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Effect of photocatalyst film geometry on radiation absorption in a solar reactor, a multiscale approach

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HIGHLIGHTS

- Monte Carlo radiative transfer simulation developed for a CPC solar reactor.
- Characteristic matrix method describes in detail the optics of catalyst films.
- Multi-tubular array of glass supporting surfaces for the catalyst is simulated.
- Radiation absorption by the reactor improved by increased absorber tube radius.
- Film thickness has to be kept inversely proportional to tube radius.

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ABSTRACT

A multiscale model is presented to describe the radiation absorption field in photocatalytic reactors with supported catalyst. The characteristic matrix method is applied at the photocatalyst layer scale, and is embedded within a Monte Carlo ray tracing method, applied at the photoreactor scale. This approach allows to account for important design parameters, such as photocatalyst layer thickness, location of supporting surfaces, and incoming radiation profiles, among others. To resolve the validity of the characteristic matrix method for the description of the optical properties of the catalyst, modeled transmittance and reflectance of the supported films is compared to experimental data. This comparison is carried out for different wavelengths and film thicknesses. Afterwards, the model is applied to a solar reactor with anatase TiO_2 catalyst films supported on multiple surfaces. The reactor consists of a compound parabolic concentrator with a tubular borosilicate glass receiver. Smaller glass tubes coated with the catalyst are located inside this receiver. With the developed model, a study is conducted to analyze the reactor optical performance as a function of two important design variables: TiO_2 film thickness and radius of the absorber tubes. The results of the model indicate directions for the improvement of the current design.

1. Introduction

Photocatalysis has been much studied for the detoxification and disinfection of water and air (Malato et al., 2009; Alfano et al., 2000; Goswami, 1997), as well as for other relevant applications, like the production of hydrogen (Jing et al., 2013a,b). Such processes require the utilization of reactors devised specifically to make the best use of radiative energy. Thus, radiative transfer analysis is a very important step in their design and optimization. This analysis requires modeling a variety of optical processes, like absorption, scattering, reflection, refraction, and transmission of light. Such processes involve the interaction of radiation with different materials and surfaces, which include metallic mirrors, glass walls or windows, catalyst particles or films, water, dissolved dyes, and other radiative participating molecules. Thus, the resulting behavior may be both complex and fascinating.

The radiative behavior of different types of photocatalytic reactors has been studied by several author (Alfano et al., 2000; Goswami, 1997; Rodríguez et al., 2004; Goswami, 1995; Salgado-Tránsito et al., 2015; Orozco et al., 2012; Arancibia-Bulnes and Cuevas, 2004; Colina-Márquez et al., 2010; Cassano and Alfano,

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2000; Villafán-Vidales et al., 2006; Li Puma and Brucato, 2007; Orozco et al., 2009; Arancibia-Bulnes et al., 2009; Valadés-Pelayo et al., 2014, 2015; Cuevas et al., 2007; Ling et al., 2004; Bandala et al., 2004; Marugán et al., 2015). Considering the variety of photoreactor designs proposed in the last years, different ways of classifying them are used, based on: (1) radiation source (lamp or solar reactors), (2) catalyst disposition (suspended particles or deposited films), and (3) irradiance levels (concentrating or non concentrating solar reactors).

Notably, over the years much more modeling effort has been dedicated to reactors where the catalyst is suspended (Arancibia-Bulnes and Cuevas, 2004; Cuevas et al., 2007; Villafán-Vidales et al., 2006; Romero et al., 2003; Li Puma and Brucato, 2007; Colina-Márquez et al., 2010; Romero et al., 1997; Giménez et al., 1997; Jing et al., 2013a; Busciglio et al., 2016; Geng et al., 2016). For these systems there is a wide acceptance among the photocatalytic reaction engineering community of the usage of continuum approximations. Such approximations are based on the radiative transfer equation (RTE) or simplified models derived from it. Spatially uniform distribution of the catalyst particles are generally assumed for simplicity, but also a non-homogeneous case has been examined by Geng et al. (2016), by using the modified differential approximation (Orozco et al., 2012).

On the other hand, for supported catalyst reactors a unified approach is lacking. Here the catalyst is usually deposited as thin films (Gelover et al., 2004) on different types of surfaces. When the supporting surfaces have a complex geometry, such as fiber meshes or monoliths, or when there are several of them, these system may become very complex to model. In such a case, there may be many different geometrical parameters to consider, with the added complexity of dealing with multiple light reflections at the photocatalyst layer scale. Most efforts to solve this problem are based on empirical or semi-empirical approaches, due to the complexity of the problem.

From a semi-empirical perspective, one possible approach to model supported reactors, is to operate within the range of validity of the RTE (Imoberdorf et al., 2008; Pozzo et al., 2006; Chiovetta et al., 2001; Loddo et al., 2007; Marugán et al., 2008). The most common supporting surface for this approach are small spheres coated with a given photocatalyst. Within this range, surface reflection can be properly considered by using a scattering model derived by geometric optics considerations (Loddo et al., 2007). However, special care must be put onto the support shapes, sizes and fluidization regime. For example, Imoberdorf et al. (2008) considered 1.18 mm diameter spheres as supports. Their model fails to predict the effective transmittance when the assumption of uniform sphere distribution breaks down.

Other studies are based on geometrical optical models, which use empirical or phenomenological parameters for the optical response of the catalyst films. These parameters are obtained from fits to experimental data (Singh et al., 2007; Chong et al., 2011; Passalia et al., 2013; Esterkin et al., 2002; Imoberdorf et al., 2010). The most common support geometries are monolits, tubes (Singh et al., 2007), flat surfaces (Passalia et al., 2013), and fibers (Esterkin et al., 2002; Imoberdorf et al., 2010). For instance, the work of Singh et al. (2007) presents a model based in a simplified reflectivity (independent of incidence angle), and which considers a certain number of lamps, and uses it to analyze the optimum monolith aspect ratio. Such models yield some orientation regarding the operating conditions and some macroscopic design parameters. However, they may be limited to a particular reactor design. It is desirable to bring more detail to the description of the supported catalyst, accounting for the spectral and angular effects on the reflectivity of the film surface, and possible sub-surface reflections; the effect of changing support morphology, photocatalyst layer thickness, or film morphology.

In this article, a multiscale model is proposed for describing the radiation absorption fields on supported photocatalytic reactors. At the mesoscopic level, the description of the optical response of the supported anatase films is based on a characteristic matrix (CM) Method. This is a widely accepted and thoroughly validated method for the description of the optical properties of thin films (Macleod, 2012; Born and Wolf, 1999; Abeles, 1967), based on the solution of the electromagnetic field equations (Born and Wolf, 1999). The obtained method is valid within the photocatalyst and support layers.

The above CM description of the catalyst film is embedded within a Monte Carlo (MC) ray trace method, which is applied at the macroscopic photoreactor scale. Monte Carlo ray tracing is a well known numerical method for the solution of radiation problems (Mahan, 2002), and has been used before to describe radiation absorption fields in supported photocatalytic reactors (Singh et al., 2007; Imoberdorf et al., 2008; Loddo et al., 2007; Busciglio et al., 2016).

One of the advantages of the proposed approach, consists on having a reduced amount of parameters to determine, all of which should be independently obtained from appropriate experiments. The parameters required by the model are: (1) the catalyst complex refraction index and (2) the reactor geometry information. However, the most important advantage actually relies in the fact that this approach allows the radiation absorption fields to account for important design parameters, such as: (1) photocatalyst layer thickness, (2) location of supporting surfaces and (3) variation of the incoming radiation profiles, among others.

To illustrate the application the CM method combined with MC simulations, a reactor developed previously by Salgado-Tránsito et al. (2015) is modeled in this work. First, the use of the CM method is validated for this case by fitting experimentally determined transmittance and reflectance curves for the anatase films utilized by these authors (Jiménez González and Gelover Santiago, 2007). Afterwards, the MC simulations are used to simulate the radiative transfer in the reactor.

The considered reactor consists of a CPC solar collector with a tubular glass receiver, inside which smaller diameter tubes support the catalyst. There are several parameters that could be varied in order to optimize this system: the number and locations of the glass tubes, their diameter, and the thickness of the anatase film, among others. However, the original design was proposed based on heuristic considerations, and no detailed parametric analysis has been undertaken so far. Here we study the effect of the diameter of the supporting tubes, and the anatase film thickness, on the optical performance of the reactor.

2. Methodology

2.1. Description of the reactor

The considered reactor is based on a CPC solar reflector with a tubular receiver, as illustrated in Fig. 1. The receiver is a glass cylinder that acts as reaction volume. Inside this cylinder, contaminated water circulates in contact with the catalyst, which is fixed on glass tubes of smaller diameter than the receiver (Fig. 1a). Water is able to circulate both inside and outside these smaller tubes, which will be denominated “absorber tubes”, to avoid confusion. Thin anatase TiO_2 films, of thickness τ, are deposited in their internal and external surfaces (Fig. 1b).

The parameters of this reactor, as given by Salgado-Tránsito et al. (Salgado-Tránsito et al., 2015) are: tubular receiver outer radius, 30 mm; absorber tubes outer radius R_t, 2 mm; absorber tube wall thickness, 1 mm; and anatase film thickness τ, 800 nm. The six tubes are distributed in a pentagonal array, with one at