

Modelling the influence of mass transfer on fixed-bed photocatalytic membrane reactors



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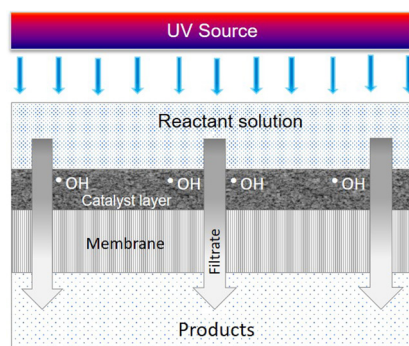
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HIGHLIGHTS

- A novel fixed-bed photocatalytic membrane reactor (FPMR) is presented.
- A quantitative model for predicting reactor performance is proposed.
- The model is experimentally verified regarding reaction kinetics and mass transfer.
- The FPMR ensures high overall mass transfer coefficient and reaction rate constant.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 23 March 2017

Received in revised form 1 July 2017

Accepted 27 July 2017

Available online 29 July 2017

Keywords:

Fixed-bed reactor

Immobilised photocatalyst

Photocatalytic membrane reactor

Microreactor

Mass transfer

Surface reaction rate constant

ABSTRACT

Photocatalytic membrane reactors have been recently considered as promising reactor types for photodegradation of organic compounds. In this work, a novel reactor concept named fixed-bed photocatalytic membrane reactor (FPMR), which relies on dead-end microfiltration of the catalyst particles, is introduced and studied. A quantitative model for evaluating the influence of mass transfer rate and intrinsic reaction rate on the overall photocatalytic degradation rate is developed and experimentally validated. The results show that the mass transfer rate contributes significantly to the overall reaction rate constant of the FPMR. They further reveal that the overall mass transfer coefficient and overall reaction rate constant of the reactor are greater than 4 s^{-1} . Those remarkably high rates are comparable to those of new photocatalytic microreactors which are two to three orders of magnitude higher than traditional photocatalytic reactors. Hence, the new reactor concept forms a powerful approach for designing photocatalytic microreactors.

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1. Introduction

Photocatalysis has been considered as a potential method for wastewater treatment. It is an advanced oxidation process which employs photocatalytic materials like TiO_2 , ZnO , CdS , etc. and illumination of suitable light energy. During the process, photocat-

alytic particles will generate strong oxidative radicals. These species are able to completely degrade toxic organic compounds, bacteria or virus (Pichat, 2013). Although there has been a huge amount of research work relating to photocatalysis, its practical applications are still limited. The main obstacle of applying photocatalysis to large scale processes is related to the efficiency of reactor design. Based on the state of catalytic particle, two types of reactors can be distinguished: reactors with suspended catalyst particles (slurry reactors) and reactors with catalyst fixed on some

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Nomenclature

A_c	area of cake layer (m^2)	S_p	specific surface area of catalyst particle (m^2/kg)
a_i	interfacial area per volume of liquid phase in FPMR (m^2/m^3)	t_M	residence time in the mixing tank (s)
C	concentration of reactant in the bulk phase (mol/m^3)	\dot{V}	flow rate (m^3/s)
C_s	concentration of reactant in the interfacial region (mol/m^3)	v_F	superficial velocity (m/s)
$c_{V,S}, c_{V,F}$	volume fraction of solid in suspension and in filtrate, respectively	x_{cum}	intensity weighted harmonic mean of the aggregate size distribution
D	diffusion coefficient (m^2/s)	z	position in the cake layer (m)
d_p	particle diameter (m)	<i>Dimensionless parameters</i>	
I	UV irradiance (W/m^2)	Re	Reynold number
I_0	UV irradiance at the top side of catalyst layer (W/m^2)	Sc	Schmidt number
k	reaction rate constant ($mol/(m^3 s)$)	Sh	Sherwood number
K_{ad}	adsorption equilibrium constant (m^3/mol)	<i>Greek letters</i>	
K_{app}	pseudo first order apparent reaction rate constant (1/s)	α	exponent reflecting influence of UV irradiance
$K_{i,0}$	combined reaction rate constant of the top layer of FPMR (1/s)	β	light absorption coefficient (1/m)
k_m	mass transfer coefficient (m/s)	ε	mean porosity of cake layer
k_{res}	intrinsic reaction rate constant per unit of cross-sectional area of the catalyst layer (m/s)	η	dynamic viscosity of fluid (Pa·s)
K_{res}	overall reaction rate constant of FPMR (1/s)	κ	light-independent reaction rate coefficient
k_s	surface reaction rate constant of the catalyst layer (m/s)	ν	kinetic viscosity (m^2/s)
L	cake thickness (m)	ρ	density of fluid (kg/m^3)
$p (\Delta p)$	pressure loss (Pa)	ρ_p	density of catalyst particle (kg/m^3)
R_M	membrane resistance (m^{-1})	τ	residence time in the loop system without mixing tank (s)
r_c	specific cake resistance (m^{-2})	φ_S	a constant of cake layer formation

materials like glass beads, ceramic foam, glass, etc. The former one offers higher activity than the latter because the illuminated surface area is typically larger. However, its major drawback is the need for an additional process step for recovering the fine catalyst particles from treated water. Coupling photocatalysis with membrane processes has been proposed as a powerful tool to cope with this (Leong et al., 2014; Molinari et al., 2017; Mozia, 2010).

Generally, the combined photocatalysis and membrane system is called *photocatalytic membrane reactor* (PMR) (Leong et al., 2014; Molinari et al., 2017; Mozia, 2010). In the initial concept, a slurry photocatalytic reactor is connected with a membrane process, which recovers the nanosized photocatalytic particles. In this configuration, photocatalytic degradation and catalyst recovering are spatially separated. It benefits from the high photocatalytic activity of slurry reactors and can be operated continuously. However, its main drawback is cake layer formation that causes a significant decrease in permeate flux. Another approach of coupling photocatalysis and membrane processes is to use photocatalytic membranes. Such membranes contain photocatalytic particles on their surface or in their structure or are made by photocatalytic materials (Aran et al., 2011; Leong et al., 2014; Molinari et al., 2004). This kind of concept has attractive advantages such as anti-fouling behaviour and compact design. Its major drawbacks are low photocatalytic activity, low stability of photocatalytic activity, and mass transfer limitation (Aran et al., 2011; Molinari et al., 2004; Zhang et al., 2014).

This study introduces a new concept for photocatalytic membrane reactors, in which a uniform pseudo-fixed catalytic layer is created by the deposition of photocatalyst particles on a membrane surface by dead-end filtration prior to photocatalysis. The corresponding catalytic coating is not really fixed, which allows an easy renewal of the photocatalytic layer when the catalytic activity decreases. Moreover, the new *fixed-bed photocatalytic membrane reactor* (FPMR) facilitates the controlled formation of a

photocatalytic coating thus the defined variation of photocatalytic activity. Besides, this reactor also offers the advantages of high surface-to-volume ratio, high mass transfer rate, simplicity and safe operation. Regarding practical aspects, it does not need to work at high pressure nor does it require aeration. Moreover, it does not require a post-separation of catalyst particles from product flow.

In order to assess the photocatalytic effectiveness of the new reactor, one can refer to several common benchmark parameters like reaction rate constant, quantum yield, and photocatalytic space-time yield (Brandi et al., 2003; Lelebici et al., 2015). The technically most important parameter is the reaction rate, which reflects both, reactor efficiency and photocatalytic activity. In publications, the *apparent reaction rate constant* K_{app} calculated by employing Langmuir-Hinshelwood kinetic is used as the reaction rate constant. Nevertheless, the apparent rate can disguise the intrinsic reaction rate and mass transfer rate (Ollis, 2005).

Mass transfer has been considered as a key influential factor in an immobilised catalytic reactor. More specifically, enhancing mass transfer is one of the main strategies for designing an effective photocatalytic reactor (Ochiai and Fujishima, 2012; Ollis, 2005; Turchi and Ollis, 1988). Some authors have figured out the influence of mass transfer in immobilised catalytic reactors. However, most of the works were carried out on photocatalytic coating layers, in which reactant flow was parallel to coating layer (Chen et al., 2000). Herz (2004) has developed kinetic models for three configurations of photocatalytic layer reactors. His results showed that the photocatalytic activity was increased when the reactant moved through the catalytic layer instead of flowing parallel to its surface. Nonetheless, the mass transfer coefficient was not involved in his models.

In the FPMR, the hydrodynamic parameters, mass transfer, effective interfacial area, etc. play a crucial role. These factors, in turn, are dependent on flow regime and bed characteristics

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