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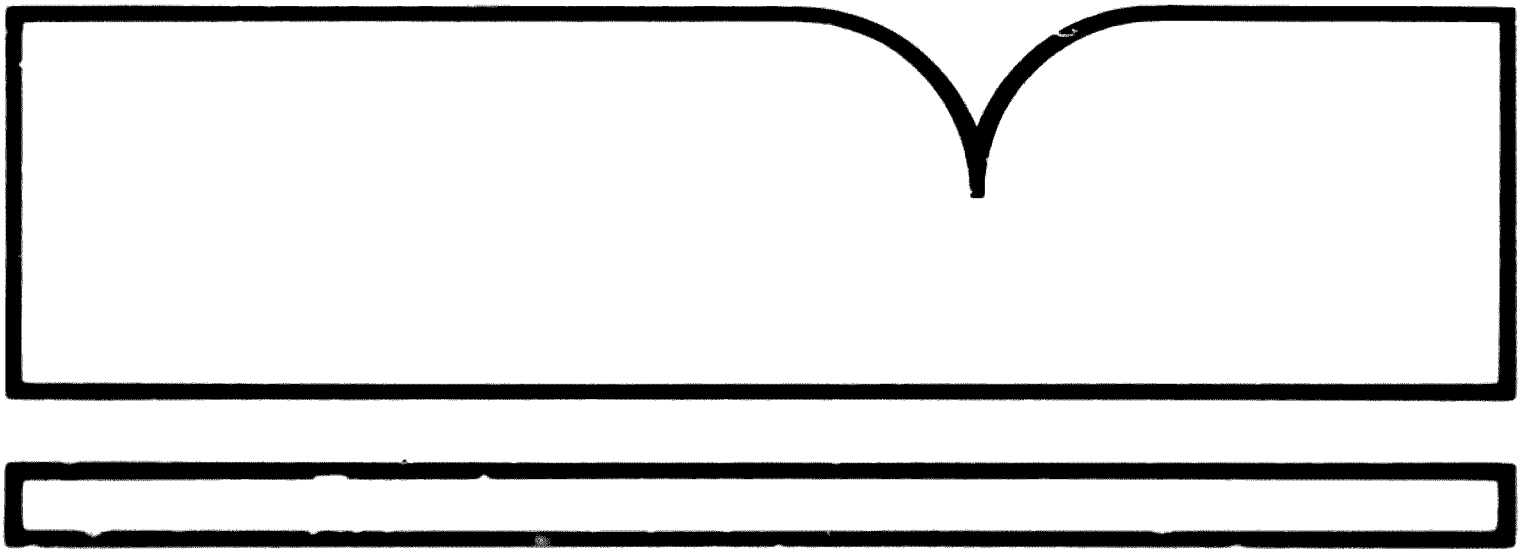

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Laminar Methane-Air Diffusion Flame with Chlorine Impurities: Preliminary Results

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ABSTRACT

Formation of products of incomplete combustion (PICs) during the thermal destruction of chlorinated compounds is of current interest. To study the fundamental processes accompanying the combustion of chlorinated hydrocarbons, exploratory tests have been conducted by adding chlorine to the fuel side of a well characterized methane-air laminar diffusion flame. The preliminary observations are concerned with the soot emissions and the flame temperature. The structure of the soot particles is examined under a Scanning Electron Microscope (SEM). The SEM studies and the temperature measurements are compared to similar studies on a methane-air diffusion flame. Results indicate no change in the temperature field between the methane-air and chlorine/methane-air diffusion flames. The SEM analysed soot indicated the absence of chlorine, although in one case the soot exhibited a porous structure possibly due to chlorine induced attack.

INTRODUCTION

Products of incomplete combustion are of concern during incineration of municipal and hazardous wastes. Chlorinated hydrocarbons and other chlorine compounds are potential sources in aiding the formation of chlorinated dioxins and furans. Therefore there is a need to conduct systematic fundamental studies on the combustion of chlorinated compounds. Although there have been studies on the flame destruction of halogenated compounds focused on kinetic studies, very little is known regarding the physical and chemical structure of soot produced during the combustion of chlorine compounds. Soot and other particles that exit the flame are condensation sites for chlorinated materials and could potentially form chlorinated dioxins and furans. We intend to conduct detailed studies connected with the formation of soot and gaseous products during the combustion of chlorinated methanes. Combustion of chlorinated compounds should result in either Cl_2 or HCl as final products. Although HCl is not known to donate Cl atoms for the formation of chlorinated PICs, researchers suspect Cl_2 to be a donor [1,2]. In such cases, besides the gaseous species in the flame and post-flame zone, the soot formed may also indicate the presence of chlorine.

Here we present preliminary results obtained by the addition of Cl_2 to the fuel side of a laminar coflow methane-air diffusion flame. Soot samples collected from various regions of the flame were analyzed for the presence of chlorine using SEM. Following this we are in the process of obtaining detailed gaseous species concentrations in the flame and post-flame regions using microsampling techniques [3].

EXPERIMENTAL METHOD

A detailed description of the experimental methods used has been reported elsewhere [3,4]. A brief description of the temperature measurement and soot sampling procedure is given here. The flame examined is a laminar coflow methane-air diffusion flame. The burner consists of two concentric tubes of 7 and 1.6 cm diameter. The fuel flows through the inner tube while air flows in the outer

tube. Before entering the burner a mixture of nitrogen and chlorine is well mixed with methane (62% N_2, 37% CH_4, and 1% Cl_2). When the fuel velocities are 3.85 and 4.025 cm/s, the respective flame heights are 4.0 and 5.5 cm. Temperatures were measured using a Pt-13%Rh/Pt thermocouple. The wire diameter of the thermocouple was 0.05 mm (bead diameter 0.1 mm), and it was coated with a uniform layer of silica. Soot samples were collected using a metallic filament 0.25 mm in diameter. Soot was allowed to aggregate on the probe for 7-8 minutes.

RESULTS AND DISCUSSION

The flames were studied in over-ventilated, laminar, and non-smoking conditions. Both the 4.0 and the 5.5 cm flames exhibited a blue zone which was 1.0 cm in height for the smaller flame and 1.5 cm in height for the larger flame. The blue zone was followed by a bright yellow-orange zone associated with the production of soot. This bright zone was followed by a light orange zone at the tip which was about 2 mm thick. The orange zone is associated with the oxidation of soot produced in the yellow-orange region, and no soot deposits occurred on the probes. Further details of the two colored soot regions have been presented by Saito et.al [5]. Soot samples were collected mainly in the yellow-orange zone. Soot was collected from the 4.0 cm flame at axial heights of 2.85 and 3.85 cm from the burner port. For the 5.5 cm flame, soot was collected from axial heights of 2.85, 4.0, and 4.4 cm. Scanning electron photographs of the soot samples magnified 7500 times are presented along with elemental analysis spectrographs (Figs. 1-5). All the spectrographs show that there is no chlorine on the soot. A peak for chromium appears since the probe contains chromium. A rather striking result is the porous structure of soot exhibited in Fig. 4. We have not observed similar porous structures while studying soot formation in other hydrocarbon flames. The SEM photographs indicate that the soot aggregates consist of individual blocks which are spherical in shape and, for a given location within the flame, of nearly uniform diameter, similar to the results reported for pure hydrocarbon flames [5,6].

Temperature measurements were obtained from the 5.5 cm flame for conditions with and without chlorine addition. Figs. 6 and 7 depict the measured temperature profiles in a three dimensional fashion. There are no significant differences in temperature between the chlorinated and non-chlorinated flames. This is consistent with observations made by other researchers, that there is no appreciable change in the flame temperature on the addition of a small amount of chlorinated hydrocarbons [2].

SUMMARY

Our preliminary investigation on the addition of Cl_2 to the methane-air diffusion flame has shown no significant effect on the temperature profiles as compared to the methane-air flame. Chlorine was not observed on the soot based on an SEM analysis. It may be speculated that Cl_2 is being converted to HCl in the flame and hence refraining from any further reactions. A detailed gaseous species analysis in progress will reveal the fate of chlorine in the flame and post-flame regions. Analysis of soot produced from other chlorinated methanes is also currently in progress. We believe that, in addition to gaseous species, studies on the history of soot from early stages in the flame to the cooler post-flame regions are important to understand the formation of solid PIC's.

ACKNOWLEDGEMENTS

The review and comments by W. P. Linak and B. K. Gullett are deeply appreciated. We wish to thank R. Gonsalez of UKCAER for SEM analyses of the soot, and J. Sievo of Princeton University for providing the excellent thermocouple. The project was supported (P.O.#9D4637NAEX) by the U. S. Environmental Protection Agency, Combuston Research Branch, Air and Energy Engineering Research Laboratory, Research Triangle Park, NC 27711.

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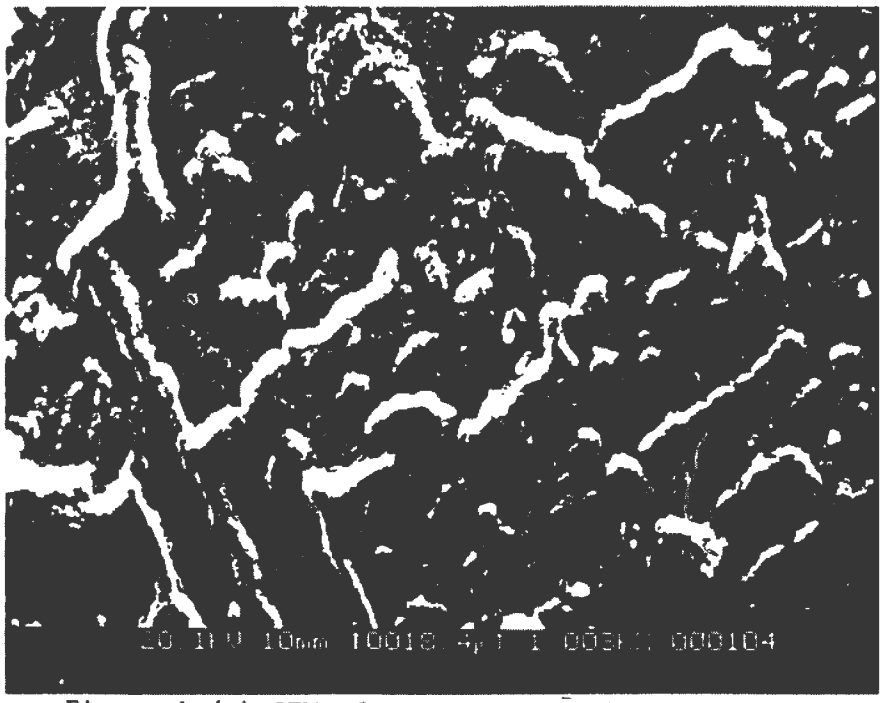


Figure 1 (a) SEM of soot collected at axial height 2.8 cm from 4.0 cm flame.

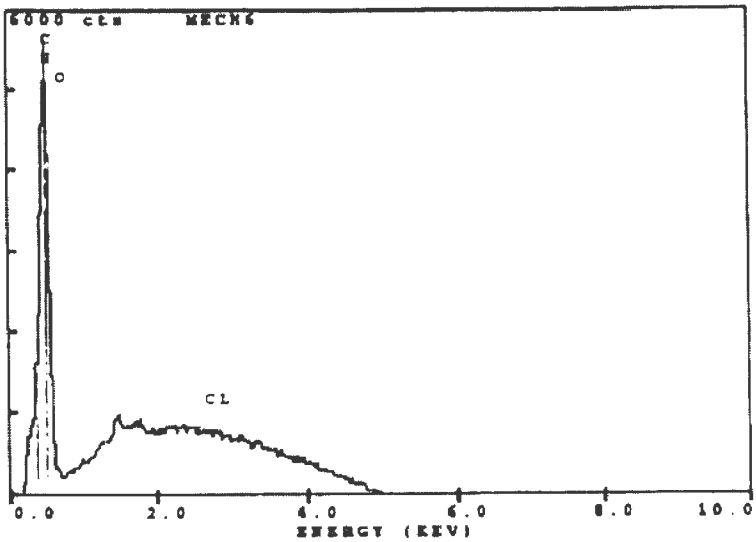


Figure 1 (b) Spectrograph of soot in Figure 1 (a)

Figure 2 (a) SEM of soot collected at axial height 2.8 cm from 5.5 cm flame.

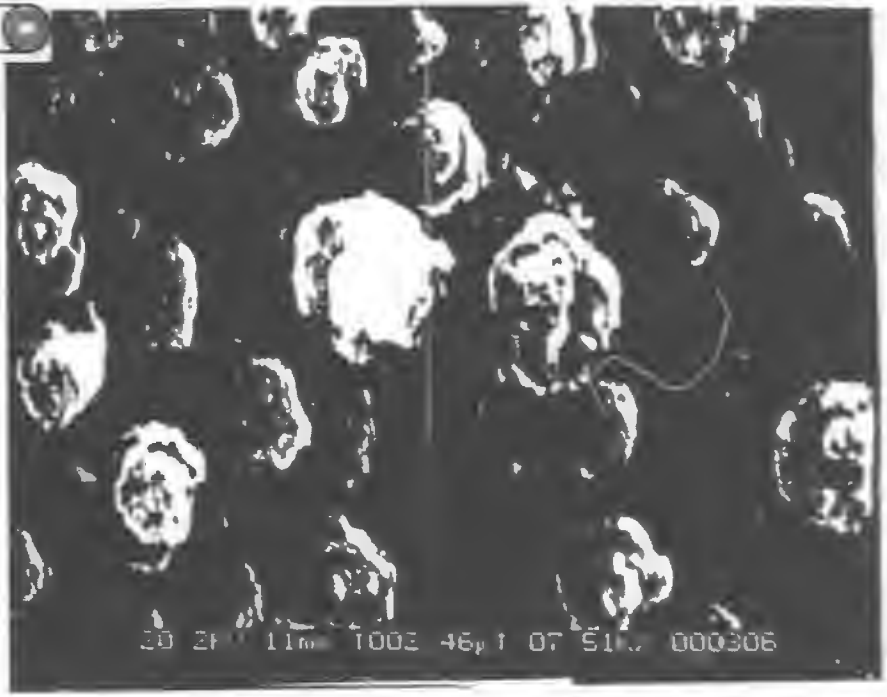


Figure 3 (a) SEM of soot collected at axial height 4.0 cm from 5.5 cm flame.

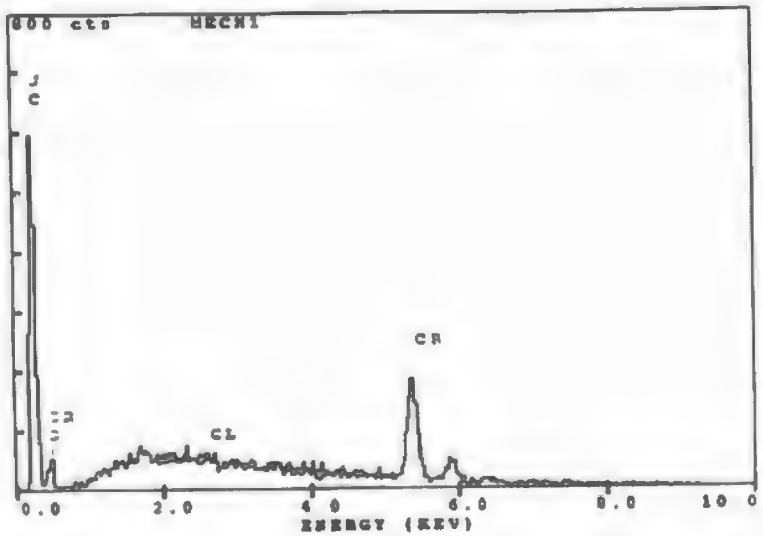


Figure 2 (b) Spectrograph of soot in Figure 2 (a).

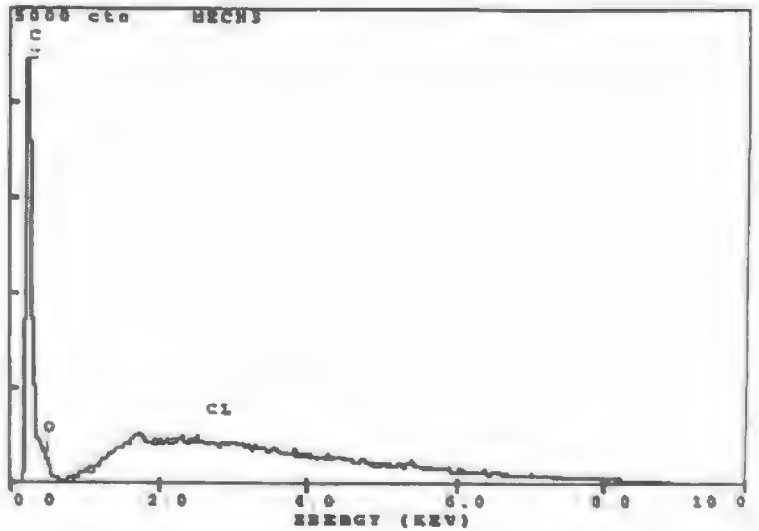
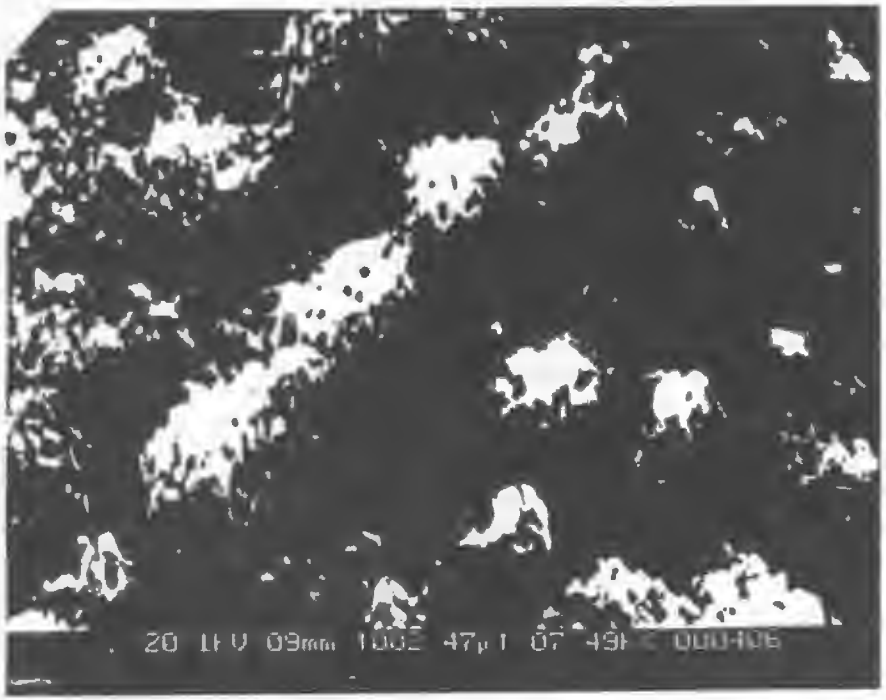


Figure 3 (b) Spectrograph of soot in Figure 3 (a).



igure 4 (a) SEM of soot collected at axial height 3.85 cm from 5.5 cm flame.

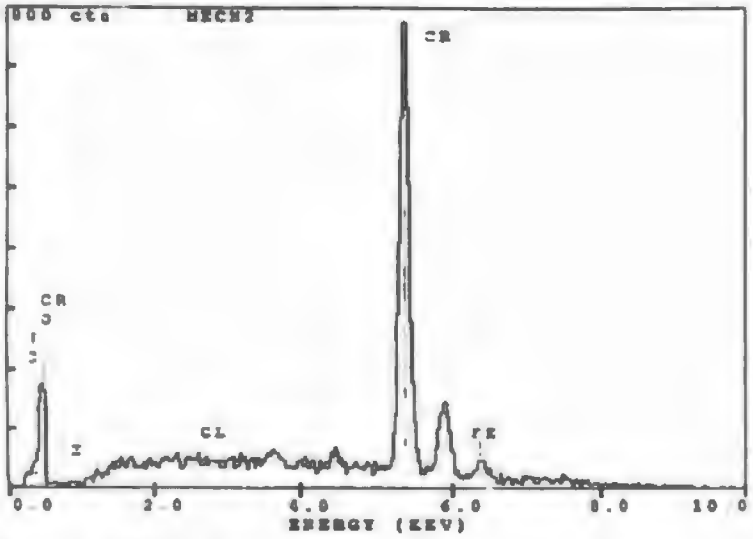


Figure 4(b) Spectrograph of soot in Figure 4 (a).

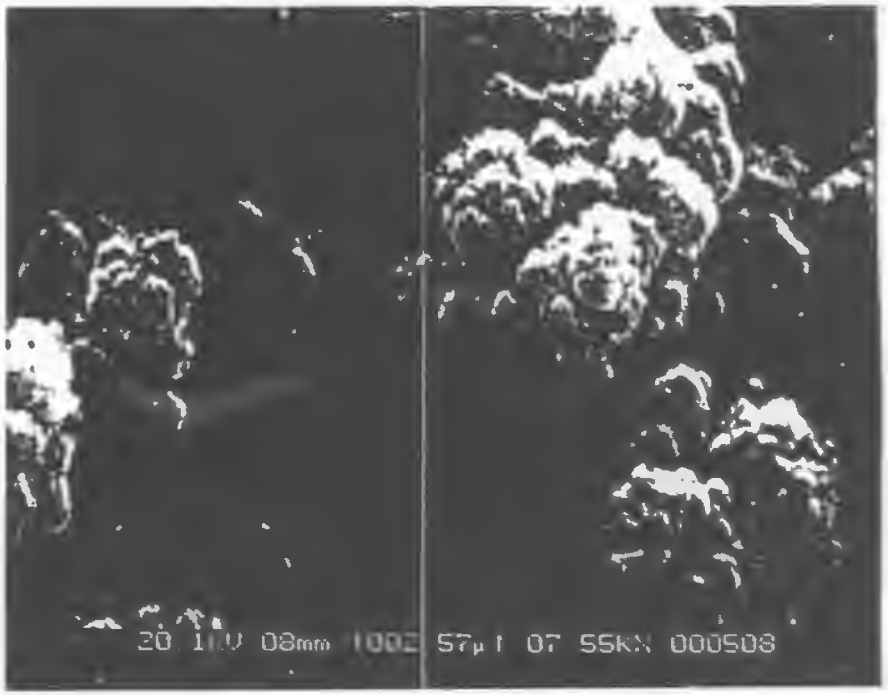


Figure 5 (a) SEM of soot collected at axial height 4.4 cm from 5.5 cm flame.

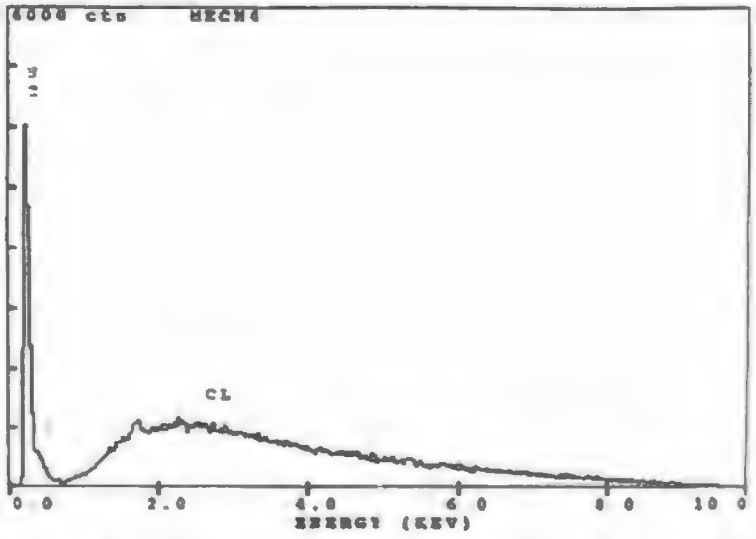


Figure 5 (b) Spectrograph of soot in Figure 5 (a).

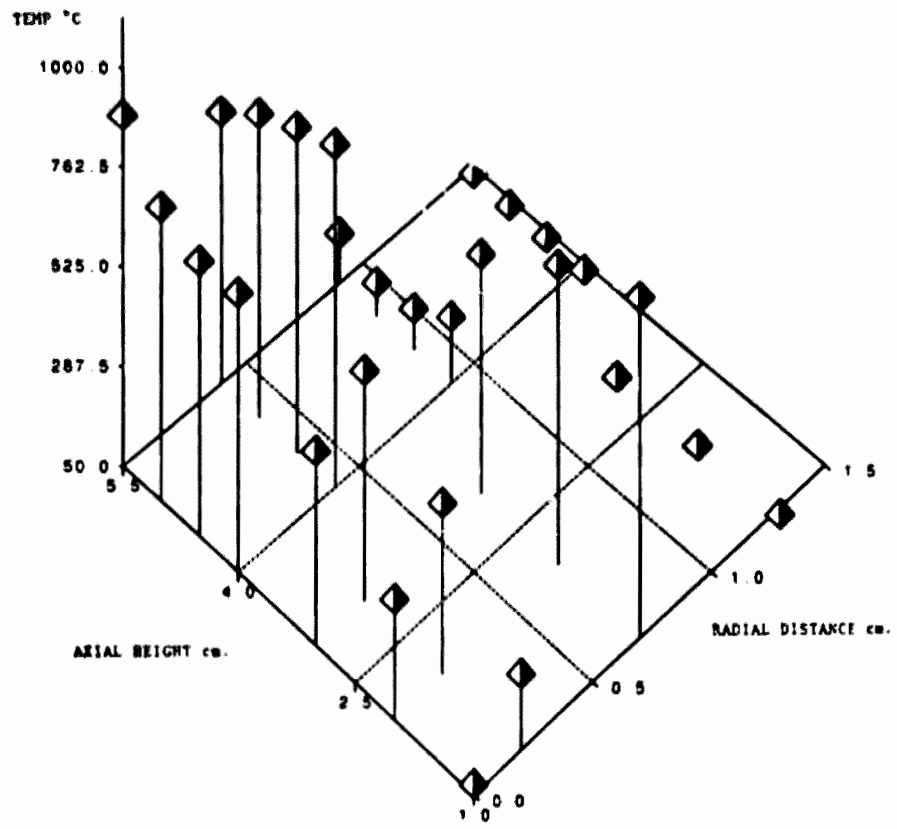


Figure 6 Temperature distribution of the methane-air diffusion flame with chlorine addition.

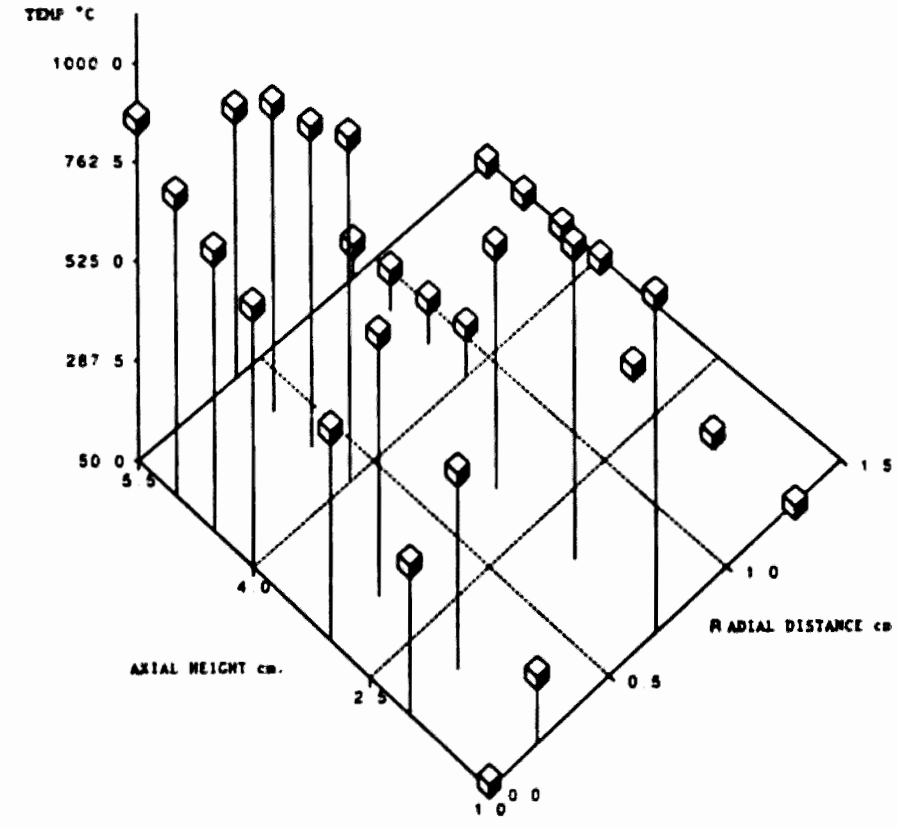


Figure 7 Temperature distribution of the methane-air diffusion flame without chlorine addition.

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16. ABSTRACT The extended abstract gives preliminary results of exploratory tests, conducted by adding chlorine to the fuel side of a well characterized methane-air flame, to study the fundamental processes accompanying the combustion of chlorinated hydrocarbons. (NOTE: Formation of products of incomplete combustion during the thermal destruction of chlorinated compounds is of current interest.) The preliminary observations are concerned with soot emissions and flame temperature. The structure of the soot particles is examined under a scanning electron microscope (SEM). The SEM studies and the temperature measurements are compared to similar studies on a methane-air diffusion flame. Results indicate no change in the temperature field between the methane-air and chlorine/methane-air diffusion flames. The SEM analyzed soot indicated the absence of chlorine, although in one case the soot exhibited a porous structure, possibly due to chlorine induced attack.

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