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Feature article

Influence of halon replacements on laminar flame speeds and extinction limits of hydrocarbon flames



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ABSTRACT

The influence of some halon replacements as the fire suppressants on premixed and non-premixed hydrocarbon flames was experimentally and numerically investigated. Unstretched flame speeds and extinction stretch rates of methane/air and propane/air flames with the addition of different loadings of C₂HF₅ (HFC-125), C₂HF₃Cl₂ (HCFC-123), and C₃H₂F₃Br (2-BTP) were measured for a range of fuel/air equivalence ratios. Experiments were conducted using the counterflow technique at ambient temperature and pressure. A newly-developed kinetic model, which was based on a C_1-C_4 hydrocarbon model and agent-inhibition sub-models, was used to compare with the measured flame speeds and extinction stretch rates. Good agreement was obtained regarding the laminar flame speeds. For the extinction rate of the non-premixed flames, the absolute experimental data were over-predicted uniformly for the entire range of conditions examined, although the model correctly predicted the trends of increasing extinction stretch rates with the increasing fuel/ $(N_2+agent)$ molar ratio. Since this new kinetic model was tested against the extinction limits for the first time, the optimization of the rate constants of the reaction(s) regarding the flame extinction is expected. Significant thermal expansion was detected in larger and brighter flames when the different agents were added to the neat mixture. This observation provided new experimental evidence of the fuel-like properties of the inhibitor. Despite the difference in the fuel type, C_2HF_5 and $C_3H_2F_3Br$ were found to be less effective at reducing the flame speeds of the lean flames as compared to that of the rich flames. However, C₂HF₃Cl₂ reduced the flame speeds for all of the investigated mixture conditions. Thermodynamic equilibrium calculations were performed to interpret the behavior of these fire suppressants in hydrocarbon flames. The extinction data for non-premixed flames revealed that the resistance of the agents to extinction decreases in the order of C2HF5, C2HF3Cl2, and C3H2F3Br, which is consistent with the suppression effect on flame speed.

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1. Introduction

As the effective fire suppressant CF₃Br (Halon 1301) has been largely phased out, a worldwide effort is directed towards the search of suitable halon replacements with certain properties, i.e., alternatives that are clean (low ozone-depleting potential, ODP) but have favorable performance comparable to Halon 1301. Several replacement agents are currently being developed for terrestrial applications and the use in the civilian aerospace industry. However, the potential halon replacements were observed unusual behavior when added at sub-inerting concentrations. According to the Federal Aviation Administration (FAA) Aerosol Can Test (ACT) report [1–4], the three tested alternatives (C_2HF_5 , HFC-125; $C_3H_2F_3Br$, 2-BTP; and $C_6F_{12}O$), caused an overpressure rise several

* Corresponding author. E-mail address: yjjiang@ustc.edu.cn (Y. Jiang). times larger than that with no agent (e.g., the maximum pressure rise was 6.4 bar for 2-BTP compared to that of 1.38 bar for the uninhibited one [4]) and thus failed the qualification test.

Enhanced combustion in the FAA-ACT has drawn particular attention (e.g., [5–12]) and the enhancement effect was reported in many laboratory-scale flames, as well as numerical simulations. Previous studies suggested that the enhancement was caused by extra heat release from the exothermic reactions of inhibitor, which increased the overall reactivity of the system. Nonetheless, these replacements have the potential to circumvent the problems of sub-inerting concentrations. Additionally, the effectiveness of the replacement agents in practical applications has been widely acknowledged. Linteris et al. [13] observed a large reduction of the burning velocity as C_2HF_5 was added to rich CH_4/air flames. The performance of $C_3H_2F_3Br$ has also been examined with great expectations, as it has a similar chemical structure to CF_3Br . In extinguishing a cup burner flame, Moore et al. [14] observed a minimum extinguishing concentration of 2.6% for $C_3H_2F_3Br$, which is

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lower than that for CF₃Br (2.9%). As a type of HCFC fire suppressant, $C_2HF_3Cl_2$ is considered as an acceptable alternative to Halon 1301 despite being listed as a controlled substance by the Montreal Protocol, because $C_2HF_3Cl_2$ has a very low ODP (0.012) and is expected to reduce the overpressure in the FAA-ACT. A recent study reported that $C_2HF_3Cl_2$ has a better performance than the C_2HF_5 agent in the hydrocarbon flame inhibition because chlorine reactions are more effective than that of fluorine reactions.

The laminar flame speed is an intrinsic property of a combustible mixture, as it provides the basic information regarding the chemical reaction rate, thermal diffusivity and heat release of the flame. Experimental data on the laminar flame speed are considered to be a useful tool for characterizing the effectiveness of fire suppressants, and the accurate knowledge of the laminar flame speed has served to validate and optimize chemical kinetic mechanisms. Regarding measurements of the laminar flame speed related to hydrocarbon gas mixtures inhibited by halogenated suppressants, few data are available in the literature. Linteris et al. [15] for the first time used the total area method to measure the burning velocity of CH₄/air mixtures with the addition of CH₂F₂, CF₃H and CF₄. Leylegian et al. [16,17] used a counterflow twinflame configuration to measure the laminar flame speeds of blends of CH_2Cl_2 and $CHCl_3$ with CH_4 /air mixtures. Saso et al. [18] investigated the laminar flame speed of the methane flames inhibited by CHF₃ using a similar configuration. Kim et al. [19] used the spherically propagating flame technique to measure the laminar flame speeds of H₂/air flames with Halon 1301 added. More recently, Osorio et al. [20] measured the laminar flame speeds of CH₄, C₂H₆ and C₃H₈ flames with Halon 1301 added using a spherically flame facility. However, very few flame speed data were available regarding the inhibition of the three potential halon replacements (C₂HF₅, C₃H₂F₃Br, and C₂HF₃Cl₂) in hydrocarbon-air mixtures until the recent study performed by Pagliaro et al. [21,22], in which spherically expanding flames were used to measure flame speeds of methane and propane flames with the addition of the agents tested in the FAA-ACT. Very useful experimental data were obtained under the condition of relatively simple methane flames and limited equivalence ratios. These flame speed data can be used to test the accuracy of the chemical kinetic model.

Given that the laminar flame speed and extinction strain rate are both fundamental combustion data, extinction strain rates can serve as a benchmark for the assessment of new Halon alternatives and as a metric for model verification. An accurate reaction model should have the ability to predict different flame kinetic parameters. The extinction limits derived from the counterflow flames add additional constraints for the validation and serve as a supplement to the laminar flame speed. Previous studies on the extinction limits of halogenated flames mainly employed the cup burner method, and the results were significantly influenced by the cup size and the burner chimney, making the comparison of different reported data and the comparison of the data with model predictions very difficult. Moreover, for numerical study, describing a cup burner flame in a two-dimensional (2-D) field requires complex coding. An alternative method for investigating the extinction mechanism involves the counterflow flames, which have a well-controlled flow field that can be more easily modeled along the one-dimensional (1-D) stagnation streamline. Thus far, the newly developed kinetic model for halon replacements in hydrocarbon-air flames has not been tested against the experimental data for extinction strain rates and further validation is needed.

To this end, the present work extends the measurements of laminar flame speeds and provides new extinction stretch rate data for CH_4 -air and C_3H_8 -air flames with added C_2HF_5 , $C_3H_2F_3Br$, and $C_2HF_3Cl_2$ over a wide range of equivalence ratios and agent loadings. The experimental values are compared to the computational

results obtained using a recently developed reaction kinetic model. The effect of the replacement agents on the opposed flow-field dynamics is examined. The objective of this study was to provide experimental data for model validation and to investigate the inhibition effectiveness of C_2HF_5 , $C_3H_2F_3Br$, and $C_2HF_3Cl_2$ with different types of fuels and combustion regimes (premixed and nonpremixed flames).

For the measurements of the flame speed and extinction limit, an opposed-jet counterflow configuration was used. It was challenging to deal with the sealing and exhausting issues with regard to the product gases because the combustion of the halogenated hydrocarbon/air mixtures was somewhat toxic (mainly released acid gas, e.g., HF and CF₂O). Compared to a constant-volume combustion bomb, counterflow configurations are more likely to cause the exposure of experimenters and are impaired by the acidic environment, hence only a very few studies were conducted using such a configuration. However, to elucidate the effects of the halon replacements on the hydrocarbon combustion, a straightforward observation of the flames would be helpful. Counterflow flames have a steady, 1-D, planar, and laminar hydrodynamic reaction zone that is easily characterized by numerical simulations. Furthermore, with the unique advantage of counterflow flames, the extinction strain rates of non-premixed flames were obtained, which are useful for chemical model validation.

2. Experimental approach

The counterflow configuration shown in Fig. 1 is based on previous designs [12,23]. Two burners that have the same construction were installed symmetrically with a certain separation distance. Each burner had a streamline contracted nozzle designed with a cycloid curve, which imposed a top-hat exit velocity profile. Nitrogen was supplied around the nozzle to shield the reactive jet from the ambient environment. When the combustible mixture was ignited, two symmetrical, laminar and planar flames could be established between the nozzles. To prevent overheating, a cooling water system kept the nozzles at ambient temperature.

The gas flow control system comprised calibrated sonic nozzles (O'Keefe Metal Orifice Assemblies) and pressure regulators (with a claimed accuracy of 0.5%). The control of the gas flow rates were achieved by fixing the upstream pressure of the sonic nozzles at a certain value, and thus the desired amount of gas was delivered. In this study, the sample gases were CH_4 (99.99%), C_3H_8 (99.5%), N_2 (99.99%), C_2HF_5 (99.5%), $C_2HF_3Cl_3$ (99.5%), and $C_3H_2F_3Br$ (99%). Air was modeled by the mixture of 21%O₂ and 79%N₂. The reactant gases were well mixed in the mixing chamber before entering the burner. A nebulizer was employed to generate seeding particles, which served as the digital particle image velocimetry (DPIV) tracers. These particles were submicron-size silicon oil droplets that have excellent traceability in the flow. The flow rate of the silicon oil was controlled using a syringe pump at small values, i.e., 0.03–0.05 mL/min.

The 2-D flow velocity field between the opposed burners was measured using the planar DPIV technique. A double-pulsed Nd:YAG laser (Litron lasers) and a CCD camera (LaVision Imager proX4M) were used. The particles in the flow were illuminated twice by a laser light sheet (532 nm, 80 mJ/pulse) within a very short time interval. Synchronized particle images were captured by external CCD camera (50 mm/F4.0); thus, the displacement of the velocity vectors of the tracers was determined using the PIV vector calculation. The delay between the laser pulses Δt (approximately 80–150 µs) was adjusted such that the maximum particle image shift in the reference flame speed was limited within a quarter of the interrogation window size. To improve the particle image quality, a 532-nm optical bandpass filter was placed on the camera lens to minimize the luminous emission

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