



Predicting the temperature and reactant concentration profiles of reacting flow in the partial oxidation of hot coke oven gas using detailed chemistry and a one-dimensional flow model

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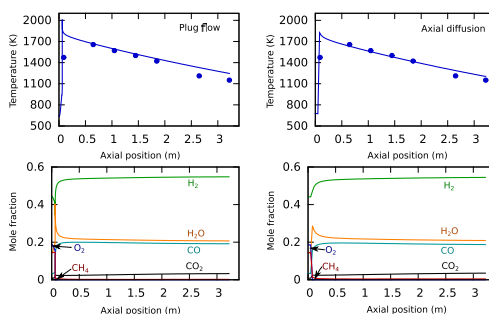
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HIGHLIGHTS

- Detailed chemical kinetic model for coke oven gas reforming.
- One-dimensional non-adiabatic reactor model with axial flow diffusion.
- Axial temperature profiles predicted from the model.
- Global sensitivity analysis for identifying dominant reactions in a reaction network.
- Comparison of model predictions with pilot-scale measurements.

GRAPHICAL ABSTRACT

Comparison of the temperature (top) and mole fraction (bottom) profiles without (left) and with (right) diffusional flow along the reactor axis during HCOG reforming via partial oxidation (run 11). The symbols indicate experimentally measured values, and the solid lines represent model predictions. The HCOG and O₂ flow rates are 62.9 N m³/h and 17.6 N m³/h, respectively.



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ABSTRACT

A numerical approach is presented for predicting the species concentrations and temperature profiles of chemically reacting flow in the non-catalytic partial oxidation of hot coke oven gas (HCOG) in a pilot-scale reformer installed on an operating coke oven. A detailed chemical kinetic model consisting of 2216 reactions with 257 species ranging in size from the hydrogen radical to coronene was used to predict the chemistries of HCOG reforming and was coupled with a plug model and one-dimensional (1D) flow with axial diffusion model. The HCOG was a multi-component gas mixture derived from coal dry distillation, and was approximated with more than 40 compounds: H₂, CO, CO₂, CH₄, C₂ hydrocarbons, H₂O, aromatic hydrocarbons such as benzene and toluene, and polycyclic aromatic hydrocarbons up to coronene. The measured gas temperature profiles were reproduced successfully by solving the energy balance equation accounting for the heat change induced by chemical reactions and heat losses to the surroundings. The approach was evaluated critically by comparing the computed results with experimental data for exit products such as H₂, CO, CO₂, and CH₄, in addition to the total exit gas flow rate. The axial

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diffusion model slightly improves the predictions of H₂, CO, and CO₂, but significantly improves those of CH₄ and total exit flow rate. The improvements in the model predictions were due primarily to the improved temperature predictions by accounting for axial diffusion in the flow model.

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1. Introduction

Efficient, cost-effective technologies to reform hydrocarbon fuels are needed to encourage future H₂ utilisation. One promising source of H₂ is coke oven gas (COG), which is a by-product in coke production, emitted at 300–350 N m³ per ton of coal [1]. The hot coke oven gas (HCOG) released from a coke oven is a multicomponent gas mixture, containing tarry compounds and steam in addition to H₂, CO, CO₂, and CH₄ [2–4]. The tar contains mono- and polycyclic aromatic hydrocarbons and comprises approximately 30 wt% of the HCOG [5]. In the conventional coke-making process, the HCOG is quenched to remove condensing components such as tar and water. The dry COG is used as fuel in steelworks and contains 54–59 mol.% H₂ and 24–28 mol.% CH₄ [6]. The amount of H₂ in dry COG can be amplified by catalytic [7,8] or non-catalytic [9,10] reforming by CO₂ or steam. Clean (tar-free) gas production by direct reforming of HCOG (without a cooling process) has been studied as an efficient process by utilising the HCOG heat [2,11,12]. The products from the partial oxidation of HCOG reforming are also suitable as feedstock for methanol production [13]. HCOG typically contains 0.3 wt% H₂S [11], which can deactivate the reforming catalysts. Catalytic deactivation by sulphur compounds [14,15] and coke [16–18] is unavoidable; these problems make non-catalytic reforming an attractive option for HCOG reforming.

Non-catalytic partial oxidation is conducted in a reactor by feeding HCOG and air in a sub-stoichiometric ratio, which results in temperature >1500 K [11]. The high temperature and steam produced by the partial oxidation process are used to sustain the endothermic reforming reactions [19,20]. There are many commercial and technical benefits of applying the numerical simulations based on the reliable reactor models, which helps in designing the reactor and optimizing the process operation. Prediction of the axial temperature profile without using empirical information is important for the design and operation of an exothermic partial oxidation process. Many flow reactor simulations have been carried out based on pre-existing/empirical temperature profiles [13,21,22] or with assumptions such as isothermal and negligible axial diffusion. For example, hydrocarbon pyrolysis experiments have been simulated numerically with a detailed kinetic model [23] coupled with a plug flow reactor model [21], where empirical temperature profiles were used as input for plug flow simulations. Sheng et al. [22] emphasised the importance of gas-phase kinetics in anode channel solid oxide fuel cells and used a model with an empirical axial temperature profile. Previously, we approximated the experimental axial temperature profile of a HCOG reformer as an empirical polynomial, validating it based on an empirical temperature profile with the assumption of negligible axial diffusion [13]. These empirical temperature profiles were developed based on limited temperature measurements and might introduce uncertainty in model predictions. Axial diffusion causes a degree of mixing in the axial direction in real reactors, the plug flow reactor model is appropriate when this effect is sufficiently small that it can be ignored. Diffusion can occur because of the concentration gradient in the axial direction. It is usually less important than bulk flow in most practical systems. However, the inclusion of axial diffusion in plug flow reactor models may give information about the deviation from ideality, which is generally present in real reactor systems [24].

The primary importance of this paper is that it estimates the axial temperature profiles for the partial oxidation of HCOG using a new numerical approach. The heat losses to the surroundings and non-ideality of the plug flow with axial diffusion are accounted for in numerical simulations. An existing kinetic model proposed by Richter and Howard [25] is used to simulate the pilot-scale HCOG reforming. The kinetic model consists of more than 2000 elementary steps, such as reactions, and is capable of accurately capturing phenomena that occur in the gas-phase reactions. The kinetic model is validated with experimental observations of the major components, such as H₂, CO, CO₂, and CH₄, and the total dry gas flow rate, in addition to the axial temperature profiles of 19 pilot-scale runs.

2. Pilot-scale test of HCOG reforming

A pilot-scale test plant for HCOG reforming with partial oxidation was installed on a platform of an operating coke oven at Kitakyushu city, Japan. The HCOG was collected from three coke chambers and introduced to the reformer at a temperature range of 625–665 K together with O₂ at room temperature. The HCOG flow rate was adjusted by the dampers, which were placed at the top of the coke oven chambers.

HCOG was fed into a horizontal cylindrical Section (0.6 m ID and 3.24 m long) at flow rates from 45 to 63.9 N m³/h and was partially oxidized by O₂ (from 14.2 to 19.4 N m³/h) from the four nozzles near the inlet. The reformer was operated at atmospheric pressure. Temperature profiles were measured with thermocouples inserted vertically at different positions inside the reformer. The gas compositions at the reformer inlet and outlet were measured using an online gas chromatography, and condensing products such as water and tar were sampled and analysed offline. The details of the pilot-scale test procedure have been reported elsewhere [13]. Pilot-scale measurements of 19 runs were used to validate the numerical approach.

The HCOG is a multicomponent gas mixture, particularly it contains around 30 wt% tar compounds that include various mono and polycyclic aromatic compounds. Due to this nature, reforming of HCOG by partial oxidation is a complex process. Chemistry and kinetics are essential to understand the complex reforming process. HCOG contains large fractions of CH₄, and H₂, and these species are participating in combustion reactions with O₂, thereby accelerating the reforming of the tar by steam. The water produced from the CH₄ and H₂ oxidations, and reverse water-gas shift reaction, in addition to these water formation reactions, HCOG also contains some fraction of H₂O, may induce reforming reactions. Most likely, these exothermic oxidation reactions were dominating at the reactor entrance, and later endothermic reforming reactions are taking place.

3. Modelling approach

A one-dimensional flow reactor model coupled with heat losses to the surroundings was used to simulate the HCOG reforming pilot-scale tests. The real reactor is shown schematically in Fig. 1. For the numerical simulations, the flow reactor was idealised as a 3.2-m-long tube reactor with a constant inside diameter of 0.6 m.

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