Dynamics of Solid Carbon Formation by Turbulent Combustion and Thermal Decomposition of Natural Gas

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EXTENDED ABSTRACT

This paper investigates the formation process of solid carbon particles in turbulent flows within fuel-rich natural gas flames. Research on carbon formation from simple hydrocarbons under real process conditions and harsh environments allows novel insights into the general understanding of soot formation. Experimental work was carried out on a small-scale gas furnace based on the design of a modern carbon black oil furnace as shown in Figure 1. The combustion system investigated is fired by a non-premixed, swirl stabilised, confined methane-air flame. The study comprises establishing conditions under which the production of solid carbon particles takes place, and progressing this towards quantification. A large number of process parameters have been investigated, and the relative role of incomplete combustion and thermal decomposition in the process of carbon particulate formation has been illustrated.

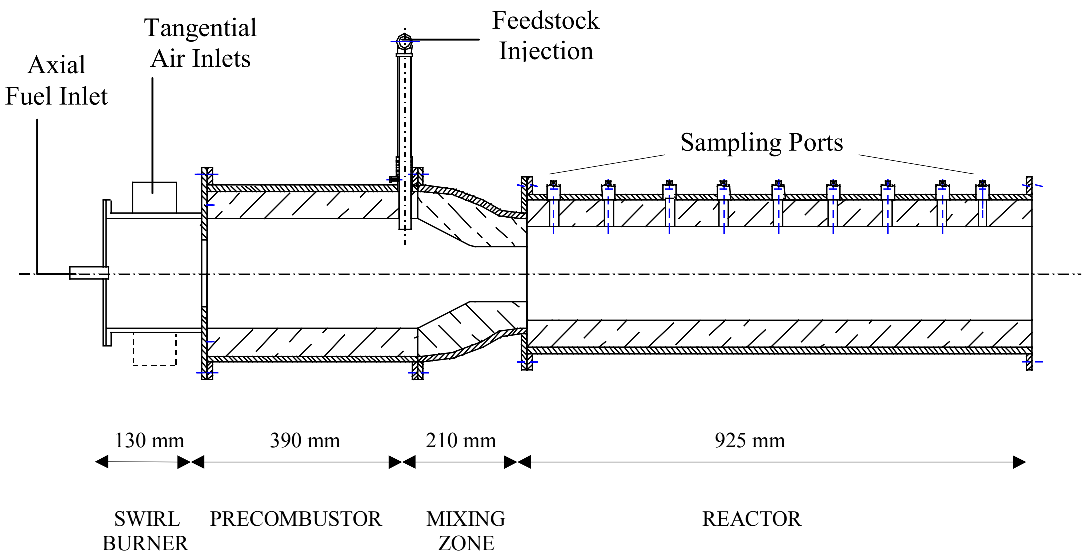

Figure 1: Illustration of small-scale gas furnace

Carbon Black Furnace and Measurements

The carbon black furnace which was used in this investigation is a simplified, versatile, small-scale axial flow gas furnace. It has been designed and manufactured on the basis of the modern oil furnace process, but using natural gas as feedstock hydrocarbon (max. performance 5-10 kg carbon black per hour). The basic geometry of the carbon black furnace consists of a precombustor, a mixing zone and a reactor. In the precombustor, the axially injected natural gas fuel burns with process air which is introduced through two tangential inlets. The highly swirling hot gases meet the feedstock natural gas (excess fuel) which is injected radially into the precombustor in the vicinity of the mixing zone. The abrupt enlargement in diameter at the exit of the choke encourages mixing of feedstock fuel with the hot gases.

The design of the furnace allows adjustment of all relevant process parameters, such as air flow rate, air/fuel ratio, air/feedstock ratio and reactant preheat temperatures. The effects of these primary parameters on process temperatures, chemical species and most importantly carbon particle concentrations are investigated. The method used to measure carbon concentrations is gravimetric sampling. This involves isokinetic sampling from the process followed by measurement of the gas volume and the carbon mass in the samples.

Results and Conclusions

Natural gas flames are generally known to produce only small amounts (virtually zero) of soot. 'Clean' combustion with regard to sooting is, however, only guaranteed for fuel lean mixture ratios. Confined methane-air flames burning under stoichiometric conditions can cause the production of large amounts of solid particle emissions. Fuel-rich burner operation or local accumulation of fuel causes the production of even larger amounts of particulate carbon. The rate of soot production is assumed to be mainly dependent on the following parameters: Equivalence ratio, furnace temperature, process air temperature and fuel temperature. The established relationship between combustor C/O ratio, mean process temperature and carbon particle concentration is shown in Figure 2.

The overall trend shows steady increase in carbon particle concentration with increasing fuel richness to a maximum followed by a sudden drop. Furthermore, process temperature has a noticeable influence on carbon production. Generally, for very rich mixtures, higher furnace temperatures cause an increase in soot output. The reverse is true for only slightly rich mixtures. This phenomenon is attributed to the two processes governing to formation of solid carbon particles in hydrocarbon flames, namely the process of incomplete combustion and the thermal decomposition of the hydrocarbon.

At high C/O ratios, the influential presence of excess fuel plus the cooling effect of the fuel injection increase soot output. As oxidising species are exhausted at these C/O ratios, increase in temperature will result in an increase in carbon production reactions. Accordingly, maximum soot yield is realised at maximum air temperature and maximum furnace temperature.

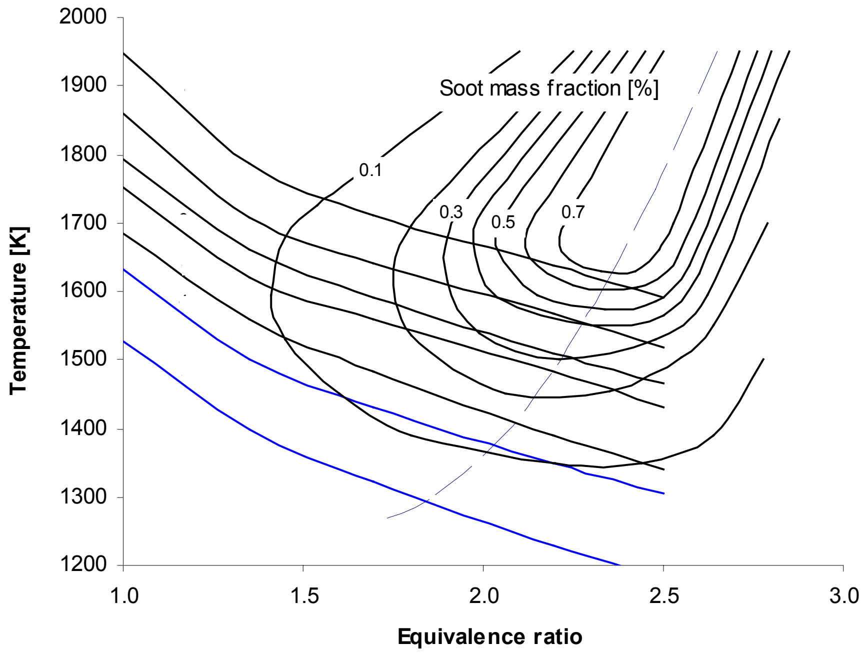

Figure 2: Plot of soot yield versus flame conditions (process temperature determined at precombustor outlet)

For only slightly rich mixtures, the increased process temperature supports oxidation reactions more than carbon production. Enhanced oxidation is reflected by an increased presence of CO in relation to CO_2 (Figure 3), which means more carbon is consumed and therefore results in the observed reduction of soot. Therefore, it can be concluded that only slightly rich conditions are governed by incomplete combustion only.

The quantification of the chemical species methane and hydrogen allows the investigation of the decomposition process. Figure 4 firstly shows the increasing amount of unburned hydrocarbons with increasing fuel richness. More importantly, the hydrogen measurements reveal that the concentration of H_2 in the reactor off-gas increases with increasing excess fuel, and reaches a maximum coinciding with the maximum of carbon particle production. Accordingly, it is concluded that changes in hydrogen mass fraction represent hydrocarbon decomposition activity.

Maximum hydrocarbon decomposition is noted at a relatively high C/O ratio, which leads to the conclusion that fuel rich combustion systems are governed by thermal decomposition processes. This is true only up to a certain limit, whereafter the increased cooling effects of high concentrations of unburned hydrocarbons reduce the mean process temperature below the required activation temperature for fuel decomposition.

First attempts to simulate the simultaneous processes of incomplete fuel combustion and fuel decomposition in turbulent combustion systems indicate reasonable agreement with experimental data. A novel combination of a chemical kinetics model and a standard soot model included in a CFD model allows the prediction of not only a large number of chemical species, but also the prediction of solid carbon particle production from both incomplete combustion and thermal decomposition.

Future work will aim to optimise particle formation processes experimentally by broadening the range of initial conditions, and computationally by progressing development of a more robust numerical model for the simulation of carbon particle formation in turbulent combustion flows.

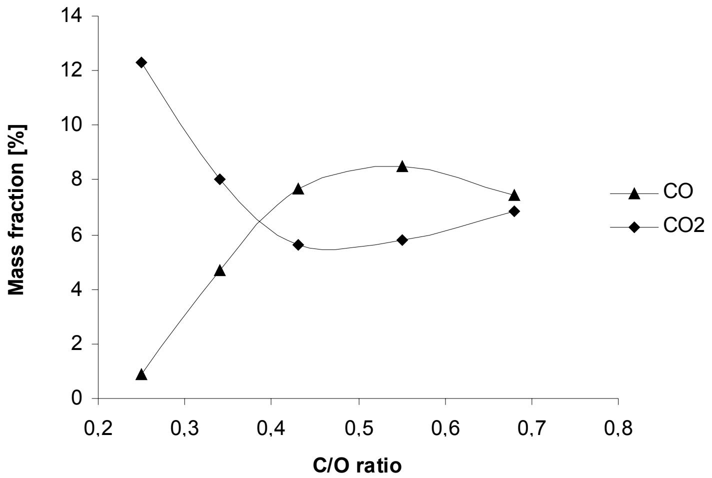

Figure 3: Effect of fuel richness on CO and CO_2 output at constant process air temperature 400°C

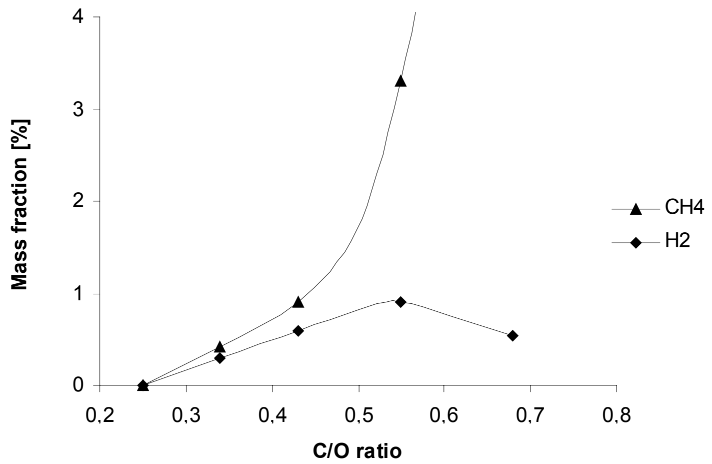

Figure 4: Effect of fuel richness on CH_4 and H_2 output at constant process air temperature 400°C