

Research Paper

Modeling of natural gas partial oxidation in an in-house developed pilot scale catalytic reactor based on local thermodynamic equilibrium concept



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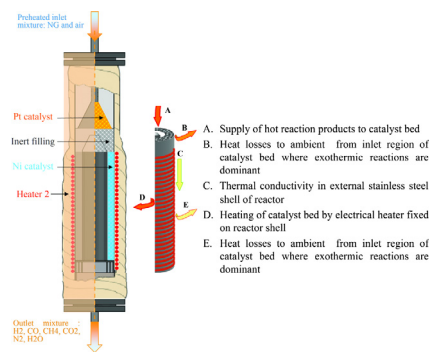
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HIGHLIGHTS

- Partial oxidation of natural gas in a pilot scale reactor is modeled.
- Developed model described heat transfer in catalyst bed and reactor shell.
- Chemical sub-model is based on assumption of local thermodynamic equilibrium.
- Model can be applied for optimization of reactor design and selecting working parameters.

GRAPHICAL ABSTRACT



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ABSTRACT

A numerical model based on the concept of local thermodynamic equilibrium was developed to describe the catalytic partial oxidation processes in reactors with performance that exceeds laboratory scale. The developed model describes heat transfer in the catalytic porous layer and steel shell of the catalytic reactor, which has external heating. Computer simulation based on this model facilitates prediction of spatial temperature profiles and reactive mixture composition inside the reactor. The predictions of the numerical model are in agreement with the mixture composition at the reactor outlet as measured during steady-state working regimes in the context of different flow rates that ranged from 1.7 to 5.1 Nm³/h.

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1. Introduction

The catalytic partial oxidation (CPOX) of hydrocarbons is considered one of the major processes for production of synthesis

gas, which is used in various industrial applications as a hydrogen source [1]. In this work, we focused on the use of synthesis gas as a protective atmosphere for chemical heat treatment of steels (hardening, carburizing, and nitriding). The basic requirements for such synthesis gas are a minimum content of steam and carbon dioxide and a controlled concentration of methane [2].

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Commercial protective atmosphere generators that run on natural gas (NG) are characterized by product content, which is typically 40% N₂, 40% H₂, 19.5–19.8% CO, 0.2–0.5% CO₂, <0.1% H₂O, and <0.1% CH₄ [3]. The performance of commercial protective atmosphere generators varies from 2.5 to 500 m³/h, and the typical performance that is desired in metal-based manufacturing industry is 15–60 m³/h of protective gas.

Industrial partial oxidation reactors operate at temperatures that typically range from 950 [2] to 1040 °C [4] in the catalytic zone to provide the desired component composition of conversion products. The motivation for this research was to optimize the design of the synthesis gas generator in terms of minimizing the volume of the catalytic bed and operation performance without the loss of conversion product quality even when the working flow rate was decreased to 20% of the nominal values. The parameters of the experimental setup were selected to approach the industrial scale, as these parameters are significantly different from those used in laboratory experiments.

In our previous work [5,6], we investigated thermal regimes and compositions of synthesis gas for two reactor designs with a range of mixture input flow rates from 1.7 to 7.1 Nm³/h and with temperatures from 650 to 1050 °C. The results of these studies led to the conclusion that the component composition of the gas mixture is close to equilibrium in the endothermic zone of the catalytic bed of industrial reactors. Moreover, such a situation was observed for all tested catalysts in our experiments [5]. Similar results about equilibrium products were found in another study [7], in which the experimental data were analyzed [8] within a temperature range of 580–650 °C; however, in these studies, the values of gas hourly space velocity (GHSV) were not indicated. In other experiments [9], temperature was maintained at 700 °C in the catalytic bed outlet, and the obtained products of methane conversion matched the amounts calculated for thermodynamic equilibrium at this temperature. The values of GHSV (8900 h⁻¹) reported for these experiments [9] were similar to the values used in our experiments [5,6], although the flow rates of reactive mixtures were much lower than those used in our experiments [6].

Comparable values of GHSV were also used in experiments conducted with a two-section cylindrical reactor with external electric heating [9]. The flow rates, which ranged from 0.1 to 0.37 Nm³/h, were closest to those used in our experiments. The composition of the products obtained in a study by Xu and colleagues was significantly different from equilibrium due to high GHSV [10]. In our studies we attributed this deviation from equilibrium to the heterogeneity of mixing when an additional oxidizing agent is admixed with the gas mixture after the first (exothermic) unit prior to feeding into the second (endothermic) catalyst unit. Temperature heterogeneity may also affect the deviation from equilibrium in the composition of CPOX products.

Considerable research has been devoted to the development of complex kinetic schemes for surface reactions [11–13]. The typical approach was described by Hickman and Schmidt [11], and a modified approach was described by Schwiedernoch and colleagues [12]. Furthermore, detailed kinetic investigations of partial oxidation and steam reforming have been performed using chemical models that have been described previously [14–16]. Typically, the experiments used to verify the models are conducted on small-scale laboratory reactors [17–19]. The conditions of heat transfer and thermal loss in laboratory equipment differ significantly from the industrial apparatus. The main difference in the industrial reactors is that the area of the exothermic processes is much smaller than the endothermic processes zone. One-dimensional modeling with a complex kinetic scheme [20] has demonstrated that the distance from the cross section of the maximum temperature in the area of exothermic processes to reaching the equilibrium characteristics is about 0.5–1 cm; however, the

scale of the endothermic processes area in the industrial reactors for methanol production comprises several meters to ensure proper heating. In this case, the thermal loss and/or heating of the endothermic part of the reactor controls the temperature field in the catalyst and thus control the quality of the conversion. Review of the literature on modeling of catalytic partial oxidation revealed no published studies in which the chemical equilibrium model was coupled with a detailed description of thermal processes.

Thus, we sought to design the systems with different performance capabilities to enable generation of a protective atmosphere (endogas) via CPOX of NG for the needs of thermal and thermochemical treatment of steel parts. In order to achieve this goal, we needed a model that would allow optimization of the main dimensions of the catalytic reactor, amount of catalyst used, temperature, energy efficiency, and, ultimately, technological simplicity of design and cost.

Based on the results obtained in the previous studies [5,6], we proposed spatial separation of the areas of exothermic and endothermic processes and tentatively use different types of catalysts in these areas. In addition, we proposed to use external heating of the endothermic region of the reactor to prevent thermal loss and to heat the catalytic bed to a target temperature of approximately 950 °C to reduce the characteristic dimensions of the catalyst bed. Furthermore, we proposed fabrication of the endothermic part of the reactor as a coaxial cylindrical catalytic bed. Developed design of reactor is tested in experiments for production of synthesis gas.

2. Materials and methods

2.1. Experimental installation

A schematic diagram of the apparatus for the production of endogas is shown in Fig. 1. The system that prepares the inlet mixture consists of two flow controllers and the mixing device, and after passing through this device, the mixture enters the heater and is then fed into a heated reactor. The conical shaped inlet region of the reactor is filled with a heat-resistant catalyst material (Pt) that ignites the working mixture. Exothermic reactions occur in this area and in the subsequent bed of inert material. The maximum temperature is reached in the exothermic reaction zone. The outer shell of the reactor is not heated, and the thermal insulation ensures that the temperature of this shell does not exceed 1100 °C. After a layer of inert material, there is a coaxial bed that contains the main working catalyst material, which may be Pt, Rd, Pd, or Ni. The shell of the reactor is heated using an electric-controlled heater (Kanthal spiral). For safety reasons, the temperature in the shell does not exceed 1100 °C. The volume of the catalyst is determined by the thickness and height of the coaxial bed. The radial dimension is the most important parameter, as this measurement determines the scale of heat transfer processes. After passing through the catalytic bed, the reaction products fall into the tubular cooler. After additional drying, part of this mixture is applied to a gas analyzer, where component concentrations are measured. The concentrations of CO, CO₂, and CH₄ are measured using optical sensors, and H₂ concentrations are measured with a Katharometer. Periodically, the composition of the outlet mixture is assessed using chromatographic measurements.

2.2. Energy characteristics of the partial oxidation process

As the experimental reactor was developed to produce CPOX products under near equilibrium conditions, we estimated the temperature regimes and power based on equilibrium characteris-

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