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Irradiance modeling in annular photoreactors using the finite-volume method

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ABSTRACT

A computational radiation field model for simulating the irradiance in single-phase annular photoreactors was developed and evaluated experimentally. The developed model included the lamp within the computational domain allowing to incorporate important interactions between the UV radiation, the quartz walls, and the Hg vapor inside the lamp. Several lamp emission models were evaluated against far- and near-field experimental data. The models with diffused radiation emission showed better overall irradiance prediction capabilities. In particular, a modification of the extensive source volumetric emission model that incorporates the high photon absorbance/re-emission effect produced by the Hg vapor in the lamp illustrated superior results. This latter model showed excellent agreement with near- and far-field experimental data indicating its suitability for integration in multi-physics models for the simulation of photoreactor performance. The advantages of this model are: it is very easy to set up; it comprises the main physical phenomena occurring in the lamp; and it allows for taking into account important lamp-sleeve interactions. Experimental results reaffirmed the importance of applying proper estimates of the lamp power output under the actual operating conditions to perform accurate simulations of radiation distribution.

1. Introduction

The use of photoreactors in water treatment applications has increased substantially over the past few years. They are commonly employed in ultraviolet (UV) disinfection and UV-based advanced oxidation processes (AOPs). A particularly emerging and promising AOP is heterogeneous photocatalysis [1–3]. Photocatalytic oxidation processes involve the use of semiconductor photocatalyst materials, predominantly titanium dioxide (TiO_2), activated by UV irradiation. One major configuration of photocatalytic reactors has the catalyst immobilized in layers to reactor inner surfaces. Since the degradation rate of contaminants in these immobilized photocatalytic reactors is directly dependent on the UV irradiance at the photocatalyst surface, the radiation field (photon distribution) within the reaction volume is one of the critical factors that determine the overall conversion and photoreactor performance. In this sense, when modeling immobilized photocatalytic reactors, the accurate prediction of the radiation field (and consequently the UV irradiance on the catalyst-coated surface) is of paramount importance.

Modeling the radiation field in a given photoreactor involves solving the radiative (photon) transfer equation (RTE) [4,5]. For monochromatic radiation, the RTE is defined as:

$$\frac{dI(\vec{r},\vec{s})}{ds} + (\kappa+\sigma)I(\vec{r},\vec{s}) = j^e(\vec{r}) + \frac{\sigma}{4\pi}\int_{4\pi} I(\vec{r},\vec{s}\,')p(\vec{s}\,'\to\vec{s})\,d\Omega' \qquad (1)$$

where I is the photon radiance, $\vec{r}$ is the position vector, $\vec{s}$ is the propagation direction vector, s is the path length, κ is the absorption coefficient, σ is the scattering coefficient, j^e is the emission (source) term, p is the phase function for the in-scattering of photons, and Ω' is the solid angle about the scattering direction vector $\vec{s}\,'$. In Eq. (1), the left-hand side terms represent the radiance change along the path length s, and the loss due to absorption and out-scattering. The right-hand side terms correspond to the radiance source and the gain in radiation radiance due to in-scattering. An analytical solution for this integro-differential equation is only possible for very simple cases [6]; for most practical engineering applications a numerical approach is necessary [7].

Several numerical methods have been developed for solving the RTE. Carvalho and Farias [7] presented a review of some of these methods. Among the proposed methods, three of them have been mostly utilized and studied in the simulation of photoreactors: the Monte Carlo (MC) method, the discrete ordinate (DO) method, and the finite-volume (FV) method [8]. In particular, the FV method has been gaining much acceptance due to very favorable characteristics. In the FV method, the RTE is integrated over both the control angle and the control volume [9], which allows for conserving radiant energy and easily incorporating the method in computational

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Nomenclature

$C_{Fe^{2+}}$	molar concentration of Fe^{2+} (mol L^{-1})
d_o	external tube outer diameter (m)
E	radiation irradiance (W m^{-2})
ESDE	extensive source superficial diffuse emission model (refer to Table 1)
ESVE	extensive source volumetric emission model (refer to Table 1)
ESVEA	modified ESVE model that accounts for photon absorption/re-emission in the lamp plasma
ESVERA	modified ESVEA model that accounts for radiation reflection, refraction and absorption at the lamp quartz envelope
ESDER	modified ESDE model that accounts for radiation reflection, refraction and absorption at the lamp quartz envelope
ESVER	modified ESVE model that accounts for radiation reflection, refraction and absorption at the lamp quartz envelope
I	photon radiance (W m^{-2} sr^{-1})
j^e	emission (source) term (W m^{-3} sr^{-1})
L	lamp arc length (m)
LSDE	line source diffuse emission model (refer to Table 1)
LSDER	modified LSDE model that accounts for radiation reflection, refraction and absorption at the lamp quartz envelope
LSSE	line source spherical emission model (refer to Table 1)
LSSS	line source spherical sources model (refer to Table 1)
MPSS	multiple point source summation model (refer to Table 1)
MSSS	multiple segment source summation model (refer to Table 1)
n	refractive index (dimensionless)
p	phase function for the in-scattering of photons (dimensionless)
P	power output of the lamp (W)
$\vec{r}$	position vector (m)
R	distance from center of lamp to the detector (m)
$\vec{s}$	propagation direction vector (m)
s	path length (m)
$\vec{s}'$	scattering direction vector (m)
t	irradiation time (s)
T	absolute temperature of the medium (K)
V_T	total volume of actinometric solution (L)
w	irradiated window width (m)
x	axial coordinate (m)
y	normal distance from the lamp center (m)

Greek letters

α	half angle subtended by the lamp at the sensor position (rad)
θ	angle from the normal axis of the lamp (rad)
κ	absorption coefficient (m^{-1})
σ	scattering coefficient (m^{-1})
σ_{S-B}	Stefan–Boltzmann constant (5.672×10^{-8} W m^{-2} K^{-4})
Φ	quantum yield of potassium ferrioxalate at 254 nm (mol $einstein^{-1}$)
Ω	solid angle about the propagation direction (sr)
Ω'	solid angle about the scattering direction vector $\vec{s}'$ (sr)

fluid dynamics (CFD) simulations. CFD is a very attractive modeling approach since it allows an integrated analysis of photoreactors through simultaneous modeling of hydrodynamics, species mass transport, chemical reaction kinetics, and photon flux distribution. Even though the FV method has been used in many CFD-based studies [8–14] few of them involved experimental evaluation of the radiation field simulations [13,14].

A key component in the development of a radiation field model is the definition of the lamp emission model (i.e., how UV radiation is emitted by the lamp). Several conceptual and mathematical models have been proposed in the literature [8,15–20]. A list with some of the lamp emission models most commonly employed in photoreactor modeling is given in Table 1. Imoberdorf et al. [15] recently proposed a more exhaustive lamp emission model that includes the reflection and refraction effects on/in the quartz envelope, so as the photon absorption/re-emission by the mercury vapor inside the lamp. The model was applied to the simulation of multi-lamp, homogeneous photoreactors, showing good agreement with the experimental data.

All the lamp emission models mentioned so far have been experimentally evaluated for lamps running in air, or inside homogeneous photoreactors [15–17,21,26–28]. Nonetheless, the radiation model evaluation close to the lamp (which is likely to be the location of the photocatalyst in the case of immobilized reactors) has been particularly challenging. Most studies utilized either a UV radiometer or a chemical actinometer for performing the irradiance/fluence rate measurements. Radiometers, however, are not reliable for measuring irradiance in close proximity to the radiation source, mainly due to the geometry and the variable response of the photometer within a nonparallel radiation field [29]. On the other hand, under high radiation fluxes, some actinometers can experience saturation of the reacting solution due to diffusional resistance effects. This phenomenon has been reported, for example, for the iodide/iodate actinometer for 254 nm radiation [17,28,30].

Another shortcoming of some evaluation studies is related to the estimation of the radiation output of the lamp under the working (experimental) conditions. In many cases, the lamp output was measured while running the lamp in air at ambient temperature. However, experiments were conducted with the lamp inside a reactor, enclosed in a quartz sleeve, and submerged in water at another temperature. Hence, the radiation outputs of the lamp during the experiments were likely different. For low-pressure mercury (Hg) lamps, fluctuations of 10 °C in the lamp temperature can produce changes in the emission efficiency as high as 50% [31–33]. In this sense, new measurement methods that address this issue have been recently proposed [34].

This investigation has focused on developing an FV-based model for predicting the irradiance inside single-phase annular photoreactors. The model is primarily intended to be applied to the CFD simulation of immobilized photocatalytic reactors. In the developed model, the UV lamp (monochromatic) is part of the computational domain. Hence, the reflection, refraction, and absorption of radiation at the air/quartz/water interface, as well as the absorption and re-emission of photons by the lamp plasma, could be considered. For defining an appropriate emission model for the lamp, several lamp emission models were evaluated against far-field (measuring position located away from the lamp) and near-field (measuring position close to the lamp) experimental data. The employed measurement techniques aimed to eliminate the previously discussed errors associated with irradiance determinations. To perform accurate evaluations, the power output of the low-pressure Hg lamp employed in the experiments was carefully measured under the operating conditions.