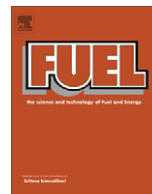




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Gasification kinetics during entrained flow gasification – Part III: Modelling and optimisation of entrained flow gasifiers

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HIGHLIGHTS

- ▶ A model framework for entrained flow gasification is presented.
- ▶ The model is validated using experimental data from various measurement setups.
- ▶ The reaction regime of char conversion in industrial scale gasifiers is analysed.
- ▶ At high temperature in the burner zone Regime III is approached.
- ▶ In the later stages the reaction occurs under Regime II and Regime I conditions.

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ABSTRACT

The paper presents a model framework that describes the reactions kinetics of entrained flow gasification. The model consists of different submodels for fuel devolatilisation, intrinsic char gasification, surface area evolution, char thermal annealing, pore diffusion, boundary layer diffusion, and particle size and density variation. These submodels are partly derived from measurements in previous publications and are partly based on theoretical derivations. The model framework is validated using fuel devolatilisation data and char gasification kinetics of a lignite that are measured in two entrained flow reactors at temperatures up to 1600 °C and pressures up to 1.0 MPa. A good correlation of model and experimental data is found for various operating conditions.

The objective of the model development is the prediction of gasification behaviour in larger scale entrained flow gasifiers. To show its capabilities the model is applied to a one-dimensional plug flow reactor that represents a simple approach for a larger scale entrained flow reactor. Gas and temperature profiles within a 500 MW gasifier are predicted and cold gas efficiency and fuel conversion for different gasifier geometries and operating conditions are shown.

Based on the model predictions the reaction regime of char conversion in entrained flow gasifiers is analysed. At very high temperature in the burner zone, char gasification approaches Regime III conditions and a concentration gradient develops in the gas boundary layer around the char particles. The observed activation energy is 62 kJ/mol. In the medium stages of char conversion the reaction occurs under Regime II conditions and then approaches Regime I conditions at high conversion and at lower temperature. This is indicated by the high observed activation energy of 239 kJ/mol in the final stages of conversion.

As all three reaction regimes are relevant, fuel properties that have an impact on diffusion (e.g. particle size and pore structure) as well as on the intrinsic char reaction (e.g. intrinsic reactivity and thermal annealing) have to be considered to determine entrained flow gasification kinetics.

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1. Introduction

Entrained flow gasification technology offers a wide range of applications for fossil as well as renewable energy sources. Solid fuels are converted at high temperature and high pressure and the

product gas can be used for power generation or as a raw material for chemical syntheses to produce liquid or gaseous fuels and chemical products. If CO₂ capture is required, a very efficient pre-combustion separation process is integrated. The possible decoupling of gasifier and final product output allows the installation of energy storage capacities that might be required due to the integration of fluctuating renewable energy sources into the electricity grid.

Entrained flow gasifiers are the most important gasification technology. According to the 2010 Worldwide Gasification

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Database [1] they account for most of the capacity installed worldwide. Solid fuels are converted at pressures of up to 7.0 MPa and temperatures in the range of 1400–1600 °C by the addition of oxygen to the reaction zone [2]. After devolatilisation, the volatiles are combusted in homogeneous gas reactions and heat is supplied for the subsequent endothermic heterogeneous char conversion. Char gasification processes with H₂O and CO₂ are the slowest reactions and determine the rate of the overall conversion process [3–5].

Since char properties influence its conversion, it is important to understand typical char properties such as composition, pore structure, morphology, and surface area [6]. The reactivity of char depends on total surface area, the number of reactive sites per unit surface area and the local gaseous reactant concentration [3]. The conversion rate of char is influenced by the intrinsic reaction rate, but also by mass transport limitations within the char porous structure and the boundary layer. For a given coal char, the degree of limitation by chemical reaction or diffusion is mainly a function of reaction temperature. The dependence of the reaction rate on temperature is usually shown as a plot of reactivity (logarithmic) versus temperature (inverse), as shown in Fig. 1. This plot is generally referred to as the Arrhenius plot and the influence of temperature is shown by the slope of the curve that gives the activation energy of reaction.

The relation between chemical reaction and mass transport limitation is classified in three regimes. In Regime I at low temperature the chemical reaction rate at the char surface is very slow and determines the overall reaction rate. The gas concentration is uniform within the char particle and the bulk gas phase. With increasing temperature the chemical reaction rate increases exponentially and becomes comparable to the pore diffusion rate within the particle. With increasing reactant consumption at the inner particle surface a concentration gradient develops within the char particle. Under ideal Regime II conditions, the reactant concentration is equal to the bulk concentration at the outer particle surface and zero at the particle centre. When the temperature is increased further, the overall rate is controlled by the mass transfer between bulk phase and particle surface and a concentration gradient is formed in the boundary layer. As diffusion processes are only slightly influenced by temperature, the slope of the curve approaches zero.

Measurements of reactions rates at low temperature under Regime I conditions are usually derived from thermogravimetric analysers, and char conversion rates of different fuels are available (e.g. [7–10]). At higher temperature, the char gasification has to

be measured in flow reactors under reaction conditions that are relevant to industrial scale entrained flow gasifiers. Only a few data sets are available that analyse char gasification at temperatures above 1000 °C.

Ahn et al. [11] investigated the gasification rate of an Indonesian sub-bituminous coal–char with CO₂ using a pressurised drop tube furnace (PDTF) at the Korea Electric Power Research Institute. Gasification rates were measured from 900 °C to 1400 °C and at 1.0 MPa reactor pressure. From 900 °C to 1000 °C the activation energy was determined to be 144 kJ/mol. At higher temperature the activation energy decreased to 71.5 kJ/mol which indicates the occurrence of mass transport limitations. The transition from Regime I to Regime II conditions occurred between 1000 °C and 1100 °C.

At the Central Research Institute of Electric Power Industry (CRIEPI), Japan, a pressurised drop tube furnace has been installed to analyse the gasification rate of coal char [12,13]. The facility was operated at up to 1500 °C and at up to 2 MPa. For H₂O gasification of the NL coal char the transition temperature (Regime I–Regime II) is 1300–1400 °C, whereas it is 1200 °C for CO₂ gasification. For the S coal char no transition in CO₂ is observed up to 1200 °C.

Matsumoto et al. [14] measured reactions rates of biomass char with CO₂ and H₂O in a pressurised drop tube furnace at the Nagasaki Research and Development Center, Japan. The PDTF was operated at a total pressure of 0.4 MPa and at temperatures from 900 °C to 1300 °C. The measured activation energies were 136 kJ/mol for the H₂O reaction and 94 kJ/mol for the CO₂ gasification, both indicating the conversion under Regime II conditions.

Coal gasification at a pressure of up to 2.0 MPa and temperatures of up to 1500 °C was measured in a pressurised entrained flow reactor at CSIRO [15,16]. The activation energies measured at 0.5 MPa CO₂ partial pressure and a total pressure of 2 MPa were 78–156 kJ/mol. These were considerably lower than the activation energies of 242 kJ/mol to 281 kJ/mol that were measured for extracted chars under Regime I conditions in a TGA and a fixed bed reactor. The transition temperature from Regime I to Regime II occurred around 1000–1100 °C dependent on char morphology.

Other experimental gasification studies are carried out in atmospheric reactors [17–19]. These measurements do not focus on the evaluation of the reaction regime and a transition temperature to Regime II is not given.

The experimental data are limited to pressures of up to 2.0 MPa and temperatures of up to 1500 °C. A transition to Regime III is not reported under these conditions. Therefore, it is assumed that char conversion occurs under Regime II conditions in entrained flow gasifiers, as shown in Fig. 1. Since the reaction temperature is usually above 1500 °C on the industrial scale, especially in the burner zone of the gasifier, a transition to Regime III is possible and has to be considered when modelling entrained flow gasification.

The objective of the paper is to measure gasification kinetics at high temperature and high pressure and extend the operation parameter range that is reported in the literature. The experimental data are combined with devolatilisation and low-temperature char gasification measurements that are available from previous publications [4,5]. Based on the experiments, a gasification model is derived and validated that describes entrained flow gasification in all three reaction regimes. Finally, the model is used to discuss the reaction behaviour within industrial scale gasifiers.

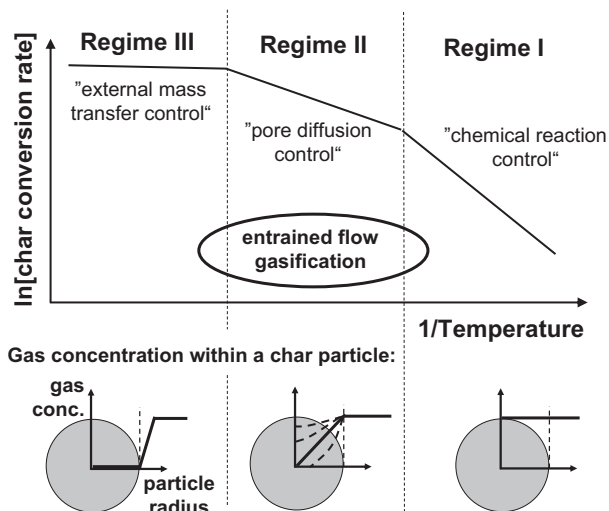


Fig. 1. Influence of the reaction temperature (inverse) on the char conversion rate (logarithmic) and on gas concentration gradients within a porous char particle.

2. Experimental methods and results

2.1. Fuel preparation

The proximate and ultimate analysis of the lignite (coal R) that is used in the analysis is shown in Table 1. The fuel is pulverised

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