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Thermochemical storage analysis of the dry reforming of methane in foam solar reactor

Xue Chen[a,b], Fuqiang Wang[c,*], Yafen Han[b], Ruitian Yu[c], Ziming Cheng[c]

[a] School of Mechatronics Engineering, Harbin Institute of Technology, 92, West Dazhi Street, Harbin 150001, PR China
[b] School of Energy and Power Engineering, Northeast Electric Power University, 169, Changchun Street, Jilin 132012, PR China
[c] School of Automobile Engineering, Harbin Institute of Technology at Weihai, 2, West Wenhua Road, Weihai 264209, PR China

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ABSTRACT

The high-temperature heat transfer and thermochemical storage performances of dry reforming of methane in a foam reactor subjected to highly concentrated solar radiation is numerically investigated. Two new correlations for the volumetric heat transfer coefficient and the pressure-drop within foam structure are proposed through several experimental tests. Temperature and species distributions inside the reactor as well as the overall methane conversion and thermochemical energy storage efficiency under various operating conditions are predicted. The results indicate that the increase in inlet velocity and CH_4/CO_2 ratio produces a reduction in methane conversion, while increasing the solid-phase thermal conductivity promotes the conversion. The energy storage efficiency exhibits non-monotonous change with inlet velocity, feed ratio, and foam structural parameters. Compared to other foam structural parameter combinations, maximum conversion and efficiency are observed at porosity of 0.9 and pore diameter of 1.5 mm. Besides, the solid-phase thermal conductivity and foam structural parameters exert negligible effect on the H_2/CO ratio in the reforming process.

1. Introduction

Developing renewable energy sources and reducing greenhouse gas emissions have become a worldwide trend and recently received great interest [1–3]. The dry reforming of methane (DRM) presents an effective way to convert the two abundant greenhouse gases (CH_4 and CO_2) into synthesis gas (CO and H_2). The products exhibit a low H_2/CO ratio and are very suitable for the further synthesis of long-chain hydrocarbons. The main chemical transformation can be represented as $CH_4 + CO_2 \rightarrow 2H_2 + 2CO$. In recent years, the DRM has been widely studied on the development of catalysts with high activity and adequate lifetime stability [4–6]. Nickel- and noble metal-based catalysts are extensively demonstrated to be highly active for the DRM reaction [7]. Furthermore, using the endothermic reactions to convert concentrated solar radiation into chemical energy is regarded as a potential method to store solar energy in a stable and transportable form [8]. As one of the emerging technologies, DRM process is feasible for the solar thermochemical storage because of large reaction heat. Solar-driven synthesis gas produced by the reforming reaction has calorific values 22–28% higher than that of a methane feedstock [9]. The performance of such reforming system (solar driven DRM) is contingent upon the catalyst as well as various other factors such as operating conditions and reactor design. The solar radiation concentration characteristics and the solar energy absorption features of reactor also significantly affect the overall performance. Besides, this endothermic reaction is also observed to strongly interact with the heat and mass transfer within the reactor [10]. Therefore, studies of the high-temperature transport phenomena and chemical reaction are essential for enhancing this technology.

As a structural and functional material, reticulated porous foams are widely used in numerous engineering applications. Foams with web-like internal structure are characterized by high effective thermal conductivity and gas permeability, superior mechanical strength and light weight [11,12]. Moreover, the irregularly-shaped geometrical structure can effectively and uniformly absorb the solar radiation and provide the required heat of reaction. Thermochemical reactor with foam material (also known as volumetric reactor) shows improved performance and efficiency [13]. This category of solar reactors has been designed and tested at laboratory scale using a solar concentrator in the CAESAR, SCR, SOLASYS, and SOLREF projects [14–16]. In these projects, alumina and SiC foams were adopted. In addition, due to the superior plasticity, metal foams are also used under the condition of low flux levels of concentrated solar radiation or low temperature, such as Ni–Cr–Al metal foam [17], 316 stainless steel foam [18], and Cu foam

* Corresponding author.
E-mail address: Wangfuqiang@hitwh.edu.cn (F. Wang).

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[19].

To gain further insight into the thermal and chemical performances of solar reforming system, heat and mass transfer in conjunction with the catalytic reaction inside the reactor should be investigated in detail and comprehensively. This objective motivates the development of numerical modeling to obtain suitable operating conditions and optimum system design [20]. Several studies have been conducted to simulate the DRM process within a packed bed reactor subjected to a heated furnace or tube wall, using one-dimensional model by Nematollahi et al. [21] and Benguerba et al. [22] and two-dimensional model by Akpan et al. [23], Yin et al. [24], and Chein et al. [25]. A pseudo-homogeneous reactor model was developed by Akpan et al. [23] and they found that the axial dispersion term should be included. Yin et al. [24] investigated the effect of catalyst particle clusters on the DRM performance and found that the reaction rate was decreased and the H_2/CO ratio was increased. Chein et al. [25] reported that DRM performance was degraded as the reaction pressure and reactant flow rate were increased in a fixed-bed tubular reactor. The thermal radiation heat transfer was not considered in the previous researches, and the solid pellet and gaseous mixture were assumed to be in local thermal equilibrium (LTE) (with identical temperature) based on the consideration of good heat transfer between the gas and solid. Although this approximation considerably facilitates the heat transfer modeling in such porous material, it fails to precisely evaluate the thermal performance [26].

By considering the convective heat transfer between the fluid and solid phases, local thermal non-equilibrium (LTNE) model should be employed by coupling the energy equations of the two phases with a volumetric heat transfer coefficient. It has been reported that the LTNE model can more effectively predict the thermal performance of porous materials. Using the LTNE model, the DRM process in a tubular and semi-cavity packed bed reactor heated by a solar dish system was simulated and experimentally validated by Yu et al. [27], the solar reactor was a type of indirectly irradiated reformer. Moreover, the microstructure of foam material is characterized by the porosity, cell shape and size, and morphology of struts, which is significantly different from the packed particles. Reasonable number of numerical investigations on the foam solar reactor is unavailable. The solid skeleton is directly irradiated with concentrated solar energy to heat the reactants up to the reaction temperature. The performance of a foam type solar thermochemical reactor was preliminarily analyzed by Wang et al. [28,29], and the effects of heat flux distribution and porosity were discussed.

The literature review indicates that notwithstanding the reporting of several catalytic DRM modeling works, extant works mainly focus on the conventional DRM process in a tubular packed bed reactor, and omit the LTNE heat transfer between the two phases and the high-temperature thermal radiation. A few experimental studies have been carried out on the chemical kinetics, catalyst development, and system test of the foam reactors; however, the detailed thermal and chemical reaction information inside the reactor could not be provided, particularly for the solar-driven reforming process. Very few studies address the numerical modeling of solar-driven DRM process in a foam reactor. Additionally, in the flow and heat transfer modeling of a gas flow and foam system, the pressure drop and volumetric heat transfer coefficient, which are the key inputs, have inspired a number of experimental measurements and direct pore-level simulations [30–32]. Numerous empirical correlations have been proposed in the literature; however, it exhibits large differences among the predicted results and an absence of general applicability [33,34].

In this study, a finite volume method coupled with thermochemical reaction kinetics is developed to investigate the heat and mass transfer, reaction performance, and thermochemical energy storage of the solar-driven DRM in a foam reactor. In the numerical model, new correlations for the pressure drop and volumetric heat transfer coefficient are used, which are derived from several experimental tests. Temperature and species distribution, methane conversion, H_2/CO ratio, and chemical energy storage efficiency are discussed under various operating conditions.

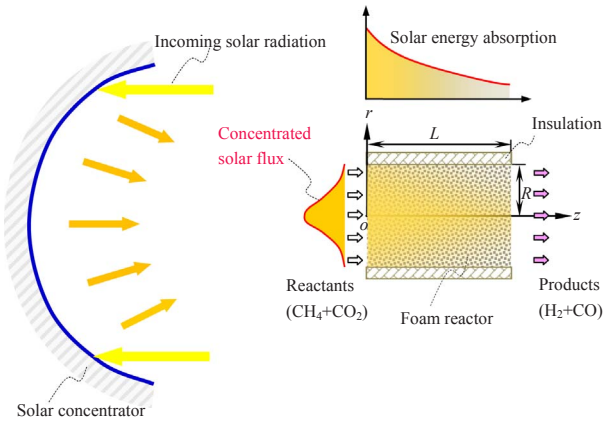

Fig. 1. Schematic of the solar-driven DRM in a foam reactor.

2. Reforming reactor description

The solar reforming system typically contains two main components: a solar concentrator and a chemical reactor, as shown in Fig. 1. The incoming solar radiation is collected and reflected by the concentrator, forming a highly intense radiative flux on the aperture of foam reactor. The concentrated solar radiation penetrates into the reactor and is directly absorbed by the catalyzed foam in the entire volume, causing a gradual temperature rise. High-temperature foam skeleton transfers the heat to the feed reactants and facilitates the endothermic chemical reaction. The reactants with a fixed-ratio are injected into the reactor, flow through the foam skeleton, react at high temperature, and produce syngas at the rear of the solar reactor.

2.1. Governing equations

Several transport processes occur simultaneously with the high-temperature chemical reaction in the solar reforming reactor. These processes can be described by the conservation equations of mass, momentum, energy, and chemical species. A 2D axisymmetric steady state model is built to study the inner heat and mass transfer and the reaction characteristics. The mass conservation equation is

$$\nabla\cdot(\rho_f \mathbf{U}) = 0 \tag{1}$$

where ρ_f denotes the fluid density and $\mathbf{U}$ is the vector of fluid velocity.

The momentum conservation equation can be described as:

$$\frac{1}{\phi}\nabla\left(\rho_f \frac{\mathbf{U}\cdot\mathbf{U}}{\phi}\right) = -\nabla p + \nabla\cdot\left(\frac{\mu_f}{\phi}\nabla\mathbf{U}\right) + \mathbf{S}_m \tag{2}$$

where $\mathbf{S}_m$ represents the momentum source term owing to the flow in the random pore structure inside the foam material, and p and μ_f are the pressure and viscosity, respectively.

The species mass balance can be expressed as

$$\nabla\cdot(\rho_f \mathbf{U} Y_i) = -\nabla\cdot\mathbf{J}_i + S_i \tag{3}$$

where Y_i, S_i, and $\mathbf{J}_i$ mean the mass fraction, net rate of production, and diffusive mass flux of the ith species, respectively.

The LTNE between two phases is considered, and the energy equations for the fluid and solid phases are expressed as follows:

$$\nabla\cdot(\rho_f c_p \mathbf{U} T_f) = \nabla\cdot(k_{f,eff}\nabla T_f - \sum_{i=1}^{n} h_i \mathbf{J}_i) + h_v(T_s - T_f) + S_{chem} \tag{4}$$

$$\nabla\cdot(k_{s,eff}\nabla T_s) + h_v(T_f - T_s) + S_r = 0 \tag{5}$$