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Gas and Particulate Matter Products Formed in a Laminar Flow Reactor: Pyrolysis of Single-Component C₂ Fuels

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Abstract

Investigations that rank the tendencies of single-component fuels to form soot have given useful insights into how molecules and local molecular structure influences the conversion of carbon in fuel to soot. These studies can provide fundamental understanding of how functional group chemistry influences the products formed during pyrolysis or combustion.

A series of experiments have been conducted which investigate the conversion of oxygenated and hydrocarbon C₂ molecules (ethanol, ethane, and ethylene) to particulate matter (PM) and other species (CO, CO₂). Single-component fuels have been tested in a laminar flow reactor at concentrations of 8000 ppmv, under pyrolysis conditions. Size and mass distributions of particulates are reported, measured by means of a differential mobility spectrometer (Cambustion DMS500).

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1. Introduction

A large number of experimental and modeling studies have been reported in literature that assess the influence that the molecular structure of a fuel has on the formation of particulates and other emissions. A number of the experimental studies reported have been carried out in practical systems such as in an engine or burner [1,2], and some have been carried out under well-controlled conditions, usually in a shock tube [3] or flow reactor [4-6].

In addition to providing some fundamental understanding of the chemical mechanisms of pollutant formation, these studies can also help inform the development of cleaner fuels. It is of interest to study to study a homologous series of hydrocarbons (ethanol, ethane, and ethylene) to understand how the functional group chemistry of the series influences conversion of those molecules to emissions,

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particularly particulate emissions. A DMS500 instrument has been used here to compare relative size and mass distributions, and total mass of particulates (smaller than 1000nm) formed from different fuels.

2. Experimental Method

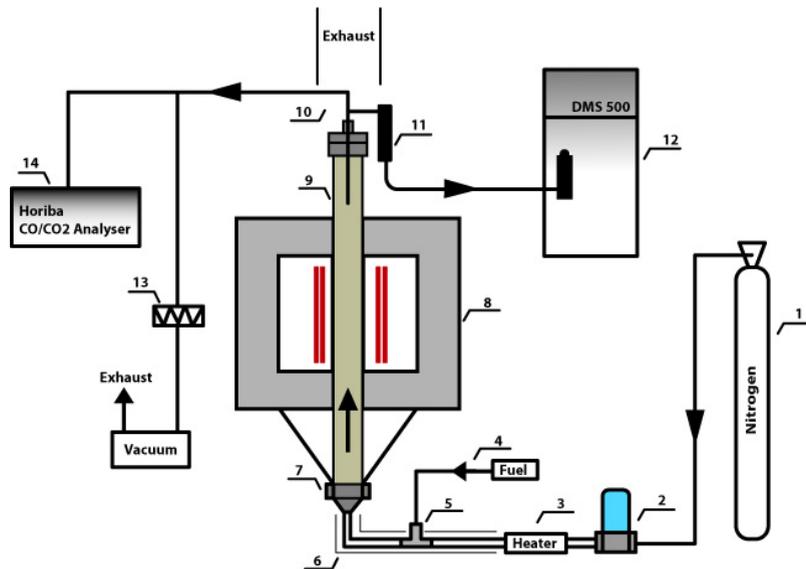


Figure 1. Flow reactor rig. 1. Inert carrier, 2. Mass flow controller, 3. Heater, 4. Fuel feed system, 5. Vapouriser, 6. Heated and insulated section, 7. Static mixer, 8. Electric oven, 9. Reactor tube, 10. Stainless steel sample line, 11. External dilution cyclone, 12. DMS 500, 13. Filter housing, 14. Horiba CO and CO₂ analyser.

Particulate matter has been generated in a flow reactor facility shown in Fig. 1, and this installation has been described in previous work [7]. The system comprises of a fuel feed system (1-7), a vertically orientated reactor (8,9), and a sampling system (10-14). The reactor can be setup for use with either liquid or gaseous fuels, which are transported by an inert carrier (N₂).

Ethanol was fed by a syringe pump through a capillary tube into a heated section of stainless steel tube, packed with borosilicate glass beads, where the ethanol was immediately vapourised and entrained in the nitrogen stream and carried to the reactor. Gaseous fuels (ethane, and ethylene) were metered by means of a mass flow controller (Bronkhorst EL-flow). In all cases the flow rate was fixed at 8000 ppmv. The inlet concentrations and radial homogeneity was verified by using a fast response flame ionization detector (Cambustion, HFR400), taking account of FID response factors as reported in [8].

The reactor tube was operated in the temperature range of 1000-1400°C, heated by an electric oven. The dimensions of the reactor tube were 104 mm i.d. an overall length of 1440mm with a heated length of 600mm in the center, which is assumed to be uniform in temperature. The flow rate of nitrogen into the reactor was fixed at 20 Lmin⁻¹, and this resulted in a laminar flow and a gas residence time of ~1s.

Measurement of particulate number and size distributions were made by a differential mobility spectrometer (Cambustion DMS500), which drew sample through a stainless steel sample tube positioned on the reactor centerline, approximately 300mm from the exit of the reactor. A gas analyser (Horiba AIA-

120) was used to sample CO and CO₂ from the same position in the reactor in the case of ethanol; CO and CO₂ were not present in pyrolysis of ethane and ethylene.

3. Results

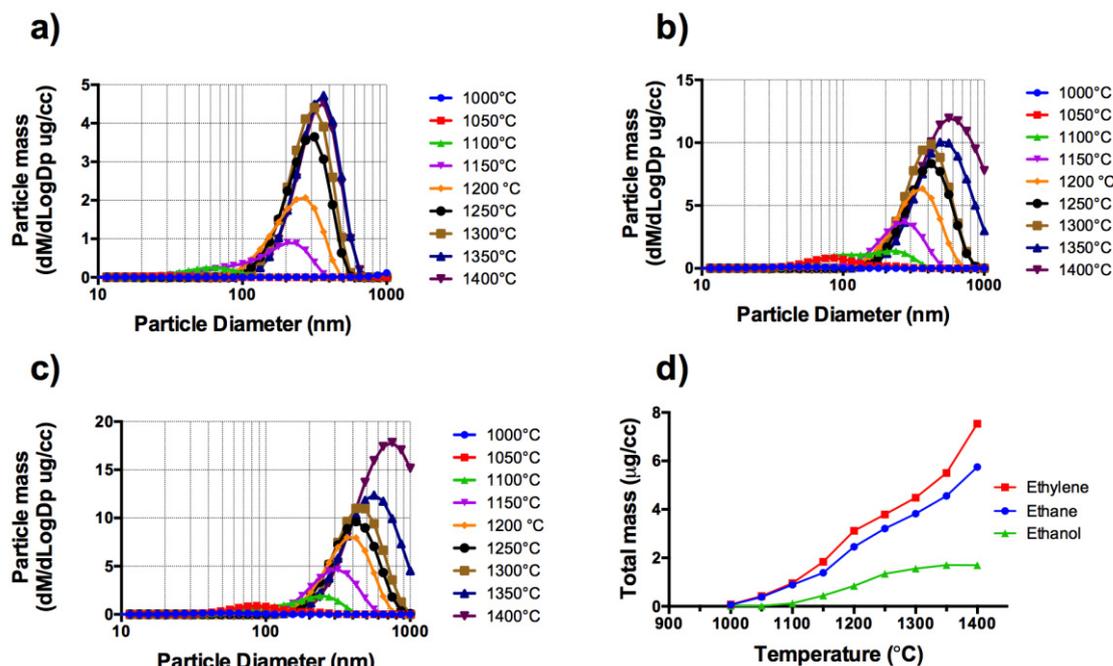


Figure 2. Particulate size and mass distribution for a) ethanol, b) ethane, c) ethylene. d) calculated mass concentrations of PM smaller than 1000nm formed from the respective fuels.

The apparent mass distributions of the particulates formed from the pyrolysis of ethanol, ethane and ethylene are shown in Fig.2 (a,b,c). Analysis of the number distributions (data not shown) shows that for all fuels tested there is a gradual shift towards larger particle diameters at increasing reactor temperatures. For example, in the case of ethanol pyrolysis, pyrolysis at 1200°C results in particulates mainly in the size range of 100–400nm, whereas at a higher temperature of 1400°C, the particulates are generally distributed in the 100–700nm range. In going from ethanol, which is oxygenated, to the hydrocarbon ethane, it is observable that at 1200°C ethane forms larger size particles than ethanol (100–600nm).

Figure 2 (d) shows that the mass concentration of particulates smaller than 1000nm produced in the reactor is found to be in the order ethanol<ethane<ethylene at any given temperature; it is also generally observed that increasing reaction temperatures gives rise to greater mass concentrations of particulates issuing from the reactor.

Finally, Fig. 3 shows the evolution of CO and CO₂ during the pyrolysis of ethanol (1000–1400°C). Formation of carbon monoxide (CO) occurs at temperatures upwards of approximately 600°C, and CO was a major product in the range 1000–1400°C. It is shown that there is negligible CO₂ formation at all temperatures tested.

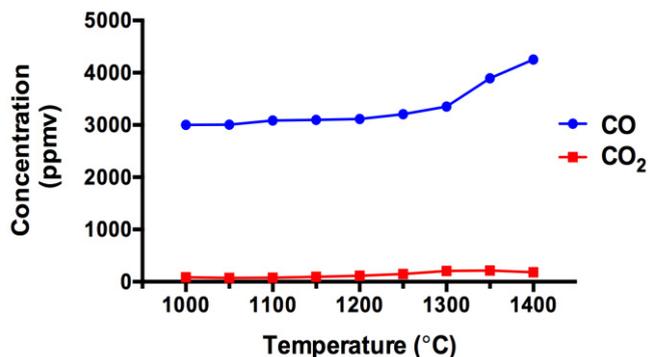


Figure 3. Concentration of CO and CO₂ formed during the pyrolysis of ethanol in the temperature range 1000-1400°C.

4. Conclusions

To conclude, size and mass concentrations have been shown for three C₂ fuels, and differences in the characteristics of the particulates formed have been reported, as measured by a differential mobility spectrometer. The results show that the overall mass of particulates formed is in the order ethanol<ethane<ethylene, which is in alignment with literature [5]. It is of interest that the presence of oxygen in ethanol is found to have a suppressed conversion to PM, with respect to ethane or ethylene, and was found to form CO, but not CO₂. This work compliments a recent isotope-tracing investigation, which reported that particulate matter formed from the pyrolysis of ethanol was derived from 68% from the ‘methyl’-carbon atom and 27% from the oxygenated carbon atom at all temperatures in the same range [7].

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