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Optimization of the radiation absorption for a scaled-up photocatalytic hydrogen production system

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ABSTRACT

The effective absorption of photons by photocatalyst in the reactor is a challenge for scaled-up application of a photocatalytic hydrogen production system. The spatial distribution of radiation and photon absorption are proposed to be strongly dependent on the photocatalyst concentration and the structure of the solar collector. However, there have been few reports of the radiation absorption properties of a cylindrical reactor based on high power solar collector that is suitable for a large-scale photocatalytic hydrogen production system. In this study, the distribution of radiation in the reactor as represented by the local volumetric rate of photon absorption was determined by adopting the modified six-flux absorption scattering model based on the compound parabolic concentrator and the surface uniform concentrator. The explanations of the differences of radiation absorption properties were comprehensively analyzed by ray-tracking technique. The structure of the solar collector and the optimum catalyst concentration are recommended based on photon absorption efficiency. This work may serve as potential reference values for the optimal reactor operating parameters for large-scale application.

1. Introduction

With the growing problems of the energy crisis and environmental pollution worldwide, the development of renewable energy has become a research hotspot in recent years. Hydrogen has promising features as a competent and safe energy carrier, which expected more in the coming years (Dincer and Acar, 2015). Numerous hydrogen production technologies have been developed, and photocatalytic water splitting is an environmentally friendly method that has received a great deal of attention as a form of clean energy (Jang et al., 2012; Jafari et al., 2016).

Currently, study on photocatalytic hydrogen production has focused on the following aspects: the preparation of photocatalyst material, the photocatalytic reaction, the photoreactor, and the evaluation of photocatalytic effect. Subdivision of areas addressing this four research directions are categorized as shown in Fig. 1. The choice of photocatalyst plays an important role in the efficiency of photocatalytic hydrogen production. To take advantage of solar energy more efficiently, it is essential to expand the spectral response range of the photocatalyst (Acar et al., 2016). The activity and stability of some photocatalysts were analyzed and compared (Serpone et al., 2016). The deactivation and regeneration of the photocatalyst were very important predictors of lifetime, and these parameters were found to be strongly dependent on the concentration of surface absorbent in several experimental studies (Zhou et al., 2010; Huang et al., 2011). The kinetic models or empirical modifications for photocatalytic degradation were summarized and categorized (Spasiano et al., 2015). However, the photocatalytic reaction rates in most of the researches were described for the particular conditions based on the laboratory experiments under simulated light source (Hisatomi et al., 2012; Vezzoli et al., 2011; Sopyan, 2007; Mills et al., 2006; Li et al., 2006; Brandi et al., 2002; Malini et al., 2016; Nomikos et al., 2014; Jallouli et al., 2016; Peng et al., 2017). The kinetic investigation and numerical modeling of the photocatalytic reaction rate for large scale system under sunlight yet a challenge to date. An overview of the design and application of photoreactors and devices was presented (Gerven et al., 2007). Quantum yield and hydrogen production rate are two main performance assessments of a photocatalytic system (Acar et al., 2016). In heterogeneous system, its evaluation is more complex due to the simultaneous existence of radiation absorption and scattering (Brandi et al., 2002). In addition to the activity of catalytic, the effective photon collecting of the reactor is also very necessary (Baniasadi et al., 2014; Le et al., 2015). So the energy conversion efficiency is one of the most important indicators in a scaled-up system (Feng et al., 2008; Hisatomi et al., 2015).

Photocatalysis technology has gradually shifted from experimental study to practical application in recent years. Although water splitting with sunlight has been extensively studied, practical application of this

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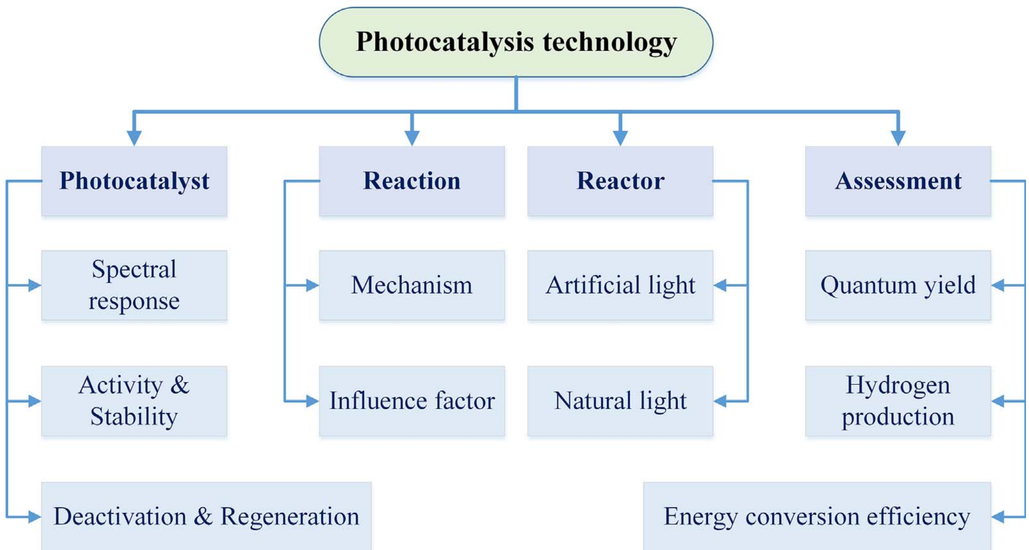

Fig. 1. Research field of photocatalysis technology.

technology remains a great challenge (Gerven et al., 2007). There are two major technical issues affecting photocatalysis technology application. The first is determining the best strategy to improve the activity and stability of photocatalysts under natural light conditions. The second is the need to design a photoreactor with simple structure, stable performance, and high efficiency (Yang et al., 2016). A cylindrical reactor combined with a compound parabolic concentrator (*CPC*) is regarded as a promising choice for large-scale applications of photocatalytic technology (Ren et al., 2016). The geometry of *CPC* has been used extensively in many pilot and full-scale studies (Blanco et al., 1999; Malato et al., 2003; Colina-Márquez et al., 2010; Polo-López et al., 2011; Alrousan et al., 2012). The spatial distribution of the radiation field in the reactor and photon absorption are key aspects in determining the success of heterogeneous photocatalytic processes (Cassano and Alfano, 2000; Zalazar et al., 2005b; Imoberdorf et al., 2007; Brandi et al., 2003). Of course, the geometric structure of the solar collector can change the direction of the light, so the design of the photoreactor may provide an approach to enhance the photon absorption of catalyst in the reactor. Therefore, photoreactor research has important theoretical and practical significance to industrial application.

Optical absorption of photon is the initial step of the photocatalytic reaction. Heterogeneous photocatalytic reaction describes the interaction of radiation absorption, transmission and reaction kinetics. The concentration of the photocatalyst has a significant impact on the distribution of radiation in the reactor because absorption, scattering, and the attenuation of photons will occur by multiphase medium. For improved application of the photocatalytic process, optimization of the radiation absorption in the reactor is an important step (Valadés-Pelayo et al., 2015).

For this reason, there have been studies of the local volumetric rate of photon absorption (*LVRPA*), one of the most important parameters in the photocatalytic reaction kinetics model (Valadés-Pelayo et al., 2015; Alfano et al., 2000). In principle, the *LVRPA* can be obtained by solving the radiative transfer equation (*RTE*). The *RTE* describes radiative transfer, the physical phenomenon of energy transfer as electromagnetic radiation. The propagation of radiation through a medium is determined by absorption, emission, and scattering processes. The exact solution of the *RTE* is typically obtained by the discrete ordinate method (*DOM*) (Cassano and Alfano, 2000). This method has been applied to a plate flat photocatalytic reactor (Cuevas and Arancibia-Bulnes, 2007), cylindrical photoreactor (Cassano and Alfano, 2000; Imoberdorf et al., 2007; Marugán et al., 2008), and parabolic concentrator reactor (Zalazar et al., 2005a). Although the radiation distribution in a heterogeneous photocatalytic system can be predicted precisely, such a high precision calculation requires significant computing time cost (Zalazar et al., 2005b; Imoberdorf et al., 2007). Additionally, this approach has limited accuracy of boundary conditions (Zalazar et al., 2005a). Other approaches used to describe the distribution of radiation in a heterogeneous photocatalytic reactor are based on the Monte Carlo method (Petrasch et al., 2011) and computational fluid dynamics (Wang et al., 2012; Jović et al., 2012; Passalía et al., 2011; Trujillo et al., 2010; Boyjoo et al., 2013). These models are as accurate as the *DOM* solution, but have not been applied to the solar scaled-up system due to the high computational time and mathematical cost. For solar photocatalytic reactor, the six-flux scattering absorption model (*SFM*) has been implemented to describe the radiation distribution and the rate of photon absorption (Colina-Márquez et al., 2010; Puma et al., 2007; Toepfer et al., 2006). This method was first proposed as a modification of the two-flue model (Brucato and Rizzuti, 1997a,b). A diffuse function was used to describe the scattered photon in the *SFM*, and the scattered photons were considered to exhibit dispersion probability following the route of the six directions in the Cartesian coordinate. This mathematical structure provides a simple and accurate method to estimate the *LVRPA* (Brucato et al., 2006).

In 2009, the first pilot demonstration of a concentrator-based, photocatalytic hydrogen production system under direct solar light was established (Jing et al., 2009). Next, the materials for photocatalysts, the catalyst-liquid slurry flow in the reactor, and the solar collector were studied, and the results indicated that the *CPC* was an efficient and necessary solar collector for the photocatalytic hydrogen system (Ren et al., 2016; Jing et al., 2010). Nevertheless, the design of photoreactor is much more complicated than that of a traditional chemical reactor. In addition to mass transport, the flow kinematics and the distribution of catalyst, the optical radiation must also be considered. *CPC* is a line-focusing solar collector, so a surface uniform concentrator (*SUC*) was next designed and tested to investigate whether the uniformity affected hydrogen production compared with the *CPC* (Yang et al., 2016). However, this work examined only surface phenomena, so that the effect of photon absorption and the influence of photocatalyst concentration were not considered. In this study, *SFM* was modified by considering the distribution of the catalyst concentration in the reactor which was obtained via *CFD*. And the spatial distributions of *LVRPA* in a cylindrical reactor were obtained by modified *SFM*, allowing