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Full Length Article

Simultaneous investigation into the yields of 22 pyrolysis gases from coal and biomass in a small-scale fluidized bed reactor

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HIGHLIGHTS

- Provision of well stirred environment and high heating rates in the FBR.
- Simultaneous measurement of 22 pyrolysis gas species with FTIR technology.
- Extensive database with temperature dependent mass yields for numerical simulations.
- Comparison between bituminous coal and torrefied biomass.
- Covering the temperature range 1023–1273 K, where data for biomass is scarce.

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ABSTRACT

Flash pyrolysis gas yields from bituminous coal and torrefied biomass have been investigated in the temperature range between 873 and 1273 K for 22 different gas species simultaneously. Recording a larger number of species gives a more detailed insight into pyrolysis of solid fuels than it is provided by other studies in literature, where typically smaller numbers of species are tracked. As processes utilizing biomass are designed for lower process temperatures, literature data for biomass pyrolysis are scarce in the temperature range between 1023 and 1273 K. This work, aiming at investigating potential fuels for pulverized solid fuel combustion, approximates pyrolysis conditions in the ignition zone of actual power plant boilers, where possible.

Thus, based on results of a literature review, an experiment was designed using a small-scale fluidized bed reactor (FBR) to simultaneously analyze the pyrolysis gas yields for 22 species and to close the temperature gap found for biomass. Small batches of pulverized torrefied biomass from poplar wood and pulverized Colombian bituminous coal (Mina Norte) were rapidly pyrolyzed in the reactor. The two fuels were chosen as exemplary conventional and alternative energy sources for pulverized fuel fired power plants. Experiments have been carried out under atmospheric pressure in pure nitrogen atmosphere.

Product gas analysis was carried out by Fourier transform infrared spectroscopy (FTIR) including two different sampling techniques. As a novelty, offline measurements have been used exclusively to improve the gas species selection procedure and reliability of detection. In case of offline measurements, the gas flow to the FTIR gas cell is stopped and the gas is enclosed in the cell allowing longer scan times of the captured gas mixture and thereby enlarging the signal-to-noise ratio (SNR). In succession, regular measurements with continuous flow (online) were performed for the simultaneous quantitative analysis of total pyrolysis yields for 22 selected gas species.

Obtained pyrolysis yields show good agreement with available literature data. The larger number of investigated species provides an extended data set also covering minor pyrolysis species, for which quantitative data are scarce. Additionally, trends in the gas yields for biomass agree well with those found by other authors outside the mentioned temperature gap between 1023 and 1273 K.

1. Introduction

In the context of this paper, the expression "pyrolysis" is used for the process of heat treating solid fuels under non-oxidizing conditions. The fuel is chemically and physically transformed in the

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process, resulting in a dry, volatile depleted, carbon rich residue which is designated as char. Data on pyrolysis mass release is employed in numerical simulations of coal and biomass combustion and gasification, where it forms the basis for the validation and improvement of computational models. Especially for determining the heat release in volatile combustion and the formation of pollutants, a quantitative knowledge of the evolved pyrolysis products in the most detailed way possible is evident. This work aims to provide such data on pyrolysis mass release for one typical and one alternative fuel used in pulverized fuel (PF) fired power plants.

In literature, the main instruments used for the experimental investigation of pyrolysis yields are entrained flow or drop tube reactors (EFR|DTR) [1–8] and thermo-gravimetric analyzers (TGA) [9–11]. Aside from these popular methods, a wide range of other reactor concepts can be found, e.g. wire mesh reactors (WMR) [11–13], fixed bed reactors [5,14] and fluidized bed reactors (FBR) [5,15–18].

For each of the studies cited, the degree of detail in the results presented varies. This can also be seen in Table 1, where an overview of selected pyrolysis experiments with coal and biomass is given. Summarizing Table 1, it is clear that a lot of work has been undertaken on the detailed investigation of char yields, its elemental composition as well as tar and total gas yields. Furthermore the main light gas components CO_2, CO, H_2O, CH_4 and H_2 were topic of a number of publications [1–4,6,9,12,16,20,21]. A special focus on nitrogen compounds can be found in [6,14,20,24–26] and on sulfur compounds in [10,15,24], respectively. Overall, all studies focus on mass specific residues of the pyrolysis process. If gaseous products are investigated, the studies usually are either focused on the small cluster of main light gases mentioned, or on an equally small group of trace gas components. For pyrolysis in nitrogen atmosphere, the largest number of simultaneously analyzed gas components was nine in the studies of Dupont et al. [2] and Septien et al. [4].

The sum of 22 analyzed gases in the present study (cmp. last column of Table 1) exceeds the numbers of all other studies listed and thus, seems to represent a novelty in pyrolysis research. With the exception of H_2, which is not IR-active, and H_2S which shows only a very weak IR-absorption, all relevant gas species are analyzed.

Additionally, yield and composition of char is determined with the fluidized bed by burning the solid residue. An analysis of tars is included, but limited to species with a medium molar mass, which, at the present partial pressure, have a condensation temperature below 453 K. Investigations were undertaken for Colombian bituminous coal and torrefied biomass from poplar wood at temperatures from 873 K to 1273 K under atmospheric pressure in a pure nitrogen atmosphere. Thereby a distinct gap in literature data concerning biomass pyrolysis yields within the range from 1023 to 1273 K [2,4,27] can be covered with the present reactor setup. Both fuels have been investigated under the same conditions. A separate investigation of the low-temperature pyrolysis of biomass below 773 K has not been undertaken, as the aim of the experiments was to approximate conditions in the ignition zone in PF fired boilers as far as the experimental setup allowed.

The small scale fluidized bed reactor employed in this study (cf. Fig. 1) is suitable for the analysis of pyrolysis yields, including primary and secondary reactions, of all solid fuels as well as reaction rates of char. The reactor allows high heating rates of 10^4 K/s (analytically approximated) [28], an unlimited residence time of fuel particles and the investigation of pyrolysis and gasification reactions in oxygen-free environments. Different atmospheres (Ar, N_2, CO_2) with O_2 concentrations between 0 and 50 vol.% can be applied and have already been investigated [28,29]. Further on, pyrolysis and char burnout can be investigated separately. Carrier gas and reaction products are monitored by either online or offline FTIR gas analysis. The usage of FTIR spectroscopy allows for the simultaneous detection of all species except H_2 with one single measurement technique. The novel application of an offline analysis method increases SNR and allows for a more reliable selection of detectable gas species.

The results of this study are presented in the most detailed way the analysis system allows for, giving concentrations and yields for single gas species. This simplifies and encourages comparisons to other experiments and to results of numerical pyrolysis models. At first, the experimental setup and the fuels investigated are described in Sections 2.1 and 2.2. Afterwards, the evaluation procedure for the calculation of mass specific pyrolysis yields is described in detail in Section 2.3. In the error analysis in Section 2.4, possible errors are discussed and their impact on pyrolysis yields is estimated. The yields are presented in Section 3 combined with a detailed comparison to literature data.

2. Experimental investigation

The experimental setup consists of three major components: a gas feeding system with thermal mass flow controllers, a small-scale fluidized bed reactor (FBR) with an inner diameter of $d = 55$ mm and a Fourier transform infrared (FTIR) spectrometer for product gas analysis. The reactor is designed to implement the concept of a well stirred reactor approximating uniform distributions of thermodynamic state variables as well as of reacting species. Small batches of pulverized solid fuel (coal and biomass) are supplied to a fluidized bed consisting of aluminum oxide (Al_2O_3) particles while simultaneously analyzing the gaseous pyrolysis products of the exhaust gas.

2.1. Experimental setup

The fluidized bed reactor shown in Fig. 1 consists of two coaxial ceramic pipes mounted in a stainless steel reactor head.

The fluidizing gas is fed in through the reactor head and flows downwards through the annular gap between the two pipes to the bottom of the reactor.

The gas enters the inner reactor pipe ($d = 55$ mm) through a porous distributor plate made from sintered silica glass and fluidizes the bed. The distributor plate with a typical pore size between 40 and 100 mm creates a homogeneous inflow and is impermeable for the Al_2O_3 bed particles, which are sieved to a mode of 115 mm, representing the particle size with the most frequent appearance. Discrete values for describing the underlying particle size distribution function are $d_{10} = 57$ μm, $d_{50} = 100$ μm and $d_{90} = 141$ μm, where the given particle size is the upper limit for mass fractions of 10%, 50% and 90% respectively. The bed height is approximately 30 mm in non-fluidized state and increases to about 70 mm under typical flowing conditions. Details on homogeneity, mixing and fluidization conditions of the bed can be found in [28]. The feed gas flow for the present experiments is 50 slph and consists of nitrogen with a purity of 99.998%. It is regulated by Vögtlin red-y thermal mass flow controllers with a precision of ±1.0% FS (full scale) and a stability of ±0.2% of the nominal value.

All ceramic parts of the reactor are located in an electrically heated furnace which can be operated up to 1553 K with a stability of ±2 K over 24 h. The bed temperature is measured separately by an S-type thermocouple placed in a ceramic shielding 50 mm above the distributor plate inside the fluidized bed.

Small amounts of fuel (15.0 ± 0.5 mg) are introduced via a lock and a vertical ceramic pipe into the reactor. The pipe was flushed with 10 mL of gas from an injection, which was filled with feed gas before, to avoid particle sticking to the feeder pipe walls and