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# Numerical investigation on photocatalytic CO<sub>2</sub> reduction by solar energy in double-skin sheet reactor



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#### ABSTRACT

Double-skin sheet reactor (DSSR) attracts attention in recent years due to the simple design and high mass transfer, but its application to photocatalytic CO<sub>2</sub> reduction with immobilized catalyst is still a new trial. In this paper, the three-dimensional models describing the photocatalytic CO<sub>2</sub> reduction in DSSR by means of solar energy were developed and numerically simulated, on the basis of the transient and continuous solar light distributions. A performance evaluation approach was proposed to guide the structural optimization of DSSR in photocatalytic CO<sub>2</sub> reduction. The effects of operation parameters and reactor structures on CH<sub>3</sub>OH concentration were analyzed and discussed. The results show that the outlet CH<sub>3</sub>OH concentration of  $2.68 \times 10^{-4}$  mol m<sup>-3</sup> in DSSR is much greater than that of  $1.77 \times 10^{-5}$  mol m<sup>-3</sup> in optical fiber monolith reactor under the same conditions. It increases as both the inlet water vapor concentration ratio and day-average light flux increase, but decreases with increasing the inlet velocity, all of which are beneficial to the CH<sub>3</sub>OH productivity. The CH<sub>3</sub>OH concentration will decrease with the increase of the number of parallel-flow channels, but increase thanks to the rise in the width–height ratio and inserting flow guiding pieces, which are recommended for higher CH<sub>3</sub>OH concentration in the design of DSSR.

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#### 1. Introduction

With fossil fuels depleting, greenhouse gases are generated, especially carbon dioxide (CO<sub>2</sub>), the main cause of greenhouse effect [1,2]. Except for selecting alternative renewable energy sources, CO<sub>2</sub> removal is equally important for relieving the greenhouse effect, which attracts wide attentions and expected more in the coming years [3]. Among various methods, photocatalytic CO<sub>2</sub> reduction combined with solar energy acts an important role in CO<sub>2</sub> removal process [4,5]. The method converts CO<sub>2</sub> into chemical fuels and has little negative impacts on environment by solar energy [6]. Though the conversion rate from CO<sub>2</sub> to fuel is relatively low [7], it is still a promising greenhouse-gas control approach for being environmentally friendly [8]. As the CO<sub>2</sub> reduction reaction carrier, photoreactors have been widely studied for improving conversion rate [9,10].

Unlike traditional chemical reactors, it is of critical importance for photoreactors to ensure that photons are collected effectively by careful physical geometry designs. A wide variety of photocatalytic reactors have been investigated so far and summarized by Tahir and McCullagh [11,12], among which the double-skin sheet reactor (DSSR) shows considerable and attractive advantages in photocatalytic performance, photon efficiency and geometric structure. DSSR was originally designed and demonstrated by Van Well et al. [13] in 1997, which was used for heterogeneous photocatalytic degradation of organic compounds. DSSR was made up of long, convoluted back and forth channels on a flat plane through which the reactant fluids and suspended photocatalyst flow. Due to transparent plexiglas used to construct DSSR, both the diffuse and direct sunlight can be used in DSSR, which is an advantage over the parabolic trough reactor. High turbulence with good mixing of catalyst and reactants is another advantage compared with the thin film fixed bed reactor (TFFBR). Although DSSR has a simple design and low investment costs, it has been proved to be highly effective in wastewater treatment. According to Dillert [14], DSSR performed at a similar efficiency as a CPC photoreactor in the degradation experiment of dichloroacetic acid, setting the two reactors at several working conditions with the ratio of lumped kinetic parameters ranging from 0.58 to 1.06 ( $k_{CPC}/k_{DSSR}$ ). Arslan et al. [15] conducted a comparison between DSSR and TFFBR, discovering that higher treatment efficiencies in terms of all investigated environmental

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#### Nomenclature

Α	area, m <sup>2</sup>	Greek le	reek letters	
С	concentration, mol m <sup>-3</sup>	3	local attenuation coefficient of catalyst film, nm <sup>-1</sup>	
D	diffusion coefficient, $m^2 s^{-1}$	η	reaction effectiveness	
Da	Damkohler number	μ	molecular weight of gas, g mol $^{-1}$	
h	convective mass transfer rate, m $s^{-1}$	ρ	density, kg m <sup><math>-3</math></sup>	
Ι	light intensity, W m <sup>-2</sup>	$\sigma$	error, mrad	
k	kinetic rate constant, m <sup>4</sup> s <sup>-1</sup> mol <sup>-2</sup>	$\phi$	quantum efficiency	
Κ	adsorption equilibrium constant, m s $^{-1}$			
Μ	molecular weight, kg kmol <sup>-1</sup>	Subscript and superscript		
NTUm	number of the mass transfer unit	a	average	
р	pressure, Pa	active	active sites	
r	reaction rate, mol $m^{-3} s^{-1}$	Α	species	
R	local apparent reaction rate coefficient, m s $^{-1}$	В	species	
Re	Reynolds number	b	bottom	
Sc	Schmidt number	с	cross-sectional area	
Sd	ratio of Sh and Da	e	east	
Sh	Sherwood number	i	inlet	
$St_m$	Stanton number of mass transfer	m	mass	
Т	temperature, K	0	outlet	
t	time, s	S	surface	
и	velocity, m s <sup>-1</sup>	w	west	
V	molar volume, cm <sup>3</sup> mol <sup>-1</sup>	*	ratio of two areas	
Ζ	axial position, m			

parameters were obtained for the suspended, heterogeneous photocatalytic DSSR, even though DSSR has a considerably lower photonic efficiency compared with TFFBR.

Though the emphasis was mainly placed on the photodegradation of organic compounds in aqueous medium by using  $TiO_2$ photocatalyst in suspension in the former studies of DSSR on wastewater treatment, the catalyst in the immobilized form was used in the simulated experiments, avoiding the filtration and separation of catalyst.  $TiO_2$  adheres strongly to a glass surface which has been explored in the degradation of contaminants [16]. In addition, when it applies to the gas reaction, immobilized form of catalyst is also a fine option.

Since DSSR has numerous advantages in photocatalytic water treatment, its application to photocatalytic  $CO_2$  reduction is expected to be attractive and promising despite the fact that few investigations were made. This work aims at a performance evaluating method of gaseous photocatalytic reaction in DSSR, based on which further performance improvement is also studied, including the structural design, flow field optimization and photocatalyst selection. Moreover, the average solar flux distribution on the reaction surface is obtained by SOLTRACE, and the effects of operating conditions such as the water vapor concentration, inflow velocity, solar flux distribution and reactor structures on the conversion from  $CO_2$  to  $CH_3OH$  are also investigated.

#### 2. Physical and mathematical models

Because all flow channels can be assumed identical in the experiments, a single unit of DSSR will be studied as shown in Fig. 1, including two channels. Each flow channel is cuboid, whose structure parameters are the length 1400 mm, the width 28 mm and the height 12 mm. The thickness of the thin plate is 4 mm. The overall internal volume including 30 channels is about 14.2 L. The species flow into the channel from the inlet, react on the inner surfaces under illumination, and flow out the unit from the outlet.

Except the above structural parameters, catalyst is also a key issue in the reaction. This study selected the catalyst  $TiO_2$  with

1% NiO/InTaO<sub>4</sub>, which was prepared by sol-gel method. The best NiO/InTaO<sub>4</sub> was calcined at 1100 °C [17], which was coated on the three inner surfaces. Comparing with pure TiO<sub>2</sub>, TiO<sub>2</sub> with noble metals or metal oxides shows a higher catalytic activity in a solid gas interface [18,19].

The commercial software COMSOL is used to investigate the photocatalytic  $CO_2$  reduction in DSSR, whose biggest advantage is coupling multiphysics. The three computational submodels, laminar flow, transport of diluted species and reaction engineering, are involved in the DSSR. Except the primary assumption that all channels are identical, the following assumptions are also made:

- (1) Steady state laminar flow of incompressible Newtonian fluid with constant physical properties.
- (2) The whole DSSR is isothermal with 298 K, which means the Arrhenius expressions are ignored in the model.
- (3) The catalyst is assumed to be stable and never deteriorated.
- (4) The reaction occurs at the three surfaces which are uniformly and fully covered with the catalyst.



Fig. 1. Schematic of one unit in DSSR.

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