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Full Length Article

Measurement of reaction rates for pulverized fuel combustion in air and oxyfuel atmosphere using a novel fluidized bed reactor setup

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HIGHLIGHTS

- A small-scale, fast fluidized bed reactor (FBR) setup is presented.
- Combustion & gasification kinetics for pulverized coal char are determined.
- The activation energy is nearly identical for N_2/O_2 and CO_2/O_2 atmosphere.
- The fastest measured reactions show a 90% carbon conversion time <3 s.
- The results of the FBR compare well with EFR and TGA type systems.

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ABSTRACT

The reaction rate of char from pulverized Columbian coal (Mina Norte) is investigated in synthetic air (N_2/O_2), oxyfuel atmosphere (CO_2/O_2) and CO_2/N_2 using a lab-scale fluidized bed reactor (FBR). Reactor temperatures range from 823 to 1273 K for combustion and 1173 to 1373 K for gasification (Boudouard). The oxygen volume concentration is varied between 15 and 30 vol.%, while gasification is investigated in a mixture of 10–75 vol.% CO_2 in nitrogen. Using an *n*th order Arrhenius approach, activation energies as well as apparent order of reaction are calculated for the combustion and gasification reactions.

It is found that the combustion reaction with this particular fuel evolves between +17% (873 K) and +75% (1223 K) faster in N_2/O_2 than in CO_2/O_2. The results of Arrhenius fit suggest that activation energy of combustion reaction does not differ significantly between synthetic air (regime I: 120.9 kJ/mol, regime II: 62.9 kJ/mol) and oxyfuel atmosphere (116.6 kJ/mol, 64.3 kJ/mol). Comparing results for oxyfuel and air, a difference of approximately 50 K in the transition temperature from regime I to regime II is observed but this finding is not statistically firm, yet. The apparent order of reaction has been calculated to $n = 0.72$ in air (combustion), $n = 0.66$ in oxyfuel (combustion) and $n = 0.49$ in CO_2/N_2 (gasification).

A comparison with available literature data confirms that the results achieved with the fluidized bed are comparable to the two most common experimental setups used in combustion research: Entrained flow reactors and thermogravimetric analyzers. The experimental setup also 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.

1. Introduction

Numerical combustion models for solid fuels utilize a simplified approach to describe a complex process, mostly divided into the four steps of particle heat-up, volatile release, homogeneous volatile combustion and heterogeneous char combustion [1]. While all steps are required for simulations of solid fuel flames, the time-determining step of this mechanism chain is the char burnout, which is also required to estimate the carbon in ash content. There is a variety of burnout models available, most of which require at least one experimentally derived characteristic value of the fuel as input [2–5]. Hence, most detailed numerical modeling approaches rely on data from char combustion experiments.

Lab scale char and coal combustion experiments are usually performed in plug flow reactor systems and thermogravimetric analyzers [6–8]. Plug flow reactors can further be grouped into entrained flow [5,9,10] and drop tube [7,11,12] type systems. Wire mesh [1] and fixed bed reactors [13] can also be found in literature, but are less commonly used.

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Nomenclature

Latin

E_a	activation energy J mol^{-1}
d	diameter m
n	order of reaction –
q_3^*	logarithmic particle size density function –
R_S	swelling ratio –
t_{90}	time to 90% conversion s
X	carbon conversion (burnout) –
t	time s
M	molar mass kg mol^{-1}
$\dot{n}$	molar flow mol s^{-1}
c	concentration vol.%
$\dot{m}$	mass flow kg s^{-1}
r	characteristic reaction rate s^{-1}
F	convolution transfer function
V	volume m^3
A	frequency factor s^{-1} Pa^{-n}
p	pressure Pa
R	universial gas constant J mol^{-1} K^{-1}
T	temperature K
R^2	coefficient of determination –
m	mass kg
c_p	specific heat capacity J kg^{-1} K^{-1}
h	specific enthalpie J kg^{-1}
A	area m^2
Nu	Nusselt number –
Pr	Prandtl number –
Re	Reynolds number –
l	length m

Greek

Δ	difference
α	coefficent of convolution transfer function –
α	heat transfer coeffient W m^{-2} K^{-1}
β	coefficent of convolution transfer function –
ρ	density kg m^{-3}
ξ	mass fraction kg kg^{-1}
σ	standard deviation
σ	Stefan–Boltzmann constant W m^{-2} K^{-4}
ε	emissivity –
ϵ	gas phase volume fraction –

Subscripts

C	carbon
char	char
coal	raw coal
exp	experimental
0	initial
10	10%
50	50%
90	90%
(s)	solid
tot	total
prod	product
URM	uniform reaction model
conv	convoluted
i	component i, timestep i
j	component j
Reg. II	regime II
Reg. I	regime I
p	particle
min	mineral matter
b	bed
max	maximum
mean	mean
dyn	dynamic
const	constant

Fluidized bed (FBR) setups are less popular than plug flow type systems within this branch of combustion research, but still more commonly employed than wire mesh or fixed bed reactors. Experimental FBR setups for investigating char combustion have been used by Fennel et al. [14,15], Scala and Chirone [16], Senneca and Cortese [8] or Saucedo et al. [17]. Bews [18] used an FBR to determine the reaction rate of pure graphite particles, comparable in size to those of this study, and Carvalho [19] and Luo et al. [20] investigated the pure Boudouard reaction using fluidized bed setups of similar dimensions as the one employed in this work. The main advantages of externally heated fluidized bed systems compared to plug flow reactors are the ability to provide an H_2O- and CO_2-free reaction environment and an unlimited residence time of fuel particles in the bed. In contrast, fluidized beds are more limited with respect to the achievable maximum operating temperature in comparison to entrained flow or drop tube reactor systems. Furthermore, the presence of inert bed particles complicate in situ measurements. Consequently, fluidized beds for combustion research are typically coupled with ex situ gas analyzers. Thereby, the largest measurable rate of change for reactions in the reactor is limited by the largest time constant of the measurement setup, which is usually determined by the gas exchange rate in the reactor. For currently known fluidized beds, this time constant is least one order of magnitude larger than the reaction times of char particles from pulverized fuel: The fastest observed reactions show an apparent 90% carbon conversion time t_{90} of ≈10 s in the analyzer. The actual combustion reaction, however, is expected to last ⩽1 s under the investigated conditions.

For the present work, a lab scale fluidized bed reactor (FBR) system optimized for fast response behavior combined with high speed Fourier-transform infrared (FTIR) spectroscopy is used to investigate the char conversion process. The presented setup successfully reduces the aforementioned disadvantages of this reactor type by increasing the gas exchange rate. It delivers reliable results up to at least 1273 K. The quickest reactions investigated to date show an apparent 90% conversion time t_{90} of 3 s in the FTIR spectrometer, which is roughly three times as fast as any other existing fluidized bed combustion analyzer setup. Further, the online FTIR gas analysis, controlled inlet mass flow and known sample masses allow for a full mass balance of all analyzed reaction products including pollutants like NO_x, SO_2, CS_2 or COS. These aspects are – in combination – unique for FBR systems and represent a novelty in combustion research.

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 to analyse exhaust gas compositions. The reactor is designed to implement the concept of a well stirred reactor with uniform distributions of thermodynamic state variables as well as of reacting species within the entire reactor. Small batches of pulverized solid fuel (coal or biomass) are fed to a fluidized bed consisting of aluminum oxide (Al_2O_3) particles.