AGENT SCREENING FOR HALON 1301 AVIATION REPLACEMENT

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ABSTRACT

A comprehensive experimental program is described in which eleven gaseous agents and sodium bicarbonate powder have been screened, so that the best three candidates for subsequent full-scale aircraft fire extinguishment evaluation can be identified. Chemicals with both a low ozone depletion potential and a high fire suppression efficiency were sought. Because the effectiveness of a fire suppression agent is known to be related to its thermodynamic properties, its behavior during two-phase flow, its interaction with flame chemistry, the timing of its release and the nature of the fire, a series of carefully designed experiments was conducted to examine each of these factors. Theoretical models were used to interpret the results, to increase our understanding of the suppression process, and to predict behavior over an expanded range of operating conditions. A pressure vessel with a rupture disc was used to discharge the agent into the atmosphere. Nine pure fluorocarbons, two mono-chlorinated fluorocarbons, a fluorocarbon azeotrope, and sodium bicarbonate powder were tested mixed with nitrogen or CF_3H. The exiting jet was photographed and laser light extinction was used to determine the extent and velocity of the jet. Four different type flame measurements were used to characterize suppression effectiveness. A description of the various flame screens is presented here. Chemical kinetics and molecular dynamic modeling were used as a means to investigate the extinction mechanism and to identify alternative gaseous chemicals likely to be superior to the original list of eleven.

BACKGROUND

Halon 1301 (CF_3Br) is currently used for in-flight protection against fires in engine nacelles and aircraft dry bay areas. After January 1, 1994, production of all halons will cease because of their high stratospheric ozone depletion potential (ODP). Low ODP alternative agents with the desirable attributes of 1301 (high suppression efficiency, low toxicity, high volatility, and satisfactory materials compatibility) need to be identified. Establishing a scientific basis upon which to chose the best alternatives was a primary objective of the research program described in this article.

The specific project objective was to screen eleven gaseous agents, $NaHCO_3$, and other promising chemicals to identify the best candidates for full-scale aircraft extinguishment tests, to be conducted at Wright Patterson AFB during 1994.

The project was broken into the following five major tasks:

1. static measurements of thermal properties and internal vessel dynamics;

2. experiments on agent dispersion and simulations of the fluid mechanics;

3. extinction measurements in a cup burner, an opposed flow diffusion flame burner, a turbulent jet spray burner, and a detonation tube;

TABLE 1. Specified chemicals evaluated, their boiling points, molecular weights and approximate critical volumes (specific volume at critical temperature and pressure of pure agent)

Compound	Formula	Boiling Pt., K	Molec.Wt.	Crit. Vol., m³/kg
Nitrogen	N_2	77	28	0.0032
FC-116	C_2F_6	195	138	0.0016
Halon 1301	CF_3Br	215	~~144~~ 149	0.0014
HFC-32/HFC-32 azeotrope	CH_2F_2/C_2HF_5	220	67	0.0021
HFC-32	CH_2F_2	221	52	0.0023
HFC-125	C_2HF_5	225	120	0.0018
HFC-227	C_3HF_7	253? ~~226~~	170	0.0016
HFC-22	CHF_2Cl	232	87	0.0019
FC-218	C_3F_8	236	188	0.0016
HFC-134a	$C_2H_2F_4$	247	102	0.0020
HCFC-124	C_2HF_4Cl	263	137	0.0018
FC-318	cyclo-C_4F_8	267	200	0.0016
HFC-236fa	$C_3H_2F_6$	273	152	0.0023
FC-3110	C_4F_{10}	273	238	0.0016

4. modeling of flame/agent chemistry; and

5. searching for additional agents for screening.

The chemicals specified by the Air Force to be examined are listed in Table 1, along with some of their key physical properties. The shear number of experimental variables 'precluded testing all agent alternatives under all conditions to be encountered. Instead, an experimental design was chosen which allowed the behavior of the different agents to be examined under conditions which isolated the influence of the many independent parameters. By identifying the physical and chemical attributes which control suppression, the performance of the agent during the full-scale testing program can be predicted in a more reliable manner. Considerable thought was given to the most appropriate means to relate the results of the individual tests to overall performance criteria. The uncertainty in each experiment was estimated by replication when appropriate. In other cases, analogy to similar experimental procedures, experience with related test methods, and theoretical calculations were used to establish error limits.

A recommendation was made to the U.S. Air Force of the best candidates for full scale evaluation, which was based on the results of the research described in this article and the results from a parallel research effort to ensure system compatibility (Gann, et al., 1993). This other study was concerned with agent residue, by-products formed during extinguishment, compatibility of the agent with metals and polymers, and environmental and toxicological issues. Details of the selection protocol are given in the final report (Grosshandler, et al., 1994).

MAJOR TASKS COMPLETED

Thermal Properties and Vessel Dynamics

Static measurements: A stainless steel pressure vessel with an internal volume of 50 ml was charged with a mixture of agent and nitrogen or CF_3H gas, and placed in an isothermal bath. Pressure measurements were made at room temperature, 213 K and 422 K. The solubility of N_2 and CF_3H at room temperature of the agents was determined, using a fill density of 1/3. The initial charge pressure at room temperature was about 4.2 MPa. Nitrogen was found to be only slightly soluble in all of the agents. The maximum pressure measured in the 1/3 filled nitrogen/agent mixtures was below 12 MPa at 422 K. The solubility of the CF_3H in all of the agents was substantial at room temperature, resulting in pressures which exceeded the design limit of the apparatus (14 MPa) at temperatures as low as 323 K, even when the vessel was initially filled to only 1/3 its capacity.

Preliminary results from these experiments were presented by Yang, *et al.* (1993); the complete thermodynamic data are available in the final project report (Grosshandler, *et al.*, 1994).

Dynamic measurements: A high pressure vessel equipped with a quick release opening and an internal volume of 500 ml was built in order to assess the discharge dynamics of agent/nitrogen mixtures. The vessel, described by Pitts, *et al.* (1993), contains a rupture disc 19 mm in diameter, mounted at the opening of the vessel. The vessel was filled initially 1/3 or 2/3 with liquid agent, and subsequently pressurized with nitrogen to within 20% of the disc burst pressure. The contents were discharged by impulsively injecting nitrogen into the vessel through a solenoid valve, causing the disc to rupture. The internal temperature and pressure of the vessel were monitored during discharge, from which the mass emptying rate was estimated. A second vessel was constructed of thick-walled plexiglass so that the emptying process could be examined with a high-speed movie camera. Dynamic measurements were carried out at 213 K and 295 K, and a number of measurements were completed at elevated temperatures up to a bottle pressure of 8.2 MPa.

Experiments were performed with different size exit orifices. Reducing the diameter from 19 to 13 mm, as expected, increased the discharge time. The most dramatic difference occurred when the diameter was further reduced to 6 mm. Discharge tests were also performed with a 0.5 m long extension tube (the same diameter as the largest exit orifice) downstream of the release mechanism to simulate flow through a short manifold in an engine nacelle application. The extension tube had only a slight retarding effect on the liquid discharge rate for the agents studied.

Agent Dispersion and Fluid Mechanics

Experiments: The overall objective of this task was to determine the rate of penetration of the eleven alternative agents into air. The efficiency of the agent dispersion and mixing is strongly dependent upon several factors, including the fluid dynamics and thermodynamics of the vessel venting process, choked flow through the orifice, and the mixing process involving a high-speed, two-phase turbulent jet. High speed photography was used to capture the complex two-phase flow in the impulsively discharged jet. Laser light extinction measured at different axial locations downstream indicated the passing of the dense agent cloud. The pressure outside of the vessel was monitored as well.

A data acquisition system was developed for simultaneously recording all of the transducer outputs ($\approx$ 25 kHz for 500 ms) during a single experiment, including laser light extinction, pressure measurements

within and external to the vessel, and thermocouple measurements for temperature within the vessel. The laser measuring system consisted of a series of five low-power helium-neon lasers and photodiode detectors which were aligned perpendicular to the centerline of the vessel containing the agent at various positions from the exit, covering a total distance of 1.2 m. By recording the times at which extinction occurred and knowing the distances between the laser beams, it was possible to measure the average velocity of the two-phase front over four downstream distances of roughly 0.3 m each.

Three piezoelectric pressure transducers were incorporated into the experimental system. One was used to measure the pressure within the vessel, located in the nitrogen gas above the liquid. A second transducer outside of the vessel was used to measure the dynamic pressure at a position near the vessel exit. The third transducer measured the dynamic pressure on the jet centerline downstream of the exit. Measurements of temperature within the liquid and gas were desired to further characterize the release of liquid from the pressurized vessels. A technique was developed utilizing a microwelder and microscope to fabricate 12.5 and 25 μm thermocouples mounted on rods which were then positioned inside the vessel at both ends (in the liquid agent and gas).

The experimental system used burst disks which ruptured at a prescribed pressure. As a result, it was necessary to add nitrogen to the vessel sufficient to reach this pressure and initiate the experiment. One of the concerns which grew out of the initial modeling effort (Cooper, 1993) was that nitrogen flowing into the vessel during a release would modify the release behavior from that typical of a self-contained vessel. The data acquisition system allowed investigation of this point by performing experiments in which the flow of nitrogen into the vessel was varied over a wide range. Conditions were characterized for which the pressure traces recorded during the release of the agents were independent of the nitrogen flow.

Pressure traces from the downward release of the 1/3 filled vessel showed two well defined behaviors: a relatively slow fall off in pressure during the first 15-20 ms of the release, followed by a much faster decrease in pressure. The timing about doubled for the 2/3 filled cases. By comparison with high-speed motion pictures of the releases within and outside of the vessel, it was clear that the two behaviors corresponded to periods when liquid was expelled from the vessel (first) and the remaining gas bled out. During the period of the release, the gas temperature within the vessel was found to decrease substantially. The pressure transducer outside of the vessel indicated two large increases in pressure during the release, which were consistent with the two flashings of the liquid observed in the high-speed motion pictures.

Average velocities of the two-phase mixture near the exit were measured to be on the order of 70 m/s for the lower boiling point agents. For the furthest position for which the measurements were recorded, the average velocity were found to decrease to the order of 50 m/s. The magnitude and shape of the jet centerline velocity profiles varied substantially with the boiling point of the agents. Measurements of upward releases were quite different than for the downward releases. High-speed motion camera pictures showed that vigorous boiling occurred within the liquid in the vessel, as opposed to the downward releases for which no boiling was observed. The releases required roughly ten time longer to expel the liquid from the vessel.

Numerical Modeling: The numerical model CONCHAS-SPRAY, developed at Los Alamos (Cloutman, *et al.*, 1982), was used to simulate the transient spray. For conditions similar to the discharge of R-22, the impact of droplet size, total mass flow rate, and temperature on the flow were investigated. Details can be found in the final report (Grosshandler, *et al.*, 1994).

Flame Extinction Measurements

Four different flame extinction experiments were designed to examine the inhibition properties of the agents over the range of conditions likely to be encountered by the aircraft in flight: a cup burner, an opposed flow diffusion flame (OFDF), a turbulent jet spray flame, and a detonation tube. The eleven gaseous suppression agents, nitrogen and halon 1301 were tested in all four experimental facilities; sodium bicarbonate was tested in all but the detonation tube. The fuels included JP-8, heptane, propane, ethene, and two hydraulic fluids.

Cup Burner: The cup burner design of Sheinson *et al.* (1989) was adapted to the multiple phase fuel and agent regime required by the current study. A 28 mm o.d. cup with a 45° ground inner edge was used in conjunction with a 120 mm o.d. chimney. The flow rates of agent and dry air were controlled by rotameters, meeting at a tee and mixing prior to entering the chimney. A calibrated, constant velocity screw was used to feed the sodium bicarbonate powder at a fixed rate.

The fuel was ignited by a small torch and was maintained at a constant level in the cup during the test (Bajpai, 1974). Measurements were performed with a constant air flow (about 20 liter/min). A one-minute burn period occurred prior to agent addition to allow the system to be preheated consistently, and the test was completed within a specified time interval to minimize preferential distillation effects. Agent was added in small increments and the flame behavior was observed for five seconds before further agent addition. The flowmeter and pressure readings at flame extinction were recorded, from which the concentration of agent was found. The experiment was repeated to insure consistent results.

The extinction concentrations of the gaseous agents are compiled on a volume and mass fraction basis in the final project report (Grosshandler *et al.*, 1994) with JP-8, heptane, propane, and the two hydraulic fluids as fuels. For conditions where reliable historical data exists, a comparison with the current study revealed no large discrepancies. Results from the powder test indicated that less $NaHCO_3$ than halon 1301 is necessary to extinguish the C_3H_8 cup burner flames, in agreement with the results of Milne, *et al.* (1970) for CH_4/air counterflowing diffusion flames.

Opposed Flow Diffusion Flame: The suppression effectiveness of the eleven gaseous agents and sodium bicarbonate powder were determined by measuring the critical concentration of suppressant needed to extinguish diffusion flames stabilized between counter-flowing streams of oxidizer and a vaporizing liquid fuel experiencing a stretched flow field (Seshadri, 1977). These experiments were conducted by K. Seshadri of the University of California at San Diego.

Unlike the cup burner, the effect of the flow field (strain rate) on the extinction process is an independently controlled parameter in the OFDF. This facilitates a more controlled comparison of the relative suppression effectiveness of the agents. The agent concentration needed to extinguish the flame was measured as a function of the oxidizer flow rate, which is directly related to the flow field strain rate. These measurements were conducted using n-heptane and JP8 as fuels over a range of global strain rates (20 s^{-1} to 200 s^{-1}). All eleven gaseous agents were tested, plus N_2 and CF_3Br. The low strain rate flames required approximately the same amount of agent for extinguishment as measured in the cup burner experiment. Significantly less agent was necessary to extinguish the high strain rate flames. The measurements of the relative suppression effectiveness for heptane are, in general, consistent with the rankings determined from extinction measurements conducted in the NIST cup burner. The agent concentrations at extinction are slightly lower for JP8 than for heptane, which is also consistent with the NIST cup burner results. The results are tabulated in the final project report (Grosshandler, *et al.*, 1994).

Turbulent Jet Diffusion Flame: An engine nacelle fire is typically a turbulent diffusion flame stabilized behind an obstruction in a high speed flow. In the facility used to simulate an engine nacelle fire in this study, the fuel (JP-8 or hydraulic fluid 83282) was injected along the centerline of a tubular burner through a pressure-jet nozzle that formed a 45° solid-cone spray. The nozzle was rated at 1.89 l/hr at a gauge pressure of 687 kPa. Experiments were also performed using hydraulic fluid 83642. Air flowed around the nozzle passage within an annular region which had an outer diameter of 50 mm. The nozzle was recessed about 80 mm from the exit of the annular casing. The flame, which was horizontal and easily viewed from the exit plane, was stabilized on a 35 mm diameter steel disk attached to the nozzle body. The air flow was monitored with a critical orifice meter, yielding mean air velocities up to 33 m/s.

Extinguishment occurred when a certain level of agent was supplied to the air upstream of the flame. The gaseous agents were injected impulsively into the air stream and dispersed uniformly across the tube before they reached the flame. At the desired moment, a computer controlled solenoid valve opened for the preselected amount of time (20 to 700 ms) and the agent flowed into the air stream. The temperature of the air was varied between ambient and 422 K. The performance of nitrogen, halon 1301, the eleven gaseous agents, and sodium bicarbonate were all evaluated. Details of the burner facility are reported elsewhere (Grosshandler *et al.*, 1993).

The unique feature of this burner is the ability to independently control the rate of injection and total mass of agent. Faster injection rates substantially reduced the amount of material necessary to extinguish the flame. The concentration of agent measured at extinguishment in the turbulent burner was less than that needed to suppress the cup burner. A detailed comparison of the results is included in the final report (Grosshandler *et al.*, 1994).

Deflagration/Detonation Tube: Dry bay protection requires that an agent respond quickly enough to suppress the transition from a deflagration to a detonation. A worst case scenario was investigated in this portion of the project, where the fuel was completely vaporized and well mixed with air. Suppression of a fully established advancing flame front was tested as a function of agent concentration. A 5 m long, 50 mm diameter, closed stainless steel tube was fitted with an ignition source at one end and a large gate valve at the other. A 2.5 m test section was attached to the downstream side of the gate valve. A 6 mm diameter stainless steel rod was formed into a spiral and inserted into the tube to act as an obstacle to the flow in order to rapidly accelerate the combustion wave (Peraldi *et al.*, 1986). Flame front progress was determined with fast time response piezoelectric pressure gauges and photodiodes which were flush mounted on the tube. Pressure and emission were monitored using a high speed digital storage oscilloscope.

The experiment proceeded as follows. Initially the gate valve was closed. The driver and test sections were filled by partial pressure. Combustible mixtures of ethene and air were used to fill the driver section and the same fuel/air mixture with various agent concentrations was used to fill the test section. The gas blends were mixed by a circulating pump. Both the fuel/air mixture on the driver side and the fuel/air/suppressant mixture on the test side were initially at room temperature. The gate valve was opened and a short time interval later ignition via a melting wire was initiated, producing a combustion wave in the driver section which accelerated towards the test section. Unlike the suppression studies done elsewhere (*e.g.*, Heinonen *et al.*, 1991), the ignition source did not influence the results since the agent did not see the flame until it was fully developed. The rate of pressure increase and acceleration of the combustion wave in the test section were monitored and compared to the case where no agent was present in the test section.

Mixtures of ethene from 5% to 9% in air were ignited in the system. Ignition was not evident when the total pressure was 10.1 kPa. The majority to the experiments took place at 100 kPa. The primary indicators of suppression were a reduction in shock Mach number and a reduction in pressure increase when compared to the zero agent concentration condition. Gmurczyk, *et al.* (1993) describe how these data were interpreted. A tabulation of the relative effectiveness of the different agents in the detonation tube facility is reported in Grosshandler, *et al.* (1994).

Flame/Agent Chemistry

The objective of this portion of the research program was to determine the role of chemical and thermal processes associated with halogenated compounds in the extinction of hydrocarbon diffusion flames through a comprehensive modeling effort. These calculations were aimed at answering the following fundamental questions: what is the relative importance of thermal vs. chemical effects on flame suppression; what is the relative effectiveness of model halogenated compounds; and what chemical characteristic of agents would be most useful in an alternative to CF_3Br?

Flame agent chemistry modeling proceeded on three fronts: compilation and evaluation of thermochemical and kinetic data, plug flow reactor modeling, and premixed flame computations. The final report (Grosshandler *et al.*, 1994) describes the methodology of each of these tasks. A brief summary of preliminary results is given here.

The development of a comprehensive mechanism for fluoro-carbon chemistry in flames was based on an initial screening mechanism, which required significant refinement in the form of addition of new species and reactions. One version of the mechanism encompassed 52 species and over 250 reactions (thirty-two C2 hydrocarbon plus twenty C1 fluorocarbon species) in which C2 fluorocarbon chemistry had been neglected. Incorporation of C2 fluorocarbon chemistry required an additional 86 species. The thermochemistry and kinetics were evaluated for this extended set.

Mechanism evaluation primarily relied on plug flow reactor computations on the C1 fluorocarbon mechanism. These computations used small concentrations (< 100 ppm) of CH_2F_2 as a means to track the fluorine chemistry under conditions where the hydrocarbon chemistry would undergo minimal perturbation. The results showed that even under lean conditions, the stability of fluorocarbon radicals lead to C2 unsaturated fluorocarbons that were not included in the working mechanism.

The premixed flame computations were somewhat more selected in scope because of the significantly greater computational resource requirements. A limited set of computations on methane or ethylene as fuel and di-and tri-fluoromethane as the agents was conducted in both the burner-stabilized and freely propagating case. The results correlated with the plug flow reactor computations.

Additional Agent Screening

The objectives of this task were to broaden the scope of agent screening beyond the 11 candidates identified in Table 1, and to develop and test structure-activity relationships (SARs) for ozone depletion potential and fire suppression efficiency. These SARs were the basis of the strategy for the identification of a new generation of environmentally safe fire extinguishants.

An exploratory list of 103 potential halon alternatives representing ten distinct chemical families was cited in NIST TN 1279 (Pitts *et al.*, 1990). This document served as the primary list of candidates. Additional possibilities were also be considered, and subjected to the screens described earlier in this article. A full list of compounds investigated is given in the final report (Grosshandler *et al.*, 1994).

A direct correlation between C-H bond dissociation energies (BDEs) and tropospheric lifetimes was demonstrated in this study. Additional agent screening was narrowed to focus on the following candidates, selected from the strategic organization of halocarbons into four working categories: (1) low ODP compounds that contain at least one Cl, Br or I (*e.g.*, $CHFCl_2$, $CF_2=CClF$, $CF_2=CFBr$, CHF_2CHFBr and CF_3I); (2) compounds which contain F but which do not contain an ozone depleting halogen (*e.g.*, $CF_2=CFCHF_2$ and $CHF=CFCHF_2$); (3) inorganic compounds which contain halogen atoms (*e.g.*, NF_3 and SiF_4); and (4) inorganic compounds which do not contain halogen atoms (*e.g.*, $NaHCO_3$ and $KHCO_3$).

Accurate calculations of BDEs for a wide range of hydrofluorocarbons and hydrochlorocarbons were completed. These calculations indicate that the presence of a fluorine or chlorine atom can influence the BDE of neighboring C-H bonds in a different manner. The effect that this has on ODP, GWP and fire suppression efficiency is discussed in the final report (Grosshandler *et al.*, 1994).

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