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Thermal and chemical analysis of methane dry reforming in a volumetric reactor under highly concentrated solar radiation

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ABSTRACT

The heat transfer and thermochemical reaction are numerically investigated for the solar driven methane dry reforming process, coupling with the solar radiation transport from concentrator to the interior of volumetric reactor. Using a Monte Carlo ray tracing method, the solar radiation concentration with a multi-dish system and absorption by the foam structure in the reforming reactor are simulated. The solar power density distribution in reforming reactor and the optical efficiency of system are determined and compared at different foam structural parameters. Then, an integrated model of transport phenomena is developed to analyze the thermal and chemical performances under different operating conditions. The results indicate that solar radiation absorption and distribution in the reactor are greatly affected by the foam structural parameters, while the optical efficiency has no significant variation. Assumption of the concentrated solar radiation as collimated leads to a temperature deviation of approximately 10%. Besides, the methane conversion increases with increasing porosity and cell size, and decreases markedly with increasing inlet velocity and CH_4/CO_2 ratio.

1. Introduction

The increasing fossil fuel depletion and the escalating greenhouse gas emissions which leads to global climate change have attracted considerable attention (Sheu et al., 2015). The development and utilization of new, renewable energy sources has become an inevitable global strategy. Solar energy is a sustainable and potential resource, because the hourly incident solar flux on the Earth is greater than the annual global energy consumption (Lewis, 2007; An et al., 2017). However, due to low energy density and the intermittent and unstable nature, the conversion of solar radiation into technically useable energy is an engineering challenge (Kodama, 2003; Zou et al., 2018). High thermal flux and high temperature can be obtained through the reflection and concentration of direct insolation (Segal and Epstein, 2003a; Levêque et al., 2017). The thermal energy from concentrated solar radiation can not only be used to generate electricity but also to synthesize chemical fuels from water and carbon dioxide (Roeb and Müller-Steinhagen, 2010). This thermochemical conversion provides an effective way to store solar energy in a long-term, stable and transportable form (Segal and Epstein, 2003b; Steinfeld, 2005).

Among the different thermochemical routes, the methane dry reforming process is highly feasible and promising for the solar energy conversion, storage and transportation. Furthermore, the product is well known as syngas which mainly contains CO and H_2. This reaction offers substantial environmental benefits: biogas utilization, removal of greenhouse gases and production of valuable syngas (Usman et al., 2015). The product has a low H_2/CO ratio, which is suitable for the synthesis of oxygenated chemicals and hydrocarbons. Additionally, the reforming system typically includes two main parts: a solar concentrator and a chemical reactor (Steinfeld and Meier, 2004). Solar reactors can be distinguished into two categories: directly and indirectly heated reactors, with the former displaying better thermal and chemical performances. As a type of directly heated reactor, volumetric reactors with foam material have been extensively developed and generally show high efficiency (Yadav and Banerjee, 2016). The incoming solar radiation is collected and redirected by the concentrator to the chemical reactor. Then, solar radiation penetrates into the reactor and is gradually absorbed by the foam structure, causing a high temperate environment and supplying the heat source to drive the endothermic reaction. Thus, there are two key processes involved in the solar reforming system: (1) solar radiation transfers from the concentrator to the reactor and distributes in the foam structure, (2)

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Nomenclature

c_p	specific heat, J kg^{-1} K^{-1}
d_c	mean cell size, mm
G_d	integrated intensity, W m^{-2}
h_i	partial enthalpy of species i, J
h_v	volumetric heat transfer coefficient, W m^{-3} K^{-1}
k	thermal conductivity, W m^{-1} K^{-1}
L	length of reactor, m
p	pressure, Pa
Q	heat rate, W
R	radius of reactor, m
S	source term
T	temperature, K
$\vec{V}$	superficial velocity, m/s
r	coordinates in cross flow region, m
z	coordinates in flow region, m

Greek symbols

ρ	density, kg m^{-3}
ϕ	porosity
ε	emissivity
η	optical efficiency
κ_a	absorption coefficient, m^{-1}
κ_s	scattering coefficient, m^{-1}
κ_β	extinction coefficient, m^{-1}
μ_f	dynamic viscosity, kg m^{-1} s^{-1}
σ	Stefan-Boltzmann constant, W m^{-2} K^{-4}

Subscripts

chem	chemical reaction
eff	effective
f	fluid phase
r	radiation
s	solid phase

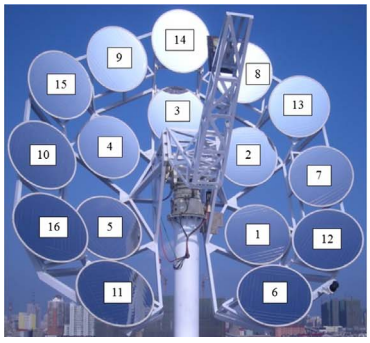
reactants pass through the foam matrix and react, along with the coupled high-temperature heat transfer. The concentration and absorption of solar radiation strongly affects the energy distribution in the chemical reactor, and consequently has a direct effect on the various transport phenomena, chemical reactions and the overall system performance. A conceptual diagram of the process is as follows (Kodama, 2003):

Solar radiation → concentrator and volumetric reactor → heat → reactants (CH_4 and CO_2) and/or catalyst → solar fuels (CO and H_2).

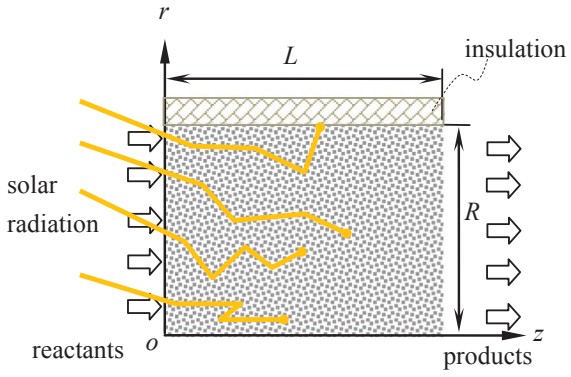
For the methane dry reforming, there are various studies focusing on thermodynamic analysis (Chein et al., 2015; Atashi et al., 2016), chemical kinetics (Gokon et al., 2009; Özkara-Aydınoğlu and Aksoylu, 2013), catalyst development (Qi et al., 2015; Abdullah et al., 2017), and system design (Said et al., 2016; Villafán-Vidales et al., 2016). In addition, several experimental studies have been conducted when the reactor is exposed to highly concentrated solar radiation, e.g. Buck et al. (1991), Wörner and Tamme (1998), Rubin and Karni (2011). However, there is a lack of detailed knowledge on the heat transfer and reaction mechanisms, temperature and product distributions inside the volumetric type of reactor. This has motivated the development of numerical models which evaluate the effects of operation conditions and other aspects to optimize the process parameters and system design (Wheeler et al., 2017).

The thermochemical process is operated at high temperature. Several transport processes occur together with chemical reactions in the chemical reactor, which show strong interactions. Nematollahi et al. (2014) proposed a one-dimensional model for the dry reforming reaction using a Rh-supported catalyst in a packed bed reactor, and found good agreement between modeling results and experimental data. Using a one-dimensional heterogeneous fixed bed reactor model, Benguerba et al. (2015) reported that the CH_4/CO_2 ratio should be close to 1.0 to maximize product yield. To gain further insight into the packed bed reforming system, a two-dimensional model was employed by Akpan et al. (2007), and they noted that the assumption of plug flow isothermal behavior is justified under certain conditions. Yin et al. (2007) investigated the effect of catalyst particle clusters on the performance of methane dry reforming in a circulating fluidized bed reformer, and observed that the reaction rate becomes smaller and the H_2/CO ratio is upgraded. A comparison between Langmuir-Hinshelwood kinetic and microkinetic models was made by Behroozsarand and Pour (2014) by simulating the dry reforming process in a micro-reactor. The results showed that the former model is more suitable. Lu et al. (2016) experimentally and numerically studied the heat transfer and thermochemical performances of dry reforming in a tubular packed reactor, and found that the efficiency can be increased by increasing the bed conductivity and decreasing heat loss. Chein et al. (2017) reported that introducing excessive CO_2 into the fixed-bed tubular reactor could enhance the system performance. These studies mainly deal with the reforming process within a tubular packed bed reactor subjected to a heated furnace or tube wall, and the two phases (gaseous mixture and bed) are assumed to be in local thermal equilibrium.

Few numerical studies involve the solar driven methane dry reforming process. Yu et al. (2017) numerically and experimentally investigated the thermochemical reforming reactions and storage performance in a tubular and semi-cavity reactor heated by a solar dish. Using a finite volume method coupled with thermochemical kinetics, a



(a) the multi-dish solar concentrator



(b) the chemical reactor

Fig. 1. Schematic of the solar reforming system considered in this study.