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Full Length Article

Numerical study of steam methane reforming over a pre-heated Ni-based catalyst with detailed fluid dynamics

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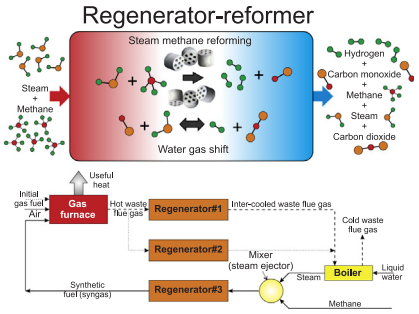
GRAPHICAL ABSTRACT



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ABSTRACT

A computational fluid dynamics (CFD) model of steam methane reforming process over a pre-heated Ni-based catalyst is developed via ANSYS Fluent for real computational domain of the reformer. The investigations are focused on determination of temperature and mole fraction of individual element distribution in the reformer at various initial conditions. The fluid dynamics of reacting flow is studied in experimental setup. It is established that the increasing of the gas flow rate leads to pressure loss increasing. For example, at velocity inlet of 1 m/s the pressure drop is 512 Pa for the investigated reformer. The results of CFD simulation are revealed that the mole fractions of each elements are constantly changing due to chemical reactions when the reaction mixture passes through catalyst bed of the steam methane reformer. When the synthesis gas composition obtained with the CFD model versus the equilibrium syngas composition is compares, it is presented that at the catalyst temperature above 1300 K a syngas composition is close to equilibrium. The distribution of the gas temperature in the reformer is determined by the developed model. The temperature gradient near the reformer inlet has a maximum value, because the temperature drop between catalyst and reaction mixture is maximum and maximum rate of steam methane reforming reaction takes place in this section of the reformer. The temperature gradient slows down to outlet of reformer, because the catalyst temperature decreases due to endothermic steam methane reforming reaction, and also due to increase of steam-to-methane ratio form inlet to outlet due to methane consumption (for inlet steam-to-methane ratio above unit). For follow initial conditions (steam-to-methane ratio is 2.0, pressure is 5 bar, residence time is 4.54 kg_{cat}·s/mol_{CH_4}, temperature of feed stock is 800 K, catalyst temperature is 1300 K), temperature of outlet synthesis gas is 849 K.

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1. Introduction

In accordance with the hydrogen scenario of the European Commission, large-scale hydrogen production in the world will begin after 2030 and will be stimulated by a significant reduction in the cost of hydrogen technologies and increased consumption of hydrogen in the transport sector [1]. Nowadays the major technology for production of hydrogen and synthesis gas (mixture of CO (carbon monoxide) and H_2 (hydrogen)) is steam reforming of hydrocarbon: methane [2–5], ethanol [6–8], methanol [9–11], biomass [12–14] and other hydrocarbons [15–17]. The production of hydrogen by natural gas reforming accounts for about 48% of the world's hydrogen production [18]. The main component of natural gas is methane; therefore steam methane reforming is the major way for hydrogen production.

The chemical reaction of steam methane reforming (SMR) is well studied. SMR process is comprised of heating the reaction mixture (steam and methane) over a various type of catalysts [2–4,19,20]. According previous investigations it can be concluded that the overall steam methane reforming process produces hydrogen, carbon monoxide, carbon dioxide and minute quantity of other chemicals. But only three reactions have substantial contribution to heat and mass balance [2,21–23]:

–steam methane reforming reaction:

$$CH_4 + H_2O = CO + 3H_2 - 206.1\ \text{kJ/mol}. \quad (1)$$

–water-gas shift reaction:

$$CO + H_2O = CO_2 + H_2 + 41.1\ \text{kJ/mol}. \quad (2)$$

–global steam methane reforming reaction:

$$CH_4 + 2H_2O = CO_2 + 4H_2 - 165.0\ \text{kJ/mol}. \quad (3)$$

Generally, the industrial reaction of steam methane reforming is carried out in reformers – large-scale furnace [24–27]. In the reformer, steam methane reforming reactions are normally taken place at moderate high temperatures 900 to 1200 K in a presence of catalysts [21]. In the industry, various types of catalysts for steam methane reforming are well characterized. Usually, Ni-based catalysts are used in industrial scale [21,28–30]. In addition, other catalysts, for example noble metals-bases, are also applied for steam methane reforming [31,32]. A large number of scientific publications and investigations concerning the study of steam reforming of methane and devices for its implementation allow to constantly improving SMR process.

Recently, numerical modeling and investigation has been actively used to study steam methane reforming. Hoang and co-workers developed 2D model of steam methane reformer and compared the obtained results with 1D model and experimental data [21]. In that study the chemical reactions over the catalyst bed, mass and heat transfers in the radial and axial directions in the steam methane reformer are considered. The optimal conditions for high energy efficiency of hydrogen production in steam methane reforming process (temperature range of 700–800 °C under steam-to-methane ratio 3–3.5) was reported by Hoang and co-authors [21]. These obtained results also are confirmed by other investigations [33–35].

Sunol and Patel presented a steady-state model of thermally coupled steam methane reformer that is consisted of three ducts [36]. Combustion of natural gas occurs in the first duct which is activated by noble metal-based catalyst and steam methane reforming process carry out in the second duct which is activated by nickel-based (Ni/Al_2O_3) catalyst. The developed model made it possible to determine the temperatures of reaction mixture and mole fractions of the reacting components in the reformer.

With considerable increase of computers computational power, CFD-simulation has become an progressively increasing way for steam methane reformer development, integrating chemical (chemical reactions) and physical (heat-and-mass transfer) models for different geometry of SMR-reformer. The main advantage of CFD-modeling is a powerful visualization of the calculation results for subsequent processing and analysis. In addition, computational fluid dynamics modeling provides versatility to modify design parameters without the expense of research equipment changes which brings large money economic and economy of time [37,38]. The CFD-modeling results of various combined chemical and physical phenomena are in well agreement with the experimental data for different reformer geometries, types of catalysts and operation conditions [39,40,21]. Various commercial and open-source software are used for modeling and simulation of steam methane reforming process, for example, ANSYS Fluent [41,42], Comsol Multiphysics [43], Autodesk Simulation CFD [44], OpenFOAM [45] (open source). Lao et al. [39] presented a computational fluid dynamics (CFD) model of an large-scale SMR process for hydrogen production. A developed CFD-model is implemented in ANSYS Fluent for real computational domain to predict temperatures and mole fractions of each element into reformer. The simulation results can be used to improve the energy efficiency of SMR process, and also can be applied to develop efficient reactors.

Dixon [40] presented a comparison of the results obtained via CFD-modeling of steam methane reforming process in Comsol Multiphysics. Author simulated the chemical reactions and temperature distribution for 1D, 2D and 3D reactor models. The most accurate data, which are in good agreement with the experimental results, are obtained for the 3D and 2D model. Also, Ku and co-workers [46] described comprehensive CFD-discrete element method of numerical model for the reactor which is implemented in open-source software OpenFOAM.

CFD-modeling studies on steam methane reforming process (mentioned above) were mainly focused on investigation of reformer, in which heat for endothermic SMR reaction supplied through the wall [39,40,21]. This paper is devoted to CFD investigation of steam methane reforming process over pre-heated Ni-based catalyst. The catalyst of reformer bed, heated by hot flue gases, is cooled through taking place a strongly endothermic steam methane reforming reaction. This method of steam methane reforming is adopted for hydrogen and synthesis gas production and also can be used for thermochemical waste-heat recovery [47–51]. Therefore, 2D CFD model of steam methane reforming process over pre-heated Ni-based catalyst is developed in ANSYS Fluent to predict the reaction and transport phenomena into reaction space of reformer. The major intention is paid to the investigation of mole fraction and temperature distribution into reformer bed for various operation conditions. In addition, much attention is paid to studying the fluid dynamics of the reacting flow, because when the reacting components pass through the catalyst bed, there are pressure losses. The results of steam methane reforming study which is implemented by Froment and Xu [2] is used for receiving equations of chemical conversion rate over a Ni-based catalyst.

2. Schematic diagram and experimental setup

2.1. Schematic diagram

Steam methane reforming over a preheated catalyst can be used not only for hydrogen and synthesis gas production. This method of SMR can be used for thermochemical waste-heat recuperation [48,49,52]. This method of improving of energy efficiency is based on the principle of transformation of the flue gases heat into the chemical energy of a synthetic fuel, which is consisting mainly of hydrogen.

Fig. 1 shows a schematic diagram of steam methane reforming process over a preheated Ni-based catalyst for thermochemical waste-heat recuperation system. A recuperation system of methane-consuming industrial furnace is consisted of at least tree regenerator (steam