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Radiation Field Optimization in Photocatalytic Monolith Reactors for Air Treatment

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The tools for optimal design of photocatalytic air purifiers are still being established. In this article, the optimal design of monolith photocatalytic reactors irradiated by cylindrical UV lamps was investigated. The radiative transfer equation in dimensionless monoliths was solved by the Monte-Carlo method to yield the radiation field in the reactor. The effect of the dimensionless design parameters, monolith aspect ratio, geometric ratios, catalyst reflectivity and number of lamps were investigated and correlated in terms of photon absorption efficiency (ϕ), uniformity of radiation distribution (η), and overall monolith photonic efficiency ($\Phi = \eta\ \phi$). Optimal design is achieved for aspect ratios of 2.5 and monolith-to-lamp-length ratios in the range from 1 to 1.1. The largest number of lamps compatible with uniform air distribution over the monoliths should always be used. The optimal monolith-to-lamp-distance ratio, $\beta_{optimum}$, was correlated as: [$\beta_{optimum} = 1.698\ (n + 1) - 0.279$], where n is the number of lamps. © 2007 American Institute of Chemical Engineers *AIChE J,* 53: 678–686, 2007
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Introduction

In most developed and many developing countries, people spend on average 90% of their time indoors,[1] where many pollutant levels are two to five times higher than outdoors. As a consequence, the US EPA and its Science Advisory Board have ranked indoor air pollution among the top five environmental risks to public health.[2] Photocatalytic oxidation (PCO) processes have been demonstrated to be a real alternative to conventional methods of indoor air purification.[3–5] In contrast to other catalytic systems of air purification, they can operate efficiently at room temperature and low pollutant concentration (ppb levels) and can eliminate the offending pollutants to levels designated by the EPA as probably harmless.

Feasibility studies have confirmed that PCO can be efficiently applied to treat air in indoor environments such as aircrafts,[6] submarine cabins,[7] and office buildings.[8] Moreover, commercial UV photocatalytic air purifiers have been made available in the market (such as Daikin (JP), ITI-Airlife (RU), Airwise (US), and Peak Pure Air (US)) that claim to be the perfect appliances to enhance indoor air quality not only in offices and homes, but also in hospitals, schools, and commercial areas. However, the tools to ensure the optimal design (that is, mathematical models) of photocatalytic air purifiers are still in the process of being established.

In general, effective photon utilization is the crucial performance criterion that, together with the pressure drop, governs the economic viability of a particular PCO reactor design.[9] As a result, structured monoliths with the catalyst coated on the walls, have been proposed as an effective reactor design due to the low pressure drop and high surface area they offer.[10]

In a photocatalytic monolith reactor, light usually enters the monolith channels from an external source and, as a result of radiation scattering and absorption, it decays

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through the channel. The radiation field in a monolith reactor can be obtained by solving the radiative transfer equation (RTE) for the monolith.[11] The distribution of the local surface rate of photon absorption (LSRPA), which in turn controls the local photocatalytic reaction rate,[12] can then be calculated by multiplying the radiation intensity at the wall by the absorption coefficient of the supported photocatalyst.

Two methods have been proposed to solve the RTE in a photocatalytic monolith reactor: the numerical and the statistical approach. The numerical method was used by Hossain and Raupp[9,13] and Hossain et al.[14] to model the radiation field in a monolith channel under the assumption of uniform illumination of the channel mouth. The statistical method based on the Monte-Carlo simulation of a significant number of photons entering the channel mouth was used by Raupp et al.[4] and Alexiadis[15] to model the radiation field in circular, square, and triangular channels. Both methods provide identical solutions and agree with experimental measurements. Hossain concluded that for a given channel type, light intensity profiles are controlled by channel aspect ratio (monolith thickness divided by channel diameter) and catalyst reflectivity. Furthermore, it was found that beyond 3 to 4 aspect ratios, the light intensity fell below 1% of the incident light. Alexiadis proved that circular geometry provides the highest degree of photon absorption compared with square and triangular geometries.

Commercial and pilot-scale monolith photocatalytic reactors are usually composed of many photocatalytic modules with cylindrical UV lamps located in between the modules. The optimal design of these reactor configurations requires the estimation of the most efficient geometry that yields the highest catalyst photon absorption efficiency and the highest degree of uniformity of distribution of radiation in all channel mouths.

In this article, we investigate the optimal design of monolith photocatalytic reactors irradiated by cylindrical UV lamps. In contrast with previous work, where all the monolith channels were considered to be identically irradiated at their entrance, a lamp emission model is introduced to consider the actual distribution of photons at the front surface of the monolith. The Monte-Carlo method is used throughout to solve the RTE in dimensionless reactor geometries to yield the radiation field in the entire reactor. The effect of the dimensionless design parameters, monolith aspect ratio, geometric ratios, catalyst reflectivity, and number of UV lamps on the distribution of radiation and optimal photon absorption efficiency was evaluated. Optimal design parameters of monolith photoreactors were determined.

Model Definitions and Assumptions

Figure 1 shows a typical commercial set-up for a monolith photocatalytic reactor. It consists of a honeycomb monolith coated throughout with the TiO_2 catalyst. One or more UV lamps are symmetrically placed in between the monoliths.

Four dimensionless geometric ratios are introduced to model the geometry of the reactor: the monolith aspect ratio, AR:

$$AR = \frac{D}{d} \tag{1}$$

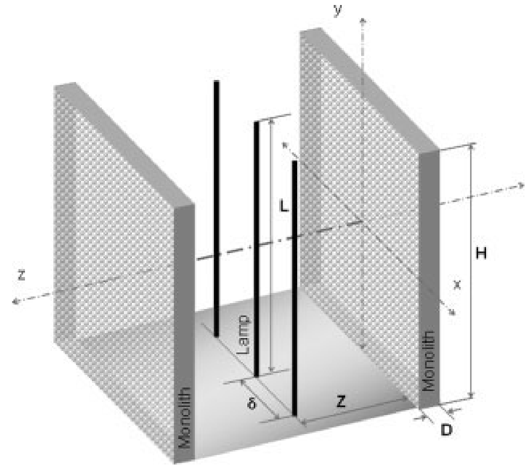

Figure 1. Standard configuration of a three lamp monolith photocatalytic reactor and definition of the coordinates of the system.

where D is the thickness of the monolith and d is the diameter of the pore; and the reactor geometric parameters, α, β, and γ:

$$\alpha = \frac{H}{L} \tag{2}$$

$$\beta = \frac{H}{Z} \tag{3}$$

$$\gamma = \frac{H}{\delta} = \frac{H}{H/(n+1)} = n + 1 \tag{4}$$

where H is the height of the monolith (normally considered square), L is the length of the lamp, Z is the distance between the lamp and the monolith, δ is the spacing between the lamps, and n is the number of lamps that are placed parallel to the monolith front surface.

Qualitatively, as α and β increase and γ decreases, the distribution of radiation on the front surface of the monolith becomes less uniform. Conversely, as the aspect ratio AR increases, more photons will be absorbed within the monolith.

Three further dimensionless performance indicators are introduced to evaluate the efficiency of a monolith photocatalytic reactor. The first is the "monolith photon absorption efficiency" (ϕ), which is defined as the ratio of the photons absorbed by the catalyst in the monolith divided by the photons emitted by the lamps. From a practical point of view, the only surfaces that are considered to be photocatalytically active are the monolith wall (the front face surface and the channels). The reactor surrounding walls are considered to be a black body that absorbs photons without reflection and catalytic activity. This assumption is supported by the fact that in an actual reactor these surfaces will probably be fixed (not replaceable, as would be the case for the monoliths) and, therefore, subject to fouling as a result of low air turbulence in the region near their walls.

Notwithstanding the inevitable decrease in radiation intensity in each channel of the monolith, the performance of a monolith reactor will also be affected by the degree of uniformity of irradiation of the front surface of the monolith. In fact, a mal distribution of the photon flux in this region would result in lower pollutant conversions compared to a situation in which all pores receive the same photon flux. Thus, the "uniformity of distribution of the dimensionless radiation intensity at the front surface of the monolith" can be defined as (η):

$$\eta = \frac{\int_A \frac{I_{(x,y)}}{I^{\max}_{(x,y)}} dA}{A_{\text{front face}}} \tag{5}$$

where $I_{(x,y)}$ is the total number of photons impinging on the differential area dA centered at the position (x, y) on the front surface of the monolith, and $I^{\max}_{(x,y)}$ is the maximum value of $I_{(x,y)}$. η is essentially a definition for the normalized total radiation intensity received by the monolith.

The final and perhaps the most important performance indicator proposed in this study is the "monolith overall photonic efficiency" (Φ), which combines the monolith photon absorption efficiency and the uniformity of distribution of radiation intensities at the front surface of the monolith. Mathematically it can be defined as:

$$\Phi = \phi\eta \tag{6}$$

Note that (ϕ), (η), and (Φ) take values between 0 and 1, with 1 being the limit for optimal design.

The following further assumptions are considered in the model:

(1) The lamps are time-invariant, diffused monochromatic light sources.

(2) The radii of the lamps are much smaller than their length; hence, the lamps are considered to be linear light emitting sources that emit photons uniformly throughout their lengths in all directions and following the Linear Source Spherical Emission Model.[16]

(3) The interaction of the light with the gas phase (absorption, scattering, and reflection) is negligible.

(4) The optical properties of the photocatalytic thin film (absorbance and reflectance) are independent of light wavelength, light incident angle, and thickness of the coating. The light scattering by the solid is considered to be elastic (energy is conserved) and perfectly diffuse.

(5) The photocatalytic thin film coating on the monolith walls is uniform and sufficiently thick that no light transmits through the thin film. Hence, the photons can only be absorbed or scattered.

(6) The thin film does not emit radiation in the spectral region of interest.

(7) If a photon encounters a lamp, it is absorbed and a new photon is re-emitted.

Monte-Carlo Simulation

Figure 2 shows the Monte-Carlo algorithm used to evaluate the efficiency of a monolith photocatalytic reactor. A statistically significant number of photons are considered to be emitted by each lamp. The fate of each photon is followed until it is either absorbed at the monolith walls or is lost on the surroundings, as shown in Figure 3. For each emitted photon, a random number sequence is initiated that locates its position on the lamp and the direction it flies. If the photon does not hit one of the monoliths on either side of the lamp, it is discarded and assumed to be lost, and the process is started again. Conversely, if it strikes the front surface of one of the monoliths, two cases can arise:

(a) It enters into one of the channels.

(b) It hits the front surface (the space between the circular channels).

When the photon hits the surface of the monolith (the front or channel wall), a random number ξ, in the domain from 0 to 1, is then generated. If ξ is higher than the reflectivity ρ of the catalytic thin film ($0 \leq \rho \leq 1$), then the photon is absorbed and the count of absorbed photons is increased by one. If $\xi < \rho$, the photon is scattered. Subsequently, the scattered photon is assumed to have a completely new random direction based on three random direction cosine variables. If subsequent scattering events result in the final absorption of the photon at the monolith walls, the count of absorbed photons is increased by one. If the photon escapes from either the entrance or the opposite exit of the channel, its flying direction is checked. If it strikes a monolith symmetrically placed on the other side of the lamp, the

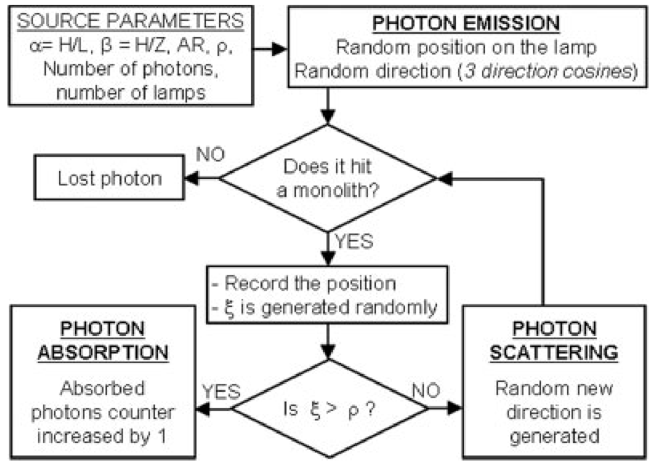

Figure 2. Simplified structure of the Monte-Carlo algorithm.

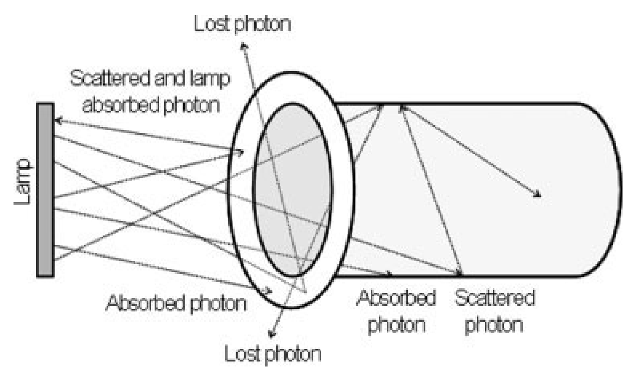

Figure 3. Possible fates of the photons in a monolith channel.

sequence of events described above is continued. If it hits the lamp, the photon is re-emitted. If it hits the surrounding walls, it is counted as lost.

At the end of the simulation, the photon efficiency (ϕ) is calculated by dividing the number of photons that were absorbed by the total number of photons emitted by all lamps. The uniformity of distribution of the dimensionless radiation intensity at the front surface of the monolith (η) was evaluated by dividing the monolith front wall in a discrete number of small domains of area dA, by counting the number of photons striking each domain and by applying Eq. 5. Under normal circumstances, the simulations were run with a total number of 10^6 photons emitted per lamp, which ensured the smoothness and invariability of the model outputs.[15]

Results and Discussion

The experimental results of dimensionless photon flux as a function of aspect ratio reported in Hossain and Raupp[9,13] for a square channeled monolith were compared with the Monte-Carlo simulations. The geometry of the monolith reactor was reproduced and simulations were run using circular channels of the same surface area as the square channels and at the same channel density (25 channels per square inch). The wall reflectivity was taken to be 0.4 as in Hossain and Raupp results. Four channels located at the center of the monolith were monitored, and the dimensionless photon-flux (number of photons escaping from the back of each channel divided by the number of photons entering each channel) was calculated at different channel aspect ratios. Figure 4 shows a slight underestimation of the experimental results by the model, which could be anticipated since monoliths with circular channel geometry yield a higher degree of photon absorption compared to monoliths with square channel geometry. The difference is, however, within the limit of confidence reported in Alexiadis[15]; thus, if one also includes the uncertainty of the experimental measurements and wall reflectivity value, the experimental results can be considered to be well represented by the model. As a result, with confidence that the Monte-Carlo method models the fundamental physics of photon transport and absorption in the monolith, further simulations were run to explore the effect of the design parameters on the monolith performance indicators (ϕ), (η), and (Φ). A realistic set of values of the design parameters were considered in the simulations, as reported in Table 1.

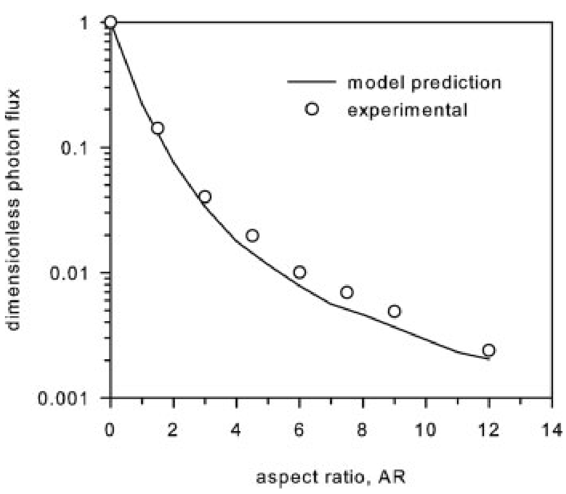

Figure 4. Dimensionless photon flux at monolith back wall as a function of aspect ratio.

Experimental results are taken from Hossain and Raupp[9,13] and refer to a square channeled monolith. Model predictions are for circular channels of the same surface area as the square channels. Channel density is 25 per square inch. Wall reflectivity is $\rho = 0.4$.

Table 1. Parameters Values Considered in the Monte-Carlo Simulations

Geometrical Parameters	
Dimensionless Parameter	Range Investigated
Ratio of height of monolith to height of lamp	$0.5 < \alpha < 2.5$
Ratio of height of monolith to distance of lamp from front face of monolith	$1 < \beta < 20$
Ratio of height of monolith to lamp spacing	$2 < \gamma < 8$
Ratio of monolith thickness to pore diameter	$0.5 < AR < 7.5$
Reflectivity of the photocatalytic thin-film	$0 < \rho < 0.9$
Number of photons emitted per lamp	$10^4 < N_{phot} < 10^7$

Catalyst reflectivity (ρ)

The reflectivity, ρ, is the parameter considered to decide the fate of a photon (absorption or scattering) once it hits the surface of the monolith. This parameter is a characteristic of the catalytic thin-film and is usually a function of wavelength.[13] A value of ρ near one means that the surface scarcely adsorbs photons and acts as a mirror; conversely, a value of ρ approaching zero means that the thin-film is similar to a black body, which absorbs all the incident radiation. This last case would be ideal for a photocatalytic surface. However, TiO_2 photocatalytic surfaces tend to have reflectivities in the range of 0.3 and 0.6, depending on the crystal composition, the particle size, and the roughness of the surface, which in turn is a function of the coating method.[13,15]

In this work we restrict the analysis to monochromatic radiation sources, although the extension of the Monte-Carlo method of solving the RTE to polychromatic sources of radiation is straightforward.[15] In most monolith photocatalytic reactors for air purification, low pressure mercury lamps emitting radiation at 253.7 nm would be the preferred choice since they would simultaneously accomplish air disinfection and detoxification. In addition, these lamps are low cost, are of high UV efficiency (40% of lamp electrical power is reemitted as UVC light) and have much longer lifetime (up to 9000 hrs) compared to UVA blacklight lamps or medium pressure mercury lamps. Low pressure mercury lamps provide relatively lower photon fluxes compared to those of medium pressure mercury lamps. As a result, multiple lamps are usually needed to irradiate the monolith, an aspect discussed later that can lead to an advantage compared to medium pressure mercury lamps.

Figure 5 shows simulation results of the influence of the wall reflectivity on the monolith photon absorption efficiency (ϕ) when β is varied at constant aspect ratio and α. The photon absorption efficiency increases with β (that is, the lamp

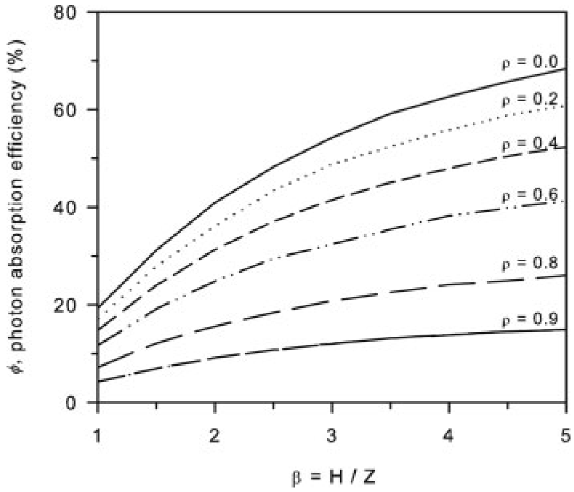

Figure 5. Photon absorption efficiency versus β at different wall reflectivities (ρ).

AR = 2.5, $\alpha = 1.5$, $\gamma = 2$.

approaching the monolith) and decreases as the wall reflectivity increases. The higher value of (ϕ) is achieved when the monolith coating behaves as a black body ($\rho = 0$) and it is placed the closest to the lamp. In practice, in small reactors, consideration of the fluid dynamics of the air flow distribution over the monolith may impede the use of such configurations.

Aspect Ratio (AR)

Raupp et al.[4] have shown that the light intensity profile in a monolith channel type is dependent on the channel aspect ratio and on the thin-film reflectivity only. Further studies[9,15] demonstrated that the photon absorption flux decays sharply to less than 1% when the monolith aspect ratio is higher than approximately 3 to 4. Therefore, typical commercial monoliths for catalytic combustions, with aspect ratios between 20 and 150,[17] are unsuitable for use in photocatalytic reactors, since the major part of the coated channel would not be sufficiently illuminated.

Figure 6 shows the Monte-Carlo simulation of the photon absorption efficiency as a function of the monolith aspect ratio at different values of the geometric parameter β. As expected, the photon absorption efficiency increases monotonically as the aspect ratio is increased, since at higher values of AR the probability of a photon to escape from the channel is diminished. It also increases with β as the lamps' position approach the channels' mouths.

Figure 6 also shows that when AR is higher than approximately 2 to 3, the efficiency tends to reach a plateau. Above this range only small increases in photon absorption efficiency can be realized, which are in practice offset by the increase of the pressure drop through the monolith at higher aspect ratios. As a result, in the present study, the aspect ratio of 2.5 was chosen as the optimum value in further Monte-Carlo simulations. These results are in agreement with the literature.[9,15]

Monolith to lamp length (α), monolith to lamp distance (β), and monolith to lamp spacing (γ)

As α increases, the lamps become shorter compared to the monolith height. As β increases, the distance between the monolith and the lamps reduces. Figure 7 shows Monte-Carlo simulations of the uniformity of the radiation distribution at the front surface of the monolith (η) when α, β, and γ were varied. As α and β increase, the distribution of radiation on the front surface of the monolith becomes less uniform. With one lamp illumination ($\gamma = 2$), halving the lamp length reduces (η) by 20% when $\beta = 1$ and by 40% when $\beta = 5$. Indeed, the closer the lamps to the monolith, the higher the effect of the lamp geometry; consequently, the radiation distribution becomes less uniform. Conversely, the distribution of radiation becomes more uniform as γ (that is, the number of lamps) increases. However, little difference can be observed when the results for γ equal to 3 and 4 are compared. Consequently, it appears that using 3 lamps should be sufficient to achieve the most uniform distribution of radiation over the monolith front surface.

The uniformity of the radiation distribution at the front surface of the monolith is an important factor in the design of a monolith photocatalytic reactor. For instance, if 60% of radiation uniformity is desired, then in the 1 lamp configuration, the lamp needs to have the same length of the monolith ($\alpha = 1$) and be located 1 monolith length away from it ($\beta = 1$). Alternatively, the same degree of radiation uniformity could be achieved using 2 lamps with the lamps half the size as above ($\alpha = 2$) or, alternatively, of the same size ($\alpha = 1$) and located 5 times closer to the monolith ($\beta = 5$). Obviously, considerations of lamp size and specifications made available by the manufacturers and of the overall compactness of the monolith reactor can dictate the design choice.

The optimization of (ϕ) and (η) in isolation cannot be used as a lone criterion for optimization of the radiation field in the reactor. Conversely, the optimal design of a monolith photocatalytic reactor would be realized by maximizing the

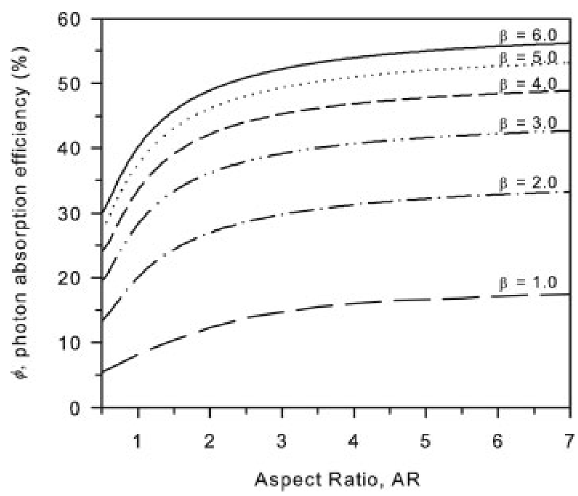

Figure 6. Photon absorption efficiency versus aspect ratio at different distances between the monolith and the lamps.

$\rho = 0.4$, $\alpha = 1.0$, $\gamma = 2$.

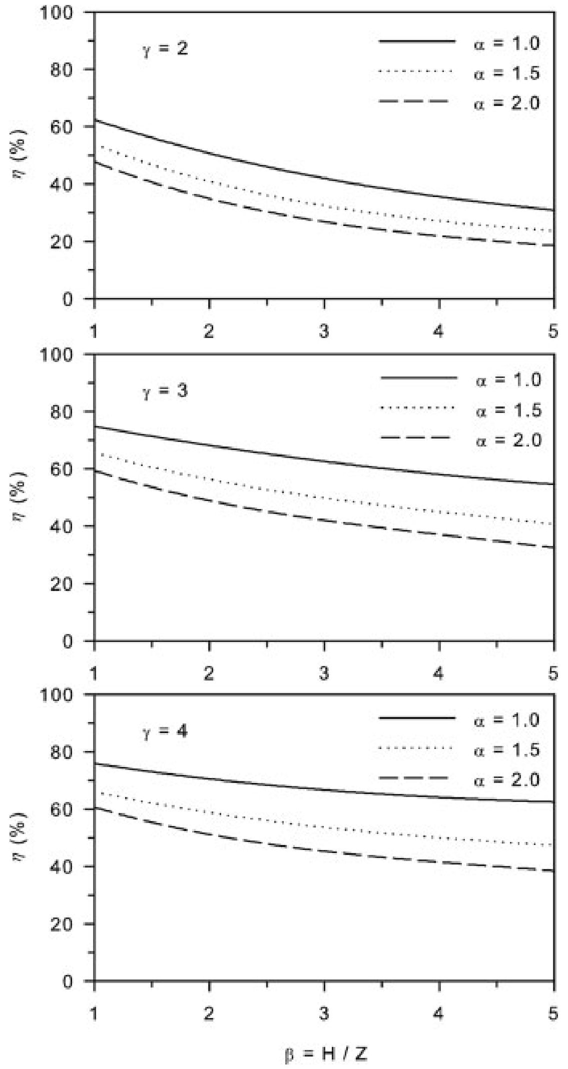

Figure 7. Influence of α and β on the uniformity of the distribution of radiation (η) for 1, 2, and 3 lamp configurations (γ = 2, γ = 3, and γ = 4, respectively).

AR = 2.5, $\rho = 0.4$.

number of photons absorbed by the monoliths and by simultaneously achieving the most uniform irradiation at the front surface of the monoliths. In this article we have introduced the concept of monolith overall photonic efficiency (Φ) to combine these two contrasting effects. Figure 8 shows the Monte-Carlo simulation of the effect of the geometric parameters α and β on the monolith overall photonic efficiency (Φ) when γ is varied from 2 to 4. For fixed α, the overall photonic efficiency initially increases as β is increased, as a result of a higher number of photons reaching the monolith surface (Figure 8a); however, as the lamps approach the surface of the monolith, the uniformity of distribution of photons on the front wall of the monolith decreases and, as a result, (Φ) decreases (Figure 8b). In all cases, a maximum in Φ is reached at specific values of α and β. When $\gamma = 2$ (that is, one lamp), the maximum overall photonic efficiency is 15.2%, which is achieved at $\alpha = 1$ and $\beta = 3.2$. When γ equals 3 or 4, higher values of (Φ) are obtained, with the maximum appearing at approximately $\alpha = 1$, and β equal to 4.2 and 6.6 respectively. The darker lines in Figures 8c and 8d correspond to the maximum (Φ); further increase in β results in lower values of (Φ). An important conclusion from the above results is that the optimal value of the design parameter α should always be found between 1 and 1.1. Further simulations showed that this is irrespective of the reflectivity of the thin-film.

Figure 9 shows the monolith overall photonic efficiency (Φ) and the photon absorption efficiency (ϕ) for different values of β and γ with $\alpha = 1$ (optimum value). Several general observations can be made. First, as γ increases, higher values of (Φ) are observed; on the contrary, (ϕ) decreases. Thus, it is clear that as the number of lamps increase, the decrease in (ϕ) is offset by a larger increase in (η). Second, it can be observed that (Φ) reaches a maximum at specific

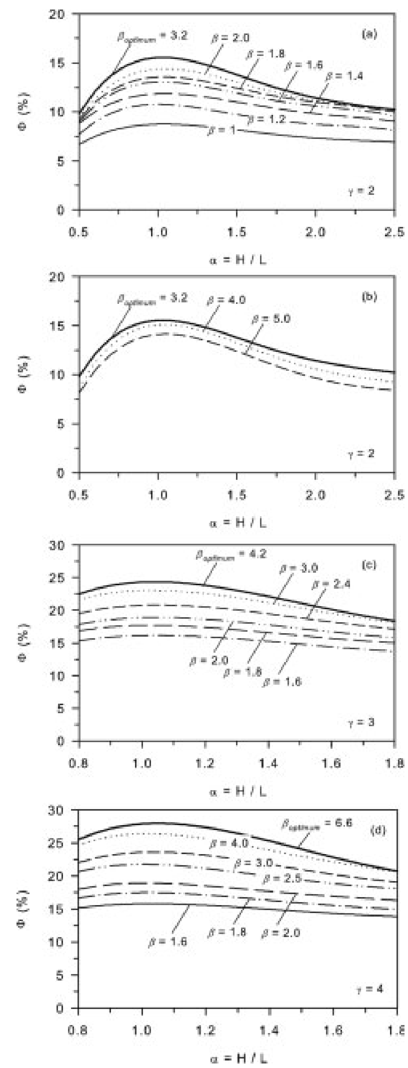

Figure 8. Monolith overall photonic efficiency (Φ) versus α at different β values, for γ = 2 (a, b), γ = 3 (c), and γ = 4 (d).

AR = 2.5, ρ = 0.4. Darker lines represent the maximum curve of (Φ).

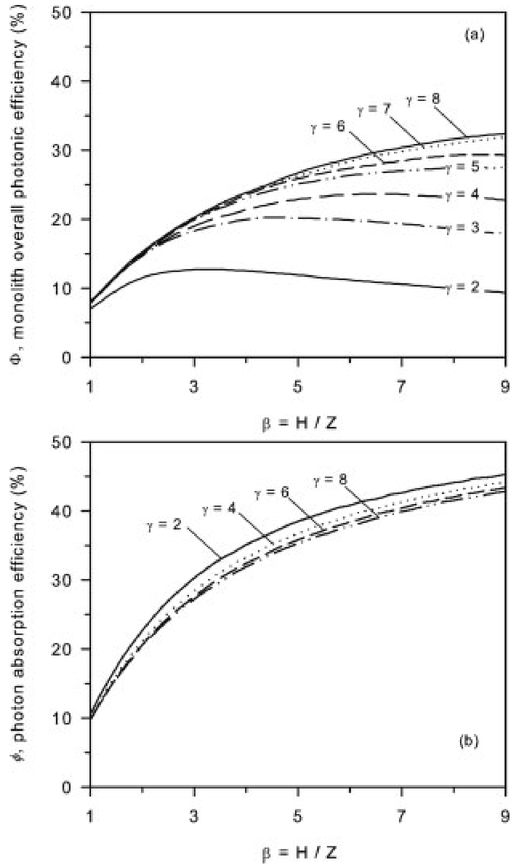

Figure 9. (a) Monolith overall photonic efficiency (Φ) versus β for different number of lamps; (b) Photon absorption efficiency (ϕ) versus β for different number of lamps.

AR = 2.5, $\rho = 0.4$, $\alpha = 1.0$.

values of β. This can be clearly identified for the case of γ equal to 2, 3, and 4. For $\gamma > 4$, the maxima are located off the figure. However, it should be observed that considering typical diameters of commercial UV lamps (say, 15 to 30 mm) and the need of distributing the fluid-flow uniformly over the monolith, large values of β (say, $\beta > 5$) can in practice be realized in monoliths of large dimensions only.

Optimal design parameters of monolith photocatalytic reactors

From the above simulation results, the optimal design parameters of monolith photocatalytic reactors can be derived, allowing the recommendation drawn from other authors[9,14,15] to be extended.

The optimum monolith aspect ratio (AR) should be approximately 2.5 and the reactor should be designed to have a monolith length approximately equal to that of the lamp ($1.0 < \alpha < 1.1$). Higher values of AR should not be used since the small increase in photonic efficiency is in practice offset by the increase of the pressure drop through the monolith. The optimum value of the design parameter β is a function of the number of lamps used, γ. Figure 10a summarizes the maximum overall photonic efficiency Φ_{max} as a function of γ and ρ. Figure 10b gives the photon absorption efficiency corresponding to the configuration yielding Φ_{max}, and Figure 10c shows the optimum value of the parameter β that results in Φ_{max}.

Figure 10a shows that Φ_{max} increases with γ and decreases with ρ, suggesting that the largest number of lamps should always be used. The optimum lamps position (β) should be determined from Figure 10c.

The reflectivity (ρ) of the catalyst plays a key role in the efficiency of the system. The efficiency decreases several fold from $\rho = 0.0$ to $\rho = 0.9$. Therefore, it is important to use photocatalytic monoliths with the lowest reflectivity. Furthermore, the gradient of Φ_{max} with the number of lamps (γ) is more pronounced at low reflectivity values, which favors

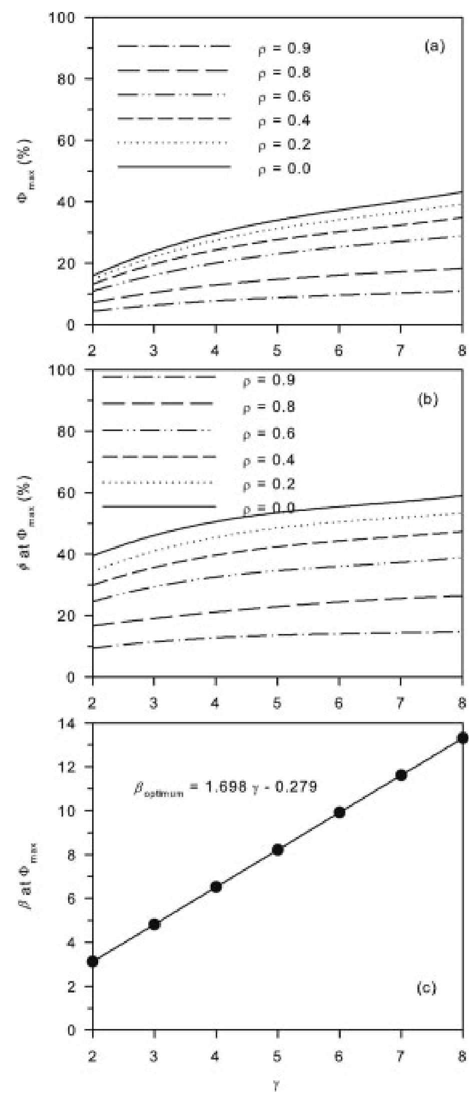

Figure 10. Optimal configuration for a circular-channeled honeycomb monolith.

(a) Maximum value of overall photonic efficiency as a function of the number of lamps γ and reflectivity of the monolith wall ρ. (b) Monolith photon absorption efficiency corresponding to the configuration yielding Φ_{max}. (c) Optimum value of β that maximizes Φ_{max} as function of γ. AR = 2.5, $\alpha = 1.0$.

the use of the largest number of lamps. Conversely, when the catalyst absorbs only 10% of the impinging light or less ($\rho = 0.9$), the increase of the number of lamps from 1 to 7 causes an increase of only 5% in the monolith overall photon efficiency (Φ).

In Figure 10c it can be observed that the lamps need to be situated closer to the monolith as their number increases. Interestingly, reflectivity does not play a role in this case. The following correlation for β is suggested for the optimal design of monolith photocatalytic reactors:

$$\beta_{\text{optimum}} = 1.698\gamma - 0.279 \tag{7}$$

Regarding the optimal number of lamps, it is interesting to note that for configurations with more than 4 lamps, the most optimal design could become impractical in small monoliths reactors, as the lamps should be located in close proximity to the monolith ($\beta > 6$), thus impeding distribution of the airflow over the monolith channels. Fluid dynamic studies would be necessary to assess the air flow behavior in the reactor. Thus, a large number of lamps could only be realized efficiently in monoliths of large dimensions.

Optimum radiation intensity

Since commercial lamps are available at fixed size and UV output, consideration of optimal photon flux over the monolith walls should be considered. The local rate of pollutant degradation in a photocatalytic surface is known to be a function of the LSRPA. The dependence has been described to be first-order on LSRPA at low irradiation intensities (reactions of electron and holes with substrates faster than recombination) and half-order at higher irradiation intensities (electron-hole recombination prevails).[18] At very high photon fluxes, the reaction rate becomes independent of photon absorption as a result of mass-transfer limitations of reactive substrates to the photocatalytic surface. Thus, optimum photon utilization and process economics would be achieved only when low irradiation intensities are used. Under this situation, the highest quantum yield (moles of pollutant reacted per Einstein of photons absorbed) would be realized.[3] The estimation of the photon flux level, which determines the departure from first-order dependence of the reaction rate from the photon flux, is not trivial and requires either experimentation or a complete model of the photocatalytic reactor.[18] It is a complex function of pollutant type and concentration, water vapor concentration, and photocatalyst spectral absorption coefficient, to name a few. Imoberdorf et al.[18] reports the criterion for determining this transition region when the kinetic parameters of VOC photocatalytic oxidation are known.

The experimental evidence of VOC oxidation on photocatalytic surfaces points towards first-order dependence of the pollutant degradation rate on the LSRPA[18–20] when UV-A blacklights were used. In a monolith channel, the photon flux decays strongly with distance, thus providing that if the photon flux at the front surface of the monolith is within the threshold limit for optimal photon utilization, then the entire reactor would be operating in the region of optimal quantum yield of pollutant degradation.

Lamp availability can quite often dictate the optimum number of lamps to be used. However, the results presented in Figure 10 should direct the designer towards selecting the optimal type of lamp.

Conclusions

In the present work, a systematic study of the efficiencies to be expected in a monolith photocatalytic reactor irradiated by cylindrical UV lamps was performed.

The effect of the dimensionless design parameters, monolith aspect ratio, geometric ratios, catalyst reflectivity, and number of UV lamps were studied and compared in terms of photon absorption efficiency (ϕ), uniformity of radiation distribution (η), and overall monolith photonic efficiency (Φ). When increasing the number of lamps, the proximity between the lamps and the monolith should also be increased in order to achieve optimal configuration. An aspect ratio of 2.5, and using lamps of approximately the same length of the monolith ($1.0 < \alpha < 1.1$), provides the highest overall photonic efficiency for all the monolith configurations. Strong dependence of the monolith overall photonic efficiency on catalyst reflectivity has also been found. Conversely, catalyst reflectivity does not seem to play a key role in defining the optimal geometry of the system.

The present article provides optimal design parameters and guidelines for the systematic design of monolith photocatalytic reactors. The results seem to suggest that there exists an optimum number of lamps that should be used, above which no improvement in performance can be expected. This depends on the reflectivity of the catalyst and the actual dimensions of the monolith and the photocatalytic reactor.

In practice, other factors related to fluid-dynamics and kinetics can come into play when designing a monolith photocatalytic reactor. The choice of the optimal number of lamps depends on further fluid-dynamics considerations mentioned earlier, especially in small reactors where fluid-flow distribution over the monolith may be obstructed by the physical dimensions of the lamps. Furthermore, kinetic limitations (such as reduced observed quantum yields as a result of mass transfer or increase in electron-hole recombination at higher photon fluxes) should be considered when selecting the lamp type and specifications.

This work contributes to a more systematic design of photocatalytic reactors for air purification since it is well documented that the operating costs of these systems are unequivocally governed by the electricity required to power the UV lamps.

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Literature Cited

1. Austin BS, Greenfield SM, Weir BR, Anderson GE, Behar JV. Modeling the indoor environment. *Environ Sci Technol*. 1992;26: 850–858.
2. EPA. Program Needs for Indoor Environments Research (PNIER). March 2005. Available at www.epa.gov/iaq/pubs/pnier.pdf.
3. Peral J, Domènech X, Ollis DF. Heterogeneous photocatalysis for purification, decontamination and deodorization of air. *J Chem Technol Biotechnol*. 1997;70:117–140.

4. Raupp GB, Alexiadis A, Hossain MD, Changrani R. First-principles modeling, scaling laws and design of structured photocatalytic oxidation reactors for air purification. *Catal Today*. 2001;69:41–49.
5. Zhao J, Yang X. Photocatalytic oxidation for indoor air purification: a literature review. *Building and Environment*. 2003;38:645–654.
6. Bendfeldt P, Hall RJ, Obee TN, Hay SO, Sangiovanni JJ. A Feasibility Study of Photocatalytic Air Purification for Commercial Passenger Airliners. UTRC. R97-1.300.9702. East Hartford: United Technologies. October 1997.
7. Toft G, Aitchison T. A breath of fresh air. *Marine*. Farnborough: Quinetiq; 2004. http://www.publicservice.co.uk/pdf/dmj/september2004/DMJ26%20Gareth%20Toft%20ATL.pdf.
8. Hodgson AT, Sullivan DP, Fisk WJ. Evaluation of Ultra-Violet Photocatalytic Oxidation (UVPCO) for Indoor Air Applications: Conversion of Volatile Organic Compounds at Low Part-Per-Billion Concentrations. Report LBLN-58936. Berkeley, CA: Lawrence Berkeley National Laboratory; 2005.
9. Hossain MM, Raupp GB. Radiation field modelling in a photocatalytic monolith reactor. *Chem Eng Sci*. 1998;53:3771–3780.
10. Sauer ML, Ollis DF. Acetone oxidation in a photocatalytic monolith reactor. *J Catal*. 1994;149:81–91.
11. Siegel R, Howell J. Thermal Radiation Heat Transfer (4th Ed.). New York:Taylor & Francis; 2002.
12. Imoberdorf GE, Irazoqui HA, Alfano OM, Cassano, AE. Scaling-up from first principles of a photocatalytic reactor for air pollution remediation. *Chem Eng Sci*. 2007;62:793–804.
13. Hossain MM, Raupp GB. Polychromatic radiation field model for a honeycomb monolith photocatalytic reactor. *Chem Eng Sci*. 1999;54:3027–3034.
14. Hossain MM, Raupp GB, Hay SO, Obee TN. Three-dimensional developing flow model for photocatalytic monolith reactors. *AIChE J*. 1999;46:1309–1321.
15. Alexiadis A. 2-D radiation field in photocatalytic channels of square, rectangular, equilateral triangular and isosceles triangular sections. *Chem Eng Sci*. 2006;61:516–525.
16. Jacob SM, Dranoff JS. Design and analysis of perfectly mixed photochemical reactors. *Chem Eng Prog Symp Ser*. 1966;89:47–63.
17. Boehman André L. Radiation heat transfer in catalytic monoliths. *AIChE J*. 1998;44:2745–2755.
18. Imoberdorf GE, Irazoqui HA, Cassano AE, Alfano OM. Photocatalytic degradation of tetrachloroethylene in gas-phase on TiO_2 films: a kinetic study. *Ind Eng Chem Res*. 2005;44:6075–6085.
19. Jacoby WA, Balke DM, Noble RD, Koval CA. Kinetics of the oxidation of trichloroethylene in air via heterogeneous photocatalysis. *J Catal*. 1995;157:87–96.
20. Raupp GB, Junio CT. Photocatalytic oxidation of oxygenated air toxics. *Appl Surf Sci*. 1993;72:321–327.

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