CARBON MONOXIDE AND SOOT FORMATION IN INVERSE DIFFUSION FLAMES

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INTRODUCTION

The objective of this project is to study carbon monoxide (CO) and soot formation in laminar, inverse diffusion flames (IDFs). The IDF is used because it is a special case of underventilated combustion. The microgravity environment is crucial for this study because buoyancy-induced instabilities impede systematic variation of IDF operating conditions in normal gravity. The project described in this paper is just beginning, and no results are available. Hence, the goals of this paper are to establish the motivation for the research, to review the IDF literature, and to briefly introduce the experimental and computational plan for the research.

MOTIVATION

Between 1972 and 1993, 73,000 people died in accidental fires in America.[1] Nearly seventy percent died from smoke inhalation, and many of them died from fires which progressed beyond flashover, the rapid change of a fire from a developing state to a fully-involved state. Flashover occurs when high-temperature fuel-rich gases accumulate within a room and ignite at a doorway or window where they mix with fresh air.[2] Carbon monoxide and soot are two hazardous components of the smoke that forms during the resulting underventilated combustion period. Hence, it is important to study the ways that these substances form in underventilated conditions.

While studies of underventilated, turbulent flames with various fuels have clearly established the importance of CO formation during underventilated burning,[3] the local conditions leading to CO (and soot) formation in these flames are difficult to deduce because theoretical predictions of turbulent reacting flows are difficult. Studying laminar flames provides a greater opportunity to predict local conditions and correlate them with global CO (and soot) measurements. This is the approach planned for the present project.

The planned research focuses on laminar IDFs because the IDF is related to the underventilated normal diffusion flame (NDF). In an IDF, the positions of the air and fuel streams are interchanged relative to a NDF. Hence, while a NDF is established at the interface between a fuel jet surrounded by air, an IDF is established at the interface between an air jet surrounded by fuel. The IDF and the underventilated NDF are related in that CO and soot which form on the fuel side of the flame can potentially escape without passing through the hot oxidative reaction zone. In the IDF, these fuel-formed species may be convected axially along the outer edge of the flame and bypass the flame tip entirely. In an underventilated NDF, these species may convect axially along the inner edge of the flame and pass through the open portion of the flame tip. In a sense, the IDF is an extreme case of the underventilated NDF, and allows the present study to focus on the type of fuel-formed species which bypass the hot oxidation zone in an underventilated flame.

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LITERATURE REVIEW

The present authors are aware of only one past study of the CO and soot amounts produced by underventilated, *laminar* flames.[4] In this experimental study of NDFs, it was discovered that the ratio of volumetric CO concentration to carbon dioxide (CO_2) concentration was about one to two in the exhaust of underventilated methane (CH_4) and ethene (C_2H_4) NDFs over a range of operating conditions. In addition, the soot from these flames was more organic and agglutinated than that typically emitted from overventilated NDFs. The CO and soot yields were *not* correlated as expected based on overventilated flame studies. This study established the need for further research on the details of CO and soot formation during underventilated burning.

Compared to the vast NDF literature, relatively few studies have been performed using the IDF configuration.[5-22] The first studies of IDFs were published by Arthur and coworkers in the 1950s.[12-15] These authors performed extensive testing on IDFs burning with air and coal gas, CH_4, C_2H_4, propane (C_3H_8), and benzene. They discovered that the soot from IDFs was stickier and more viscous than that from normal flames, and that the hydrogen (H_2) content of the IDF soot was higher than that of NDF soot.[13] This soot structure is qualitatively similar to that discovered in underventilated NDFs.[4] In some cases, because Arthur *et al.* found that IDFs were difficult to stabilize between pure air and pure fuel, they studied flames between central O_2 jets and surrounding mixtures of (fuel + N_2).[12] Several years later, in 1979, Walker reported that, for air/natural gas flames, the IDF tip was more rounded than the NDF tip, the IDF was less luminous than the NDF, and the IDF soot was visible only near the flame tip.[11] This structure is shown schematically in Fig. 1 and in excellent color photographs in Refs 7 and 8.

Kent and Wagner performed a 1984 study of air/C_2H_4 IDFs surrounded by a glass tube.[8] They reported that their flames were stabilized by strong, buoyancy-induced recirculation zones in the annular region between the flame and the glass tube. Hence, it became apparent that IDFs formed between pure air and pure fuel were not independently stable over a wide range of operating conditions. Wu and Essenhigh addressed this issue in 1984.[5] They published stability maps of air/CH_4 IDFs obtained by systematically varying fuel and air inlet velocities. They found five types of flames, ranging from unstable, buoyancy-sensitive blue flames at very low fuel and air velocities, to stable, weak blue flames at low air and moderate fuel velocities, to stronger blue flames with orange-yellow caps at higher air velocities. Interestingly, these authors reported the existence of an unoxidized "pool" of CO and H_2 at the IDF tip.

When Sidebotham and Glassman published a 1992 study of air/C_2H_4 IDFs, they used a heavily-diluted fuel stream (92% N_2) and an oxygen-enriched air stream (36% O_2).[6] These authors determined that CO was formed on the fuel side of the temperature peak and did not penetrate across the reaction zone into the central air jet. They also found that the CO diffused radially outward into the surrounding fuel stream. They stated that the outwardly-diffusing CO could possibly quench in the cool fuel stream and be swept out of the flame as a net emission. Hence, this scenario, along with the flame-tip CO "pool" found by Wu and

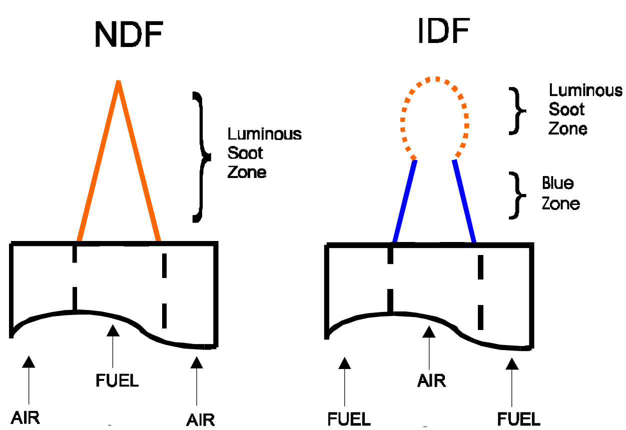

Fig. 1. Schematic showing visible appearance of sooting normal diffusion flame (NDF) and sooting inverse diffusion flame (IDF).

Essenhigh, supports a hypothesis that IDFs will emit large amounts of unoxidized CO, in a manner which is similar to underventilated NDFs.[4]

In 1994, Makel and Kennedy studied air/ C_2H_4 IDFs stabilized by a H_2 pilot flame.[9] They found that the integrated axial soot volume fraction in each flame increased and leveled off to a constant value with increasing axial distance, in contrast to the increasing and then decreasing trend measured in C_2H_4/air overventilated NDFs.[24] The IDF soot volume fractions presumably did not decrease because the soot did not pass through an oxidizing zone at the flame tip. This supports the hypothesis that IDF soot follows a pathway which is similar to that followed by underventilated NDF soot (i.e., it is not fully oxidized before exiting the flame).

Kang *et al*. published a 1997 study of air/C_3H_8 IDFs with diluted fuel (70% N_2) and oxygen-enriched air (40% O_2).[10] These authors demonstrated that the soot in an IDF experiences a temperature-time history similar to that of soot in a counterflow flame with the flame on the oxidizer side of the stagnation plane. They coined the phrase “soot formation flame” to describe these types of flames, as opposed to the phrase “soot formation-oxidation flame,” which was used to describe NDFs and counterflow flames established on the fuel side of the stagnation plane.

The studies discussed above demonstrated that (1) buoyancy-induced instabilities impede systematic variation of pure IDF operating conditions in normal gravity, and (2) IDF CO and soot formation is similar to that in underventilated NDFs. The use of microgravity should eliminate the buoyancy-induced instabilities and allow a systematic study of underventilated-flame CO and soot formation.

To the present authors’ knowledge, only two groups have previously studied microgravity IDFs.[25,26] Law *et al.* studied IDF structure in a low-pressure (1/4 atm) chamber.[25] These authors stabilized a burner-supported, spherical IDF by issuing a mixture of 10% O_2 and 90% N_2 through a porous sphere into a still environment containing H_2 mixed with about 2% CH_4. These authors did not report visible soot in their flames. Sunderland *et al*. found that drop-tower stabilized spherical IDFs of air issuing into C_2H_4 produced large amounts of soot which exited the flame without passing through the hot oxidation zone.[26] It is interesting to note that low-flow-rate CH_4/air and C_3H_8/air NDFs established in the NASA 2.2-Second Drop Tower had open tips similar to underventilated flames.[27,28]

RESEARCH PLAN

The specific project tasks are (1) to attempt to stabilize IDFs in a series of 2.2-second drop-tower experiments and capture the events with a video camera for assessment of their stability, (2) to gain an understanding of CO and soot formation in IDFs by analyzing the post-combustion products of CH_4 and C_2H_4 flames under normal and low gravity conditions, (3) to use a fiber-optic-coupled CO sensor to monitor the IDF tips during microgravity combustion, and (4) to model the flow field and soot particle temperature-time histories in both normal and low gravity conditions using an existing NIST computer program.[29] Post-flame amounts of CO and soot will be collected and measured. The soot will be analyzed for organic and graphitic content, for carbon and hydrogen content, and for primary sphere size, agglomerate size, and structure. The fiber-optic CO sensor will be based on near-infrared tunable diode laser absorption. The computer code features a particle transport model which includes the effects of inertial, thermophoretic, and gravitational forces.[30] The local computed flow conditions will be combined with the measured CO and soot information to gain insight into the conditions leading to the formation of these species during underventilated combustion.

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