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Experimental investigation of the flue gas thermochemical composition of an oxy-fuel swirl burner

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ABSTRACT

The level of understanding of coal combustion compared to the requirements of predictive engineering is still insufficient due to the complexity of the processes and breadth of scales involved. In particular, in modern oxy-coal firing, where the fuel is burned in an oxygen/carbon-dioxide atmosphere to reduce the emission of greenhouse gases and pollutants, both experimental investigations and detailed numerical simulations are sparse. To understand and model the phenomena involved, the community follows a stepwise approach from generic to close-to-application combustion systems by steadily increasing the size, complexity and thermal power of the investigated burners as well as the complexity of the fuel. Here, an investigation of the flue gas of an intermediate range oxy-fuel burner is presented. To simplify the fuel in a first step, the combustion of methane in air and two oxygen/carbon dioxide atmospheres (oxy-fuel) is investigated. The concentrations of multiple species as well as the path-integrated temperatures are measured using three independent spectrometer systems based on tunable diode laser absorption spectroscopy (TDLAS). The rate of burnout and fuel-slip in the flue gas is studied through measurements of the concentration of CH_4, whereas a quasi-simultaneously applied OH-measurement system allows for the detection of remaining combustion zones in the flue gas. An acetylene (C_2H_2) measurement as a chemical precursor provides insights to the formation of soot. The O_2-concentration is measured for an investigation of the oxygen-excess, and, in combination with measurements of the CO, CO_2 and H_2O concentration at various positions allows to detect the main components and pollutants of the oxy-fuel flue gas. An Allan-Werle variance is utilized to study the time scales occurring in the combustion process.

A quasi-simultaneous measurement of the path-integrated H_2O and CO_2 temperature allows for investigating the progression of mixing of the different flows within the combustion chamber. This measurement reveals, that a complete mixing of all flows is only achieved close to the outlet.

1. Introduction

The combustion of fossil fuels for power generation, in particular of coal with its high carbon content, is one of the major sources of the anthropogenic greenhouse effect through the emission of carbon dioxide (CO_2). Even though the global share of renewable energies on the overall power generation is on the rise, the combustion of fossil fuels continues to grow and will continue to give an important contribution to power generation [1]. To reduce the emissions of CO_2 of existing and future coal and gas power plants, the approach of firing fossil fuels under oxy-fuel atmosphere is a promising technique. During oxy-fuel combustion, air is replaced by a mixture of oxygen (O_2) and CO_2 as oxidizer. As the flue gas of this process is mainly composed of CO_2 and water vapor (H_2O), the CO_2 can be easier separated, captured, liquefied and used in chemical processes (*carbon capture and use*) or stored underground (*carbon capture and storage*). Additionally, emissions of nitrogen oxides (NO_x) are reduced [2] and the stoichiometric flame temperature can be influenced through variation of the oxygen content, which can have a positive influence on the plant thermodynamics. However, the combustion of fossil fuels under oxy-fuel atmosphere is accompanied by major changes in the combustion behavior. Compared to air combustion, the oxy-fuel atmosphere has an increased molar heat capacity which leads to a lower flame temperature and a decreased laminar flame speed, which eventually influences flame stability. Additionally, the mass diffusion coefficient of oxygen is lowered, causing an increased Lewis number and a need for higher oxygen concentrations [3]. Due to radiation of tri-atomic molecules, the radiative heat flux is increased [4]. Furthermore, in contrast to nitrogen, carbon dioxide is not inert during the combustion as it reacts to CO at higher temperatures.

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Hence, for the development of predictive engineering tools for designing future coal or gas power plants, detailed studies of the coal and gas combustion under oxy-fuel atmosphere, accompanied by a model development for all processes involved, are necessary. The research community, in particular the collaborative research center/transregio 129 "Oxy-Flame", follows a stepwise approach from generic to close-to-application combustion systems by steadily increasing the size, complexity and thermal power of the investigated burners as well as the complexity of the fuel. The power of burners under investigations range from single particle combustion [5–8], < 1 kW_{th} [9], 2–3 kW_{th} [8,10–12], 20–60 kW_{th} [3,13–17], 200–800 kW_{th} [18–20] up to 30 MW_{th} [21] and the fuels under investigation cover acetylene [9], methane [10,11,13,19,22–24], methane-assisted coal [5,6,8,14,15] and coal [3,8,16,18,21]. However, in the intermediate range of 10–100 kW thermal power, detailed and comprehensive experimental and numerical investigations of the oxy-fuel combustion are still sparse. In particular, investigations of multiple quantities, such as flow field, flame structure and temperature, flue gas composition and temperature in similar combustion systems in terms of thermal power, geometry, fuel and oxygen content are very rare. Balusamy et al. [14,15] measured the flow field, soot distribution and reaction zone of a 20 kW_{th} gas-assisted oxy-coal swirl burner using laser Doppler velocimetry, particle image velocimetry (PIV), laser induced fluorescence (LIF) and laser-induced incandescence. Hees et al. [3,16] measured the flame structure using chemiluminescence, species concentration within the reaction zone and flue gas composition in a 60 kW_{th} coal burner under air and oxy-fuel atmosphere.

For a validation of simulations and a detailed understanding of the process, besides the flow field and flame structure in the near burner region, an analysis of the flue gas composition and temperature are of high relevance. However, a direct experimental investigation of oxy-fuel combustion is challenging, as most conventional temperature and species concentration measurement techniques fail under operation of the high temperature environments of combustion chambers, which include high radiation levels, eventually high particle load and ongoing chemical reactions. Additionally, most conventional measurement techniques are not capable of resolving the high fluctuations of the oxy-fuel combustion systems. An ex-situ concentration analysis of the process gases is commonly performed using Fourier transform infrared spectrometry [16], gas chromatography [25] or mass spectrometers [26]. The flue gas is often analyzed using conventional gas analyzers based on chemical absorption of the gases, UV photometry and non-dispersive infrared absorption [3,11]. Even though these methods allow highly accurate measurements, they require complex gas sampling, which leads to loss of information on process-related fluctuations, short-living intermediate species and, through the necessity of suction probes, may affect the flow field and flame structure. Hence, contactless measurement techniques are favorable. He et al. [27] qualitatively measured the sodium (Na) and potassium (K) concentrations emerging from coal combustion in the flue gas of a lean $CH_4/CO_2/O_2$ flame using laser-induced breakdown spectroscopy and Köser et al. investigated the qualitative OH-concentration of coal particles burning in the flue gas of a similar burner [8] with laser induced fluorescence of the OH-molecule. In terms of temperature measurement, conventional tactile methods, such as thermocouples, have to be radiation corrected in the flue gas of oxy-fuel combustion, due to the presence of high concentrations of hot CO_2 and H_2O and therefore enhanced heat transfer through radiation. This process is prone to large errors. Additionally, most laser based techniques for temperature and species concentration measurements, such as coherent anti-Stokes Raman spectroscopy [8], Rayleigh scattering, Raman spectrometry [28] or laser induced grating spectroscopy [29] are not suitable for investigations of oxy-coal combustion, due to high particle loads.

Tunable diode laser absorption spectroscopy (TDLAS), detecting the resonant absorption of laser light from discrete gas absorption lines in the infrared spectrum, can be set up to measure independently from broadband losses caused by particles. Hence, it is increasingly utilized to measure species concentrations and/or temperature in coal [30–33] and gas combustion [34–37] environments. However, it has rarely been used for investigations in oxy-fuel environments. Drasek et al. used TDLAS to measure the temperature, O_2 and CO-concentration in the exhaust of a 700 kW_{th} natural gas/O_2 burner [20]. In Ref. [12], we used TDLAS to study a 3 kW_{th} laminar, non-premixed methane oxy-fuel flame, and in Ref. [38] to measure the residence time distribution.

In this work, to close the gap and simultaneously provide experimental validation data for numerical simulations, we present an experimental investigation of species concentrations and temperatures in the flue gas of a turbulent, partially premixed 20 kW_{th} oxy-fuel swirl burner, which is designed for both gas and gas-assisted coal firing. We are limiting the measurements to oxy-gas combustion of methane (CH_4) for a stepwise increase of complexity. The operating conditions (air, $25\%_V$ $O_2/75\%_V$ CO_2 and $30\%_V$ $O_2/70\%_V$ CO_2 atmosphere) of this burner have been subject to various investigations [17,38,39]. A set of three spectrometer systems based on TDLAS are applied to the optically accessible combustion chamber of the burner. The spectrometer systems are capable of measuring the absolute species concentrations of O_2, H_2O, CO_2, CO, CH_4, C_2H_2 and OH as well as the flue gas temperature. The rate of burnout and fuel-slip in the flue gas is studied through measurements of the concentration of CH_4, whereas a quasi-simultaneously operating OH-measurement system allows for the detection of remaining combustion zones. The O_2-concentration is measured for an investigation of the oxygen-excess, and, in combination with measurements of the CO_2 and H_2O concentration at various positions, allows to detect the main components of the oxy-fuel flue gas. A CO measurement allowed for a detection of the main pollutant, whereas an acetylene measurement allows an estimation of the upper C_2H_2 concentration limit, which is a precursor of soot. The Allan-Werle variance of these spectrometers were not only used to identify spectrometer drifts, but also to analyze time scales within the combustion chamber. Combining multiple spectrometers to quasi-simultaneous measurements allows for an investigation of the mixture progress of species within the combustion chamber.

In the following, we will first present the burner and the spectrometer systems. Subsequently, the influence of the atmospheric composition on the combustion based on experimental findings of temperature and species concentrations is discussed.

2. Experimental setup

2.1. Combustor

The combustor under investigation is a generic intermediate-scale (20 kW_{th}) combustor, which is suitable for operating gas flames as well as gas-assisted coal flames (Fig. 1a). It is designed for operation under both air and oxy-fuel atmosphere and aims for closing the gap between unconfined laboratory-scale burners [14,15] and confined industrial-scale combustors [3,16]. The burner itself (Fig. 1b) is designed to mimic the features of a coal firing burner [40] and is down-firing into the combustion chamber of 420 × 420 × 600 mm^3 volume. Through the aid of guidance plates, the corners of the combustion chamber are rounded. Wedged fused silica windows in the walls allow spacious optical access. The measurements in this work are performed in the flue gas in the lower quarter of the combustion chamber. Similar to industrial coal burners, the gas issues from the burner through a partially premixed (fuel and oxidizer), unswirled primary flow and a secondary, swirled oxidizer flow. While the ratio between the primary and secondary flow allows for a variation of the local, near burner air-fuel ratio and swirl number, a tertiary flow close to the windows allows for a variation of the global air-fuel ratio and a cooling of the windows. The primary and swirled secondary flow issue from two concentric annular orifices atop of a quarl in which the two flows mix and a flame can be stabilized. This quarl is of particular relevance as the coal particles are