# ON THE SUITABILITY OF $CF_3BR$ AS A BENCHMARK FOR REPLACEMENT FIRE SUPPRESSANTS

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## ABSTRACT

We investigate the rationale for the use of $CF_3Br$ as a reference standard for alternative fire suppresants. Stating efficiencies of alternative agents on a relative basis to that of $CF_3Br$ is well founded from an engineering standpoint, but less so from a mechanistic standpoint. The behavior of $CF_3Br$ is, in several significant respects, atypical of fire suppressants, which act by catalytic scavenging of flame radicals. In particular, saturation effects, dependence of inhibition efficiency on flame temperature, and changes in flame structure are significantly different for $CF_3Br$ than for other catalytic substances including iron and alkali metals. For this reason, the use of $CF_3Br$ as an archetype for catalytic suppressants must be qualified to account for these differences.

## INTRODUCTION

The SERPD Next-Generation Fire Suppression Technology Program Element 2A, "Mechanisms of Ultra-High Efficiency Chemical Suppressants" seeks to understand how key chemical reactions or processes of combustion are affected by "superagent" suppressants and how these effects differ from those of Halon 1301 ($CF_3Br$). In this context, a "superagent" is defined as a substance whose fire suppression effectiveness is equal to or greater than $CF_3Br$.

$CF_3Br$ is often used as a benchmark for alternative fire suppressants, based on two primary motivations: (1) since much of the research and development in this area is focused on finding environmentally acceptable alternatives to Halon 1301 to provide equivalent protection against a specified fire threat, the effectiveness of the substitute agent relative to 1301 is an important parameter for system engineering; (2) inhibition properties and chemical kinetics of $CF_3Br$ relevant to fire suppression have been extensively studied for many years. The number of experimental and computational studies conducted on $CF_3Br$ has greatly increased since its manufacture was prohibited in developed countries at the end of 1993.

These considerations raise the question of how applicable the knowledge gained in recent years on $CF_3Br$ will be in identifying alternative suppressants. The questions raised include the following: Is $CF_3Br$ "typical" of efficient fire suppressants in general? In other words, will all promising replacements for $CF_3Br$ have properties that are "similar" in some respect? To what extent is commonality of properties with $CF_3Br$ a useful guide in the search for alternatives? To this end, we have performed modeling studies of premixed flames containing substances representative of different classes of inhibitors, including inert gases, fluorocarbons, and catalytic scavengers of flame radicals. We find that the behavior of $CF_3Br$ differs in several significant respects from that of other catalytic agents.

## TYPES OF SUPPRESSANTS

Suppressants may be grouped into various categories, based on the means by which they suppress combustion. Physical suppressants (such as nitrogen, argon, $CF_4$, and water) do not participate in flame chemistry to a significant extent, and they inhibit combustion by adding heat capacity and diluting the reactants. Chemical suppressants also participate in the flame chemistry and may be subdivided into two groups: catalytic suppressants (including bromine, iodine, and various metallic elements) reduce concentrations of flame radicals through a regenerative cycle in which one molecule of suppressant recombines several radicals. Noncatalytic chemical suppressants (e.g., fluorocarbons) reduce the concentrations of flame radicals by scavenging, but do not have a catalytic cycle and are generally less effective. $CF_3Br$ is primarily a catalytic suppressant [1], due to an H + H recombination cycle involving the bromine atom, although the fluorine atoms add some noncatalytic suppression as well.

Water mist having a drop size of less than 50 µm **is** comparable to or more effective than $CF_3Br$ on a mass basis at both reducing burning velocity of premixed flames and extinction strain rates of nonpremixed flames. This is a consequence of the high heat capacity and enthalpy of vaporization per unit mass of water. With this one exception, **all** other superagents identified are believed to be catalytic scavengers. Thus, the combustion chemistry of superagents is essentially that of catalytic scavenging cycles.

The effectiveness of an inhibitor at reducing burning velocity in a fuel/oxidizer mixture may be placed on a quantitative basis in terms of an "inhibition parameter" $\Phi$ first proposed by Rosser et al. *[2]* and modified by Noto et **al.** [3]:

$$\Phi = \ln(U_0/U_i)\,(X_{O2}/X_i) \qquad (1)$$

where $U_0$ and $U_i$ are the burning velocities of the uninhibited and inhibited flames, respectively, and $X_{O_2}$ and $X_i$ are the reactant mole fractions of oxygen and inhibitor. **In** Noto et **al.** [3], the burning velocity of a given fuel/air mixture was shown to exhibit an exponential dependence on the inhibitor concentration. This exponential dependence, by which the inhibition parameter was defined, was shown to hold for the inert agents $N_2$ and $CF_4$, several hydrofluorocarbons, and $CF_3Br$.

Catalytic scavengers necessarily manifest saturation effects, because concentrations of flame radicals, which typically exist in superequilibrium, cannot be reduced below the thermal equilibrium values at the local flame temperature. The existence of a similarity relationship describing inhibition by $CF_3Br$ as well as noncatalytic agents is thus somewhat unexpected. Saso et **al.** **[4]** modeled the combined effect of mixtures of $CF_3Br$ and inert inhibitors and found synergism, meaning that the two agents in combination had a greater inhibiting effect than the additive effect of the agents in isolation. The synergism was attributed to a temperature effect on the inhibition effectiveness of $CF_3Br$, rather than a saturation phenomenon. Over a range of adiabatic flame temperatures, Saso et al. found $CF_3Br$ to have virtually identical inhibition parameters at concentrations of 0.5% and 1% in methane/oxygen/inert mixtures, indicating the absence of significant saturation effects.

## MODELING RESULTS: BURNING VELOCITIES AND SPECIES PROFILES IN INHIBITED PREMIXED FLAMES

In the calculations described below, the **PREMIX** code [5] was used to compute burning velocities and flame structures of atmospheric pressure flames. An **85** cm domain was used in the calculations. Comparison with thermal equilibrium calculations showed that at the final grid point, temperatures were generally within 5K and radical concentrations within a few percent of their equilibrium values. Flames were stoichiometric, atmospheric pressure methane/oxygen, with various inhibitors added as indicated below.

For the hydrocarbon chemistry, GRIMech 2.1 1 was used **as** the kinetic mechanism. Fluorocarbon chemistry was based on mechanism refinement previously performed in our laboratory [6,7]. Bromine chemistry used for $CF_3Br$ was that developed by Noto et **al.** [3]. The kinetic mechanism for sodium was based on that of Zamansky et **al.** **[8],** while that for $Fe(CO)_5$ was described by Rumminger and Linteris **[9]**.

Changes in burning velocity, adiabatic flame temperature, radical superequilibrium, as well as the inhibition parameter $\Phi$ as defined by Noto et al., are plotted as a function of $(NaOH)_i$ concentration (Figure 1) and $CF_3Br$ concentration (Figure *2).* A constant value of $\Phi$ indicates that the exponential dependence of burning velocity on inhibitor concentration is well observed. Since sodium is known to be a catalytic agent with suppression efficiency some 10 times that of bromine [10], a comparison of changes in flame structure between the two agents tests which effects of $CF_3Br$ are generic **to** catalytic inhibitors in general.

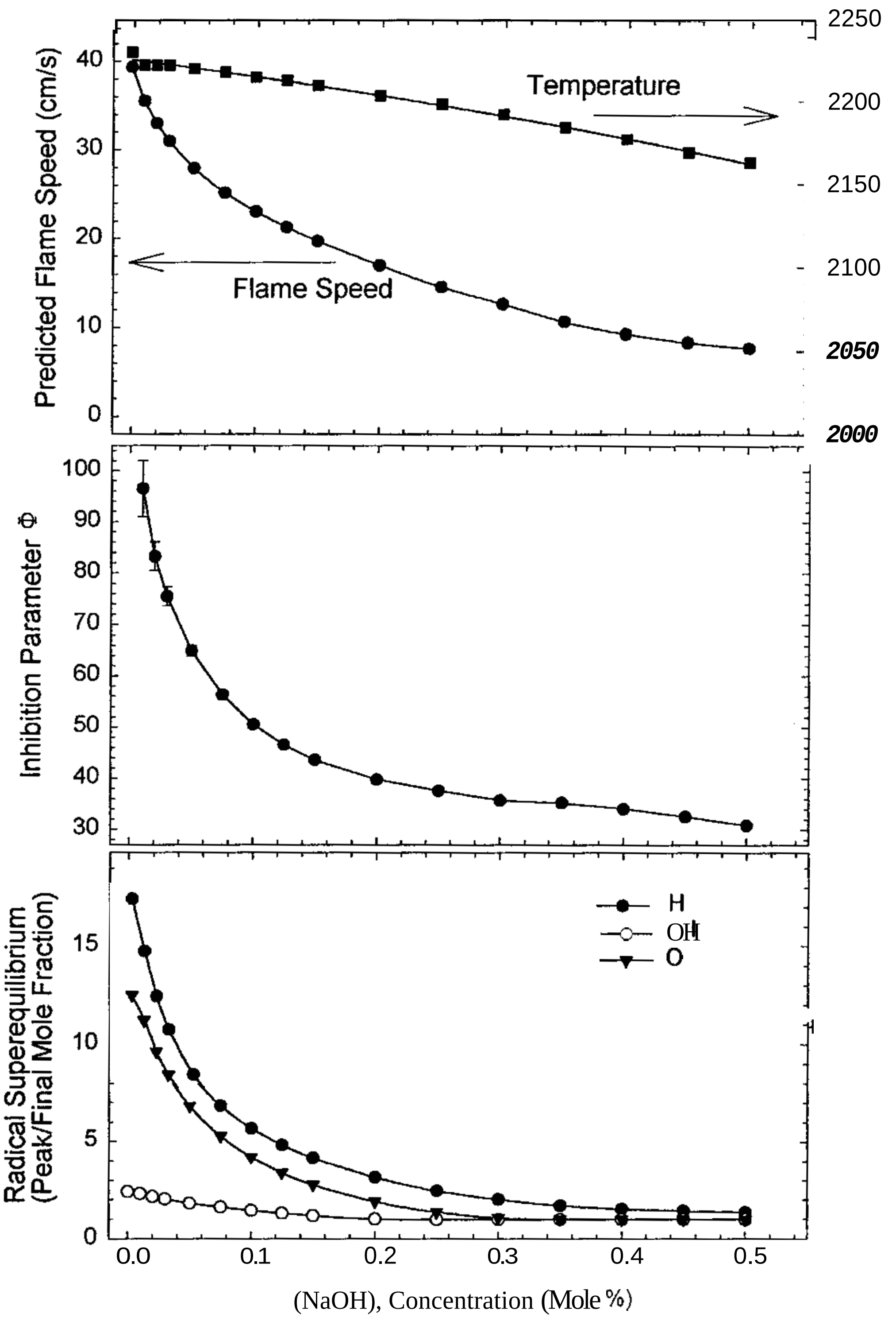

Figure 1. Flame speed, final flame temperature, inhibition parameter (defined in Noto et al.[3]), and superequilibrium concentrations of flame radicals computed for atmospheric pressure stoichiometric methane/air mixtures inhibited by sodium hydroxide. To simulate the evaporation process, NaOH was added to the reactants as a dimer and required to undergo an endothermic decomposition process before inhibition chemistry could occur.

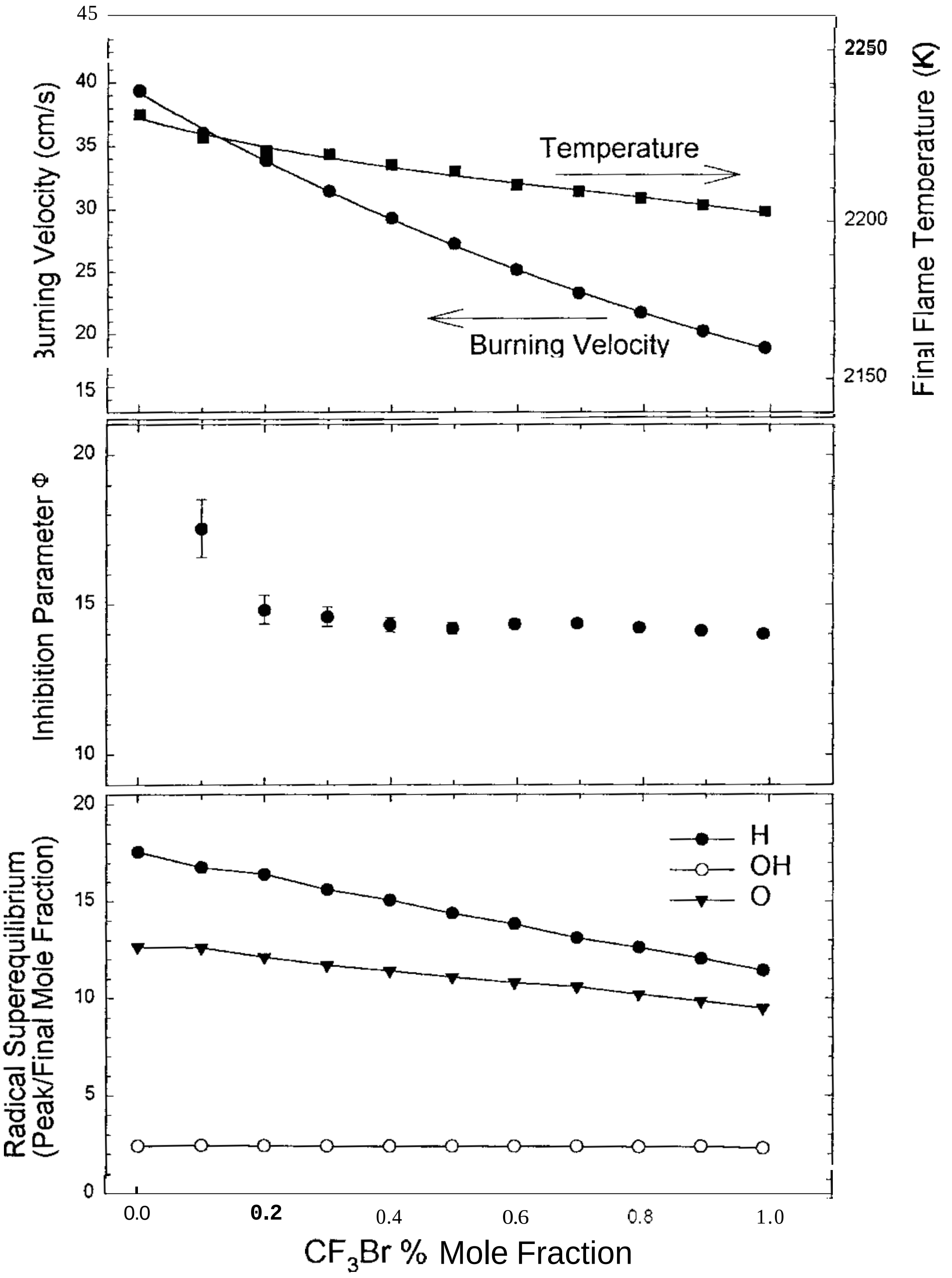

Figure **2.** Flame speed, final flame temperature, inhibition parameter (defined in Noto et al. [3]), and superequilibrium concentrations of flame radicals computed for atmospheric pressure stoichiometric methane/air mixtures inhibited by $CF_3Br$.

In the flame inhibited by NaOH, the radical superequilibrium is drastically reduced as the inhibitor is added, eventually reaching unity for all primary flame radicals for a sufficiently large inhibitor concentration (meaning that the radical concentrations never overshoot their equilibrium values). For inert agents, by contrast, the radical superequilibrium is increased as the flame is inhibited, even though the absolute concentrations decrease, because the predominant effect is the lowering of the final flame temperature. Notably, the inhibition parameter is not constant as a function of inhibitor concentration, but varies by more than a factor of three over the range of sodium concentrations considered here. The similarity relationship identified by Noto et al. **[3]** does not hold for sodium.

For inhibition by $CF_3Br$, the degree of radical superequilibrium is reduced with increasing agent concentration, though not as dramatically as **for** sodium. By this measure, $CF_3Br$ shows behavior typical of catalytic agents. The inhibition parameter, unlike **for** sodium, is nearly constant as a function of inhibitor concentration (except **for** the initial data point at an inhibitor concentration of 0.1%, which has a high uncertainty due to the small differences in flame speeds). This nearly constant inhibition parameter was found by Noto et al., although the modifications to the fluorocarbon kinetics yield somewhat higher inhibition parameters, which is in excellent agreement with the experimental value of 14.0 [3].

This raises the question of why the exponential relationship between burning velocity and inhibitor concentration is observed for $CF_3Br$ but not for other chemical inhibitors. The present results support the conclusion of Saso et al. [4] that saturation is a minor effect in the suppression effectiveness **of** $CF_3Br$/inert mixtures. One point is that for inhibition by $CF_3Br$, the H atom superequilibrium is only reduced by some **30%** for a 50% reduction in flame speed, whereas for NaOH the H atom superequilibrium is reduced by nearly **80%** for the same reduction in flame speed. Thus saturation is not as pronounced for $CF_3Br$ in part because the peak radical concentrations are still far out of equilibrium even when the burning velocity has been substantially reduced.

The other important point for $CF_3Br$ is that the bromine catalytic cycle involves the sequence of reactions [11]

$$\mathrm{Bp} + \mathrm{B}\rho + \mathrm{M} => \mathrm{B}\rho 2 + \mathrm{M} \qquad \text{(R1)}$$

$$\mathrm{H} + \mathrm{B}\rho 2 => \mathrm{HB}\rho + \mathrm{Bp} \qquad \text{(R2)}$$

as an important pathway in regeneration of HBr, since the direct reaction

$$\mathrm{H} + \mathrm{Br} + \mathrm{M} => \mathrm{HBr} + \mathrm{M} \qquad \text{(R3)}$$

has rather slow kinetics. This has two consequences: the increasing importance of Reaction (R1) (since it has a second-order dependence on the inhibitor concentration) compensates for the saturation effect as the inhibitor concentration **is** increased. **Also,** a much higher concentration **of** bromine is required **for** this reaction to be significant. Compared to sodium and iron, whose suppression kinetics are not thought to depend significantly on any second-order pathways, bromine is a relatively inefficient scavenger.

## COMBINED EFFECTS OF MIXTURES OF AGENTS

Since various studies have described the synergism of combinations of $CF_3Br$ and inert agents, it is worth exploring the extent to which the observations carry over to other combinations of agents. Furthermore, is there any advantage to combining different chemical scavengers that act independently (i.e., recombine different flame radicals)?

To this end, inhibition by iron was modeled in combination with both nitrogen and sodium. For this modeling, iron was considered to participate in a three-step mechanism involving only an **O + O** recombination. This pathway was identified by Rumminger and Linteris [9]; however, it is usually secondary in importance to an H + H recombination pathway. Using this simplified model, sodium and iron do not tion cycle. This model is *not* intended as an accurate description of iron's combustion chemistry but to investigate the combined effect of scavengers that operate through different pathways.

In Table 1, the effects of iron and sodium both separately and in combination, are given. The effectiveness of the inhibitors when combined is described by their "differential inhibition parameters." The differential inhibition parameter of substance A is determined by comparing the burning velocity of a mixture inhibited by A and B in combination to that of the mixture inhibited by B alone. As seen in Table 1, the differential inhibition parameters of iron and sodium in combination are much less than those of the two elements taken separately. Even though the two elements do not directly compete for the same flame radicals, the rapid exchange between H, O, and OH means that the two suppressants reduce each other's effectiveness. Furthermore, comparison between different scavenging cycles (for instance, the O + O scavenging cycle of iron and the H + OH scavenging cycle of sodium [Table 2]) shows that, for a comparable reduction in flame speed, the peak H atom and **O** atom concentrations are essentially independent of which scavenging cycle is occurring.

TABLE 1. INHIBITION BY TWO CATALYTIC CHEMICAL AGENTS IN COMBINATION.

Flame: Stoichiometric methane/air, atmospheric pressure

| Agent | Flame Speed (cm/s) | Inhibition Parameter Φ [Eq. (1)] |
|---|---|---|
| None | 39.6 | ---- |
| 0.2% $(NaOH)_2$ | 17.0 | 40.1[a] |
| 0.1% $FeO_2$ | 29.5 | 56.1 |
| 0.1% Fe **O,** + 0.2%$(NaOH)_2$ | 14.9 | 24.9(Fe), 32.4(Na)[b] |

[a] per sodium atom

[b] differential effect **of** specified inhibitor relative to other inhibitor alone

TABLE 2. COMPARISON OF RADICAL CONCENTRATIONS FOR FLAMES INHIBITED BY DIFFERENT SCAVENGING CYCLES.

| Inhibitor | Major Scavenging Cycle | Burning Velocity | Superequilibrium Factors H | Superequilibrium Factors **O** | Superequilibrium Factors OH |
|---|---|---|---|---|---|
| 500 ppm $FeO_2$ | 0 + 0 | 33.3 cm/s | 13.1 | 9.6 | 2.2 |
| 200 ppm $(NaOH)_2$ | H + OH | 33.0 cm/s | 12.6 | 9.8 | 2.2 |

Iron, in combination with nitrogen, does exhibit synergy. Figure 3 shows the burning velocity, flame temperature, and differential inhibition parameter of iron (using the **O + O** recombination mechanism) as a function of nitrogen addition to a methane/air mixture. The differential inhibition parameter of 500 ppm $FeO_2$ increases by some 23% as the flame temperature is lowered by nitrogen addition from 2230 K to 2010 K. These results may be compared to those of Saso et al. [4], who found that the inhibition parameter of $CF_3Br$ increased by some **45%** (at both 0.5% and 1.0% mole fractions of agent) over the same range of final flame temperatures.

Of the three reactions making up the O + **O** catalytic cycle of iron:

$$\Phi\varepsilon + O_2 + M \Rightarrow \Phi\varepsilon O_2 + M \qquad (R4)$$

$$\Phi\varepsilon O_2 + O \Rightarrow \Phi\varepsilon O + O_2 \qquad (R5)$$

$$FeO + O \Rightarrow Fe + O_2 \qquad (R6)$$

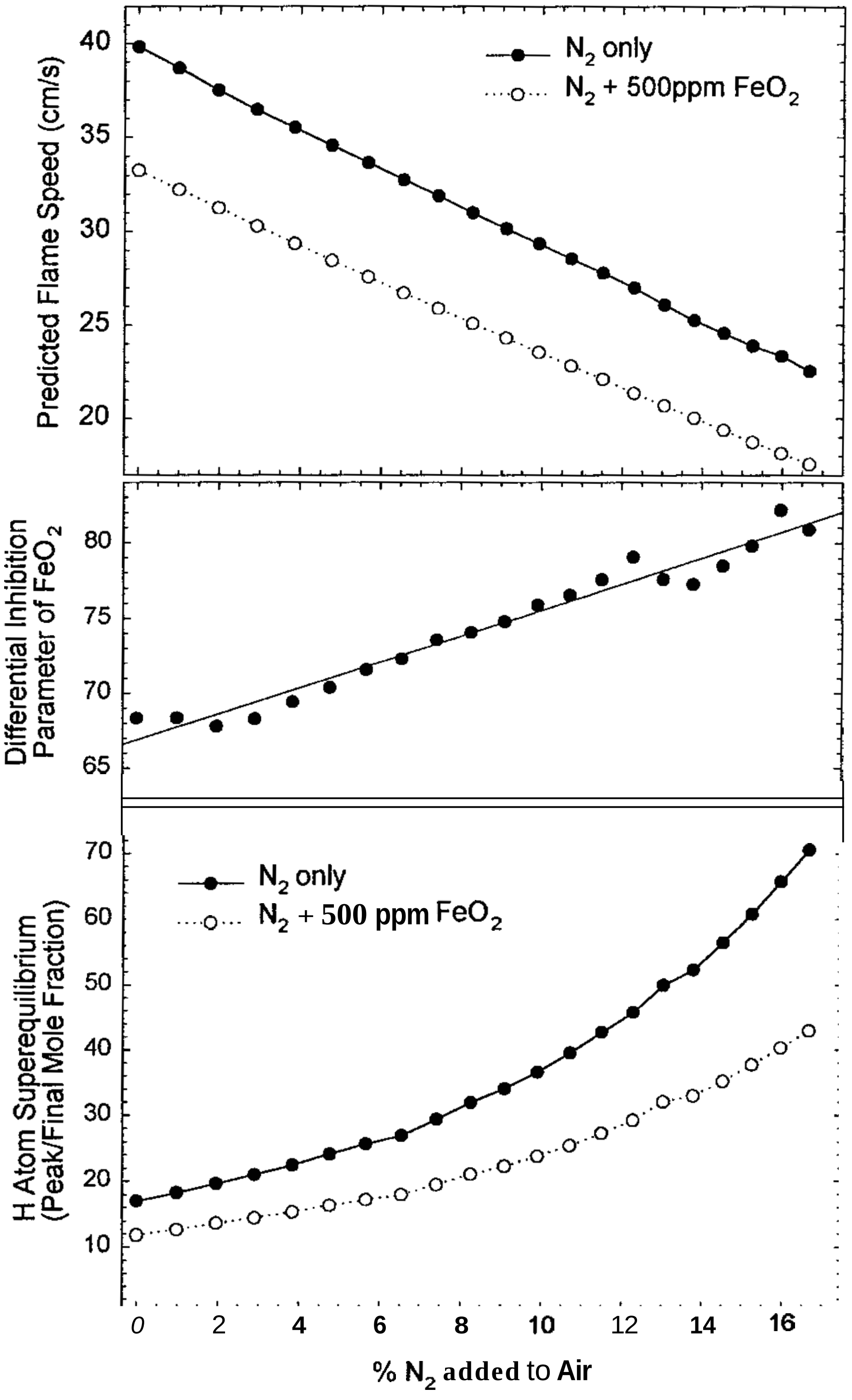

**Figure 3.** Flame speed, differential inhibition parameter (see text), and superequilibrium concentrations of atomic hydrogen computed for atmospheric pressure stoichiometric methane/air mixtures inhibited by nitrogen and $FeO_2$ in combination.

(R5) and (R6) are assumed to have rate coefficients independent of temperature. (R4) has a slight increase in rate with increasing temperature, but the product of the rate coefficient and the number density of third body colliders varies by **less** than 10% over the temperature range from 1400 K and 2500 K. Thus the kinetics of this scavenging cycle have no explicit dependence on temperature and changes in efficiency are due to variations in the radical pool with the addition of the inert agent. While the findings of synergism agree qualitatively between $CF_3Br$/inert and Fe/inert mixtures, the final flame temperature influences the inhibition parameter of $CF_3Br$ twice as much as that of the **O + O** recombination cycle of iron. Therefore while synergism in catalytic/inert mixtures may be a general characteristic, its magnitude can vary considerably depending on the kinetics of the catalytic component.

## CORRELATION OF BURNING VELOCITIES, FINAL FLAME TEMPERATURE, AND RADICAL CONCENTRATIONS

According to laminar flame theory, the burning velocity of a premixed flame is proportional to the square root of the overall reaction rate. In modeling the burning velocity of a large number of inhibited flames, the burning velocity correlates in almost **all** cases with the product of the peak H atom mole fraction and global activation energy:

$$S_L^2 = AX_H\exp(-E_a/kT_f). \qquad (2)$$

where **S,** is the laminar burning velocity, $X_H$ the peak mole fraction of atomic hydrogen, and T, the final flame temperature. The fitted parameters A and **E,,** obtained by considering the flame inhibited by nitrogen, are A=7940 cm/s, $E_a$=24.7 kcal/mol. The same empirical correlation holds for inhibition by other inert gases, by iron and sodium, by nitrogen and iron in combination (Figure 3), and by artificially increasing the H+ OH recombination rate. For the catalytic agents, which do not cause appreciable changes in the final flame temperature, the flame speed correlates with the peak mole fraction of atomic hydrogen, but not with that of atomic oxygen or OH radicals.

The observation that atomic hydrogen controls the burning velocity is not unexpected, due to the high diffusivity and reactivity of this species. Nevertheless, some noteworthy conclusions can be made: (1) this correlation can quantitatively describe the effect of a wide variety of inhibitors, both individually and in combination; and (2) catalytic agents reduce the flame speed by reducing the atomic hydrogen concentration, whether or not the scavenging cycle directly involves H atom recombination. The partial equilibrium between H, **O,** and OH is not disrupted in most inhibited flames.

Table **3** compares the burning velocities estimated using Eq. (2) to the calculated values **for** a variety of flame inhibitors. In almost **all** cases, the reduction in burning velocity relative to the uninhibited flame, predicted by Eq. (2) is within 10% of the actual value using the full kinetics. This relationship between burning velocity, final flame temperature, and peak H atom mole fraction holds for many agents (catalytic and inert). **At** the same time, there are a few notable exceptions: the fluorocarbons CHF, and $C_3HF_7$ are much better inhibitors (by nearly a factor of two) than **Eq.** (2) would predict, and so are $CF_3Br$ and HBr. Flame structure modeling indicates that the breakdown of this relationship involving the peak H atom concentration is a consequence of these agents reducing the H atom concentration mostly in the early part of the flame, as demonstrated below.

## CHANGES IN FLAME STRUCTURE DUE TO DIFFERENT AGENTS

The explanation for why the correlation between flame speed, H atom concentration, and temperature does not hold for either fluorocarbons or bromine containing compounds can be gained from examination of the effects of these compounds on flame structure. In Figure 4 the mole fraction of atomic hydrogen is plotted against the local temperature for flames inhibited by the inert agent N,, the catalytic agent $Fe(CO)_5$, the noncatalytic fluorocarbon $CF_3CHFCF_3$, and $CF_3Br$. All the inhibited flames have burning

TABLE 3. COMPARISON OF CALCULATED FLAME SPEEDS TO VALUES FROM EQ. (2)

| $S_{eqn} = A(X_{H,max}*exp(-E_a/kT_{ad}))^{1/2}$ | A=7940 cm/s | | $E_a$=24.7 kcal/mol | | |
|---|---|---|---|---|---|
| Flame Condition | $T_{adiabatic}$ | $X_H$max | $S_{premix}$ | $S_{eqn}$ | $\Delta S_{premix}/\Delta S_{eqn}$ [a] |
| $CH_4$/air (uninhibited) | 2234 | 6.64e-3 | 39.8 | 39.8 | ----- |
| +3.85%$N_2$ | 2187 | 5.96e-3 | 35.5 | 35.6 | 1.01 |
| +9.09%$N_2$ | 2121 | 5.12e-3 | 30.2 | 30.2 | 1.00 |
| +13.04%$N_2$ | 2065 | 4.45e-3 | 26.1 | 26.0 | 0.99 |
| +16.67%$N_2$ | 2015 | 3.94e-3 | 22.6 | 22.7 | 1.01 |
| +8.26%$CF_4$ (inert) | 1968 | 3.50e-3 | 18.4 | 19.9 | 1.07 |
| +500ppm$FeO_2$ (O+O cycle) | 2220 | 2.52e-3 | 33.3 | 32.7 | 0.92 |
| +1.0%$CF_3Br$ | 2203 | 3.42e-3 | 18.9 | 27.6 | 1.71 |
| +0.5%HBr (H+H cycle) | 2221 | 5.36e-3 | 30.3 | 35.2 | 2.08 |
| +0.5%HBr (H+OH cycle) | 2220 | 5.47e-3 | 33.3 | 35.5 | 1.52 |
| +5.2%$CHF_3$ (phi=1.1) | 2036 | 4.04e-3 | 16.3 | 23.7 | 1.46 |
| +3%$C_3HF_7$ (phi=0.9) | 2161 | 3.86e-3 | 17.8 | 27.7 | **1.81** |
| +0.05% (NaOH), | 2219 | 2.97e-3 | 28.0 | 26.2 | 0.87 |
| **+0.40%$(NaOH)_2$** | 2177 | 3.31e-4 | 9.4 | 8.3 | 0.97 |
| Increase H + OH + M Rate: | | | | | |
| x10 | 2234 | 4.04e-3 | 32.0 | 31.1 | 0.90 |
| x100 | 2234 | 1.35e-3 | 19.8 | 18.0 | 0.92 |
| x1000 | 2234 | 3.64e-4 | 11.2 | 9.3 | 0.94 |

[a] $(39.8-S_{premix})/(39.8-S_{eqn})$

velocities approximately 50% of that of the uninhibited flame, whose structure is also plotted for comparison. For all the flames considered here, temperature overshoot does not occur; the temperature monotonically increases with position passing from reactants to products.

The way in which the relationship between H atom mole fraction and local temperature is altered is characteristic of each type of agent. Other agents modeled (not shown on the plot for clarity) produce H atom profiles that closely resemble each other within the same class: $CF_4$ and N,, NaOH and $Fe(CO)_5$, and $CH_2FCF_3$ and $CF_3CHFCF_3$. The physical agents reduce the final flame temperature, but the H atom mole fraction at a given isotherm (above approximately 1300K) is changed very little from its value in the uninhibited flame. Both iron and sodium reduce the H atom mole fraction by a relatively constant factor throughout the reaction zone, in other words the inhibition occurs throughout the flame. The fluorocarbons, on the other hand, reduce the H atom mole fraction early in the flame (in the region below about 1200 K) but have relatively little impact on the peak concentration. The same situation occurs for $CF_3Br$; in this respect $CF_3Br$ bears more resemblance to nonbrominated fluorocarbons than it does to other catalytic agents such as iron and sodium. It is noteworthy that all the flames inhibited by chemically active suppressants show nearly identical H atom concentrations at 1700 K.

The depletion of radical species early in the flame has a marked influence on the flame speed. It is for this reason that fluorocarbons and bromine compounds are better inhibitors than the changes in temperature and peak H atom concentrations would predict. This observation implies that agents that deplete radicals in high temperature regions but not early in the flame are likely to be less effective inhibitors

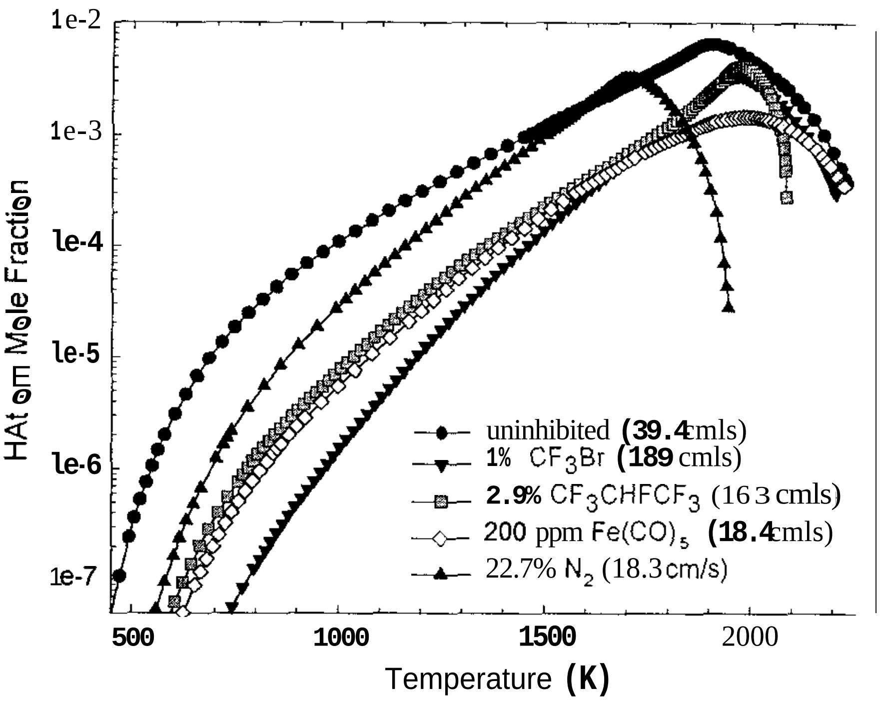

Figure 4. Mole fraction of atomic hydrogen as a function of local flame temperature for an uninhibited atmospheric pressure methane/air flame, and the same flame inhibited by various classes of suppression agents.

than would otherwise be expected. This may be the case, for instance, for condensed-phase agents which must undergo a vaporization process before inhibition chemistry can begin.

## CONCLUSIONS

In several respects, $CF_3Br$ is not representative of catalytic fire suppressants in general. Several features of its kinetics, including the strong temperature dependence of the catalytic suppression cycle, the apparent absence of significant saturation effects for a burning velocity reduction of more than 50%, and the preferential reduction of atomic hydrogen concentrations in low temperature regions of the flame, are not shared by other catalytic suppressants. At the same time, catalytic suppressants all appear to reduce burning velocity by reducing concentrations of atomic hydrogen, either directly *or* indirectly.

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