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Extinction of Nonpremixed Opposed-Flow Hydrocarbon Flames by Chemically-Passive Fire Suppressants

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[Abstract] Effects of chemically-passive fire suppressants on the critical strain rate required to extinguish hydrocarbon-air opposed-flow diffusion flames were investigated by combined use of experiments and computations. Extinction strain rates were determined for fuel streams consisting of pure CH_4, C_2H_6, C_3H_8 or C_2H_4 and oxidizer streams composed of air with 0-30% volume fractions of Ar, N_2 or CO_2 as inert suppressants. Relative suppressant effectiveness increased for all fuels in order from Ar to N_2 to CO_2, consistent with the increase in specific heat due to the resulting suppressant concentration in a stoichiometric mixture of the fuel and oxidizer streams. The higher suppressant effectiveness in the C_2H_4 flames relative to previous measurements in wet-CO flames reflects the role of peak H-radical concentrations in the flame extinction process. Accordingly, although extinction strain rates for all fuels and suppressants correlate well with peak temperature just below the extinction limit, better correlation is found with the peak value of H radical concentration and with corresponding H+OH and H+O radical concentrations. However in contrast to suppressant effects in premixed flames, the present results do not indicate that the chain-terminating three-body recombination reaction $H+O_2+M \rightarrow HO_2+M$ controls the suppressant effectiveness in diffusion flames.

I. Introduction

EFFECTIVE fire-suppression systems are essential for terrestrial as well as spacecraft fire safety. Traditionally, chemically-active agents such as Halon 1301 (CF3Br) have been used for fire extinction [1], and the underlying suppression mechanism that determines the effectiveness of these compounds has been understood for some time [2-4]. However because these compounds contribute to stratospheric ozone depletion, their use is being increasingly restricted. Moreover in closed spacecraft environments, the reaction products they generate can be dangerous to health and life-support systems [5-6]. As a result, there is considerable interest in understanding the relative effectiveness of alternative chemically-passive fire suppressants, and in understanding the fundamental mechanisms that determine the effectiveness of

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any given suppressant for any given fuel. The present study thus investigates the underlying suppression mechanism that leads to the relative effectiveness of certain chemically-passive fire suppressants on the extinction of diffusion flames.

Prior fundamental investigations of fire suppressants have been conducted in both premixed [7, 8] and diffusion [9, 10] flames, but have focused primarily on the effects of chemically-active fluorinated or brominated agents. The present study uses a steady laminar opposed-flow diffusion flame configuration to investigate fundamental aspects associated with the effectiveness of chemically-passive suppressants on the critical extinction strain rate for several key hydrocarbon fuels burning in air. The opposed-flow diffusion flame allows measurements under carefully controllable conditions, as well as corresponding one-dimensional calculations of flame properties to assist in interpreting the underlying extinction mechanisms. Previous studies of laminar opposed-flow diffusion flames without suppressants have shown that extinction occurs as a result of conductive heat losses from the reaction zone as the strain rate is increased [11-16]. Additionally, radiative heat losses from the reaction zone can also contribute to extinction, and when the nominal strain rate is low buoyancy can produce additional strain that can lead to extinction [17]. In the present study, effects of buoyancy and radiation have been kept sufficiently small to allow investigation of the purely strain-induced extinction of opposed-flow diffusion flames in the presence of chemically-passive fire suppressants.

While most opposed-flow diffusion flames studies have focused on extinction of unsuppressed flames, several prior investigations have included limited results for the effects of inert diluents on the extinction limits of such flames. In particular, Milne et al [9] and Ishizuka and Tsuji [18] together have shown that increasing concentrations of He, Ar, N_2 and CO_2 in the fuel or oxidizer stream act to reduce the peak flame temperature until an extinction limit is reached. Puri and Seshadri [19] examined the effect of N_2 dilution level on the extinction strain rates of methane-air and propane-air flames. Their results showed a large reduction in extinction strain rate as the N_2 dilution level was increased, but was not directed at different fuels and suppressants to identify the underlying mechanism responsible for the effectiveness of various such inert species as fire suppressants for various types of fuels.

The present study specifically investigates the effects of Ar, N_2 and CO_2 as chemically-passive monatomic, diatomic and triatomic fire suppressants supplied at concentrations up to 30% by volume in air to the oxidizer stream in a steady laminar opposed-flow diffusion flame. Extinction strain rates are experimentally measured for fuel streams consisting of pure CH_4, C_2H_6, C_3H_8 or C_2H_4. These fuels allow comparisons of results between CH_4 and C_2H_4 and between C_2H_4 and C_2H_6 to separately investigate effects of changing only the number of carbon or hydrogen atoms, and comparisons of results between CH_4, C_2H_6 and C_3H_8 to investigate the effects of simultaneously changing the number of carbon and hydrogen atoms. Collectively these results address key aspects of a wide range of practical combustible materials. For the twelve fuel and suppressant combinations, the extinction strain rates were measured at up to four different suppressant concentrations. For each case, corresponding OPPDIF calculations with the GRI-Mech 3.0 reaction mechanism were used to provide temperature and radical concentrations at strain rates immediately below the respective extinction limits. The results have been used to examine various general correlations that seek to identify the underlying mechanisms that control the suppressant effectiveness. Such correlations in principle would allow a priori assessments of the effectiveness of any other chemically-passive suppressant for any other hydrocarbon fuel.

II. Flame Extinction Measurements

A. Apparatus

A stable, planar, laminar, opposed-flow diffusion flame was established at the center of an 8-mm-wide vertical gap separating the exit planes of a lower fuel tube and an upper oxidizer tube. The fuel and oxidizer each issued through central tubes having an inner diameter of 10.3 mm, a wall thickness of 0.4 mm, and a length of 200 mm. The diffusion flame was positioned at the center of the vertical gap by adjusting the momentum of the fuel and oxidizer streams. The fuel and oxidizer streams were each surrounded by an annular coflowing nitrogen stream that issued from a coaxial outer tube having an inner diameter of 18.3 mm and a wall thickness of 0.4 mm. The coflowing nitrogen stream velocities were each matched to the respective fuel and oxidizer stream velocities. The nitrogen coflow eliminated entrainment into the fuel and oxidizer streams and minimized disturbances from the surrounding environment. To smooth the velocity profiles issuing from the tubes, each of the two inner tubes contained two 25-mm-long honeycomb flow straighteners having 1.6 mm cell size and located 70 mm and 140 mm upstream of the exit. The fuel, oxidizer and nitrogen streams were metered and controlled using pressure regulators and precision rotameters that had been calibrated with either wet-test or bubble flow meters.

B. Reactant Stream Compositions

The hydrocarbon fuels were supplied from high-purity cylinders of methane, ethane, ethylene or propane. The oxidant stream consisted of mixtures of air and suppressant (Ar, N_2 or CO_2) supplied from commercially-prepared high-pressure cylinders premixed to 0%, 10%, 20% or 30% suppressant by volume in order to minimize variability from oxidizer mixture blending by in-line flow metering. For each fuel and suppressant type, the suppressant concentrations investigated are listed in Table 1. Prepared cylinders of air mixed with suppressant allowed the fuel and oxidant flow rates to the opposed-flow diffusion flame to be gradually increased until the extinction strain rate was reached, while at the same time keeping the oxidant mixture composition constant and maintaining the flame position at the center of the gap. The fuel and oxidizer flow rates at the extinction limit, together with the 8 mm gap width, determined the extinction strain rate aq [20].

Table 1. Test conditions for hydrocarbon fuels

Fuel Type	Suppressant	X_s
CH_4	Ar	0, 0.1, 0.2, 0.3
	N_2	0, 0.1, 0.2
	CO_2	0, 0.1
C_2H_4	Ar	0, 0.1, 0.2, 0.3
	N_2	0, 0.1, 0.2, 0.3
	CO_2	0, 0.1, 0.2
C_2H_6	Ar	0, 0.1, 0.2, 0.3
	N_2	0, 0.1, 0.2, 0.3
	CO_2	0, 0.1, 0.2
C_3H_8	Ar	0, 0.1, 0.2, 0.3
	N_2	0, 0.1, 0.2, 0.3
	CO_2	0, 0.1

III. Flame Calculations

For each case in Table 1, the extinction strain rate and the flame structure immediately below the extinction limit were calculated using the OPPDIF one-dimensional opposed-flow diffusion flame code [21]. The CHEMKIN package was used to evaluate chemical reaction rates and thermodynamic and transport properties. The GRI-Mech 3.0 detailed chemical reaction mechanism, consisting of 325 elementary reactions involving 53 species, was used as the first part of the mechanism describes the reaction chemistry of CH_4. The second part of the mechanism was taken from Ref. 35 and involves the reactions of C2-C3 hydrocarbon fuels considered herein. Selected cases were computed separately both with and without the optically-thin radiation model. Results indicated that radiation losses were negligible for the present conditions, and thus all of the computed results included herein are from radiation-free calculations.

To allow comparisons between calculated and measured values for the extinction strain rates a_q, the strain rate in the calculations was defined from the maximum absolute value of the velocity gradient on the oxidizer side; note that this is also the point where the temperature starts to increase sharply. The resulting strain rate values are consistent with the essentially uniform velocity gradient that results from OPPDIF across the interior of the gap for the corresponding nonreacting case [22]. This way of defining the strain rate is also consistent with the widely used choice of the velocity gradient immediately ahead of the preheat zone on the oxidizer side [23, 24].

IV. Flame Extinction Strain Rates

Flame extinction strain rates aq were measured for the four hydrocarbon fuels CH_4, C_2H_4, C_2H_6, and C3H8 and for the three chemically-passive suppressants Ar, N_2 and CO_2 at each of the concentrations shown in Table 1. For each fuel and all suppressants, the critical extinction strain rate decreased with increasing suppressant concentration. Figure 1 shows the effect of varying the suppressant type on the measured extinction strain rate for a given fuel, in this case showing results for the C_2H_4-flames. Corresponding results for the other fuels were qualitatively similar, though the extinction strain rate at zero suppressant concentration, here denoted a_{q0}, depends on the particular fuel. For all these fuels, the relative suppressant effectiveness $1-(a_q / a_{q0})$, giving the fractional reduction in extinction strain rate relative to the unsuppressed value, increased in order from Ar to N_2 to CO_2, with the latter being the most effective. The observed ordering from the monatomic to the triatomic suppressant is consistent with the simple increase in the specific heat of the non-fuel gases per unit oxygen concentration in a stoichiometric mixture of the fuel and oxidizer streams. Huggett [25] has shown that a wide variety of fuels will not burn in oxygen-containing atmospheres if the heat capacity per gmol of oxygen in the atmosphere exceeds a critical value. This specific heat effect is also consistent with previous observations of suppressant type and concentration on laminar burning velocities of premixed flames [26, 27]. In the present diffusion flames, the increased specific heat due to the suppressant leads to a corresponding reduction of the reaction zone temperature in these flames. This in turn contributes to an associated decrease in the reaction rates, and thereby leads to a reduction in the strain rate at which extinction occurs. The effect of suppressant type and concentration on the extinction mechanism will be examined in Section 5.

Figure 2 shows the effect of varying the fuel type for a given suppressant, in this case showing results obtained for Ar as the suppressant. Corresponding results for the other suppressants were qualitatively similar. For any given concentration of the suppressant, the measurements show the flames to become increasingly more difficult to extinguish in order from CH_4, C_3H_8, C_2H_6 to C_2H_4, with the latter having the highest extinction strain rate. This ordering is consistent with the relative reactivity of each fuel as characterized by its peak laminar burning velocity in a corresponding unsuppressed premixed flame. Ethylene has the highest laminar burning velocity among these fuels, and methane the lowest. Moreover among the alkane fuels a further indication of the relative reactivity is given by the inverse of the ignition temperature, which increases in order from CH_4 to C_2H_6 to C_3H_8 [28, 29]. The effect of this will also be examined in Section 5.

The above observations of suppressant effects on the extinction strain rates of hydrocarbon-air flames suggest a comparison with recent results for suppressant effects on diffusion flames of wet-CO fuel burning in air. Figure 3 shows the present measured values of suppressant effectiveness in terms of a_q / a_{q0} for the most reactive fuel, C_2H_4, as well as corresponding measured values [30] for a fuel composed of 95% CO with 5% H_2, as the suppressant concentration increased. The relative suppressant effectiveness can be seen to be higher in the C_2H_4 flames. Owing to the higher H-C ratio, the reduction in peak flame temperature with increasing suppressant concentration suggests a larger reduction in H-radical concentration within the reaction zone for the C_2H_4 flame than for the wet-CO flame. The larger reduction in H-radicals should in turn lead to a proportionally larger reduction in the rate of chain branching through the reaction $H+O_2 \rightarrow O+OH$, which reduces radical concentrations in the reaction zone and thereby reduces the strain rate at which extinction can occur.

Figure 4 shows comparisons between measured and computed values of the extinction strain rates for the most reactive (C_2H_4) and least reactive (CH_4) cases among the hydrocarbon fuels considered here. The computed aq values for all fuels over the entire range of suppressant concentrations follow the same trends as the measured values, but underpredict the absolute extinction strain rates. While previous measurements of chemically-passive suppressant effects in near-limit premixed flames [27] have indicated possible uncertainties in third-body efficiencies of key reactions relevant to extinction, here the differences between computed and measured extinction limits are more likely due to small differences in the flow field assumed in OPPDIF and that obtained in the experiments, and the fact that strain rate values in the experiments were obtained indirectly using the formulation of Seshadri and Williams [20]. In the following section we thus use the computed results for the flame structure to obtain insights into suppressant effects on the extinction limit.

V. Mechanism for Suppressant Effectiveness

A sufficiently fundamental understanding of the mechanism by which chemically-inert suppressants affect the extinction strain rate of diffusion flames would in principle allow initial a priori assessment of the relative effectiveness of essentially any suppressant for essentially any fuel. The results in Fig. 1 have already indicated that the effectiveness of Ar, N_2 and CO_2 are consistent with their effect on the specific heat of the stoichiometric fuel-air mixture. This in turn suggests that there will be a correlation of relative suppressant effectiveness with peak flame temperature. To test this, Fig. 5a shows the measured

a_q / a_{q0} values versus peak flame temperature for all fuels, suppressant types, and suppressant concentrations. The temperature values for each case are from the OPPDIF calculations at incipient extinction, and are shown referenced to the 300K temperature of the fuel and oxidizer streams. It is apparent that, as expected from the simple specific-heat mechanism, there is considerable correlation between the effectiveness of any suppressant and its effect on reducing the peak flame temperature. However, it is also apparent that there are additional effects that play a significant role in determining the suppressant effectiveness. In particular, the results for CH_4 and C_2H_4 in Fig. 5a appear to follow a substantially different correlation than do those for C_2H_6 and C_3H_8. Thus while these results verify the dominant role of the simple specific heat effect, they suggest that additional chemical mechanisms must be accounted for in the flame extinction mechanism to permit a broadly useful assessment of the fire-suppressant effectiveness of any given chemically-inert suppressant for any given fuel.

An obvious chemical effect of suppressant addition is to reduce the oxygen availability in the flame. The oxygen index has been widely used in premixed flame suppression to specify requirements for inerting any given atmosphere, including spacecraft atmospheres having reduced pressure but elevated oxygen concentration, to prevent fires or extinguish flames [31, 32]. This suggests that oxygen index may also provide an effective means for correlating suppressant effects in diffusion flames. Consistent with the results in Fig. 1, oxygen index values for premixed flame suppression using CO_2 are typically larger than those for suppression with N_2, requiring lower suppressant concentrations to achieve flame extinction.

The above considerations indicate that the peak concentration of key radical species may provide a better correlation of suppressant effectiveness than does the simple temperature effect examined above. In particular, it is well known that H, O, and OH radicals play key roles in the chain-branching and chain-terminating reactions that ultimately determine the extinction strain rate in unsuppressed hydrogen-air and hydrocarbon-air flames. This suggests that the effect of suppressants in altering the concentrations of these radicals may provide a more fundamental correlation for the relative effectiveness of any given suppressant for any given fuel. Since these radical concentrations can be expected to decrease with decreasing peak flame temperature, a correlation with radical concentration will include the simple temperature effect, but may also permit better correlation of extinction strain rate for different fuel types.

In particular, the decrease in peak flame temperature due to suppressant addition can be expected to lead to reduced H-radical concentrations, which will reduce the chain-branching reaction rate $H+O_2 \rightarrow O+OH$. This in turn will lead to reductions in concentrations of O and OH radicals. Based on these considerations, Figs. 5b-d examine the correlation of measured a_q / a_{q0} values with the peak value of H radical concentration and with corresponding H+OH and H+O radical concentrations for all fuels, suppressant types, and suppressant concentrations. The radical concentrations for each case are from OPPDIF calculations at incipient extinction. It is apparent that there is considerable correlation between the suppressant effectiveness and these radical concentrations. Moreover, in contrast to the simple temperature correlation in Fig. 5a, all three radical correlations in Fig. 5b-d show the results for CH_4 to be in substantially better agreement with those of the other alkane fuels C_2H_6 and C_3H_8. All three alkane fuels can be seen to follow essentially the same correlation of relative suppressant effectiveness with radical concentrations. The degree of correlation is about equally good for all three radical groups, but is highest for H+OH. This appears consistent with the findings that suppressant effects on laminar burning velocities in premixed flames correlate well with the peak H-radical concentration in the reaction zone [33, 34], but that the strongest correlation is found with the peak H+OH concentration [26].

While the three alkane fuels correlate relatively well in Figs. 5b-d, the correlation for the alkene fuel C_2H_4 differs substantially from that for the alkanes. In particular, for ethylene the same relative suppressant effectiveness a_q / a_{q0} occurs at larger values of H-radical concentration. This is consistent with the expected peak H-radical concentrations for the alkane and alkene fuels based on the fast chain-initiating reactions for each fuel type. For the alkene fuel, the higher energy of the double carbon bond leads to H_2 and H-radical production at the chain-initiation step, while for the three alkane fuels the lower energy of the single carbon bonds do not produce H_2 or H-radicals at the initiation step. Accordingly, the same relative suppressant effectiveness in Figs. 5b-d corresponds to the same H-radical concentration for all three alkane fuels, but corresponds to higher H-radical levels for C_2H_4.

Studies of suppressant effects in premixed flames have identified the important role of the three-body recombination reaction $H+O_2+M \rightarrow HO_2+M$, which competes with the chain-branching reaction to maintain the flame [26, 27]. This might suggest a comparably important role for this reaction in determining suppressant effectiveness in diffusion flames. Figure 6 thus examines the effects of the three chemically-passive suppressants Ar, N_2 and CO_2 on the reaction rates for the chain-branching reaction $H+O_2 \rightarrow O+OH$ in Fig. 6a and chain-terminating reaction $H+O_2+M \rightarrow HO_2+M$ in Fig. 6b. Results are shown from OPPDIF calculations of the C_2H_4 flames in Fig. 1 without suppressant and with 20% mole fraction of each suppressant, with the calculation in each case done at a strain rate corresponding to incipient extinction. In Fig. 6a it is apparent that the suppressants reduce the peak value of the chain-branching reaction rate in order of increasing effectiveness from Ar to N_2 and CO_2. This is the same ordering as the suppressant effect on a_q / a_{q0}, suggesting that this chain-branching reaction plays a key role in diffusion flame extinction by suppressant addition.

By comparison, Fig. 6b shows corresponding results for suppressant effects on the chain-terminating reaction $H+O_2+M \rightarrow HO_2+M$. Here it is apparent that CO_2 produces a large increase in the rate of this reaction, consistent with CO2 being the most effective suppressant in Fig. 1. However, the chain-terminating reaction rates for Ar and N_2 are seen in Fig. 6b to be lower than those in the unsuppressed flame, rather than higher, despite the fact that both suppressants reduced the critical strain rate needed to extinguish the diffusion flame in Fig. 1. This indicates that the reaction $H+O_2+M \rightarrow HO_2+M$ does not appear to control the suppressant effectiveness in these diffusion flames.

Further evidence of this can be found in Fig. 7, where reaction rates are shown for the same C2H4 diffusion flame without suppressant and with 20% mole fraction of each suppressant, but with the calculation now done at the same strain rate for all cases. Rates for the chain-branching reaction $H+O_2 \rightarrow O+OH$ are shown in Fig. 7a and for the chain-terminating reaction $H+O_2+M \rightarrow HO_2+M$ in Fig. 7b. For the chain-branching reaction the order in which the total reaction rate decreases among the suppressants is consistent with the relative effectiveness of each suppressant. However, for the chain-terminating reaction the total reaction rate for Ar and N_2 suppressants are again smaller than in the unsuppressed flame.

Collectively these results suggest that while the $H+O_2+M \rightarrow HO_2+M$ reaction may be potentially significant contributor to the higher effectiveness of the CO_2 suppressant, it does not play a significant role in the effectiveness of the Ar and N_2 suppressants. Instead, the effect of each suppressant in reducing the chain-branching reaction $H+O_2 \rightarrow O+OH$ appears to a far more significant factor in determining the relative effectiveness of these chemically-passive suppressants.

VI. Conclusions

An opposed-flow diffusion flame has been used to determine effects of chemically-passive fire suppressants on the critical strain rate needed to extinguish hydrocarbon flames of pure CH_4, C_2H_6, C_3H_8 or C_2H_4 burning with an oxidizer stream consisting of air containing 0-30% volume fraction of Ar, N_2 or CO_2 as inert suppressants. Results show that for all fuels these suppressants perform in order of increasing effectiveness from argon to nitrogen and carbon dioxide, consistent with the increase in specific heat due to the resulting suppressant concentration in a stoichiometric mixture of the fuel and oxidizer streams. A substantially higher suppressant effectiveness is observed in the present C_2H_4 flames than in previous measurements of wet-CO flames, consistent with the role of H-radical concentrations in the flame extinction process and the suppressant effect in reducing peak levels of H-radicals. Measured extinction strain rates for all fuels and suppressants correlate well with peak temperature just below the extinction limit. Substantially better correlation is found with the peak value of H-radical concentration, and with corresponding concentrations of H+OH and H+O radicals, especially for the alkane fuels. In contrast to suppressant effects in premixed flames, the present results do not indicate that the chain-terminating three-body recombination reaction $H+O_2+M \rightarrow HO_2+M$ controls the suppressant effectiveness in diffusion flames.

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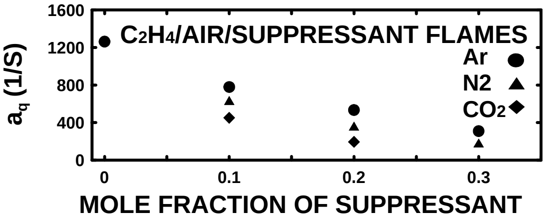

Fig. 1. Measured extinction strain rates as a function of mole fraction of suppressants for C_2H_4-air opposed-flow diffusion flames.

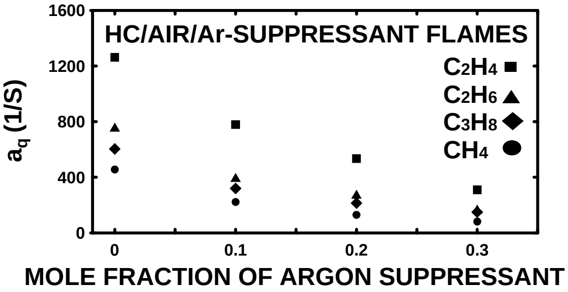

Fig. 2. Effect of fuel type on measured extinction strain rates for hydrocarbon-air diffusion flames with varying concentrations of Ar suppressant.

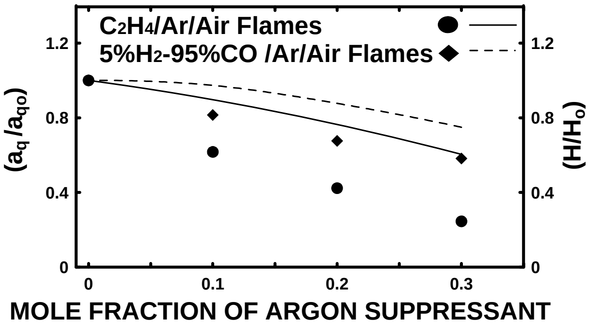

Fig. 3. Relative effectiveness of Ar suppressant in present C_2H_4-air flames and previous wet-CO flames [30], showing extinction strain rates (symbols, left axis) and peak H-radical concentration (lines, right axis), both normalized by corresponding unsuppressed values.

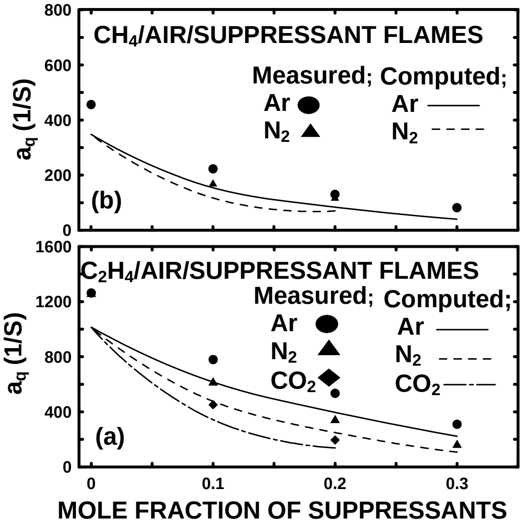

Fig. 4. Measured and calculated flame extinction strain rates as functions of suppressant concentration for *(a)* C_2H_4 flames and *(b)* CH_4 flames.

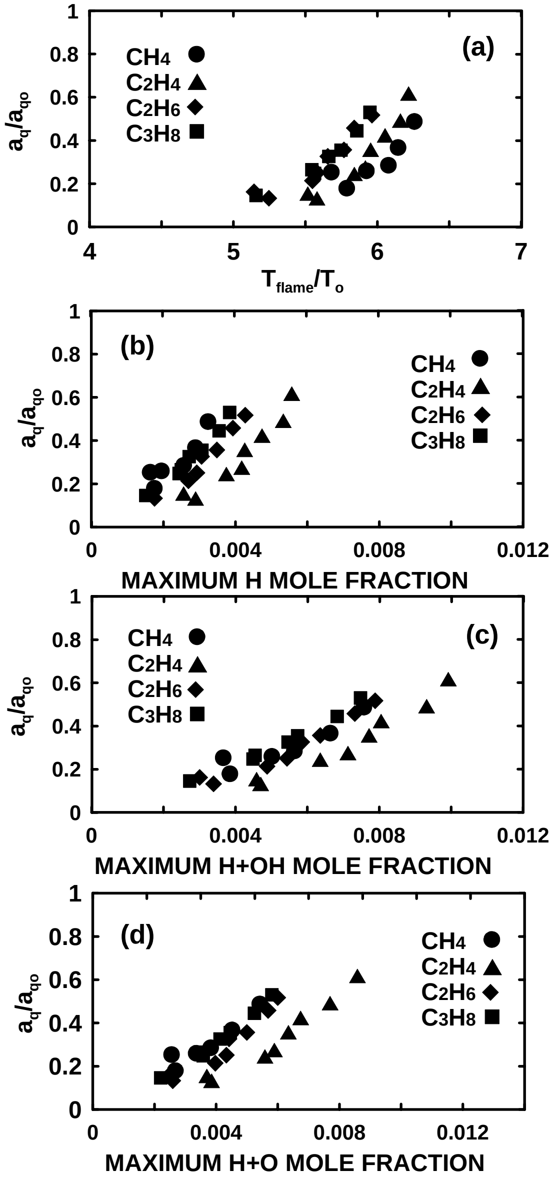

Fig. 5. Correlation of relative suppressant effectiveness for all fuels (CH_4, C_2H_4, C_2H_6, C_3H_8) and suppressant types (Ar, N_2, CO_2) and concentrations with peak values at incipient extinction of *(a)* temperature, and mole fractions of *(b)* H, *(c)* H+OH, and *(d)* H+O radicals at location of maximum H-radical concentration.

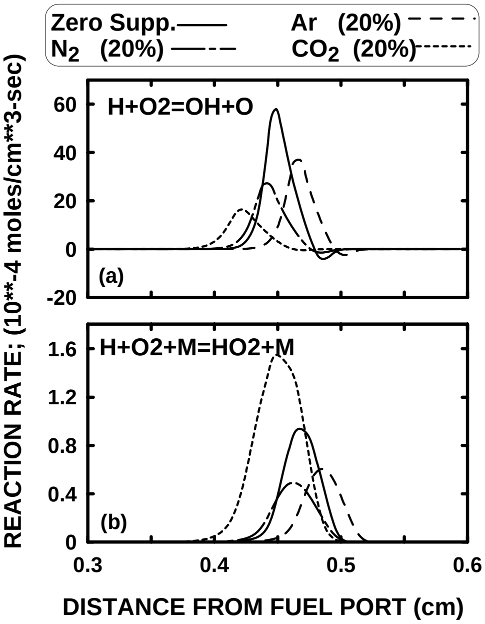

Fig. 6. **Effect of suppressant type on reaction rates of H+O2→OH+O and H+O2+M→HO2+M for C2H4-air diffusion flames, showing results at incipient extinction strain for each case.**

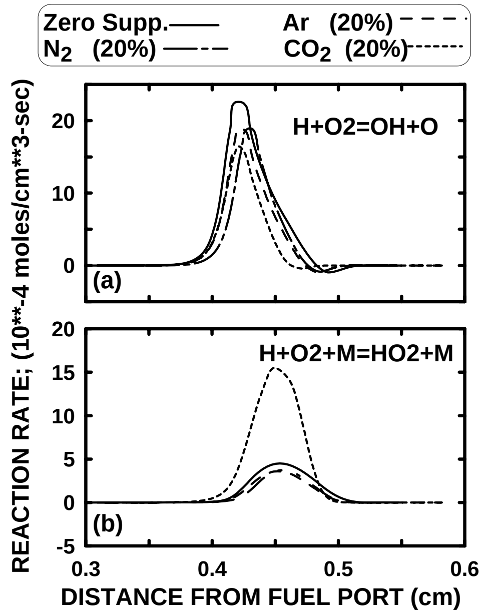

Fig. 7. **Similar to Fig. 6, but comparing effects of suppressant type on reaction rates at the same strain rate.**