

THE EXTINCTION OF INHIBITED, STRETCHED AND
DILUTED COUNTERFLOWING FLAMES

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1. FLAME SUPPRESSION

It has been shown that a criteria for flame extinction can be described by the Damkohler Number which is the ratio of a characteristic flow or diffusion time to a characteristic chemical reaction time [Linan, 1974]. As the Damkohler Number is reduced the fuel burning rate gradually decreases until a critical value is attained when the flame will abruptly extinguish.

The Damkohler Number criteria suggests a number of strategies for extinguishing fires including: (1) cooling the reactants or the combustion zone, (2) reactant isolation or removal, (3) blow-out, or (4) chemical inhibition [Williams, 1974]. The addition of an inert agent, such as N_2 or Ar, to the flow field serves to isolate the reactants from each other as well as to decrease the flame temperature. Extinction by flame stretch occurs when the strain rate in the flow field is increased, decreasing the Damkohler Number, until flame blow-out transpires. The addition of halogenated species inhibits gas-phase chemical reactions during hydrocarbon combustion. The halogenated compounds act as radical scavengers by influencing the O-H chemistry and inhibiting key chemical reactions. Although several studies have investigated the effect of halogenated agents on flames, they have not focussed on the detailed changes in flame structure which arise upon addition of these agents to the oxidizer side of a non-premixed flame.

Counterflow flames result when reactant streams flow towards each other in a stagnation point flow. The flame structure is steady and one-dimensional in temperature and species concentration making it amenable to mathematical analysis and facilitating computational simulation. The flow is characterized by either a local or a global stretch rate which is related to the inverse of a local flow time and a Damkohler number. Extinction takes place if this number reaches a critical minimum value [Linan, 1974].

The objective of this study is to investigate differences in the detailed chemical mechanisms which control the kinetics of non-premixed flames as they approach extinction by stretch, inhibition, and dilution.

2. METHODOLOGY

The various test cases for which the numerical simulations were conducted are listed in the Table. The species N_2 , H_2O (vapor), CH_3Cl , HCl , Cl_2 and CH_4 termed "agents" were introduced into the oxidizer stream of a CH_4 -Air non-premixed flame that is referred to as the "base" flame. The flame structure was numerically simulated for stretch rates of 98 s^{-1}

Combustion Institute/Central and Eastern States Section.
Combustion Fundamentals and Applications. Joint
Technical Meeting. March 15-17, 1993, New Orleans, LA,
56-60 pp, 1993.

in all cases, except when computations simulated blow out of the base flame. Extinction conditions were measured and directly compared to the computational results for N_2 and CH_3Cl . Temperature profiles were measured for the base flame and for the N_2 and CH_3Cl cases.

2.1 Numerical Method

Numerical simulations of laminar counterflow flames were performed using a previously developed computer code [Rogg, 1991]. The code calculates flame structure using detailed models of molecular transport and chemical kinetics. The kinetic scheme used here is amalgamated from Miller and Bowman [1989] for methane (CH_4) and from Barat et al. [1990] for methyl chloride (CH_3Cl) combustion. The scheme includes 179 elementary forward reactions and involves 38 gas-phase species including C_2 species. The backward rates are calculated from thermodynamic equilibrium [Rogg, 1991]. A complete description of the computation is described elsewhere [Lee, 1992]. The kinetic scheme has been employed to successfully predict the flame speed of unstretched premixed flames burning CH_3Cl -air mixtures [Lee, 1992].

2.2 Experimental Method

The experimental set-up has been previously described [Lee, 1992; Yang, 1992]. Extinction experiments were performed with N_2 and CH_3Cl agents. Temperature profiles were measured using Pt/Pt-10% Rh thermocouples (0.076 mm diameter) coated with Yttrium Oxide to avoid catalytic effects.

3. RESULTS AND DISCUSSION

3.1 Extinction

The numerical computation converged for CH_3Cl additions to the oxidizer stream of 8 mole percent, but did not converge for 9% CH_3Cl additions. Non-convergence is interpreted as flame extinction. Thus, the table notes that flame extinction was simulated for approximately 9% CH_3Cl and approximately 22% N_2 . Experimentally measured extinction concentrations were in close agreement, approximately 9% for CH_3Cl and 20% for N_2 . The experimental result supports the validity of the kinetic scheme and the numerical simulations in general. Simulations of H_2O addition gives flame extinction at approximately 19 mole percent.

In order to distinguish between the thermal and kinetic effects due to CH_3Cl , extinction calculations were repeated with all chemical reactions involving CH_3Cl removed, effectively treating it as an inert. These calculations predict that 16% "inert" CH_3Cl is required to extinguish the flame. This suggests that chemical effects increase CH_3Cl inhibition effectiveness by roughly 50%. Suppressants such as halogenated molecules exert both chemical kinetic and physical effects.

3.2 Effect on Temperature

Figure 1 shows the computed temperature as a function of location (Y) in the reaction zone for 0, 5, 7, 8 and 9% CH_3Cl (by volume) added to the oxidizer side of the base flame. The oxidizer and fuel ducts are separated by 15 mm and the position $Y=0$ is the location of the stagnation point (the oxidizer side is for $Y>0$). CH_3Cl addition broadens the temperature profile, increasing the maximum temperature and shifting the spatial location of the peak towards the oxidizer side of the flame. The increased flame temperature is attributed to the increased enthalpy associated with CH_3Cl addition. The simulated temperature profiles (not presented) for N_2 addition shows that the peak flame temperature decreases, whereas the spatial location of the maximum is unaltered. The

effect of increasing stretch in the base flame is to decrease the peak flame temperature, until a critical stretch rate ($\approx 290 \text{ s}^{-1}$) is achieved which leads to extinction. The peak temperature just before extinction in the stretched case ($\approx 1740 \text{ K}$) is approximately equal to the peak temperature just before extinction in the nitrogen addition case (≈ 1770).

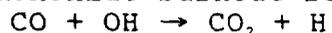
Computations with H_2O addition to the oxidizer stream impacts the temperature profile in a manner similar to N_2 addition. CH_4 addition to the oxidizer yields broadened temperature profiles and increases the peak temperature. Cl_2 addition is similar to CH_3Cl addition, the peak temperature increases and the location of the temperature maxima shifts towards the oxidizer duct. This is in contrast to HCl which has little influence on the temperature profile. This is not surprising as HCl is a fairly stable molecule.

3.3 Calculated Effect on Radical Species

The effect of N_2 addition to the oxidizer stream is to decrease the OH , H and O radical concentrations in the flame, while not shifting the location of the maxima. This is attributable to the decreased flame temperature with N_2 addition. CH_4 addition accomplishes the opposite result, yielding increased flame temperatures and radical concentrations due to the enthalpy addition to the flame. In contrast, Fig. 2 shows that the OH radical concentration decreases with increasing amounts of CH_3Cl addition, even though the temperature profiles in the CH_3Cl addition flames are higher than the base flame. For 9% CH_3Cl addition (close to extinction) the peak OH mole fraction is much larger than in the 20% N_2 addition case (also near extinction). Analogous results are calculated for O atoms in the CH_3Cl addition cases. The H atom profiles for CH_3Cl addition are presented in Fig. 3. The addition of 5% CH_3Cl to the oxidizer side significantly decreases the H atom concentration, but subsequent addition of this agent (7, 8, and 9%) has negligible effect on the maximum H atom concentration. When the flame is extinguished by flame stretch, the radical concentration monotonically decreases as the strain rate increases.

3.4 Effect on Chemistry

Thermal, kinetic, and stretch effects impact flame chemistry by suppressing the base flame. Decreases in radical concentrations induced thermally, kinetically, or by stretch, lead to decreases in the rates of important bimolecular reactions involving radicals. For instance, the rate of the carbon monoxide burnout reaction:



which is a major source of heat release in the flame, is shown in Fig. 4 for CH_3Cl additions to the oxidizer stream. As CH_3Cl is added, decreases in the OH radical concentration leads to decreases in the rate of this reaction.

The addition of CH_4 to the oxidizer stream produces a partially premixed flame. This type of configuration has been shown to be more resistant to extinction than the base flame [Hamins et al., 1985]. Although both CH_4 and CH_3Cl add enthalpy to the flame leading to increased flame temperatures, the Cl atom acts as a radical scavenger, significantly disrupting the radical pool and destabilizing the flame.

From the computations it is clear that N_2 and H_2O act physically. The chlorinated compounds act both physically and chemically, altering the flame structure by influencing the concentration distribution of radical species in the flame through radical scavenging.

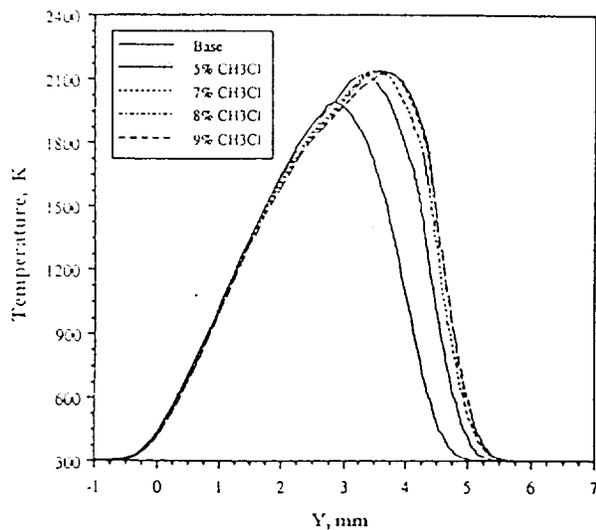
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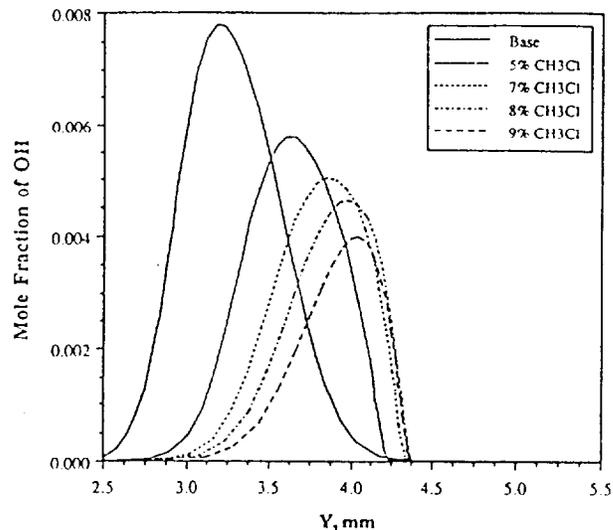
TABLE. Mole percent of species added to the oxidizer side of the flame for stretch rates equal to $\approx 98^{-1}$ in all cases.

| Species | Mole Percent Added to Oxidizer Stream |
|------------------------------|---------------------------------------|
| N_2 | 5, 10, 15, 20, 22* |
| H_2O | 5, 10, 15, 19* |
| HCl | 5 |
| Cl_2 | 2.5, 5 |
| CH_3Cl | 5, 7, 8, 9* |
| inert CH_3Cl | 10, 15, 16* |
| CH_4 | 5 |

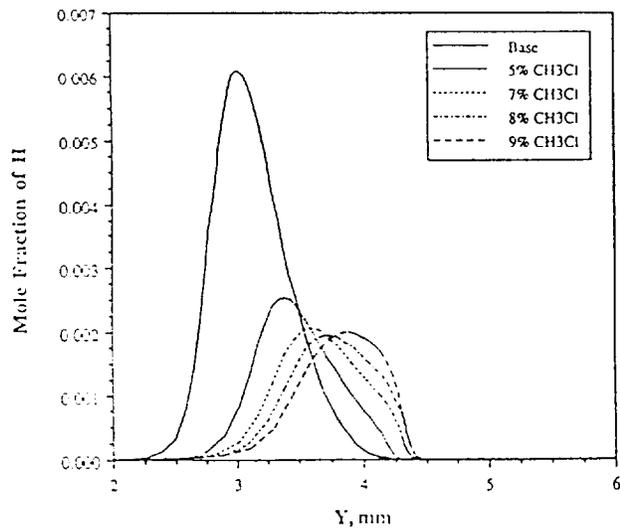
* Conditions calculated to be near extinction.



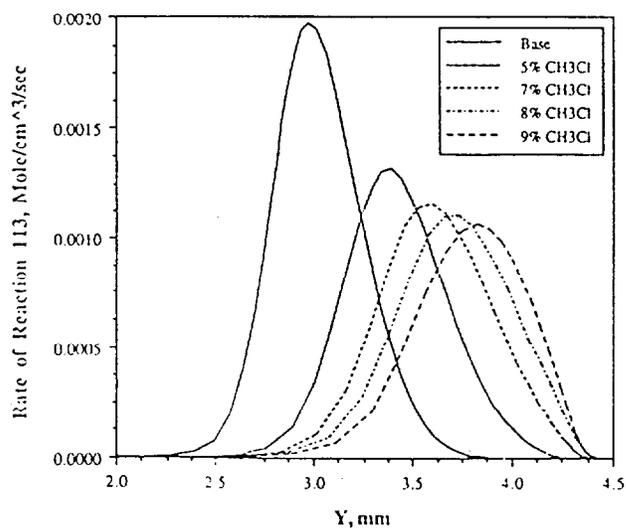
The computed temperature profiles with various amounts of CH₃Cl added to the oxidizer stream of the base flame.



The computed OH radical concentration profiles with various amounts of CH₃Cl added to the oxidizer stream of the base flame.



The computed H atom concentration profiles with various amounts of CH₃Cl added to the oxidizer stream of the base flame.



The rate of the CO burnout reaction with various amounts of CH₃Cl added to the oxidizer stream of the base flame.