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SUPPRESSION EFFECTIVENESS STUDIES OF HALON-ALTERNATIVE AGENTS IN A DETONATION/DEFLAGRATION TUBE

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Experimental studies of the effect of the presence of halon-alternative agents on the suppression of premixed high-speed turbulent flames and quasi-detonations have been carried out in a 7.5 m long, 50 mm diameter tube. Lean and stoichiometric C_2H_4/air mixtures in the absence of any halocarbon, initially at 100 kPa and 295 K, constitute the reference states. A primary objective of the work has been to determine the relative suppression efficiencies of different agents under highly dynamic situations, without the undue influence of either the ignition event of the mixing of the agent into the flame front. This was accomplished by generating a highly turbulent flame/quasi-detonation in the driver section, which contained no suppressant, followed by measurements of the velocity and pressure ratio as the wave front entered the test section of the tube, which contained suppressant premixed with the same fuel/air combination. A turbulence generator in the form of a spiral obstruction was used in the tube to broaden the gas dynamic conditions attainable by the flame. Flame and shock wave velocities up to 1300 m/s, pressure ratios across the shock fronts over 26:1, and shock wave/flame spacings of the order of 10 cm were measured with piezo-electric pressure transducers and fast photodiodes. The experimental facility was successfully employed to clearly discriminate among the dynamic characteristics of the five compounds, revealing behavior distinct from what was observed in companion studies using atmospheric non-premixed flames. The suppression process is strongly influenced by the concentration of an agent, the structure and composition of an agent molecule, and the composition of the combustible mixture itself. The results were compared with the situation when an agent is fully premixed with the combustible mixture in the tube.

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ABSTRACT

The paper constitutes a brief report on the research done under highly dynamic conditions to find alternatives to halon 1301 for application to aircraft in-flight fire protection. Experimental investigations of the effect of the presence of halon-alternative agents on the suppression of premixed high-speed turbulent flames/quasi-detonations have been carried out in a 7.5 m long, 50 mm diameter tube. Lean and stoichiometric C_2H_4/air mixtures in the absence of any halocarbon, initially at 100 kPa and 295 K constitute the reference states. A primary objective of the work has been to determine the relative suppression effectiveness of different agents under highly dynamic situations, without the undue influence of either the ignition event or the mixing of the agent into the flame front. This was accomplished by generating a highly turbulent flame/quasi-detonation in the driver section, which contained no suppressant, followed by measurements of the velocity and pressure ratio as the shock/flame wave front entered the test section of the tube, which contained suppressant premixed with the same fuel/air combination. A turbulence generator in the form of a spiral obstruction was used in the tube to broaden the gas dynamic conditions attainable by the flame. Flame and shock wave velocities up to 1300 m/s, pressure ratios across the shock fronts over 26:1, and shock wave/flame spacings of the order of 10 cm were measured with piezo-electric pressure transducers and fast photodiodes. The experimental facility was successfully employed to clearly discriminate among the dynamic characteristics of the alternative compounds, revealing behavior distinct from what was observed in companion studies using atmospheric non-premixed flames. The suppression process is strongly influenced by the concentration of an agent, the structure and composition of an agent molecule, and the composition of the combustible mixture itself.

INTRODUCTION

The work is part of a large research program (Grosshandler *et al.*, 1994) focused on finding alternatives to halon 1301 for application to aircraft engine nacelle (Grosshandler *et al.*, 1993) and dry bay (Gmurczyk *et al.*, 1993, 1994) in-flight fire protection. A malfunction of an aircraft equipment or an anti-aircraft device entering the aircraft body could lead to a situation in which a vaporizing hydrocarbon spray (resulting from leaking fuel, hydraulic or lubrication lines) produces a combustible mixture which is then ignited by a hot or glowing fragment. If the space were confined, the pressure would increase behind the reaction front, accelerating the flame. A transition to turbulence would likely occur as the flame encounters clutter in the dry bay. If the ventilation is insufficient to relieve the pressure build up, the possibility of a supersonic detonation would exist, leading to destructive over-pressures in the dry bay.

A supersonic flame is distinct from the flames simulated in the cup, OFDF (opposed flow diffusion flame) and spray burners. As a result, the effectiveness of an agent in preventing a detonation depends upon different chemical and physical mechanisms. A shock wave precedes the supersonic flame. Obstructions in the flow promote intense mixing of the fresh reactants with the combustion products and cause the pressure waves to interact with the mixing region. Given enough distance, the flame can accelerate dramatically, increasing the temperature of the reaction zone behind the shock and further adding to the heat release rate. Depending upon the geometric details, the wave can approach its theoretical Chapman-Jouguet velocity and accompanying high pressure ratio. Even a slight variation in composition of the reactants near the limit of detonation can cause a dramatic change in the wave velocity and cause destructive pressures to be attained.

There exists an extensive literature describing the kinetics and dynamics of flame/shock wave systems formed within classical detonation tubes (*e.g.*, Lefebvre *et al.*, 1992; Nettleton, 1987; Lee, 1984; Baker *et al.*, 1983; Westbrook, 1982). Chapman and Wheeler (1926) were the first to note that a methane/air flame could be accelerated to a terminal velocity in a shorter distance within a circular tube by placing obstacles into the flow. Lee, *et al.* (1984) built on this observation to study quasi-detonations in hydrogen/air and hydrocarbon/air mixtures. A quasi-detonation propagates more slowly than a true detonation due to pressure losses in the flow, but its structure is more complex than a true detonation, and the mechanism of its propagation is not fully understood. Although an obstructed flow is more difficult to analyze than the flow in a smooth-walled tube, the complex arrangement was chosen for the current study because it more closely simulates a potentially damaging condition in the dry bay. The desire to rapidly suppress a flame and the associated pressure build up in such a situation is the primary motivation behind this study.

EXPERIMENTAL FACILITY

The effectiveness of a fire fighting agent in suppressing a high speed, premixed flame or quasi-detonation can be rated by the extent to which it decelerates the propagating wave and simultaneously attenuates the hazardous shock which is always ahead of the flame. Because the fire extinguishant is unlikely to be released prior to the establishment of a turbulent flame, the traditional experiment in which the flame inhibitor is premixed with the fuel and air prior to ignition (*e.g.*, Das, 1986) does not replicate the chemistry critical to the actual situation. Each dry bay on an aircraft has a different geometry, and the release of the agent once a fire is detected is highly variable. Heinonen *et al.* (1991) injected suppressant into a chamber shortly following the ignition of a fuel spray in air, but had difficulty controlling the mixing and in duplicating the process. The complexities and biases associated with the fluid dynamics of release can be avoided by premixing the agent with the fuel and air in a portion of the tube distinct from where the flame is initiated.

The two-section, deflagration/detonation tube shown in Figure 1 was designed to produce the desired environment for both the flame initiation and flame suppression regimes. A repeatable, uninhibited turbulent flame was fully established in the driver section, the design of which was based directly upon the work of Peraldi *et al.* (1986). They found that a 50 mm inner diameter tube with a blockage ratio of 0.43 could be used to create repeatable, high-speed flames and quasi-detonations within the first several meters of an 18 m tube. By varying the equivalence ratio of ethene/air mixtures from 0.5 to 2.1, they were able to produce flame velocities between about 600 and 1300 m/s.

The driver section was 5 m long and was equipped at the closed end with a spark plug. This section was filled with the combustible mixture of ethene and air. The gas handling system consisted of a vacuum pumping network; pressurized gas cylinders for the fuel, oxidizer and agent; and a dual

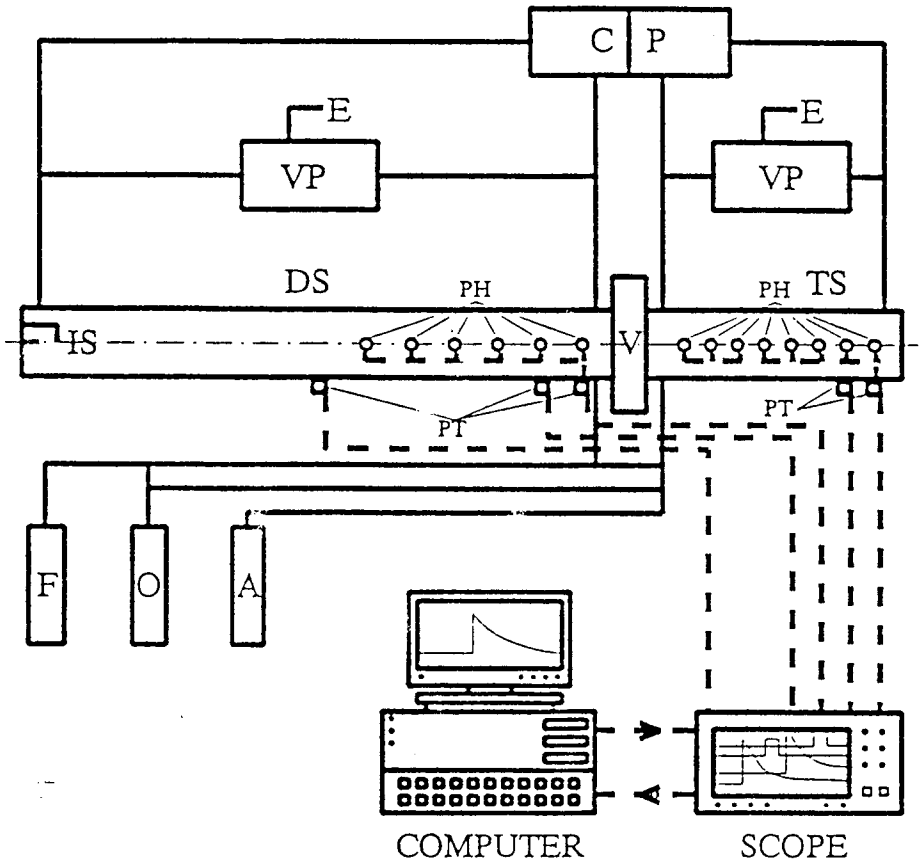

FIGURE 1. Schematic of the detonation tube facility installed at NIST. DS - driver section, TS - test section, V - gate valve, CP - dual circulating pump, VP - rotary vacuum pump, E - exhaust, IS - ignition source, F - fuel, O - oxidizer, A - agent, PT - piezo-electric pressure transducers, PH - visible radiation fast photodiodes, SCOPE + COMPUTER - collect measurement signals from pressure transducers and photodiodes.

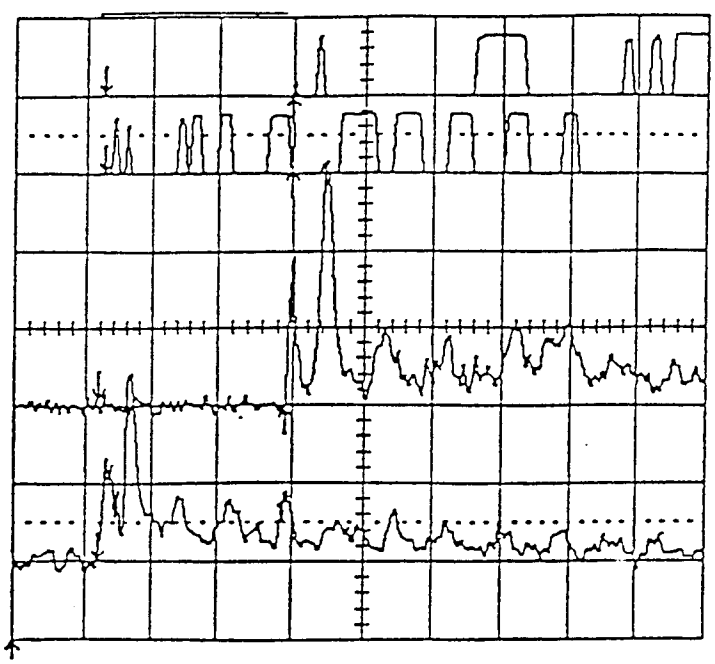

FIGURE 2. Suppression of 0.75 C_2H_4/AIR mixture with 1% vol. C_4F_{10}. Initial temperature 295 K and pressure 100 kPa. The two lower signals come from the pressure transducers; the two upper ones from the photodiodes.

circulating pump. The ignition energy was delivered in a microexplosion of a tin droplet short-circuiting the tips of nichrome electrodes connected to an 80 V power supply. Spiral-shaped obstructions made of 6.4 mm stainless steel rods with a pitch equal to the inner diameter of the tube were inserted into the tube, to produce an area blockage ratio of 44%, close to the value shown by Lee *et al.* (1984) to promote a quasi-detonation in their facility. The second section of the deflagration/detonation tube contained the gaseous agent along with the same fuel/air mixture used in the driver section. The diameter was the same and its length was 2.5 m. An identical spiral insert was used to maintain a high level of mixing. The two sections were separated from each other by a 50 mm inner diameter, stainless steel, high vacuum gate valve, which remained closed until just before ignition.

Pressure transducers and photodiodes were located along the test section to monitor the strength and speed of the combustion wave. Their output was recorded on a fast, multi-channel, digital storage oscilloscope coupled with a computer.

The whole system was evacuated to 10^{-1} Pa before filling the two sections separately with the desired mixtures, which were attained through the method of static partial pressures. The fuel/air ratio and total pressures were held constant across the gate valve. The initial temperature was the ambient value, 22 $\pm$ 3 °C. The oxidizer used in all experiments was breathing grade air. Ethene (CP grade, 99.5% volume purity) was chosen as the fuel because it had been demonstrated (Lee *et al.*, 1984) that subsonic flames, quasi-detonations, and full detonations all could be obtained in a tube of this geometry simply by varying the stoichiometry. After filling, the gases were homogenized independently using a double, spark-free circulating pump, recirculating the entire tube volume a total of 20 times. The mixtures were left for five minutes to become quiescent. About ten seconds prior to ignition, the gate valve was opened manually.

After ignition, the flame propagated into the driver section and accelerated quickly due to the intense turbulence created by the interactions of the flow with the obstacles, generating a shock wave ahead of it. After passing through the open gate valve the flame/shock system encountered the same combustible mixture and a certain amount of agent in the test section. Depending on the concentration of the agent, the flame was or was not extinguished and the pressure wave attenuated.

EXPERIMENTAL RESULTS

Baseline Study

Mach number and pressure ratio were the two dependent parameters which were measured as a means to characterize the extent of flame suppression. The Mach number was based upon the time it took for the pressure wave to travel the distance between the two pressure transducers, normalized by the sonic velocity of the reactant gases in the test section. The pressure ratio was evaluated from the average amplitude of the first pressure pulse to be recorded by each transducer, normalized by the initial pressure. Consecutive pressure jumps occurred, as can be seen in Figure 2, indicating that localized explosions in the mixing region between the spirals were present. Individual runs were concluded before the shock wave reflected from the end plate arrived back at the pressure transducers. The distance between the leading shock wave and the flame front was measured in some of the experiments from the time lag between the photodiode and pressure transducer response at the same location. The primary independent variables were agent type and concentration. The fuel/air equivalence ratio and the system absolute pressure were also varied for a number of agent experiments.

Experiments using 5% ethene in air mixtures were run under a variety of conditions to assess the operation of the system. Figure 2 is an example of the pressure trace when the agent partial pressure is insufficient to extinguish the flame radiation of an ethene/air mixture at a total initial pressure of 100 kPa. Knowing the distance between the pressure transducers, the shock speed was determined from the time lag between the pressure rises. The pressure ratio was tabulated from the initial pressure in the tube and either the first peak or the maximum pressure increase. The sensitivity of the flame speed and pressure ratio to the voltage of the ignition system, the mixing time of the components before ignition, the presence or absence of the gate valve, the speed of opening the gate valve, and cleaning the tube between runs were all investigated.

Experiments were conducted with 100% nitrogen in the test section, a 5% ethene in air mixture in the driver section, and the total pressure equal to 20, 50 and 100 kPa. The incident shock wave velocity measured 2.2 m beyond the gate valve and 0.3 m from the end of the tube was 420 $\pm$ 8 m/s at all three total pressures. The pressure ratio (P_1/P_0) based upon the initial pressure rise was 2.5 $\pm$ 0.5, and about 3.0 $\pm$ 1.0 based upon the peak increase.

No significant changes in shock speed occurred for a partial pressure fraction (which is approximately equal to the mole fraction) of nitrogen in air greater than 40%. The region of no change was extended down to 30% for the 20 kPa case. When no suppressing nitrogen was added to the test section the shock velocities attained values of 780 and 1170 m/s, respectively, for the 20 kPa and 100 kPa experiments. The pressure ratio increased dramatically at the same point as the velocity when the partial pressure of nitrogen was decreased, reaching maxima of 26 and 18, respectively, for the 20 kPa and 100 kPa initial conditions when no nitrogen was added to the ethene/air mixture. The peak pressure ratio, which normally did not correspond to the initial pressure pulse, exceeded 50:1 for the 20 kPa and 50 kPa experiments. The 100 kPa experiments generated peak pressure ratios around 30:1.

The results of the preliminary parameter assessment led to an experimental protocol which yielded flame speeds which were reproducible from run to run within about $\pm$ 2% (maximum deviation). Pressures downstream of the shock wave had a higher variability of $\pm$ 20% (maximum deviation) because of the complex shock structures created by interactions with the spiral rod inserts.

The bromine atom in CF_3Br is known to inhibit laminar flames by scavenging hydrogen atoms from the chain-branching radical pool. Experiments using halon 1301 were run to compare to an inerting agent like N_2, and to determine the suitability of the facility for assessing the effectiveness of a wide range of agents for suppressing high speed turbulent flames and detonations. The shock Mach numbers and the respective pressure ratios measured at three different total pressures as a function of the partial pressure fraction of CF_3Br in the test section have been measured. The largest effect of total pressure occurred between 50 and 100 kPa for halon, compared to between 20 and 50 kPa for N_2. Halon 1301 suppressed the flame at a partial pressure fraction of 10% to the same extent as if the test section had been completely filled with nitrogen. An unusual behavior occurred in the 100 kPa experiments when the concentration was between 2% and 3%. Both the Mach number and pressure ratio increased with the amount of CF_3Br, followed by the expected decrease for large concentrations. The reversal, while small, was greater than the uncertainty in the data.

Halocarbons, unlike nitrogen, are known to promote the production of soot. To determine the sensitivity of the shock velocity to soot contamination, a series of experiments with a 5% ethene/air mixture at a total pressure of 20 kPa was run for CF_3Br partial pressure fractions between 0 and 10% with and without cleaning the tube and spirals in the test section. The performance was similar, with the maximum deviation between the cleaned and uncleaned results amounting to less than 35 m/s.

Lean Mixtures at 100 kPa

Attenuation of the shock speed and pressure increase by the core agents and CF_3I was measured with the ethene/air equivalence ratio fixed at 0.75 (5.0% by volume C_2H_4), an initial absolute pressure of 100 kPa, and an initial temperature of 22 °C. It was found that the amplitude and speed of the pressure wave, and the speed of the trailing flame, were all strongly dependent on the agent type and concentration. The flame always followed the shock wave in such a way that both speeds were equal. However, when the amount of the agent in the mixture was increased, the distance between the shock and flame increased as well, up to around 100 mm as full suppression was approached. At the extinguishing concentration the radiation disappeared, which indicated the absence of the flame. In that situation the pressure wave amplitude was attenuated by a factor of eight and the wave speed by a factor of three, similar to the results for nitrogen.

The results for all the alternative agents are compared in Figures 3-4. The amount of agent is expressed both as a mass fraction and as a partial pressure fraction. The Mach numbers and pressure ratios at zero represent the pure combustible mixture with no flame suppressing agent present. One can see that the concentrations necessary for total extinguishment for all the compounds are between 40 and 50% by mass. However, at low concentrations the Mach numbers and pressure ratios are higher for some agents than even the value for the pure combustible mixture. Because of the sheer number of data, it is instructive to examine the results by the class of compound; *i.e.*, FCs, HFCs, HCFCs and the IFC (CF_3I).

The **FCs**, as a class, were generally the best performers on both a partial pressure and mass fraction basis. In fact, FC-116 was superior to CF_3Br, and FC-318 was about equivalent. FC-218 and FC-3110 slightly enhanced the pressure ratio in low concentration. FC-218 reduced the pressure ratio to less than 5:1 at a mass fraction of 0.29, which was better than with CF_3Br. Adding hydrogen to the molecule had a significant effect on the performance of the **HFCs**. The HFC-32/125 mixture produced peak pressures more than double the value for no suppressant. The Mach number was increased to its highest value of 4.1 when the mass fraction was 11%. It wasn't until the mass fraction exceeded 38% that the HFC-32/125 mixture became as effective as nitrogen in reducing the speed and pressure build-up of the shock wave. The two fluoropropanes, HFC-227 and HFC-236 produced the lowest pressure build-up of the HFCs and did a good job of suppressing the shock speed.

The chlorine atom in the two **HCFCs** created an additional complexity because chlorine is a strong oxidizer. As it has been found HCFC-22 was the least effective on a mass basis of all of the agents in fully suppressing the combustion wave Mach number, requiring a mass fraction of over 50% in the test section. The maximum pressure ratio for HCFC-124 was 32:1 at a 23% mass fraction, which was exceeded only by HFC-125 and the HFC-32/125 mixture. The one **IFC** tested was CF_3I. The Mach number and pressure build-up were cut about in half with partial pressure fractions in the test section of only 15 to 20%. None of the other chemicals, including CF_3Br, were able to accomplish that. Unfortunately, when the mass fraction was increased to 30%, the Mach number shot back up and the pressure ratio attained a value of 21:1, which was greater than when no CF_3I was present. This reversal, which was slight in the bromine-containing halon 1301, changed what at first appeared to be the most effective suppressing agent into one of the least effective agents. It is known that iodine atoms may cause a catalytic effect in some reactions by lowering an overall activation energy. It is not out of the question that in the case under consideration, in the intermediate concentration regime, those effects became important.

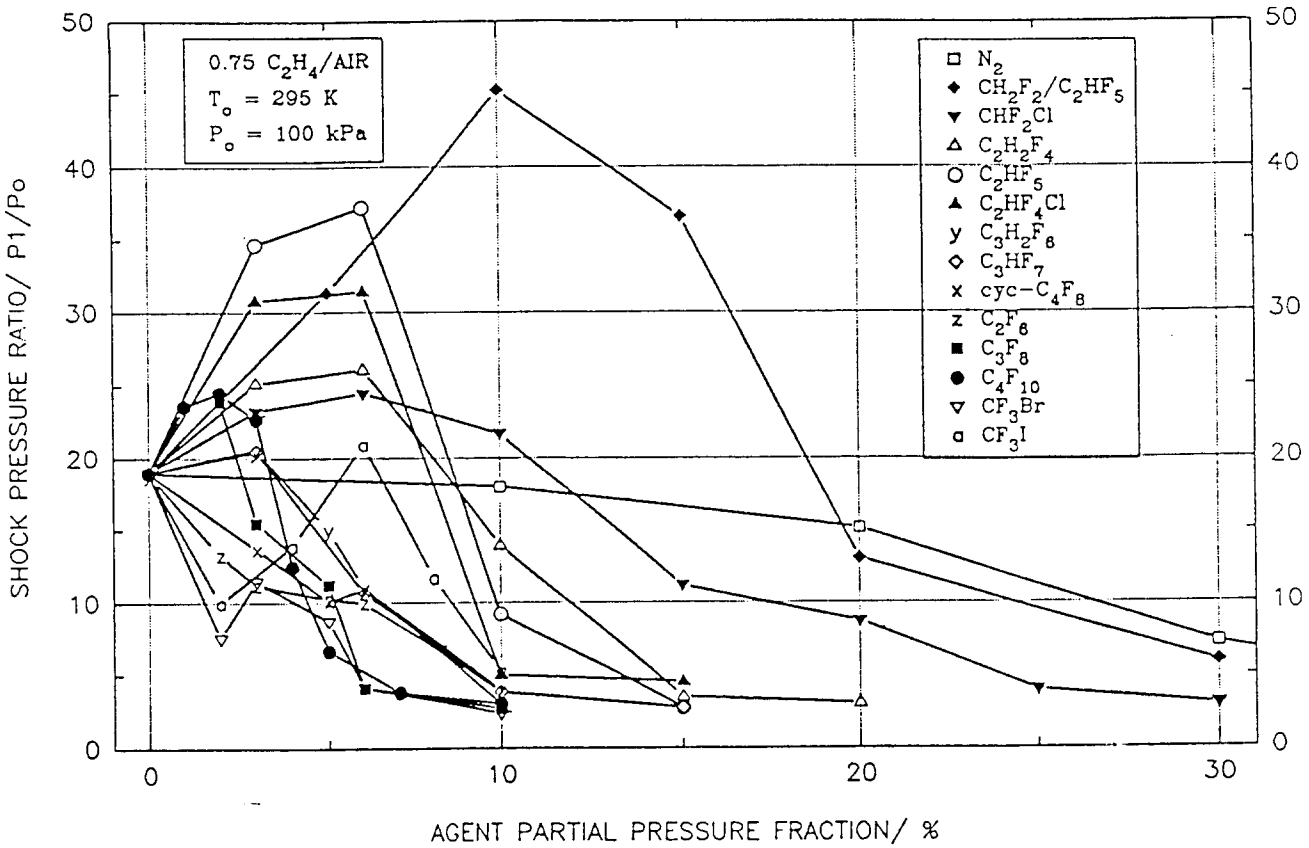

FIGURE 3. Suppression effectiveness of the agents in terms of shock pressure ratio versus partial pressure fraction.

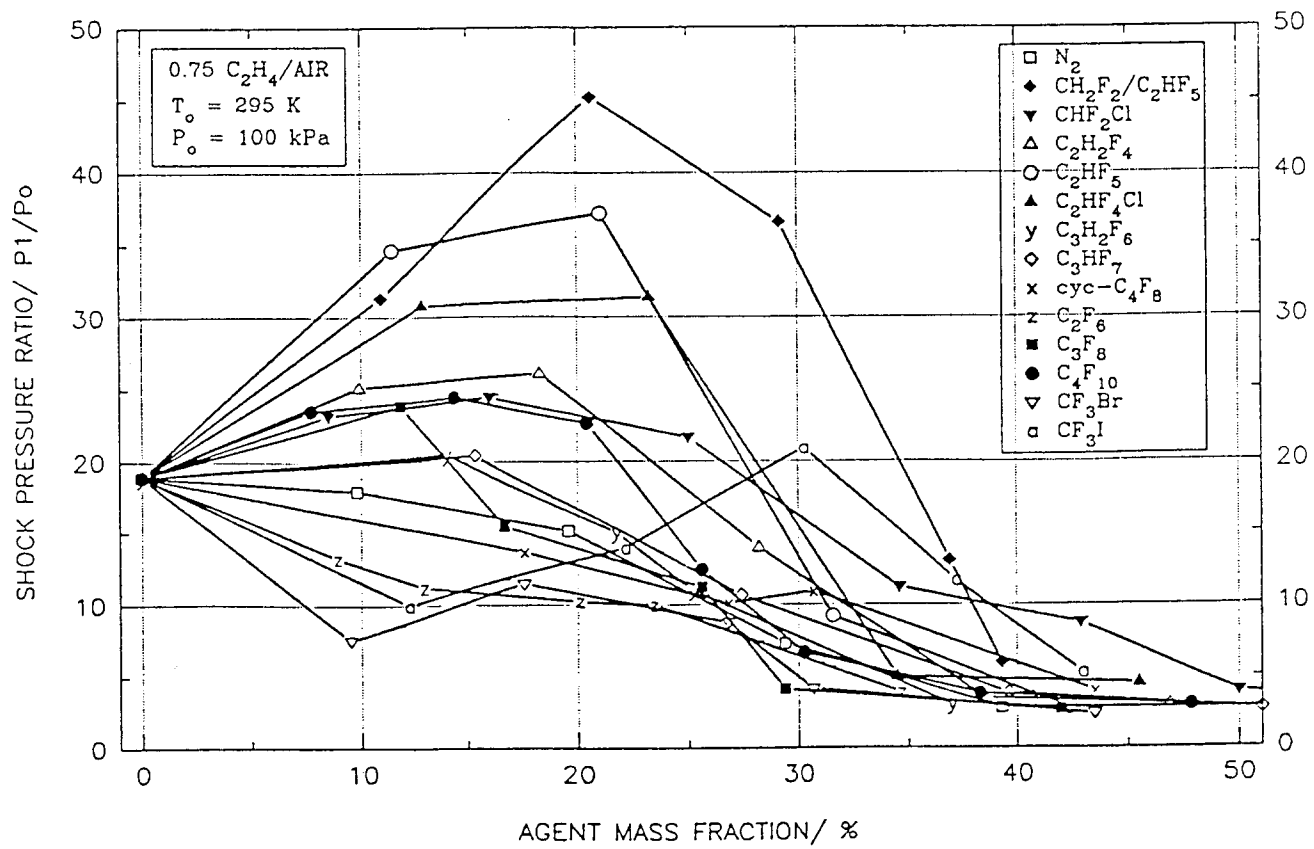

FIGURE 4. Suppression effectiveness of the agents in terms of shock pressure ratio versus mass fraction.

Stoichiometric mixtures at 100 kPa

Changing the fuel/air mixture (*i.e.*, the equivalence ratio) changed the flame temperature and radical concentration significantly. Figures 5-7 demonstrate the impact of equivalence ratio on the shock Mach number and pressure ratio with no agent in the test section. To see if the relative performance of the agents was dependent upon the fuel/air ratio a number of experiments were run under stoichiometric conditions. The initial pressure and temperature remained the same for the stoichiometric experiments: 100 kPa and 22 °C.

The results for the FCs are plotted in Figure 5. In all cases, the Mach number and pressure ratio increased as the fuel/air mixture was changed from lean to stoichiometric. The shapes of the curves, in the most part, were not significantly altered. The impact was greatest at low concentrations, decreasing as the partial pressure fraction reached 8%. For FC-116, the biggest difference occurred at a partial pressure fraction of 3%.

A distinctly different behavior occurred with the two HFCs shown in Figure 6. The hydrogen atoms attached to the agent molecules had less of an enhancing effect under stoichiometric conditions. The over-pressure was greatly reduced when the equivalence ratio was increased, leading to a cross-over condition where both the Mach number and pressure ratio became less for the stoichiometric condition. The implication of these data is that HFC-125 and the HFC-32/125 mixture increase in relative effectiveness under stoichiometric conditions.

Trifluoroiodomethane behaved no differently in suppressing the shock speed and pressure ratio under lean and stoichiometric conditions. From the data in Figure 7 it can be seen that the Mach number and pressure ratio increased uniformly over the range of partial pressure fractions investigated at an equivalence ratio of 1.0. The reversal of suppression effectiveness at partial pressure fractions between 3% and 6% observed at an equivalence ratio of 0.75 also occurred for stoichiometric combustion.

Ranking the Agents

The results which were gathered indicated the complexity of the suppression process in the deflagration/detonation tube. Because one does not know *a priori* the conditions in an actual dry bay fire zone, and because different intial conditions (*i.e.*, pressure, fuel/air ratio) affected the amount of agent required for suppression to varying degrees, a specific set of initial conditions was chosen at which all the agents were compared: 100 kPa, 22 °C, and 0.75 ethene/air equivalence ratio. The pressure ratio rather than the Mach number was chosen as the measure of suppression because of its direct impact on the structural integrity of the dry bay. The amount of halon 1301 required to reduce the pressure (a) totally and (b) to one-half the maximum increase was used to normalize the results.

Figure 8 displays three different performance parameters calculated under these conditions. The flame suppression number (FSN) is defined as the mass fraction of an agent required for suppression divided by the required mass fraction of halon 1301. The volume factor (VF) is defined as the storage volume of the alternative agent necessary for suppression divided by the storage volume of the CF_3Br. The saturated liquid density of the agents at 20°C was used to convert mass to volume. Halon 1301 has a VF and FSN of unity; the smaller these values, the better the agent.

While there are some reversals depending upon the basis of evaluation, Figure 8 shows that the perfluorocarbons were clearly the best performers and the HFC-32/125 mixture was the worst. HCFC-124 was the best of the non-perfluorocarbons. By comparison, the FSN and VF for nitrogen were found to be 1.2 and 32, respectively.

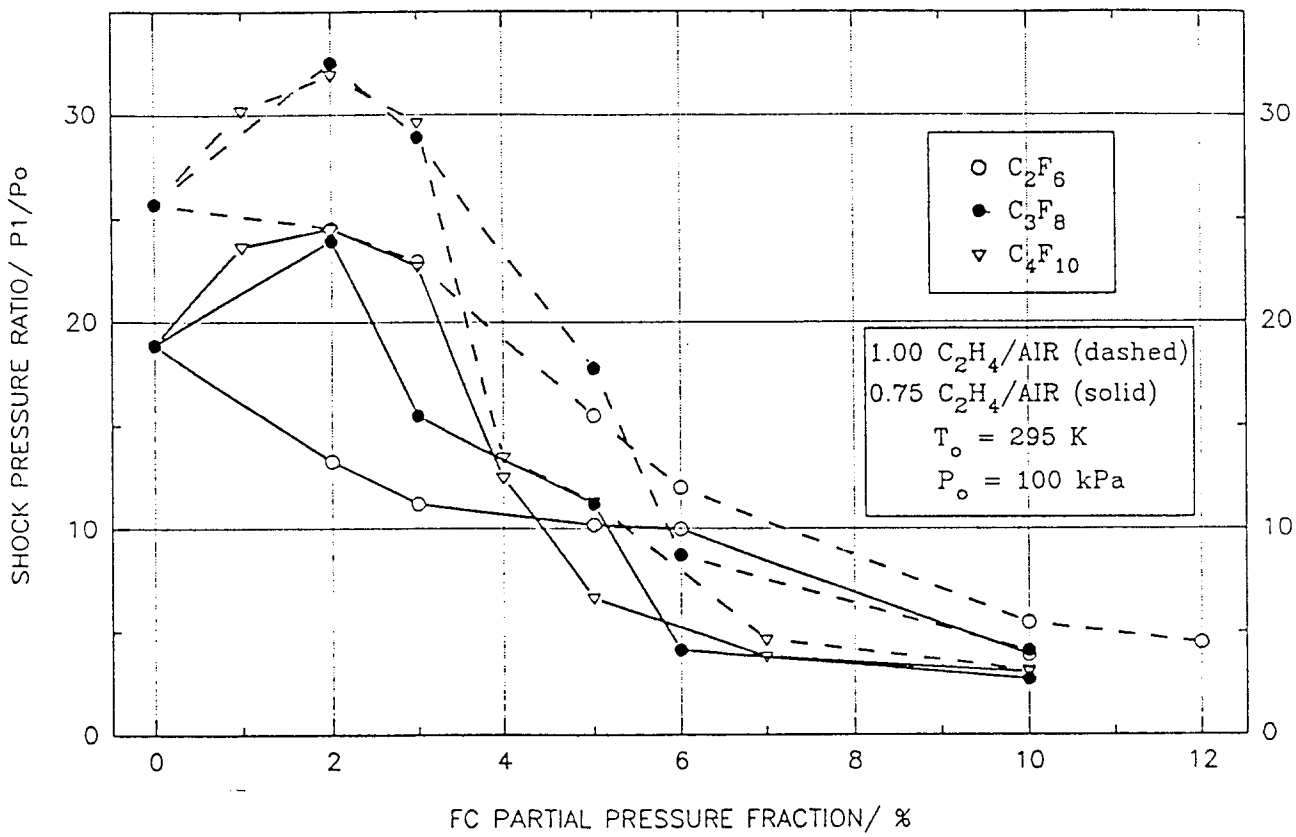

FIGURE 5. Suppression effectiveness of the FCs in terms of shock pressure ratio versus partial pressure fraction.

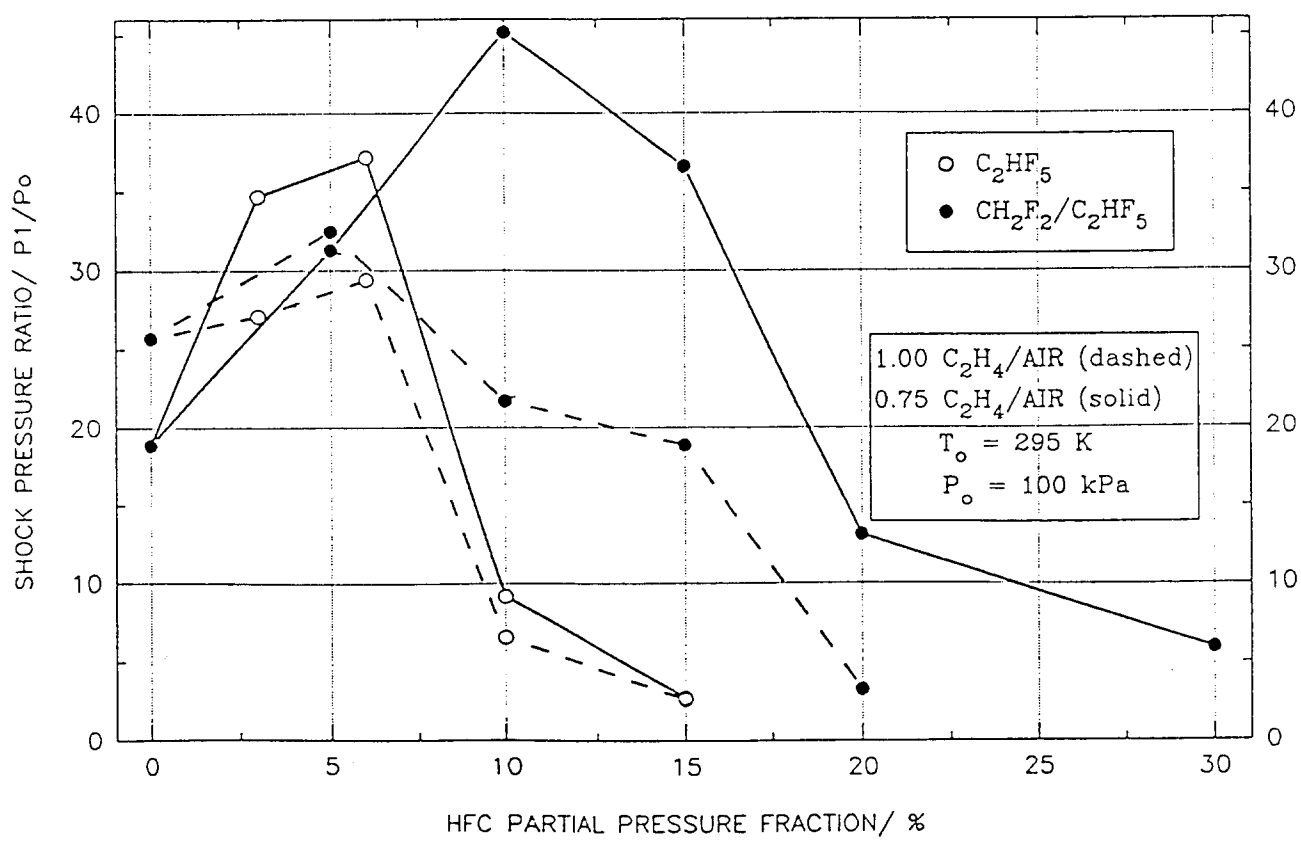

FIGURE 6. Suppression effectiveness of the HFCs in terms of shock pressure ratio versus partial pressure fraction.

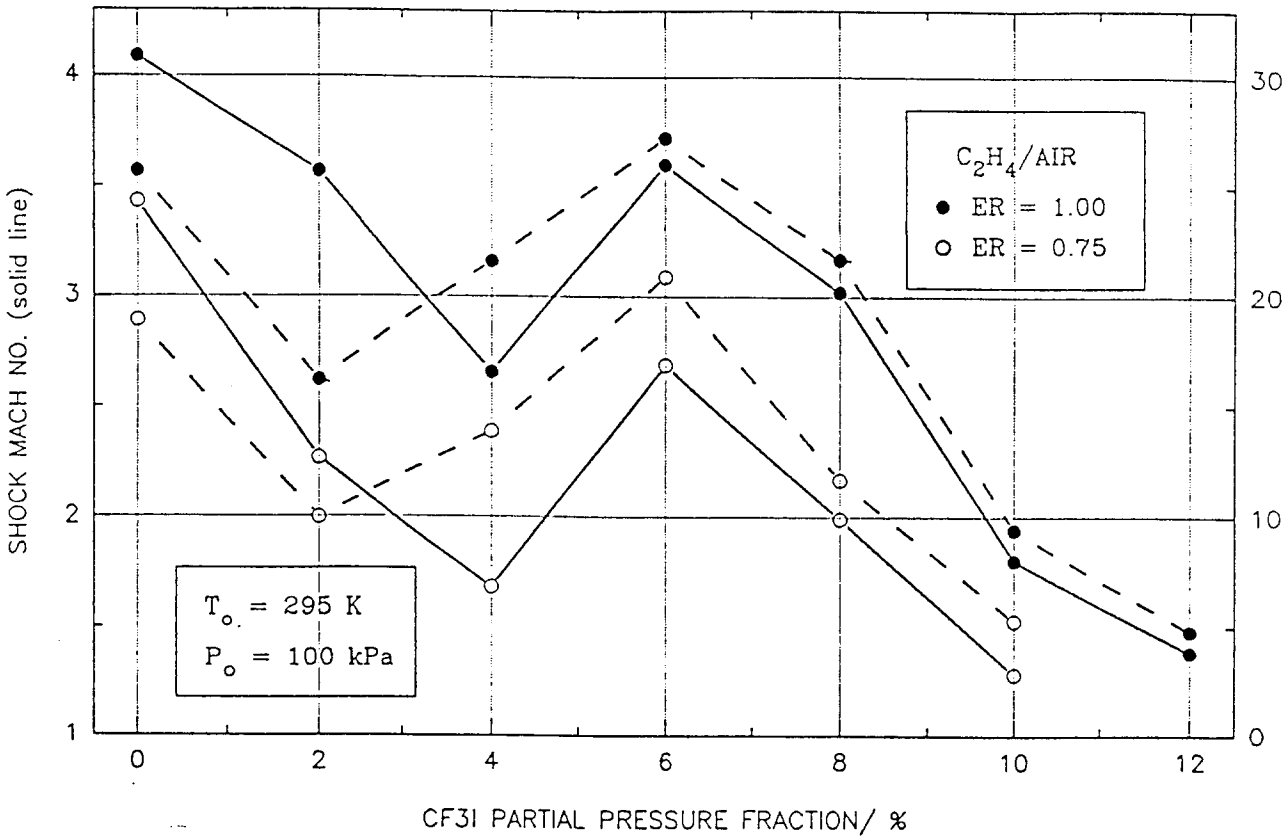

FIGURE 7. Suppression effectiveness of the IFC in terms of shock pressure ratio versus partial pressure fraction

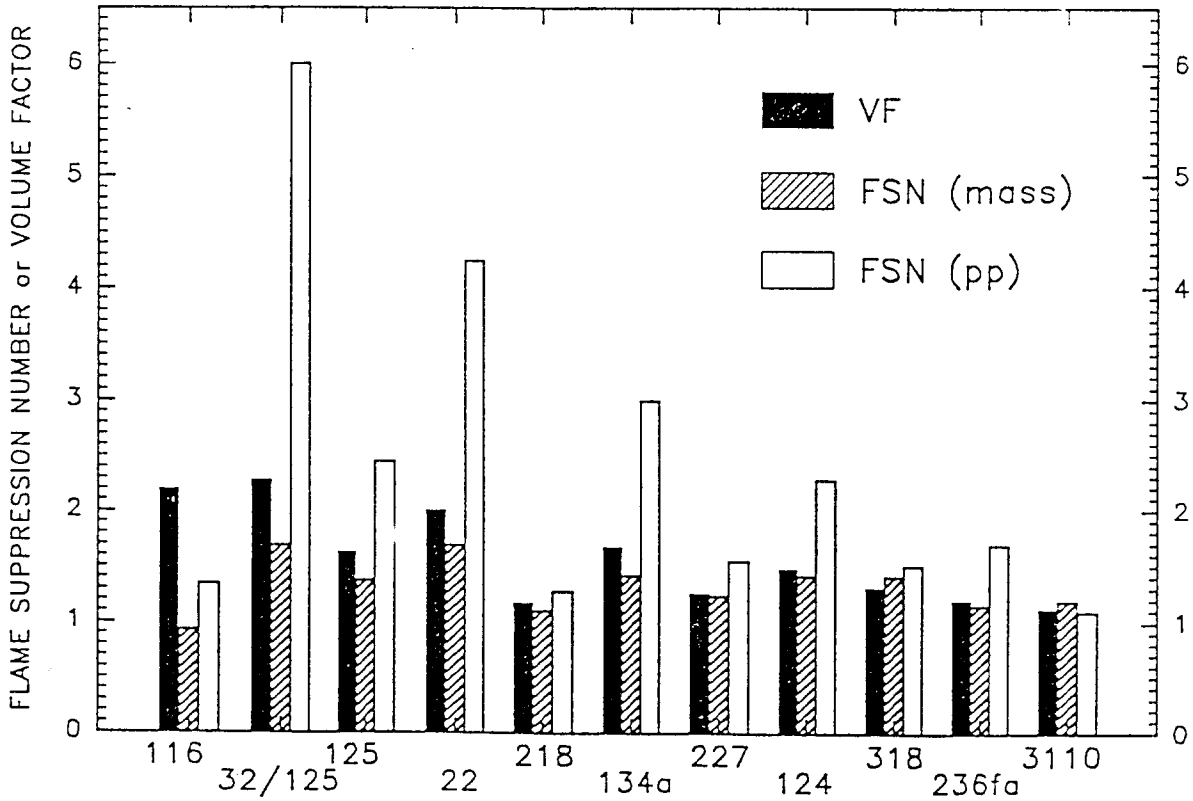

FIGURE 8. Volume factor and flame suppression numbers (based upon mass and partial pressure) versus alternative agents. The agents are ordered according to increasing boiling point.

CONCLUSIONS

It is necessary to emphasize that the experimental conditions in the deflagration/detonation tube differred significantly from those used in the cup burner, opposed-flow diffusion flame burner and turbulent spray flame burner. The main qualitative difference was the occurrence of a strong shock wave ahead of the flame. That wave influenced the gasdynamic, thermodynamic and chemical state of the pure combustible mixture in the driver section and the mixture containing an agent in the test section. Another feature was a high level of turbulence within the flame due to its high speed and the interactions with the spiral obstruction. The quantitative difference was a supersonic regime (relative to the undisturbed mixture) of flame/shock propagation and strong pressure changes (due to confinement and shock) during the process. Thus the oxidizer, fuel and agent molecules underwent preliminary processes before entering the flame zone.

The following conclusions can be drawn based on the results obtained:

1) Suppression of highly dynamic flames can be effectively studied in the two-sectional tube, permitting clear discrimination of performance among various alternative extinguishing agents.

2) The high-speed turbulent flame and the flame in the quasi-detonation wave under suppression strictly follows the shock wave which is always ahead of it in such a way that its velocity is the same as the shock velocity. The distance between the flame and the shock increases with the amount of an extinguishing agent. At extinguishment the flame disappears while the residual shock still exists.

3) The suppression process of the lean ethene/air mixture is pressure dependent both for chemically inert nitrogen and chemically active halon 1301. That may be related to the fact that the oxidation mechanism of ethene is known to be pressure dependent.

4) The analysis of the suppression data for the lean ethene/air mixture leads to the division of the alternative compounds into four general categories. These are, in order of decreasing effectiveness: perfluorocarbons; hydrofluorocarbons; and hydrochlorofluorocarbons (CF_3I itself constitutes a separate category discussed below). Within each of the categories the agents can be ordered (on a mole fraction basis) approximately inversely according to the molecular weight of the molecule.

5) The extinguishing concentrations of the more effective agents are around 10% by volume and 40% by weight, while less effective ones are 15-30% by volume and 40-50% by weight. The least effective agent is the HFC-32/125 mixture, giving unusually high pressure ratios. HCFC-22 requires the highest extinguishing concentration of all the alternatives.

6) The presence of a hydrogen-containing suppressant in the combustible mixture results in a significant increase in pressure ratio relative to that for the pure combustible mixture. The phenomenon occurs also for the compounds not containing hydrogen atoms at relatively lower concentrations but the impact is not so dramatic. The impact is generally weaker for stoichiometric relative to lean mixtures. That may be related to the fact that the agent acts as an extra fuel, causing the mixture to be richer. It is suspected that the effect may be associated with the release of hydrogen during the process which enhances the branching steps in the ethene oxidation mechanism. It is also not out of the question that a homogeneous autocatalysis occurs lowering the overall activation energy and enhancing the oxidation mechanism.

7) The behavior of CF_3I is different from the other agents. At lower concentrations (2-3% by volume) the performance is the best of all the alternatives. However, at intermediate concentrations (3-7% by volume) the performance is worsened significantly. Eventually at higher concentrations, up to the extinguishing value 10%, the performance is comparable with the perfluorocarbons and CF_3Br. The phenomenon is independent of the equivalence ratio of the combustible mixture. The behavior at the intermediate concentrations may be attributed to the catalytic effect caused by the iodine atoms.

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