Asymptotic and Numerical Techniques for Evaluating the Chemical and Thermal Influences of Fire-Suppressants on Nonpremixed Flames

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Abstract

Analytical and numerical approaches for evaluating the chemical and thermal influence of fire suppressants on laminar nonpremixed flames are described here. The analytical approaches employ asymptotic techniques **for** evaluating the chemical and thermal influences of suppressants on flames. In asymptotic analysis the Damkohler number $\delta \equiv \tau_f / \tau_c$ is used to characterize the structure of the flame. Here τ_f and τ_c are respectively the characteristic flow time and the characteristic chemical reaction time. The results of asymptotic analysis show flame extinction to take place at a critical value of the Damkohler number represented by 6,. Combustion takes place for values of 6 greater than 6.. Chemical influences of any agent increases the value of 6.. On the other hand thermal influences do not change the value of 6,. Using this criteria CF_3Br is found to have significant influence on flame chemistry and N_2 is found to extinguish the flame by thermal mechanisms. The thermal and chemical influences of a number of replacement agents are evaluated using this criteria and the results are compared with those for CF_3Br and N_2.

A numerical and analytical technique is described for evaluating the effectiveness of a given agent in extinguishing nonpremixed flames when it is mixed with air with its effectiveness when it is mixed with fuel. This technique is applied to evaluate the effectiveness of CF_3Br and N_2 in extinguishing methane flames. The results show CF_3Br to be more effective in extinguishing the kame when it is mixed with air in comparison with its effectiveness when it is mixed with fuel.

1 Introduction

Numerous agents are currently being tested to find out if they can replace Halon 1301(CF_3Br) for firefighting. These replacement agents extinguish fires by chemical and thermal mechanisms. It is of practical interest to evaluate the degree of chemical and thermal influences of these agents on fires. Here previously used experimental, numerical and analytical approaches for evaluating the levels of chemical and thermal influences of any given fire-suppressant on flames are described [1–3].

The experimental studies involve measurements on nonpremixed flames stabilized in the counterflowing configuration. This configuration provides a convenient geometry for detailed, fundamental studies of the structure and mechanisms of extinction of nonpremixed flames. In the counterflowing configuration the reactant streams (fuel stream and the oxidizer stream) flow toward each other, and a stagnation plane is formed where the value of the normal component of the flow velocity **is** zero. For hydrocarbon fuels, the stoichiometry of the overall combustion process is **such** that the flame is stabilized on the oxidizer side of the stagnation plane. In the counterflowing configuration, the flame front (defined as the position where the temperature attains a maximum value) is approximately parallel to the stagnation plane, and the structure of the flame in the vicinity of the stagnation streamline is nearly one-dimensional. Also in the counterflowing configuration, the characteristic residence time for the reactants in the flame can be easily related to the flow velocity of the reactants. This simplifies theoretical interpretation of the experimental results. Fire suppressants that extinguish flames by predominantly chemical mechanisms have different effectiveness when added to the fuel stream or to the oxidizer stream of the nonpremixed flame. Here previously used experimental, numerical and analytical approaches for comparing the effectiveness of chemical fire suppressants when it is added to the oxidizer stream with its effectiveness when added to the fuel stream is also discussed [4,5].

2 Thermal and Chemical Influences of Flame Suppressants

Experimental data showing extinction characteristics of various suppressants can be used to distinguish between their thermal and chemical influences on the flame. Recent studies [2,3] have examined the chemical and thermal influences of various halogenated agents on the structure of nonpremixed flames. These agents were tested to find out if they are suitable substitutes for for Halon 1301 (CF_3Br). The agents considered were $CF_3CH_2CF_3$ (HFC-236fa), $CHFClCF_3$ (HCFC-124), CHF_2CF_3 (HFC-125), C_3HF_7 (HFC-227ea), C_3F_8 (FC-218), CHF_2Cl (HCFC-22), CH_2FCF_3 (HFC-134a), C_4F_{10} (FC-31-10), CH_2F_2/CHF_2CF_3 (60%/40% by volume) (HFC-32/HFC-125), cyclo-C_4F_8 (FC-318) and C_2F_6 (FC-116). The counterflowing configuration was employed and the fuels used were heptane and JP-8 [2,3]. The effectiveness of these agents in extinguishing nonpremixed flames burning these fuels were compared with those of N_2 and CF_3Br. the oxidizing gas was a mixture of air and the agent. Experimental results were interpreted using one-step, activation-energy asymptotic theories [6–9]. Although this is a poor approximation for the chemistry taking place in the reaction zone, it provides a rough indication of the thermal and chemical influence of these agents on the flame structure.

2.1 Experimental Results

The counterflow burner employed in the present study is described in detail elsewhere [3]. It consists of a fuel cup which has a diameter of 45 mm and a depth of 18 mm, and an oxidizer duct with an inner diameter of 50.1 mm through which the oxidizing stream containing the gaseous agent to be tested is introduced into the flame. The oxidizer duct has approximately five layers of fine wire screens to reduce turbulence and ensure a flat velocity profile at the exit of the duct. Experiments were performed with $L = 1$ cm, where L is the separation distance between the surface of the liquid pool and the exit of the oxidizer duct. The flowrates of air, nitrogen and the gaseous agent were measured accurately by use of variable area flowmeters. The volumetric flow rates of the gases measured using these flowmeters are rated at an accuracy of 3%. Experiments were performed with the oxidizer stream at an initial temperature of 298K. To obtain the critical conditions of extinction the flow rate of air in the oxidizer duct was maintained at some predetermined value and the liquid pool was ignited. The agent was then added gradually to the oxidizer stream until the flame extinguished. The flowrates of air and the agent at extinction were recorded. The experiments were repeated for a different value of the flowrate of air. The velocity of the oxidizer stream at the exit of the oxidizer duct U was presumed to be equal to the ratio of the total flow rate to the cross sectional area of the duct. The characteristic strain rate at extinction was calculated using the expression $a = 2U/L$ [10].

Figure 1 shows the mass fraction of the agents at extinction as a function of the strain rate.

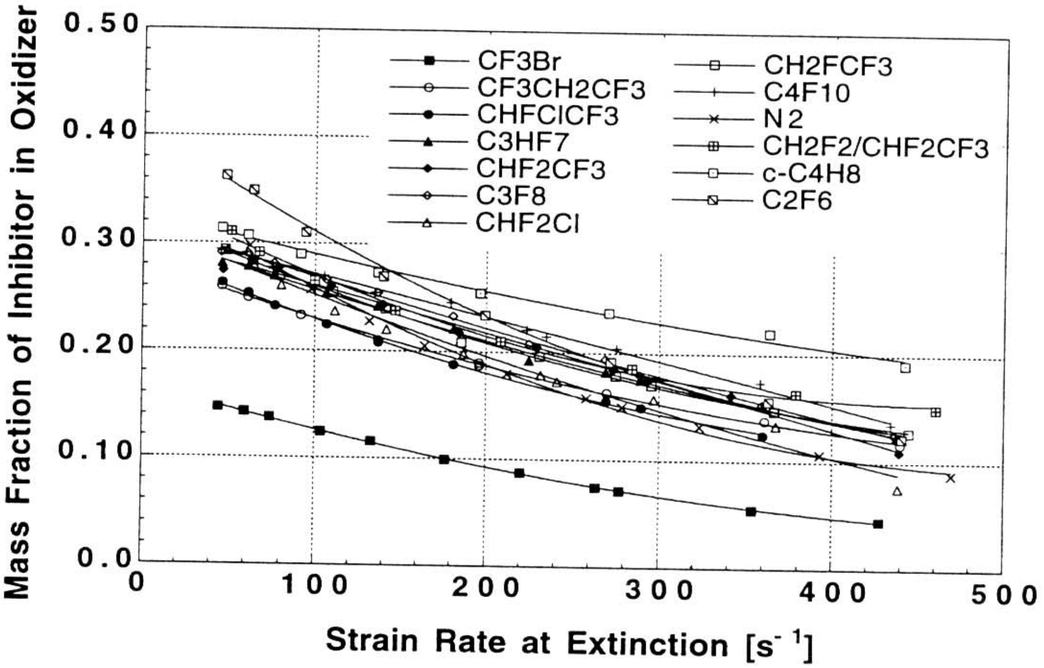

Figure 1: Mass fraction of various agents as a function of the strain rate (s^{-1}) at extinction. The fuel tested was heptane and the initial temperature of the oxidizer stream was 25°C [2].

For a given agent, the region above the curve represents flammable mixtures. At values of a below those shown in Fig. 1, it was not possible to stabilize a steady flame in the counterflow burner. Figure 1 shows that for any given value of a, the value of the mass fraction of CF_3Br required to extinguish the flame is lower than the mass fraction of all other agents. Therefore on a mass basis CF_3Br is considerably more effective in extinguishing the flame than all the agents tested. Figure 1 shows that the relative effectiveness of various agents in extinguishing the flame changes for different values of a. At low strain rates $CF_3CH_2CF_3$ is most effective and C_2F_6 least effective in extinguishing the flame, whereas at high strain rates CHF_2Cl is most effective and cyclo-C_4F_8 least effective in extinguishing the flame. In fact at high strain rates only CF_3Br is more effective than N_2 in extinguishing the flame and the effectiveness of CHF_2Cl is nearly the same as that of N_2. Figure 1 shows that, except for CF_3Br, the mass-based effectiveness of the various agents in extinguishing the flame does not differ much from that of nitrogen. If the results in Fig. 1 are plotted showing the mole fractions of the agents at extinction as a function of a, then the relative effectiveness of the agents ranked on a mole basis is found to be different from their mass-based effectiveness. On a mole basis CF_3Br is found to be most effective and N_2 least effective [3].

2.2 Interpretation of the Experimental Results Using Activation-Energy Asymptotic Theories

Activation-energy asymptotic theories of flame extinction [6-9], where the gas-phase chemical reaction is approximated as a one-step process have predicted the value of the Damkohler number at extinction δ_e, which is defined as the ratio of τ_f to τ_c, where τ_f is the characteristic flow time and τ_c the characteristic chemical reaction time. For a given chemical system, the value of the τ_c will depend on the relative concentrations of the various reactants including that of the agent and the local gas temperature, and the value of τ_f will depend on the strain rate a. For a given fuel-oxidizer system, the effectiveness of an agent in interfering in the chemical reaction between the fuel and oxygen can be roughly estimated by examining the value of 6,. For a given value of τ_f, the value of τ_c at extinction can be expected to decrease and the corresponding value of 6, to increase with increasing chemical inhibition of the agent. In absence of temperature measurements in the flame at conditions close to flame extinction, the calculated adiabatic flame temperature T_f, can be expected to provide a rough measure of the characteristic chemical time. The value of τ_c decreases with increasing values of T_f.

Measurements of critical conditions of heptane flames with some selected agents shown in Fig. 1 were used to calculate the adiabatic flame temperature and the results are plotted in Fig. 2. In the calculations, the agents were presumed to be inert, and the gas-phase chemical reaction was presumed to occur as a one-step overall process with stoichiometric amounts of fuel reacting with oxygen to form the products CO_2 and H_2O. Figure 2 shows that for any given value of the strain rate a, the value of T_f calculated for flames inhibited with CF_3Br is higher than the value of T_f calculated for flames inhibited with the other agents. Therefore for any value of τ_f, the critical conditions of extinction measured with CF_3Br yield values of τ_c which is smaller, than those for the other agents. Hence the values of δ_e for extinction of flames with CF3Br is higher than those for extinction of flames with the other agents. Therefore CF_3Br appears to interfere more with the flame chemistry than the other agents and cannot be presumed to be inert. Also, except CF_3Br, the values of T_f calculated using the measured mass fractions of the other agents at extinction do not differ much from those calculated using the measured mass fraction of N_2 at extinction. Since the influence of N_2 on the flame is primarily thermal, it appears that with the exception of CF_3Br, all the agents have a significant thermal influence on the flame.

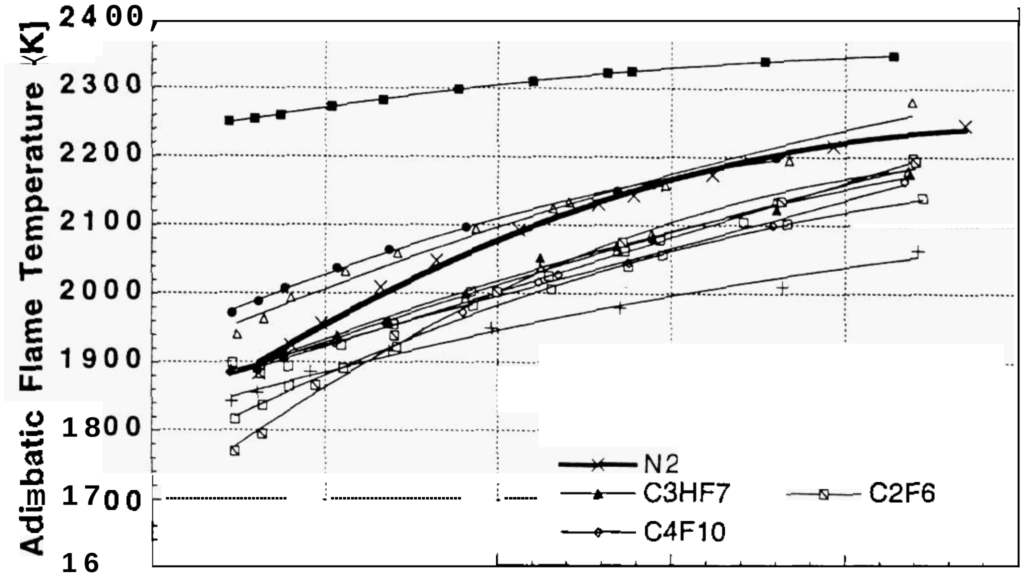

Figure 2: Calculated adiabatic flame temperature as a function of the strain rate (s^{-1}) at extinction [2] .

The calculated values of T_f are used to interpret the extinction measurements using predictions of activation-energy asymptotic analysis [6-9]. The agents, including CF_3Br, presumed to be chemically inert. The rate of gas-phase chemical reaction between the fuel and oxygen is represented by the Arrhenius expression $k = B\exp\left[-E_0/(RT)\right]$ where k is the rate constant of the reaction, B the frequency factor, E_0 the activation energy, R the gas constant and T the gas temperature. The activation energy of the chemical reaction is presumed to be large in comparison to the thermal energy in the flame. Results of activation-energy asymptotic analysis predict that at flame extinction [8]

$$Fa/T_f^2 = \rho_\infty T_\infty B_0 R^3 \exp\left[-E_0/(RT_f)\right] / \left(2W_F E_0^3\right) \tag{1}$$

where ρ_∞ and T_∞ represent respectively the density and temperature of the oxidizer stream at the exit of the gas duct, W_F is the molecular weight of the fuel, and B_0 represents the frequency factor for molar rate of consumption of oxygen. The value of the quantity F depends on the flow field, and Krishnamurthy et al [8] have outlined methods for estimating F for the counterflow geometry. It follows that a plot of $\ln\left(Fa/T_f^2\right)$ versus $1/T_f$ represents an Arrhenius type plot, and the slope of this plot can be used to deduce the activation energy of the chemical reaction between the fuel and oxygen.

Figure 3 shows Arrhenius diagrams plotted using the measurements of critical conditions of extinction of heptane flames with some selected agents shown in Fig. 1. The value of E_0

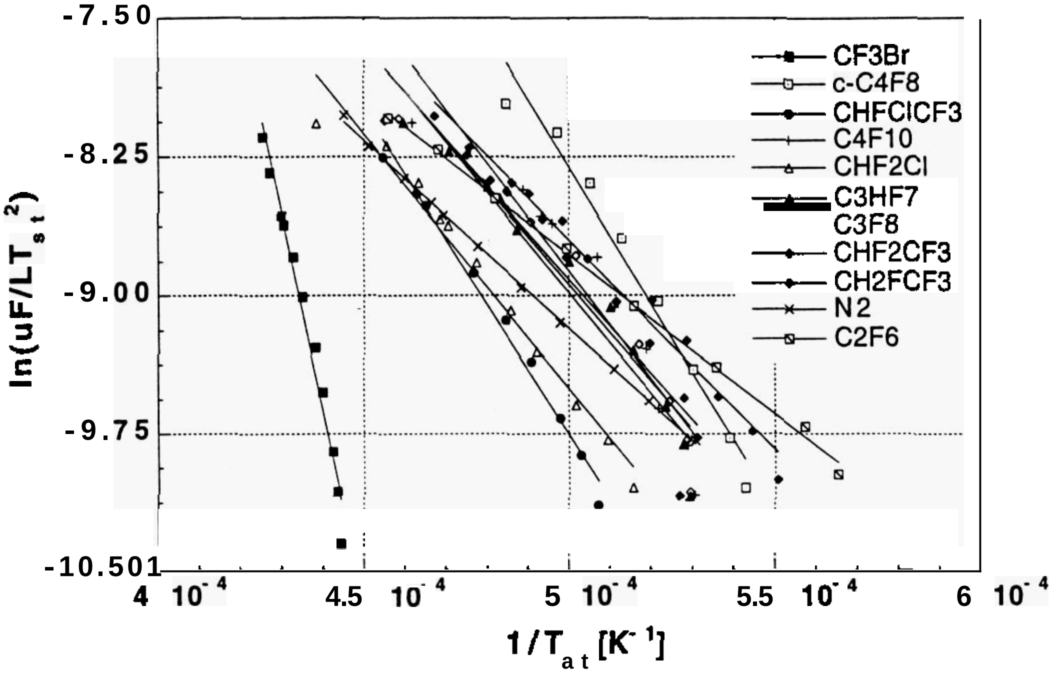

Figure 3: Arrhenius plots obtained using the extinction data shown in Fig. 1 [2] .

deduced from the slopes of the lines in Fig. 3 is shown in Table 1.

The overall activation energy for one step reaction between heptane vapors and oxygen is 168 kJ/mole. Since the influence of N_2 on the flame is primarily thermal deviations of the value of E_0 calculated using the extinction data obtained with other agents form that of N_2 is a rough measure of the chemical influence of these agents. Since the value of E_0 calculated using extinction data with CF_3Br is unrealistically high, clearly the one-step approximation is not valid, and CF_3Br dearly appears to have a significant influence on the flame chemistry. However, in comparison to CF_3Br, the other agents appear to have considerably less influence on the flame chemistry.

3 Comparison of the Effectiveness of CF_3Br When Added to the Oxidizer Stream With Its Effectiveness When Added to the Fuel Stream

A detailed experimental, numerical and analytical study was conducted previously to clarify the influence of CF_3Br on the structure and critical conditions of extinction of nonpremixed methane flames [4,5,11,12]. In some of these previous studies the effectiveness of CF_3Br in extinguishing methane flames when it is added to the fuel stream was compared with its effectiveness when it is added to the oxidizer stream [4,5]. The counterflowing configuration was used. The burner used in the experiments is described elsewhere [5]. It comprised two opposing ducts with inner diameters of 22.2 mm through which the gaseous reactants were introduced. The distance L between the exits of the ducts was 10 mm. Seshadri and Williams [10] have shown that for

Agent	Chemical Formula	E_0 in kJ/mole
Halon 1301	CF_3Br	899
FC-318	cyclo-C_4F_8	311
HCFC-124	$CHFClCF_3$	294
FC-31-10	C_4F_{10}	244
HCFC-22	CHF_2Cl	231
HFC-227ea	C_3HF_7	237
FC-218	C_3F_8	218
HFC-125	CHF_2CF_3	214
HFC-134a	CH_2FCF_3	189
Nitrogen	N_2	168
FC-116	C_2F_6	143

laminar flow in the counterflowing configuration, if the Reynolds number is large enough, and if the values of the tangential component of the flow velocities at the exits of ducts are zero, then the flow field comprises two inviscid rotational regions on either side of a thin viscous region formed in the vicinity of the stagnation plane. The strain rate a, defined as the normal gradient of the normal component of the flow velocity in the inviscid region on the oxidizer side of the stagnation plane, is given by the expression [10]

$$a = \frac{2|V_2|}{L}\left(1 + \frac{|V_1|\sqrt{\rho_1}}{|V_2|\sqrt{\rho_2}}\right), \tag{2}$$

where V is the velocity, and ρ the density. Subscripts 1 and 2 respectively denote conditions in the fuel stream and in the oxidizer stream at the exit plane of the ducts. The value of the strain rate calculated from **Eq.** 2 was taken as a measure of the inverse of the characteristic flow time of the reactants in the flame. The value of the strain rate at extinction is denoted by a_q.

Numerical calculations were performed to determine the critical conditions of extinction and the results were compared with measurements [4,5]. In the numerical calculations the oxidation of methane was described using reactions **1–35,** 37–40 shown in Table 1 of Ref. [13]. This mechanism excludes reactions in which compounds comprising two or more carbon atoms participate. Westbrook [14] has proposed a detailed chemical-kinetic mechanism for describing the influence of CF_3Br on the structure of premixed flames of hydrogen, methane, methanol, and ethene. The inhibition chemistry was described by reactions B1–B10, B15, B17, B18, B21–B36, B40–B46, B48–B56 taken from Table II of Ref. [14]. For a given value of the strain rate two converged solutions can be obtained. The solution with the higher value of the maximum flame temperature is stable and the other is unstable. The calculations thus yield the classical "C-shaped" curve when the maximum value of the flame temperature is plotted as a function of the strain rate. For values of the strain rate greater than a critical value a, no solution is obtained. **This** critical value a, is the strain rate at extinction.

In Fig. 4 the strain rate a_q **is** plotted **as** a function of the mass fraction of the inhibitor, $Y_{I,2}$, in the oxidizer stream, where the subscript Z denotes CF_3Br. The points in Fig. **4** represent measurements and the lines are results of numerical Calculation with detailed chemistry. In this figure the region below any curve represent flammable mixtures. The fuel stream and the oxidizer stream were diluted with nitrogen. The measurements were made with the value of the ratio $Y^0_{O_2,2}/Y^0_{F,1}$ fixed at **0.233** and for values of $Y^0_{O_2,2}$ equal to **0.233, 0.227, 0.220,** and **0.209.** Here $Y^0_{O_2,2}$ is the mass fraction of oxygen at the oxidizer boundary prior to addition of the inhibitor, and $Y^0_{F,1}$ is the **mass** fraction of fuel at the fuel boundary prior to addition of the inhibitor. The numerical calculations were performed with the composition of the reactants streams identical to those used in the experiments and for various values of the strain rates [5]. Figure **4** shows the results of numerical calculations to agree very well with the measurements. In general at a given value of $Y_{I,2}$ the calculated value of a, is larger than its measured value. However for high inhibitor concentrations (low strain rates) at a given value of $Y_{I,2}$ the calculated value of a, is very close to its measured value.

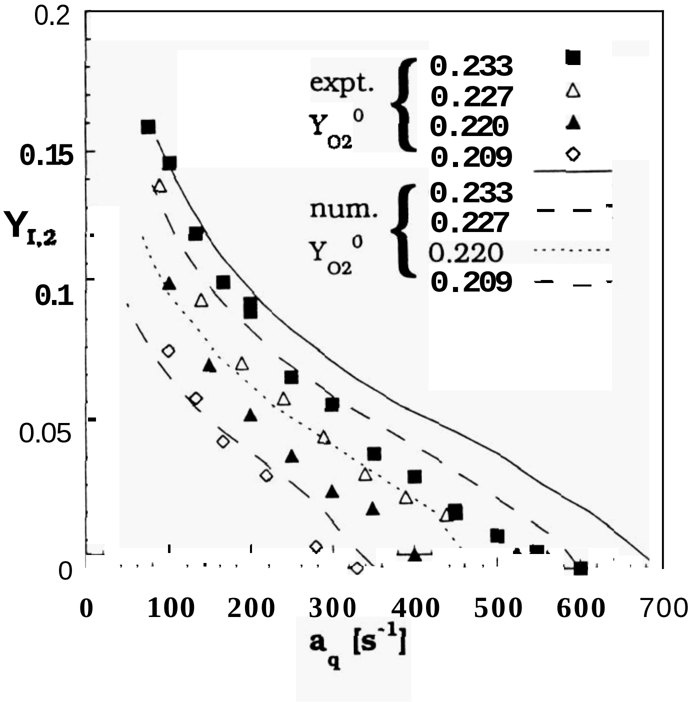

Figure **4:** Comparison of experimental measurements (points) of critical conditions of extinction with results of numerical calculation (lines) with the inhibitor added to the oxidizer stream. Measurements are made for values of $Y^0_{O_2,2}/Y^0_{F,1}$ equal to **0.233.** The values of $Y^0_{O_2,2}$ prior to addition of the inhibitor is shown **[5] .**

Figure **5** shows a_q as a function of the **mass** fraction of CF_3Br in the fuel stream, $Y_{I,1}$ [5]. The points in Fig. **5** represent measurements and the lines are results of numerical calculation with detailed chemistry. The measurements are made with the value of $Y^0_{O_2,2}/Y^0_{F,1}$ fixed at

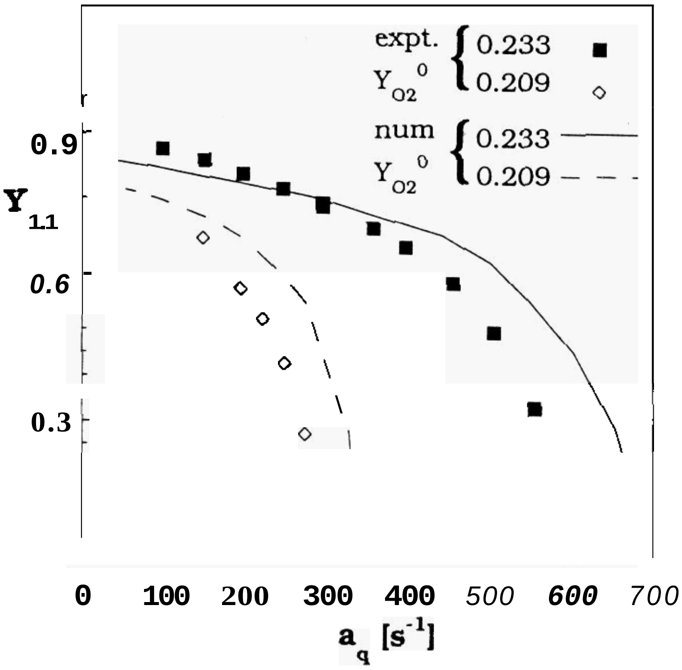

Figure 5: Comparison of experimental measurements (points) of critical conditions of extinction with results of numerical calculation (lines) with the inhibitor added to the fuel stream. Measurements are made for values of $Y^0_{O_2,2}/Y^0_{F,1}$ fixed at 0.233. The values of $Y^0_{O_2,2}$ prior to addition of the inhibitor is shown [5] .

0.233 and for values of $Y^0_{O_2,2}$ equal to 0.233 and 0.209. Figure 5 shows the results of numerical calculations to agree very well with the measurements.

A comparison of the results in Figures 4 and 5 show that for a given value of *a*, the measured and calculated values of $Y_{I,1}$ are significantly larger than those of $Y_{I,2}$. To compare the effectiveness of CF_3Br in extinguishing the flame when it is added to the oxidizer stream with its effectiveness when added to the fuel stream, the flux of the inhibitor entering the reaction zone must be calculated. At the flamesheet which is presumed to be located at ξ_{st}, by definition, the mass fractions of fuel and oxygen calculated assuming frozen flow, must be in stoichiometric proportions [9,15]. Here ξ is a conserved scalar quantity and its value is 1 at the fuel boundary and 0 at the oxidizer boundary [9,15]. In a previous asymptotic study of chemical inhibition of nonpremixed methane flames by CF_3Br [11], it was presumed that in the flamesheet approximation [9], CH_4 and CF_3Br are consumed in two different thin zones. The global chemistry at these thin zones were represented by [11] $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$, and $CF_3Br + 2H_2O \rightarrow HBr + 3HF + CO_2$. It is noteworthy that oxygen does not appear in the global step that consumes CF_3Br. To the leading order it was shown that the position of the primary reaction zone ξ_{st} where fuel and oxygen are completely consumed is given by

$$\xi_{st} = [1 + 2Y_{F,1}W_{O_2}/(Y_{O_2,2}W_F)]^{-1}, \tag{3}$$

where Y_i and W_i represent the mass fraction and molecular weight of species i and the subscripts F and O_2 denote the fuel and oxygen respectively. The mass fraction of the inhibitor does not appear in the expression for ξ_{st} because neither the fuel nor oxygen appears in the global step that consumes CF_3Br.

Following a previous study [16] the mass fraction of the inhibitor at the flamesheet, $Y_{I,st}$ can be calculated from the expression $Y_{I,st} = Y_{I,2} + (Y_{I,1}\text{-}Y_{I,2})\xi_{st}$, assuming frozen flow and neglecting preferential diffusion, can be used to measure the effectiveness of the inhibitor. The values of $Y_{I,st}$ calculated using the experimental results for $Y^0_{O_2,2} = 0.233$ in Figs. 4 and 5 are shown in Fig. 6. For a given value of strain rate the value of $Y_{I,st}$ when the inhibitor is added to the oxidizer stream is significantly lower than the value of $Y_{I,st}$ when the inhibitor is added to the fuel stream. Therefore the experimental results show CF_3Br to be more effective in extingnishing the flame when it is added to the oxidizer stream. In Fig. 6 the values of $Y_{I,st}$ calculated from results of previous experimental studies on laminar [17,18] and turbulent [16] flames stabilized in the coflowing configuration are also shown. In the coflow configuration the strain rate cannot be easily calculated from the velocities of the reactants, therefore the experimental results obtained from these previous studies are represented by straight lines in Fig. 6. The experiments of Creitz [17] and Kubin et al [18] were conducted on methane flames and the results show CF_3Br to be more effective in extinguishing the flames when it is added to the oxidizer stream. The experiments of Masri [16] were conducted on natural gas flames containing 90% CH_4 by volume and show that the effectiveness of CF_3Br when it is added to

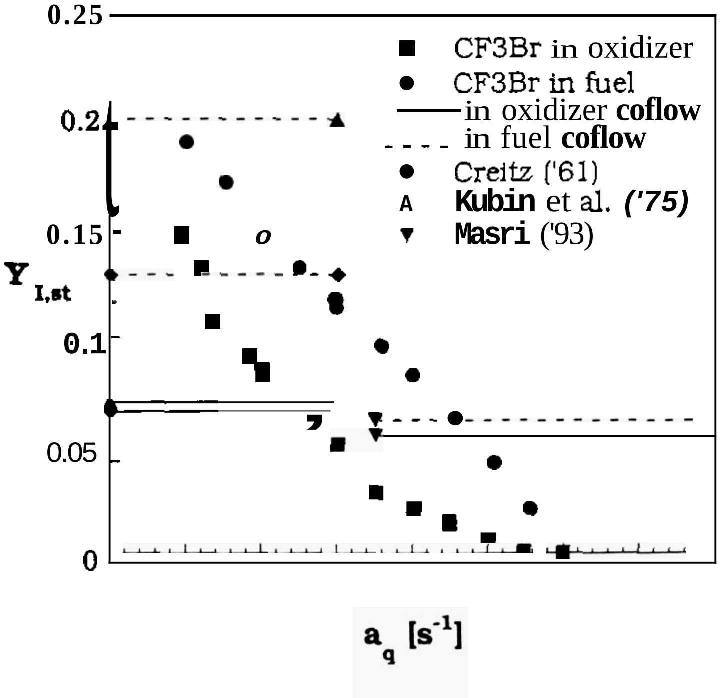

Figure 6: The strain rate at extinction a, as a function of $Y_{I,st}$ calculated using the extinction results for $Y^0_{O_2,2} = 0.233$.

the oxidizer stream is nearly equal to its effectiveness when added to the fuel stream. The qualitative differences between the results obtained by Masri [16] and the present experimental results and those of Crietz [17] and Kubin et al [18] are attributed to differences in the flow field (turbulent flow versus laminar flow) and to the type of fuel (natural gas versus methane). These results clearly show CF_3Br to be more effective in extinguishing methane flames when it is added to the oxidizer stream

4 Summary

Experimental and analytical studies on mechanisms of extinction of heptane flames with a number of agents show that

1. On **a** mass and mole basis CF_3Br is considerable more effective in extinguishing the flames than all the substitute agents considered. With the exception of CF_3Br, on a mass basis the effectiveness of **all** the other agents in extinguishing nonpremixed flames was nearly the same as that of nitrogen.
2. Interpretation of the results based on one-step activation energy asymptotic theories shows that CF_3Br has a significant influence on the flame chemistry. The other agents tested appear to have a significant thermal and some chemical influence on the Aame.

Experimental and numerical studies on mechanisms of extinction of methane flames with CF_3Br show that

1. The chemical kinetic mechanisms proposed by Westbrook [14] is found to provide a realistic description of inhibition of nonpremixed methane flames by CF_3Br.
2. CF_3Br is more effective in extinguishing nonpremixed methane flames when it is added to the oxidizer stream in comparison to its effectiveness when added to the fuel stream. This result is in agreement with previous studies [17, 18].

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