



Comprehensive multiphysics modeling of photocatalytic processes by computational fluid dynamics based on intrinsic kinetic parameters determined in a differential photoreactor



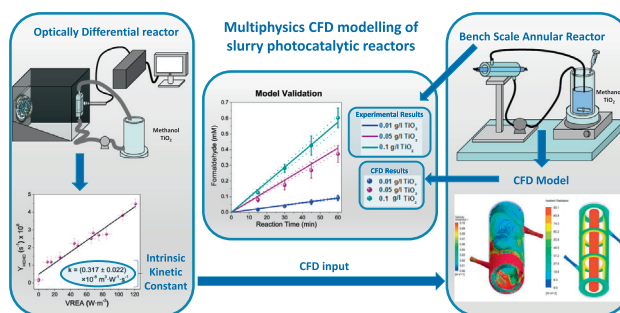
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HIGHLIGHTS

- Intrinsic kinetic parameters determined in an optically differential photoreactor.
- Predictive multiphysics CFD simulation of an annular photocatalytic reactor.
- Good correlation between experimental and simulation results for velocity field.
- Optimal TiO_2 concentration calculated by radiation absorption simulation.
- Successful experimental validation of CFD conversion predictions with error <10%.

GRAPHICAL ABSTRACT



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ABSTRACT

This work describes the procedure for the simulation of the operation of a photocatalytic reactor by using a multiphysics computational fluid dynamics (CFD) model based on the determination of the intrinsic kinetics parameters in an optically differential photoreactor. The model includes the rigorous description of the hydrodynamics, radiation transfer, mass transport and chemical reaction rate based on a mechanistic kinetic model. Possible existence of dead and recirculation zones has been identified from the flow field, showing a non-uniform flow through the reactor domain. The theoretical laminar profile is not reached due to the short length of the annular core and the departure from the ideal models has been quantified. The predicted velocity field has been experimentally validated with good agreement by injecting a tracer. The radiation field was simulated for slurry TiO_2 suspensions with concentrations between 0.005 and 5 $\text{g}\cdot\text{L}^{-1}$, showing an optimum catalyst loading around 0.1–0.2 $\text{g}\cdot\text{L}^{-1}$. Above this value, the increase in the absorption of radiation is negligible, whereas a more non-uniform radiation profile develops, keeping the most external regions of the reactor in the dark. The results of photocatalytic activity, using methanol oxidation as test reaction, showed good agreement between model predictions and experimental data, with errors between 2% and 10% depending on the catalyst concentration. The successful validation confirms not only the scientific background of the model, but also supports its applicability for engineering purposes in the design and optimization of large scale photocatalytic reactor to overcome some of limitations hindering the industrial development of this technology.

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Nomenclature

ALSPR	annular laboratory scale photoreactor	α_2	kinetic parameter ($\text{m}^3 \text{W}^{-1}$)
a_v	catalyst surface area per unit volume (m^{-1})	α_3	kinetic parameter (dimensionless)
C_{cat}	catalyst loading ($\text{g} \cdot \text{m}^{-3}$)	β	volumetric extinction coefficient (m^{-1})
CFD	computational fluid dynamics	κ	volumetric absorption coefficient (m^{-1})
$D_{i,m}$	diffusion coefficient of species i in the mixture ($\text{m}^2 \cdot \text{s}^{-1}$)	λ	wavelength (nm)
DOM	discrete ordinate method	ρ	density ($\text{kg} \cdot \text{m}^{-3}$)
\vec{g}	gravitational acceleration ($\text{m} \cdot \text{s}^{-2}$)	σ	volumetric scattering coefficient (m^{-1})
g	asymmetry factor of the Henyey-Greentein's phase function (dimensionless)	τ	optical thickness (dimensionless)
I	radiation intensity (Einstein $\text{m}^{-2} \text{s}^{-1} \text{sr}^{-1}$)	$\bar{\tau}$	stress tensor ($\text{N} \cdot \text{m}^{-2}$)
J_i	diffusive flux species i ($\text{kg} \cdot \text{m}^{-2} \text{s}^{-1}$)	Φ	wavelength averaged primary quantum yield (mol Einstein^{-1})
k	kinetic constant ($\text{m}^3 \text{kmol}^{-1} \text{s}^{-1}$)	Ω	solid angle of radiation propagation about the direction $\underline{\Omega}$ (sr)
p	Henyey-Greentein scattering phase function (dimensionless)	$\underline{\Omega}$	unit vector in the direction of radiation propagation
P	pressure ($\text{N} \cdot \text{m}^{-2}$)		
ODPR	optically differential photoreactor		
R_i	mass rate of production/depletion of species i ($\text{kg} \cdot \text{m}^{-2} \text{s}^{-1}$)	<i>Subscripts</i>	
r_i	rate of production/depletion of species i ($\text{kmol} \cdot \text{m}^{-3} \text{s}^{-1}$)	HCHO	relative to formaldehyde
RT	residence time (s)	i	relative to species i
RTD	residence time distribution	i_{out}	relative to species i at the reactor inlet
RTE	radiative transfer equation	i_{in}	relative to species i at the reactor inlet
S_g	TiO ₂ specific surface area of the catalyst ($\text{m}^2 \text{kg}^{-1}$)	Tank	relative to reservoir tank
ST	space time (s^{-1})	V_R	relative to reactor volume
t	time	λ	indicates dependence on wavelength
\vec{v}	velocity vector ($\text{m} \cdot \text{s}^{-1}$)	$\underline{\Omega}$	indicates a directional dependence
V_R	reactor volume (m^3)		
V_T	total liquid volume (m^3)	<i>Superscripts</i>	
VREA, e^a	volumetric rate of energy absorption ($\text{W} \cdot \text{m}^{-3}$)	s	relative to the reaction rate per unit surface area
Y_i	mass fraction of species i in the mixture (dimensionless)		
		<i>Special symbols</i>	
<i>Greek letters</i>		-	indicates a vectorial magnitude
α	kinetic parameter ($\text{kmol} \cdot \text{W}^{-1} \text{s}^{-1}$)	$\langle \rangle$	indicates average value
α_1	kinetic parameter ($\text{kmol} \cdot \text{m}^{-3} \text{s}^{-1}$)		

1. Introduction

Despite the many advantages of photocatalysis for water purification [1,2] and the extensive laboratory research done in this field, including 13,500 papers, reviews and reference work over the last 38 years [3], photocatalytic technologies for water remediation are not fully industrially developed yet. Tremendous efforts have been devoted to improve photocatalytic efficiency of TiO₂ by doping with metals or non-metals or to the development on new photocatalytic materials. Materials improvement has been accompanied with extensive research to identify reaction mechanisms and obtain appropriate kinetic models, but there are several challenges preventing the development of this technology to the commercial scale [1]: mass transfer limitations, catalyst deactivation, generation of intermediate products and by-products and especially low quantum efficiency requiring materials, reactor and light source optimization [2,4,5]. For this purpose, computational fluid dynamics has been shown to be a very promising tool in the design, optimization, and scaling-up of photocatalytic systems for different applications [4–7], saving time, costs and efforts.

Annular reactors have been widely investigated in CFD modeling, as they are the most popular slurry reactors because of the advantageous basic features of this geometry [7,8]. Previous investigations have reported modeling related to hydrodynamics and mass transfer [8–12], irradiance [13,14], and chemical reactions [15–17]. Some studies have also reported the scaling-up of processes to bench scale based on the kinetic constants determined

under controlled conditions at lab scale [18,19]. Validation of model predictions with experimental data has been also reported. Passalía et al. [18] determined kinetic parameters experimentally in a lab scale flat plate TiO₂ coated reactor (25.6 cm³), under kinetic control regime, and used them in the model of a bench scale corrugated plate type coated with TiO₂ as catalyst (1800 cm³) in the gas phase (HCHO as model for indoor pollution control). Also, Elyasi et al. [19] applied the photoreaction rate of the homogeneous UV/H₂O₂ degradation of rhodamine WT measured in a bench-scale photoreactor under controlled conditions for the model of a pilot scale photoreactor. Similar methodological approaches have also been reported for immobilized TiO₂ [1,15,20].

Rigorous kinetic description of the photocatalytic process requires the explicit inclusion in the model of the photon absorption rate. However, due to the intrinsic nature of photoactivated processes, it is not possible to achieve a uniform light intensity along the whole reactor volume. Therefore, an appropriate knowledge of the irradiance distribution inside the reactor is required through the rigorous resolution of the Radiative Transfer Equation (RTE). Resolution of the RTE in homogeneous systems such as UV/H₂O₂ reactors is relatively simple, as scattering of radiation can be neglected and only absorption should be considered. On the other hand, calculation of radiant fluxes in immobilized TiO₂ surfaces is also relatively easy to accomplish, as the gas or liquid filling the reactor can be considered usually as non-absorbing media, concentrating radiation absorption on the thin TiO₂ layer. In contrast, modeling of the radiation transport in heterogeneous media such

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