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Design and validation of a LED-based high intensity photocatalytic reactor for quantifying activity measurements

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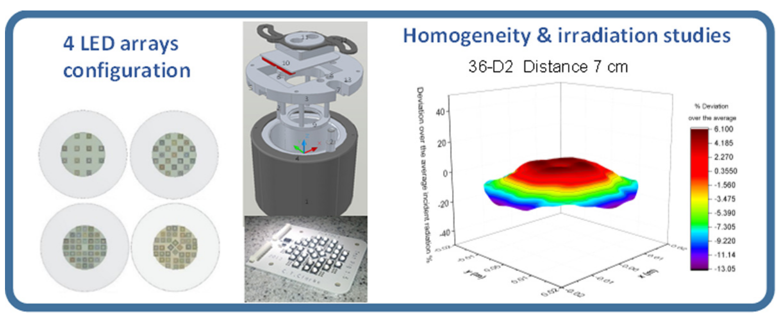
HIGHLIGHTS

- Radiation homogeneity over a catalyst surface studied for a range of LED arrays.
- Construction of an optimized LED-based photocatalytic reactor for catalyst testing.
- Novel design for assessment and characterization of photocatalytic materials.
- Radiation model successfully validated with experimental measurements.
- Determination of absorbed energy and optimal concentration for TiO_2 suspensions.

GRAPHICAL ABSTRACT



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ABSTRACT

Ultraviolet light emitting diodes (UV-LEDs) are attracting the interest of researchers for the design of compact photoreactors due to their energy efficiency, life expectancy, design flexibility, and easily tuned intensity and emission wavelength. However, due to the quasi-point source nature and viewing angle dependence of these illumination sources, the light distribution in LED based reactors can be highly inhomogeneous if the locations of the LEDs in the reactor are not carefully designed. This work describes the design of a novel standardized reactor for accurate measurements of the efficiency of photocatalytic materials under well-controlled lighting conditions. For standardized kinetic studies, it is necessary to ensure that a homogeneous radiation distribution is achieved over the catalyst surface. UV irradiation calculations involving rigorous solution of the radiative transport equation have been performed to compute the incident radiation at each point of the reactor geometry. Homogeneity calculations over the catalytic surface have been analysed for a range of LED configurations, diameter and distance of the catalyst surface with excellent agreement with measurements. We demonstrate that for many of the configurations and distances examined a poor homogeneity over the catalyst surface is obtained if the LED configuration is not carefully designed. The optimized reactor was built and predictions of the numerical model were validated against spectrophotometric measurements. The designed reactor can be also operated for the determination of the activity of photocatalytic materials in a slurry under very high radiation fluxes. The reactor model was validated with rigorous inclusion of absorption and scattering phenomena under

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highly demanding conditions of high incident radiation intensities. The developed design provides a novel route for quantitative assessment of photocatalytic materials and reactions.

1. Introduction

During the last decades, photocatalysis has been shown to be a promising technology, and its principles and potential applications have been investigated in depth. However, there remain a number of challenges in its development for water treatment applications on a high volume and high throughput scale. Mass transfer limitations, elucidation of intermediate products of degradation, low quantum efficiency or deactivation of catalyst are just some of challenges [1]. However, the optimum design and operational conditions of photoreactors remains a major concern for the development of industrial-scale photocatalytic processes [2–4].

An efficient photoreactor requires an efficient and durable light source. The recent rapid advances in light emitting diode (LED) technology, including high brightness devices emitting ultraviolet (UV) light has made possible a new paradigm for a wide range of lighting applications including photocatalysis [5,6]. The energy conversion efficiency of visible range LED has increased exponentially and they now compete with traditional light sources in most applications. Ultraviolet LED (UV-LED) technology is also following this development path and are increasing in efficiency and decreasing in price every year [7,8]. There are several advantages of UV-LEDs technology over the classical ultraviolet light irradiation sources [9]. They avoid the problem of mercury pollution of UV-A black lights and UV-C germicidal lamps, they have higher energy efficiency, are more robust and have longer life time. Although the emission power can decay with time (ca. 5% per operating year), the life expectancy of LEDs based on maintaining at least 50% of the original light output is approximately 50,000 h, which is five times longer life than Hg-vapour lamps [10]. LED heat production is low compared with traditional lamps [11], and they also offer a good linearity of the emitted light intensity with electrical current and therefore energy consumption. Moreover, in terms of reactor design, UV-LEDs offer interesting options in terms of design flexibility since they are available with monochromatic irradiation, can be used for operation in a pulsed regime at high frequencies and are easily programmable [4,12,13]. The use of UV-LEDs has therefore attracted the interest of researchers and have been used in the design of compact photoreactors for gaseous and aqueous applications [14–17], thereby suggesting that LEDs will be the future of illumination in photocatalytic reactors.

However, due to the limited view angle of these emission sources and their quasi-point source nature, the light distribution obtained in reactors illuminated by LEDs can be highly inhomogeneous if the distribution of LEDs within the reactor is not carefully designed. Levine et al. [10] reported that a lower level of photodegradation was obtained in a reactor working with LEDs in comparison with a conventional black-light fluorescent lamp, with comparable emission, attributing this behaviour to the poorer homogeneity of the LED sources. Similar conclusions have been also reported in a recent work [18] analysing the photonic efficiency of different LED configurations in comparison with a diffuse UV-A mercury fluorescent lamp of similar emission power and spectrum. The work showed that if light homogeneity is not carefully considered in reactor design the improvement in energy efficiency based on the higher electrical energy to light conversion of an LED can be counteracted by a decrease in the photonic efficiency of the photocatalytic process.

The lack of knowledge about the homogeneity of the light distribution present in a photoreactor can lead to erroneous analysis of the reaction rates obtained in photocatalytic processes. For slurry systems, the absorption profile present in a given volume is inherent to the nature of light inside the reacting media and must be known for correct interpretation of the degradation rates within the reactor. When the catalyst particles, for example TiO_2, are immobilized on a surface, the challenge in designing an efficient photocatalytic reactor becomes one of simultaneously optimizing both the area covered by photocatalytic particles and the light distribution. LEDs currently available in the market can have very high irradiance, which may lead to a reduction in the reaction time needed to reach a degradation target, however the dependence of the photocatalytic reaction rate on the absorbed energy can become non-linear at high incident optical power densities.

To analyse these factors, computational fluid dynamics (CFD) is a promising instrument for the design, optimization, and scale-up of photocatalytic systems. In previous work [19], a lab-scale annular reactor was modelled and integrated the hydrodynamics, species mass transport, light intensity distribution and reaction kinetics within the reactor with results in good agreement with experimental data. CFD capabilities have been also demonstrated by other research groups using traditional UV-light sources [20–22]. More recently, published reports have included the application of CFD tools for the evaluation of irradiance distribution with LEDs arrays sources and LED-based photoreactor designs [23–27]. Most of these reports have focused of TiO_2 supported reactors and the use of simplified models for light distribution calculations. In this work we present a model-based design and optimization of a novel photocatalytic reactor using numerical simulations based on rigorous solution of the Radiative Transport Equation (RTE) to calculate the UV incident radiation at each point of the geometry. The presented reactor has been designed for standardized measurements of immobilized photocatalyst and photocatalytic films. This application requires not only an optimum efficiency in the use of light but also a well-defined, and finely tuned light output over a wide range of intensities with an homogeneous irradiation of the complete catalyst surface being tested. The model-based optimization of the reactor will allow the determination of the optimal geometry of the reactor, catalyst support and the LED array configuration, and the predictions are successfully validated by experimental spectroradiometric measurements. In addition, the designed reactor can be also operated for the determination of the activity of photocatalytic materials in slurry under very high radiation fluxes. For this application, the reactor model has been also validated with a rigorous resolution of the absorption and scattering phenomena under highly demanding conditions of high incident radiation intensities.

2. Methods

2.1. LED board design and homogeneity tests

In order to design a LED array that homogeneously illuminates the supported catalyst, four different configurations of LEDs were considered, see Fig. 1. The selected configurations were chosen based in several constraints: i) the catalyst support will have a