Applied Energy 230 (2018) 1403–1413

Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Enhanced photocatalytic reduction of carbon dioxide in optical fiber monolith reactor with transparent glass balls

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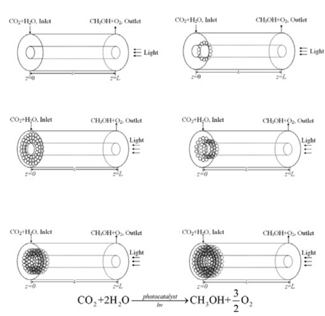
HIGHLIGHTS

- Photocatalytic CO_2 reduction in OFMR with transparent glass balls is studied.
- For single circle and layer model, CH_3OH concentration increases with ball number.
- The closer the distance to fiber and inlet is, the higher the CH_3OH production is.
- CH_3OH concentration rises with increasing the circle and layer numbers.
- 3-circle and 5-layer balls configuration is recommended in the design of OFMR.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Photocatalytic
Carbon dioxide reduction
Methanol production
Optical fiber monolith reactor
Glass balls

ABSTRACT

Photocatalytic reduction of carbon dioxide to produce methanol is a promising approach to restrain greenhouse gases emissions and mitigate energy shortage, which attracts extensive concerns in recent years. The optical fiber monolith reactor with solid glass balls for photocatalytic carbon dioxide reduction is proposed in this work to increase the product concentration, and the glass balls are transparent and coated with photocatalysts evenly to absorb light. The photocatalytic reduction of carbon dioxide in optical fiber monolith reactor is numerically investigated, by which the effects of glass ball number, location, circle and layer on the production are analyzed. The results show that in the single-circle and single-layer model, the outlet methanol concentration increases with increasing the ball number. The closer to the fiber and reactor inlet the balls keep, the higher the methanol production is. As the circle and layer numbers increase, the methanol concentration also increases. The outlet methanol average concentration of the optical fiber monolith reactor with 3-circle and 5-layer balls gets 11.43% higher than the case without glass balls.

1. Introduction

The continuous development of world economy and progress of human civilization since the industrial revolution of mankind basically depend on the adequate energy supply, generally fossil fuels [1]. However, the non-renewable characteristics of fossil fuels compel human explore the renewable sources aiming at sustainable development [2]. Besides, the carbon flow between the ocean and the atmosphere should be natural while human activity adds too much CO_2 to that cycle [3]. Typically, the burning of fossil fuels produces a large

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https://doi.org/10.1016/j.apenergy.2018.09.081
Received 13 May 2018; Received in revised form 28 August 2018; Accepted 8 September 2018
0306-2619/

Nomenclature

C	concentration, mol/m^3
d	diameter, m
D	diffusion coefficient, m^2 s^{-1}
f_θ	fraction of incident light with incident angle less than 90°
I	light intensity, W m^{-2}
k	kinetic rate constant, m^4 s^{-1} mol^{-2}
K	adsorption equilibrium constant
l	distance, mm
L	reactor length, mm
M	relative molecular mass
n	number
p	pressure, Pa
P	total pressure, Pa
r	reaction rate, mol/m^3 s^{-1}
R	radius, mm
T	temperature, K
u	velocity, m s^{-1}
V	molar volume, cm^3 mol^{-1}
z	axial position, mm

Greek letters

α	refractive loss coefficient, cm^{-1}
β	attenuation coefficient of the tip light flux, cm^{-1}
δ	catalyst film thickness, nm
ε	local attenuation coefficient of catalyst thin-film, cm^{-1}
η	energy conversion efficiency
μ	molecular weight of gas
ν	viscosity, m^2 s^{-1}
ρ	density, kg m^{-3}
θ	circumferential coordinate, rad
σ	fractional surface coverage
λ	overall factor
ϕ	quantum efficiency

Subscript and superscript

f	optical fiber
fc	fiber coating
l	layer
m	monolith
mc	monolith coating
gs	glass ball
gsc	glass ball coating
w	monolith channel wall
r	radius direction in cylindrical coordinate system
θ	circumferential direction in cylindrical coordinate system
z	axial direction in cylindrical coordinate system

amount of greenhouse gases, mainly carbon dioxide [4], resulting in many environmental issues such as global warming [5]. Therefore, the great challenge for the mankind is to take powerful measures to develop alternative renewable energy resources [6] and reduce atmospheric CO_2 concentrations [7]. Among various CO_2 capture technologies [8], it seems practical and alternative to take advantage of the MEA (monoethanolamine) and ammonia aqueous solution absorption in packed absorbing columns [9,10] or bubble columns [11], hydrate-based biogas upgrading with CO_2 valorization [12] and artificial photosynthesis in photoreactors [13], etc. Besides, using coal fly ash is an available and potential way to store carbon dioxide [14], and the conversion to fuels as well as the utilization of CO_2 also attracts extensive attention [15]. It is potential that methanol synthesis using captured CO_2 could reduce CO_2 emission and produce the clean fuel [16]. With the efforts of many scholars, the technologies of photocatalytic reduction of CO_2 have made great progress [17], among which the CO_2 photo-hydrogenation by mimicking photosynthesis is one of the best routes of obtaining renewable hydrocarbon fuels under the sunlight and action of photocatalyst in photoreactors [3]. This method is effective to reduce the atmospheric CO_2 concentration and relieve global energy shortage [18]. However, the hydrocarbon yield is commonly low [19] due to the limited reaction surface area, so many researchers aim at improving the photoreactor structure to promote the CO_2 reduction [20] and hydrocarbon conversion [21].

In previous studies, various photoreactors such as slurry reactor, fixed bed reactor, surface coated reactor, twin reactor [22], optical fiber photocatalytic reactor [23], annular and bubble flow reactors [24] were proposed, for which the structures play an important role in the CO_2 photoreaction. For optical fiber monolith reactor (OFMR), the optical fiber is inserted into the parallel reaction channels, and the catalyst is coated on the fiber and monolith surfaces to convert the CO_2 to methanol under the artificial light [25,26]. This type of photoreactor shows many advantages over other photoreactors [27], such as the efficient and uniform light distribution, good interaction between the catalyst layer and photons, high conversion efficiency and product yield, low pressure drop and operational costs [28], so it seems more promising for the photocatalytic CO_2 reduction [29].

For the OFMR, the solar energy utilization ratio for the CO_2 photoreduction is only around 3% and the reaction surface area is not large enough, which shows a wide space to increase the solar energy conversion efficiency and improve the photocatalytic CO_2 reduction [20]. In the aforementioned studies, more emphases were placed upon the energy conversion efficiency and modified effective photocatalysts, while the photoreactor structure is still worthy of further investigation [23].

As a credible way of OFMR, a bundle of optical fibers like honeycomb is commonly used to transmit the light with solar concentrating devices and increase the reaction surface area. In this work, a filling transparent solid glass ball model is proposed in the light of OFMR, where the surfaces of the fiber, monolith, and the glass balls are all coated with photocatalysts to adequately absorb photons for the photocatalytic CO_2 reduction and improve the energy conversion efficiency. By this way, the photoreaction surface area is enlarged without destroying the central symmetric structure of the reactor and the disturbance to the flow is minimized as much as possible. The filling ball model is simulated in this paper using the commercial multi-physics software COMSOL. The numerical simulation can not only verify the reliability and accuracy of the experimental results after the experiments, but also predict the feasibility before the experiments. The yield and the average outlet concentration of the product, the CO_2 concentration and velocity distribution, are all obtained and compared with the photoreaction in the traditional OFMR, which can contribute to the optimal design and energy efficient operation of photocatalytic CO_2 reduction in optical fiber monolith reactor.

2. Multi-physics modeling

Due to the identity of each flow-path, only one reactor unit, namely one channel, is taken into account in this work. The comprehensive and rigorous model is developed, which consists of three computational modules of chemistry, transport of dilute species and creeping flow.

2.1. Physical model

The traditional and proposed models are schematically shown in Fig. 1. In the traditional model, the light source is set at the inlet of