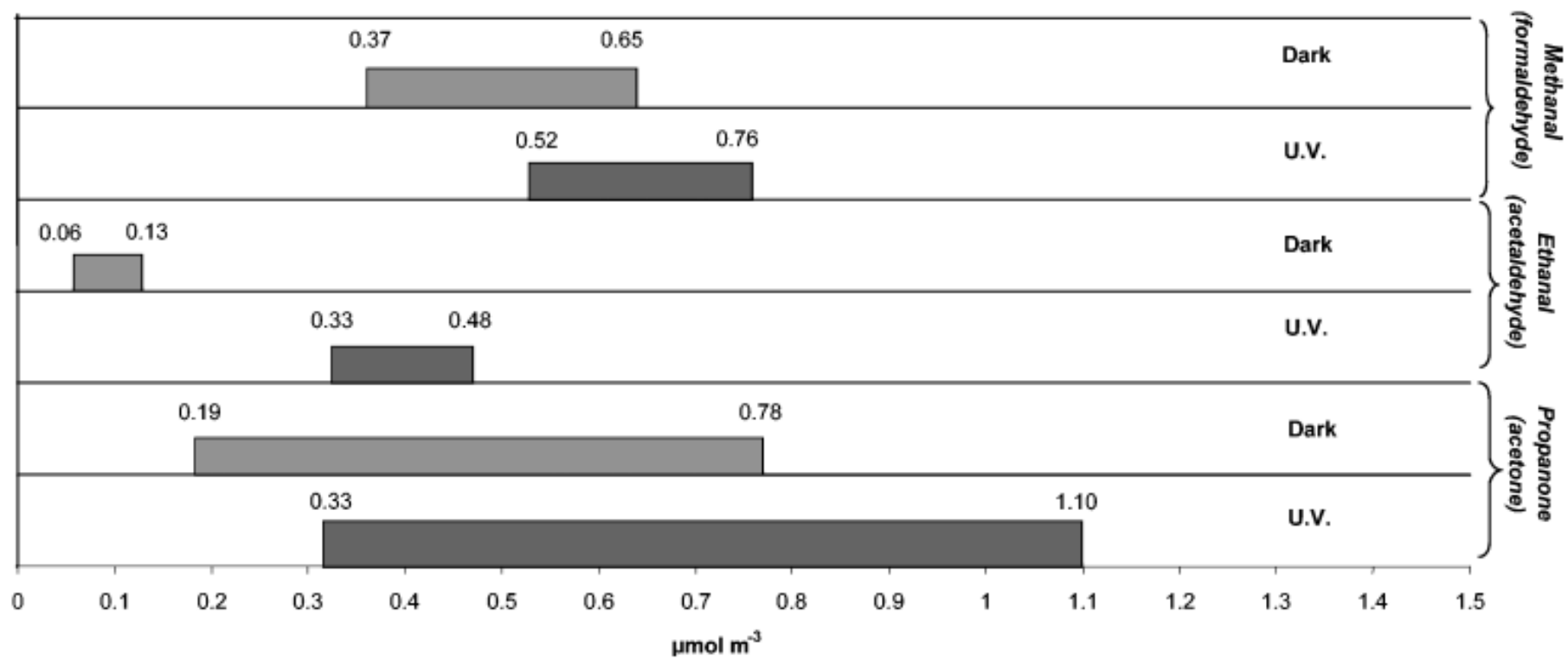


Fig. 4

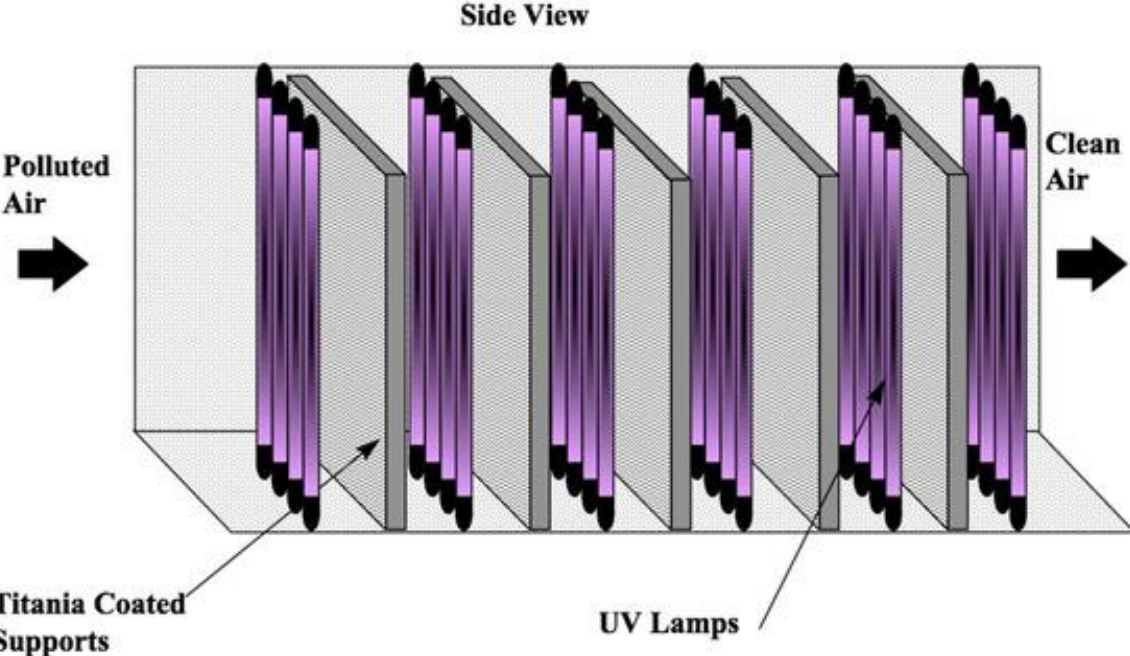


Fig. 5

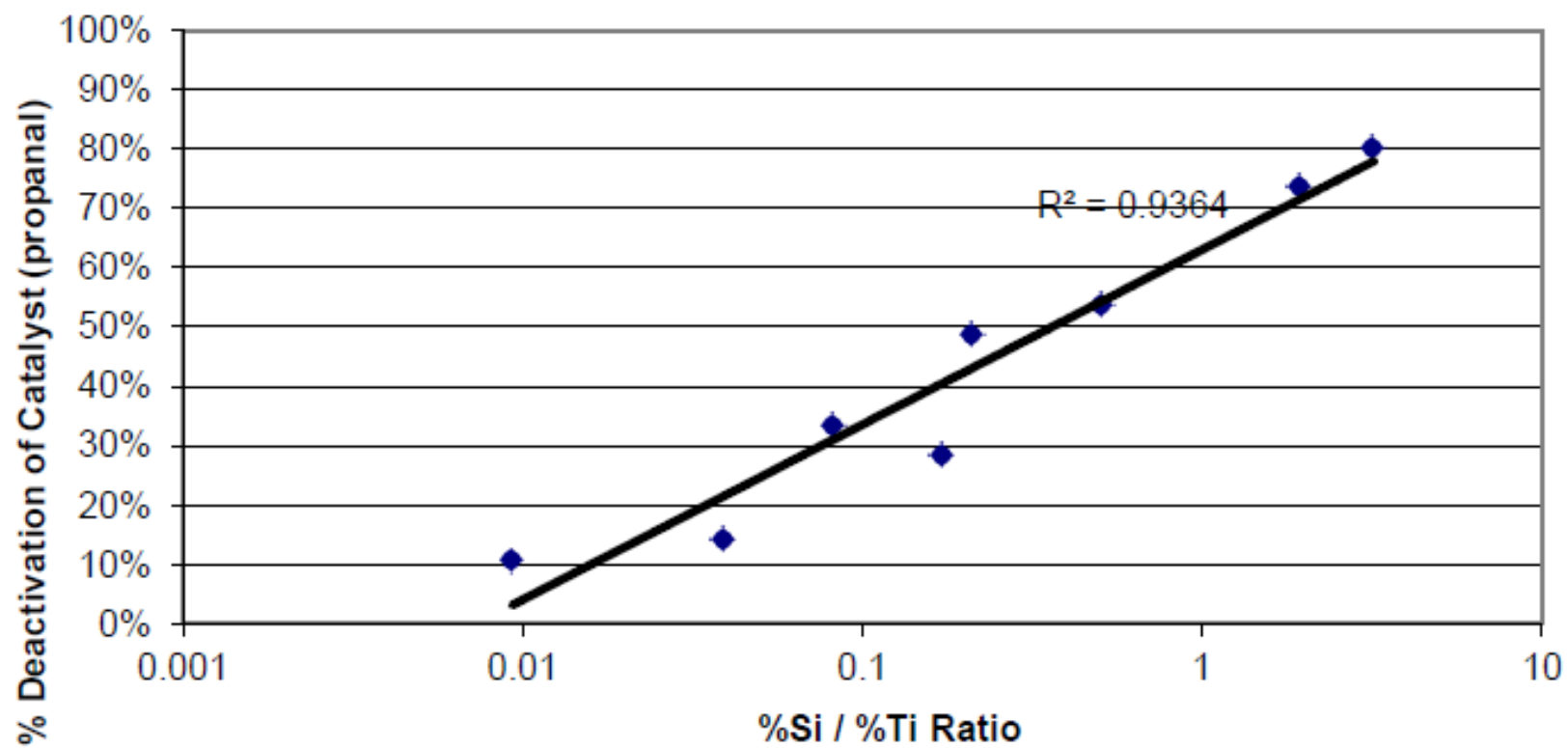


Fig. 6

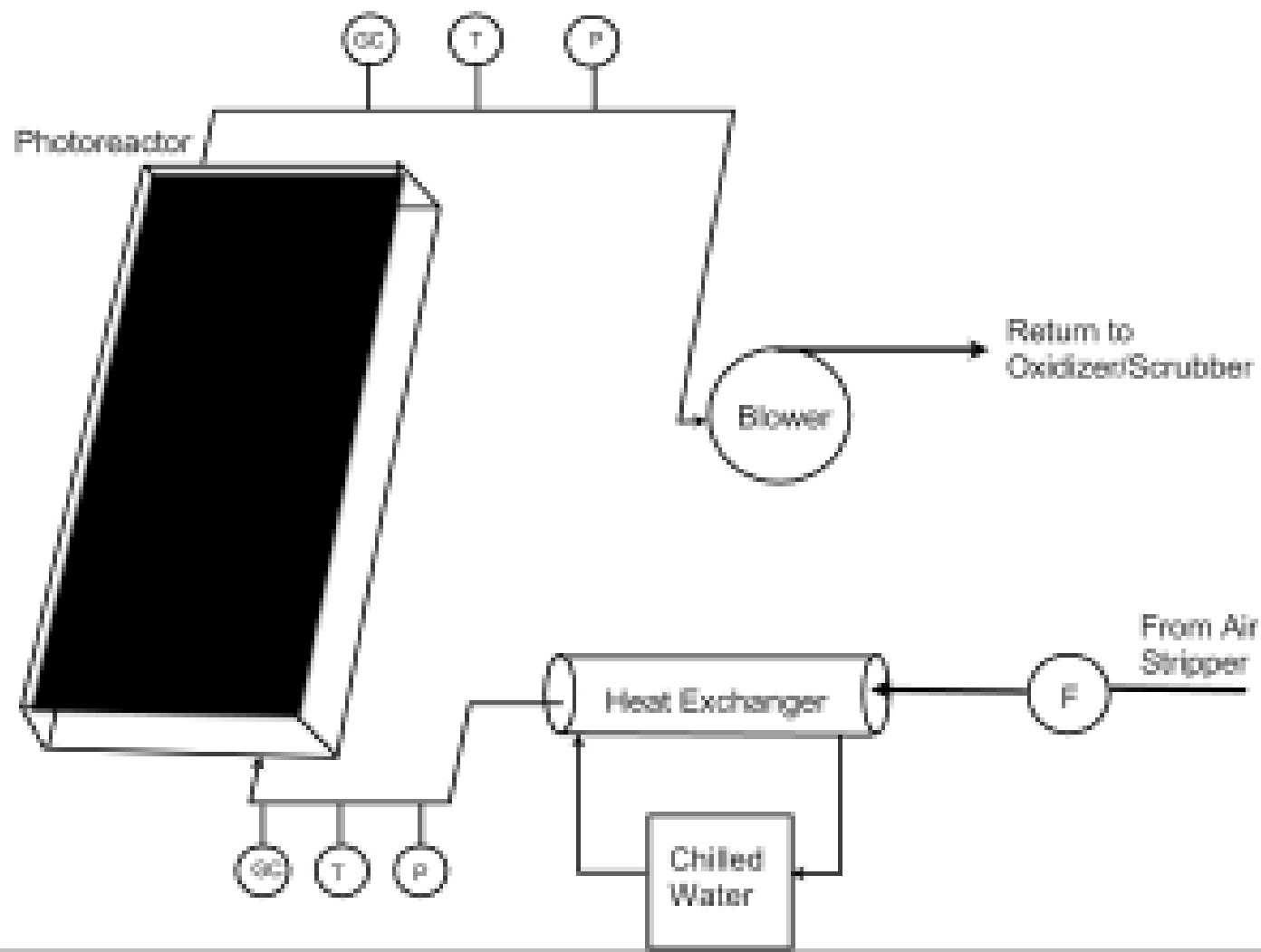
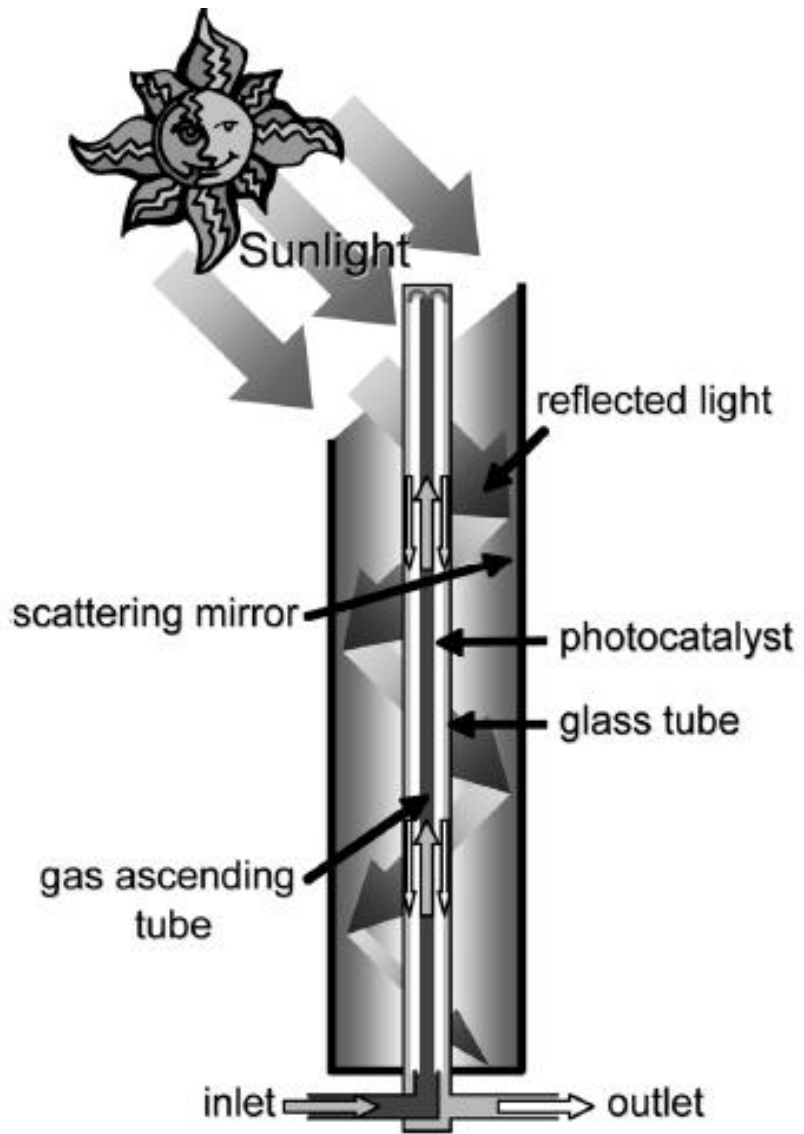


Fig. 7



Levelized Annual Cost (\$/cfm)

Fig. 8

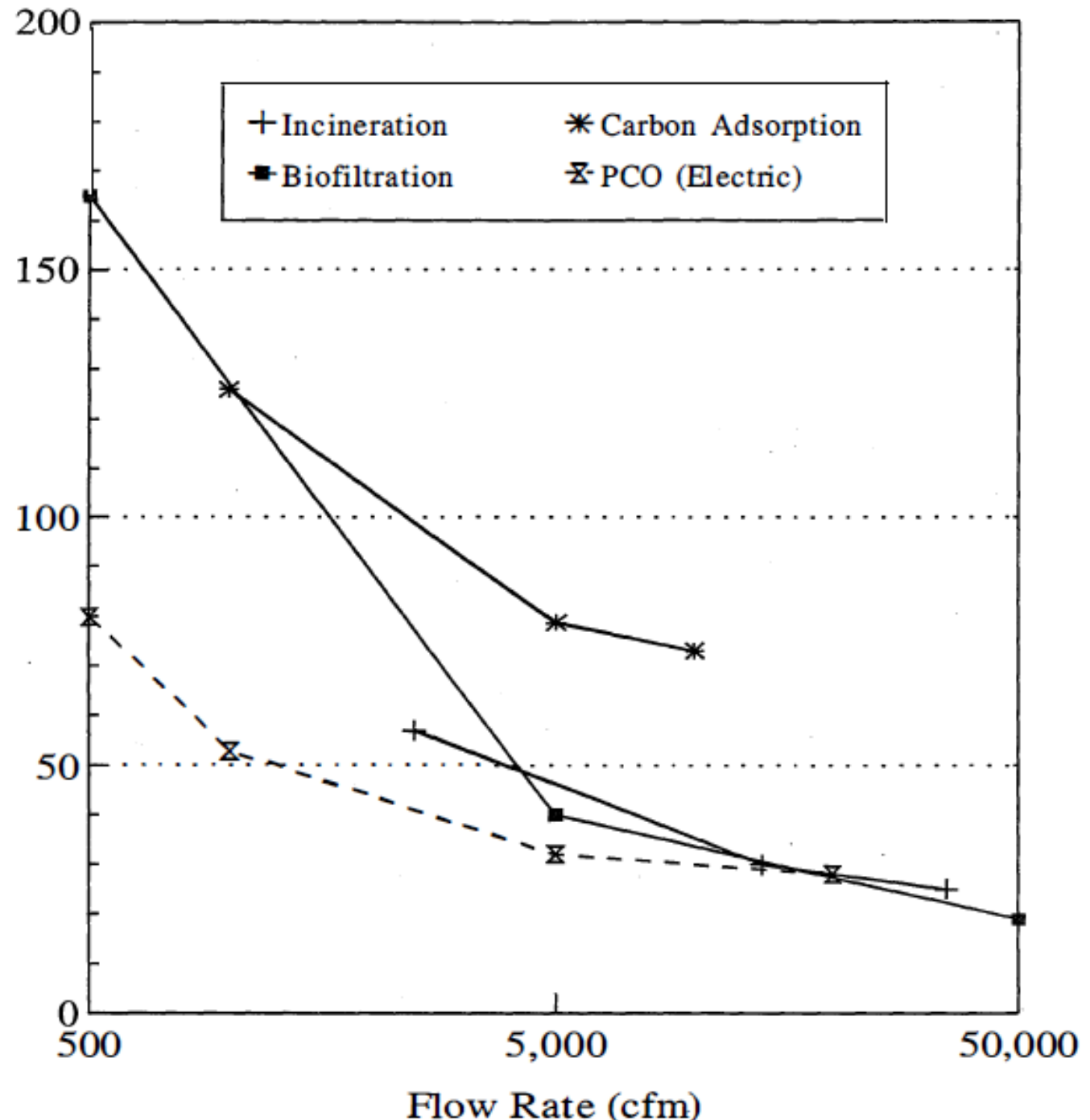


Fig. 9



