



Gasification kinetics of a bituminous coal at elevated pressures: Entrained flow experiments and numerical simulations



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HIGHLIGHTS

- Entrained flow gasification experiments with a bituminous coal.
- Focus on the impact of total pressure on conversion.
- Modeling approach based on measured intrinsic reaction rates.
- Taking into account thermal annealing and diffusion limitations.
- Good correlation of the simulation results with experimental data.

ARTICLE INFO

Article history:

Received 6 April 2016

Received in revised form 20 December 2016

Accepted 27 January 2017

Available online 7 February 2017

Keywords:

Entrained flow gasification kinetics

Increased total pressures

CFD modeling

Intrinsic reaction rates

Char particle conversion

ABSTRACT

Predicting conversion rates of solid fuels in entrained flow gasifiers, operating at increased temperatures and pressures, and better understanding the underlying reaction kinetics are of major interest for all industrial gasification applications. Numerical simulations in combination with experiments in lab- and pilot-scale entrained flow reactors help to understand the occurring reaction processes and can be used for gasifier optimization. The presented model is based on the software Ansys Fluent 16.0 and is validated for a bituminous coal with focus on the impact of total pressure. An *n*th order effectiveness factor approach with measured intrinsic reaction kinetics is applied in order to take diffusion limitations into account, and a thermal annealing model is included in order to account for the influence of decreased reactivities of the char surface due to deactivation, both being relevant at increased operating temperatures. The required model input parameters are derived from pyrolysis experiments and laboratory analyses. The simulation results are in good agreement with experimental data obtained from a pressurized entrained flow reactor operated at the Technische Universität München. The validation experiments are carried out at an operating temperature of 1200 °C, at total pressures of 0.5 MPa, 1.0 MPa and 2.0 MPa, and with a constant molar *O/C* ratio of one.

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1. Introduction

Entrained flow gasification processes transform a solid fuel into a combustible gas, composed primarily of carbon monoxide and hydrogen. This syngas can be converted to various chemical products or directly used for electricity production in an Integrated Gasification Combined Cycle (IGCC) [1]. Pressurized entrained flow gasifiers are used in IGCC power plants, because of the high power density, the production of a tar-free syngas as well as fuel flexibility [1]. The prediction of conversion rates for different feedstocks in

entrained flow gasifiers are of major interest, especially at operating conditions relevant to large-scale industrial applications, i.e. comparable heating rates, increased total pressures, and high operating temperatures (typically in the range of 1200–1600 °C) [2].

Numerical simulations in combination with experiments in entrained flow reactors help to understand the occurring reaction processes and can be used for the optimization of entrained flow gasifiers. The implementation of a detailed modeling approach for entrained flow gasification kinetics in a CFD code has only been reported by a small number of researchers [3–5], and many of the published simulations of entrained flow gasifiers lack validation against experimental data. This results from the fact that available validation test cases are scarce, in particular at increased and vari-

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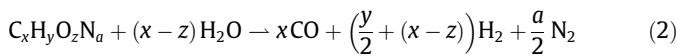
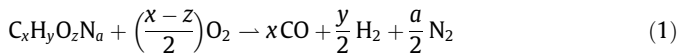
able total pressures. The modeling approach applied in this work has already been successfully validated for a lignite at 0.5 MPa [5]. The presented work is motivated by extending the validation domain for feedstocks of other rank and for higher total pressures, with focus on the impact of total pressure on conversion under entrained flow conditions. Therefore, a bituminous coal at increased total pressures is investigated. The presented validation test case and set of model parameters extend the available data, and can also be used by other researchers for model validation and simulation of entrained flow gasifiers at elevated pressures.

2. Modeling approach

This section includes a brief description of the modeling approach, with focus on the modeling of char reaction kinetics.

The CFD model is developed as a three-dimensional steady-state RANS analysis based on the software *Ansys Fluent 16.0* [6]. The two-phase mixture, i.e. gas flow and solid particles, is calculated with a Eulerian-Lagrangian approach. Turbulence-chemistry interaction is computed using the *Eddy Dissipation Concept*. Several submodels for char particle conversion are implemented by means of User Defined Functions (UDF). A detailed discussion of the applied submodels can be found in an earlier publication by the authors [5].

The key steps in a gasification process are particle drying, volatile release, and heterogeneous char reactions, with the heterogeneous gasification reactions being the rate determining step. These steps are calculated consecutively in the modeling approach. First, the injected fuel particles are heated up and the moisture content is released. The dry particle then undergoes pyrolysis and is converted to a char particle, assumed to only consist of solid carbon and mineral matter. The released volatiles are modeled as $C_xH_yO_zN_a$ molecules that are reformed by subsequent gas phase reactions, as shown in Eqs. (1) and (2). Pyrolysis kinetics is modeled in a simplified manner as a function of time and temperature by means of an empirical single-rate model, with rate parameters taken from the *Ansys Fluent* database [6]. Tar and soot formation are not explicitly modeled due to missing experimental data and an increased model complexity. However, it can play a role at the investigated operating conditions, in particular for bituminous coals [7]. Gas phase reactions are modeled using the global Jones-Lindstedt mechanism for hydrocarbon combustion [8], as shown in Eqs. (1)–(4). The reaction that primarily determines the gas composition in entrained flow gasifiers is the water-gas shift reaction (4) [2].



After pyrolysis, the solid carbon in the char particle is converted by heterogeneous reactions with O_2 , CO_2 and H_2O , shown in Eqs. (5)–(7).



Intrinsic reactivities of pyrolysis chars are used in combination with the modeling of diffusion limitations, i.e. pore diffusion within the particle and molecular diffusion in the boundary layer of the particle, in order to apply the measured intrinsic data (regime I) to higher temperatures (regime II and III). An nth order effectiveness factor approach is applied for this purpose [9] and the resulting reaction rate r_j is calculated for each char reaction as shown in Eqs. (8) and (9) [5].

$$r_j = (\eta_j A_{t,in,j} + A_{t,ex,j}) \cdot r_{intr,j} \quad (8)$$

$$r_{intr,j} = k_{0,j} \cdot \exp\left(\frac{-E_A}{R \cdot T_{p,j}}\right) \cdot p_{s,j}^n \quad (9)$$

Here, $r_{intr,j}$ is the intrinsic reaction rate, $p_{s,j}$ is the partial pressure of the reactant gas on the particle surface, n is the reaction order, $k_{0,j}$ is the intrinsic pre-exponential factor, $T_{p,j}$ is the particle temperature, R is the universal gas constant, E_A is the activation energy, η_j is the effectiveness factor, and j indicates the particle time step. The partial pressures on the particle surface $p_{s,j}$ are calculated for each particle time step by applying a mass balance for each reactant gas: the diffusion rate of the reactant gas through the particle boundary layer must be equal to the consumption rate of the reactant gas by the corresponding heterogeneous surface reaction. In regime I, the partial pressure inside the particle is equal to the partial pressure in the surrounding atmosphere. The specific char surface area $A_{t,j}$ is considered to consist of an internal part $A_{t,in,j}$ and an external part $A_{t,ex,j}$. The effectiveness factor η_j sets the diffusion limited rate in relation to the chemical reaction rate by reducing the available internal surface area $A_{t,in,j}$ depending on the reaction regime. It describes a concentration gradient of the reactant gas in the particle and is a function of the Thiele modulus [9]. The Thiele modulus depends on particle and pore structure, as well as on diffusion and reaction parameters [9]. As these values change individually for each particle over residence time, the effectiveness factor varies as well.

The power law approach is widely used for describing char reaction rates [3,5,10], although it is a simplification compared to Langmuir-Hinshelwood kinetics and should not be applied over a wide range of operating conditions, in particular if saturation and inhibition effects could be significant [11,12]. However, in this work the simpler power law approach is implemented in the CFD code, because the implementation of Langmuir-Hinshelwood kinetics combined with the modeling of pore diffusion is mathematically complex and leads to increased simulation times, in addition to larger experimental effort.

The structure of char particles changes during conversion. In this work, a recently developed pore structure model for entrained flow gasification is applied [5] that models char particles as microporous spheres with a uniform distribution of cylindrical pores and mineral matter. Furthermore, thermal annealing (or char deactivation), i.e. a decrease in reactivity of the char surface with time at high temperatures, is included in the model, taking into account the thermal history of each individual particle track. Thermal annealing is modeled by reducing the intrinsic pre-exponential factor $k_{0,j}$, as shown in Eqs. (10) and (11) [13]. In general, different explanations for thermal annealing are discussed in literature, e.g. the loss of catalytically active mineral matter, the loss of active sites on the char surface, or the graphitization of the carbon structure. The presented approach does not differentiate between these reasons, and only models the resulting effect on intrinsic reactivity.

$$k_{0,j} = k_{0,j,deact} \cdot [1 + f_{an,j} \cdot (r_{max} - 1)] \quad (10)$$

$$df_{an,j} = -k_{0,an} \cdot \exp\left(\frac{-E_{A,an}}{R \cdot T_{p,j}}\right) \cdot f_{an,j} dt \quad (11)$$

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