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Full Length Article

Development of a rapidly responding fluidized bed reactor by theoretical and experimental evaluation of combustion reactions

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ABSTRACT

The development of a rapidly responding lab-scale fluidized bed reactor (FBR) optimized for the investigation into combustion reactions of solid fuels is presented. The experimental setup represents a novelty in FBR systems, as it quantitatively captures reactions with an apparent 90% carbon conversion time t_{90} of < 3 s, which is a third of the time of comparable setups described in literature. The reactor design is discussed in detail and improvements undertaken to shorten the systems' response time are explained. The shortened response time results in the possibility to investigate reactions at higher temperatures.

Original and improved design are modeled with a 1-D finite volume approach to gain insights into the ideal operating conditions and operating limits. In a second step, the theoretical reaction rate limits are compared with experimental combustion experiments. Reaction rate measurements with chars from two pulverized Colombian coals (Calenturitas, Mina Norte) and one pulverized biomass (beech wood) are undertaken in N_2/O_2 atmosphere at different temperatures ranging from 823 to 1273 K.

It is found that results of the 1-D simulation are a good guidance for actual experiments.

1. Introduction

Lab-scale fluidized bed reactors are a versatile tool for combustion experiments of solid fuels [1–11]. As an implementation of a well-stirred reactor, approximating perfectly-stirred conditions, small fluidized beds provide close to quasi-constant and identical boundary conditions for all reacting particles, which simplifies some steps of the data evaluation process [12].

The main advantages of the presented fluidized bed reactor (FBR) are high particle heating rates (approximately 10^4 K/s [12]), unlimited residence time, flexible fluidizing gas composition (N_2, CO_2, CO, O_2, Ar, up to three gases simultaneously), the ability to close the carbon mass balance and to measure rates of reaction across five orders of magnitude ($5{\cdot}10^{-5}$–$5{\cdot}10^{0}$ 1/s). The batch-operated reactor combines external electric heating with ex-situ gas analysis by Fourier-transform infrared (FTIR) spectrometry.

However, the FBR concept also exhibits some limitations, foremost the response time. The latter constitutes an important limitation of most FBR setups compared to other reactor concepts like entrained flow type systems. The response time is defined as the time it takes the signal in the measurement instrument to increase from 0 to a fixed fraction (typically 50 or 90%) of its original sampled value when a Heaviside function of the measured quantity is applied at the sampling location. In gas analysis, this time is typically close to zero for in situ type measurement systems like laser absorption spectroscopy, but not for those of ex-situ type. While in situ optical gas analysis has advantages in this regard, it is difficult to realize inside fluidized beds because of the bed material which can be very abrasive and blocks the optical path through the reactor in the zone of interest. FBR reactor concepts therefore typically rely on ex-situ gas analysis [1–13]. In this case, response time is dominated by gas exchange in the reactor, piping and measurement instrument.

Table 1 shows an overview of different studies investigating char combustion reactions in fluidized bed reactor systems. The studies can be divided into two particle size groups. The group of large particles includes studies with particle sizes larger than 1000 µm, which is typical for fluidized bed combustors, while the group of small particles (74–120 µm) represents particle sizes for pulverized fuel combustion. With up to 1223 K bed temperature and 20.9% O_2 in the experimental investigation of Saucedo et al. [7], the typical range of operating conditions for fluidized bed combustors is covered. In contrast, the investigated temperatures up to 1173 K in the small particle group are below typical operating temperatures of pulverized fuel boilers. Investigations at higher temperatures therefore would be preferable, but cannot be found in literature. Especially, the transition from kinetic controlled regime I to pore diffusion controlled regime II is of certain

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Table 1
Overview of char combustion investigations in fluidized bed reactor setups.

		Study	Fuel	Particle size d_P [µm]	Temperature T_{max} [K]	Atmosphere	App. conv. time t_{min} [s]
Particle size group	large	Morin et al. [1]	Biomass char	4000 – 7000	643	Air	10000
		Bu et al. [2]	Wood char	6000	1088	40 % O_2 in N_2	≈ 100
		Scala and Chirone [3]	Bituminous coal char	6000 – 7000	1123	8 % O_2 in CO_2	≈1500
		Wang et al. [4]	Bituminous coal char	4000	1123	10 % O_2 in N_2	≈800
		Konttinen et al. [5]	Coals and biomass	3000 – 6000	1123	5 % O_2 in Ar	≈40
		di Celso et al. [6]	Wood char	5000	1173	Air	≈60[1]
		Saucedo et al. [7]	Lignite char	1000 – 1400	1223	20.9 % O_2 in CO_2	≈ 40
	small	Fang et al. [8]	Chinese coal chars	74 – 100	1003	20 % O_2 in Ar	≈90
		Guo et al. [9]	Herb residue	80 – 120	1073	20 % O_2 in Ar	8.9 [1]
		Fennell et al. [10]	Coal char	75 – 106	1173	20 % O_2 in N_2	≈8

[1] Based on CO_2 signal only.

interest. A transition from regime I to II for pulverized fuel particles (≈100 µm) is typically expected to occur at or above the maximum investigated temperatures, which makes investigations up to higher temperatures necessary. For the design of experimental setups used for determination of combustion kinetics at higher temperatures, the minimum apparent conversion time t_{min} can be used as a guideline.

The apparent conversion time describes the elapsed time until the signal has completely passed the ex-situ measurement device. This time depends on the chemical reaction itself and transport processes from reaction zone (fluidized bed) to analyzer. To ensure the reliable measurement of kinetic parameters, effects of gas transport and analyzer precision have to be taken into account. Transportation of gases from reaction zone to the analyzer can be characterized with the response time of the experimental system. The shortest apparent conversion times (40 s) in the large particle group are obtained by Konttinen et al.[5] and Saucedo et al. [7], which are still expected to be higher than the response time of the system. In contrast, minimum apparent conversion times in the small particle group (8 s) come close to the response time of the experimental systems. This observation was also remarked by Fennell et al. [10]. This means, that investigations at higher temperatures will result in actual conversion times below the response time. In this case, observed time dependent concentration curves of burnout products originate mostly from the applied gas exchange rate in the system and not from the reaction chemistry itself. Estimation of kinetic data from those profiles will result in misleading parameters, if no correction of those effects is considered. This also restricts the usable temperature range, as for combustion, higher temperatures result in lower conversion times. To overcome those limitations, constructional changes are required to reduce the response time.

The purpose of this work is to present an experimental setup for a fluidized bed reactor which is capable of measuring reactions with an apparent 90% carbon conversion time of $t_{90} < 3$ s and to show which modifications have been undertaken to achieve this goal. In the first section the experimental setup is presented and modifications are listed which were undertaken to improve the response time of the system. In the second section, a detailed analysis of the system's limitations regarding flow rate, sample mass and reaction rate is given. Optimum operating points within those boundaries are calculated based on a maximization of the signal-to-noise ratio (SNR). The final section summarizes the gained insights.

2. Experimental investigation

In the following two subsections, an experimental setup of FBR type designed to implement the concept of a well stirred reactor is discussed in detail. First, the original reactor is presented and afterwards, improvements made to the setup to shorten its response time are illustrated.

2.1. Original experimental setup

The fluidized bed reactor consists of three major components: a gas feeding system with thermal mass flow controllers, a small-scale fluidized bed reactor inside an electrically heated furnace shown in Fig. 1a and a Fourier-transform infrared (FTIR) spectrometer to analyze exhaust gas compositions. As reactor, two coaxial ceramic pipes are mounted in a stainless steel 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. During this passage, it is heated to oven temperature and then flows through a sintered silica glass distributor plate into the inner pipe (d = 55 mm), where it fluidizes the Al_2O_3 bed. This distributor plate with a typical pore size between 40 and 100 µm creates a homogeneous inflow and is impermeable for bed particles.

The bed height is 30 mm in non-fluidized state and increases to approximately 70 mm under typical experimental conditions. Details on homogeneity, mixing and fluidization of the bed have been presented in [12]. Two thermal mass flow controllers (model Vögtlin red-Y) are used to regulate the inflow of N_2 and O_2. The controllers operate with a precision of ± 1% of 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 1573 K with a stability of ± 2 K. 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 batches (1–15 mg) of pulverized solid fuel (char from coal or biomass) are introduced through a lock and a vertical ceramic pipe (d = 6 mm) into the reactor. The fuel is transported onto the fluidized bed by gravity.

The particle heating rate has been analytically approximated to 10^4 K/s [12]. To promote fast and homogeneous mixing of fuel and bed particles over the entire bed height and to prevent the less dense fraction from being entrained in the outflowing gas stream, a similar Archimedes number for fuel and bed particles is aspired. Therefore, the required particle size of the bed material is calculated based on the fuel size and density. A sieved Al_2O_3 fraction with fitting size distribution is then chosen for the experiment.

All gases (product gases of the reaction and fluidization gas) flow through a sampling line (d = 6 mm) and a filter into the measurement cell of the FTIR spectrometer. The entire sampling system is heated to 453 K to prevent condensation. Due to pressure loss in the sampling line, filter and FTIR gas cell, the reactor is operated slightly above atmospheric pressure. The absolute pressure for a single experiment depends on ambient pressure and differential pressure, which itself is a function of volume flow, reactor temperature and gas composition. Absolute pressure fluctuates by less than ± 0.2 kPa during a single experiment and is accounted for during evaluation. For all experiments