Soot formation in diffusion flames in oxy-fuel atmospheres

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Oxy-fuel combustion has been recognized as one of the most promising CO_2 capture technologies because of the potentially high capture rate and relative simplicity of the process. Several large scale pilot and demonstration projects for oxy-fuel coal combustion are taking place around the world [1]. Although the main focus has been on oxy-fuel coal combustion, oxy-fuel natural gas combustion is also a very promising technology for CO_2 capture. This involves natural gas fired gas turbines and several other natural gas fired industrial processes. The main approach when using the oxy-fuel technology is to replace the air with a mixture of oxygen and recycled exhaust gas containing mainly CO_2. One of the challenges implementing the oxy-fuel technology is that this CO_2/O_2 mixture has very different physical properties compared to air. CO_2 has a high specific heat capacity and also absorbs and emits radiation in specific wavelength bands. CO_2 also participates in the chemical reactions of the combustion process. This makes it necessary to special design combustion chambers for oxy-fuel operation. As part of the development of natural gas oxy-fuel combustion, this work deals with the formation of soot in oxy-fuel flames. The importance of understanding the soot formation is not restricted to predict the particulate matter emission, but it also strongly influences the radiation of heat from the flame. Experiments with methane and ethylene flames in different O_2 enriched atmospheres of both CO_2 and air have been conducted in a jet in co-flow burner (diffusion flame). The fuel was introduced through a 5 mm I.D. nozzle and the oxidizer through a 90 mm I.D. co-flow tube placed inside a square 20x20 cm combustion chamber. The co-flow velocity was 0.25 m/s and the methane velocity was 7.6 m/s. The laser beam extinction method followed by Abel transformation of the data was used to measure the soot volume fraction shown in figure 1 and 2. Measurements were also performed on ethylene flames using the Laser Induced Incandescence (LII) technique. The main results are shown in figure 3 and 4. LII is generated when soot particles are irradiated with a high energy laser light.

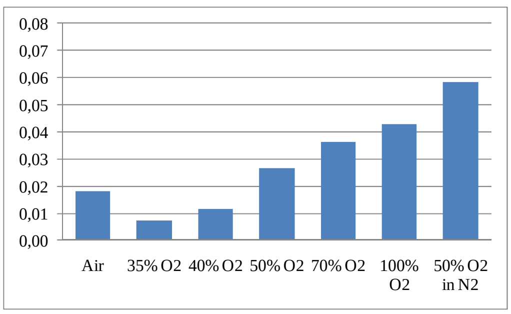

Figure 1 Peak soot volume fraction in ppm(v) in CH_4 flame 50 mm above nozzle

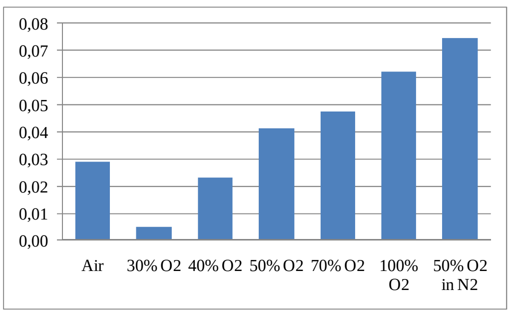

Figure 2 Peak soot volume fraction in ppm(v) in CH_4 flame, 100 mm above nozzle

The soot particles absorb the laser light and are heated to a temperature much higher than the surrounding gas temperature. The heated particles emit black body radiation according to the increased temperature which is at or close to the evaporation temperature of carbon, about 4000 K. The soot volume fraction is found by capturing the radiated light in an intensified CCD camera and using the laser beam extinction technique for calibration.

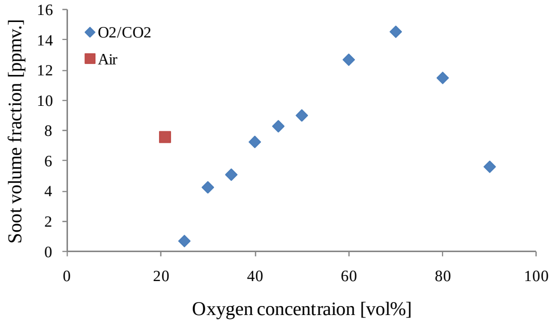

Figure 3 Peak soot concentrations as function of oxygen concentration in oxidizer, C_2H_4 flame

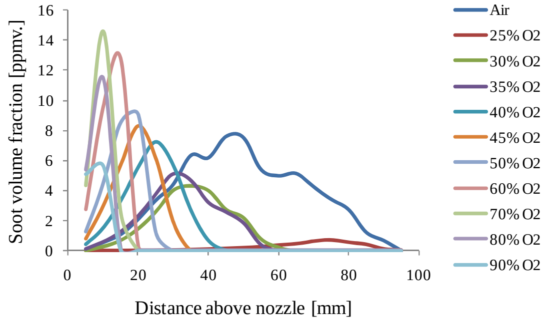

Figure 4 Soot concentration as function of height above nozzle, C_2H_4 flame

The experiments revealed that the CO_2 concentration strongly influences the soot formation. The soot formation process in diffusion flames is controlled by the flame structure, i.e. the relationship between the local temperature and the local species profiles. High temperature in fuel rich regions leads to rapid formation of soot precursor species and the available time at high temperature controls the nuclei and particle growth process [2]. This corresponds well with the experiments as the flame temperature as well as the soot volume fractions increase with increasing oxygen content. The O_2 enriched air co-flow results in higher soot concentrations compared to O_2/CO_2 atmospheres corresponding to the same adiabatic flame temperatures. This can be explained by the chemical effect of CO_2 on the soot formation process found by Liu et al. [3] among others. The OH concentration increase which leads to increased oxidation of the soot nuclei/precursors in the soot forming regions.
The results show that the difference in soot formation between oxy-combustion and air combustion should be considered when designing combustors for oxy-fuel combustion.

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