

FUEL EFFECTS ON THE EXTINGUISHMENT OF LAMINAR DIFFUSION FLAMES BY THERMAL AGENTS

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INTRODUCTION

As part of the National effort to identify effective replacements for halon fire suppressants [1], the National Institute of Standards and Technology (NIST) has been investigating whether highly effective thermal agents are feasible. Thermal agents are defined as those that obtain their effectiveness solely by heat extraction and dilution. Excluded from investigation are species that directly or indirectly disrupt the combustion chemistry, such as halons, which derive much of their effectiveness by the release of bromine atoms that catalytically remove hydrogen atoms in the flame zone.

A paper presented during the 1999 Halon Options Technical Working Conference (HOTWC) described a study designed to better understand the effects of thermal agents on diffusion flames [2]. Detailed chemical-kinetic modeling was used to predict the effectiveness of nitrogen (N_2), carbon dioxide (CO_2), argon (Ar), and helium (He) on methane opposed-jet flames. Surrogate agents having idealized properties were utilized to demonstrate that the effectiveness of a thermal agent is independent of the location of heat extraction relative to the flame front as long as the gases are convected into the flame. It was also shown that an agent having zero heat capacity (i.e., a pure diluent) weakens a flame by slowing the overall rate of the combustion reactions. During HOTWC 2000, extinguishing concentrations were reported for two liquid agents, methoxy-nonafluorobutane (HFE7100) and lactic acid/water mixtures, measured using the NIST Dispersed Liquid Agent Fire Suppression Screen (DLAFSS) with a propane flame stabilized on an opposed-flow porous cylindrical burner (a.k.a. a Tsuji burner) and for gaseous thermal agents using methane flames with two diffusion-flame burners, the DLAFSS and a modified coflow burner known as a Santoro burner [3].

In this paper, experimental extinguishment measurements and detailed chemical-kinetic modeling investigations are extended to propane diffusion flames. The results are compared with earlier findings for methane flames in order to better understand the role of fuel variations in flame extinguishment behavior.

EXPERIMENTAL AND DETAILED CHEMICAL-KINETIC MODELING APPROACHES

EXPERIMENTAL

Two experimental systems were used to study extinguishment of diffusion flames by gaseous thermal agents. The first of these was the DLAFSS, which incorporates a Tsuji-type cylindrical porous burner located in a test section through which a conditioned flow of oxidizer passes. A detailed description of the system is given elsewhere, so a brief summary is provided here [3,4,5]. Even though it was designed primarily for evaluating liquid fire suppression agents, the system can also be used for gaseous agents.

Typically, the Tsuji burner is operated at considerably higher oxidizer flow velocities than required here, and the resulting strain rates are much greater than found in buoyancy-dominated flames. To generate the low oxidizer velocities required to investigate buoyancy-dominated flames, the blower used to induce the flow in the DLAFSS was operated at the lowest setting possible. The thermal agents were then added to the induced flow. Note that the oxidizer flow velocity is expected to vary as the agent volume fraction increases.

The flow rate of agent was controlled using a needle valve, and the volume flow rate was measured using a rotameter (Fischer Porter, Model **10A1755***). Even though the agent volume flow rate was known, it was discovered that it was not possible to predict the agent volume fraction using only the assumed air-flow rate. This observation was attributed to variations in the total flow volume due to changes in the blower efficiency as agent was added. An alternate approach was adopted to estimate the total flow rate and thus the local flow velocity at the flame. The agent concentration was determined by measuring the oxygen volume fraction in the oxidizer stream at the center of the settling chamber of the wind tunnel using a paramagnetic oxygen meter (Servomex Oxygen Analyzer Model **570A**) in conjunction with a small sampling pump (Gilian Hi Flow Sampler, Model HFS **513A**). Since the volume flow rate for the agent was known, knowledge of the degree of air dilution allowed the total flow rate to be calculated.

Suppression tests were performed by gradually adding the gaseous inhibitor to the oxidizer (air) stream until blow-off occurred. The volumetric flow rates of the suppressant at blow-off were recorded. The oxygen concentration was then noted for the same flow conditions. Measurements were made using methane and propane as fuels and N₂, CO₂, Ar, and He as suppressants.

The second experimental system was a coflow diffusion flame burner based on a design originally developed by Santoro et al. and often referred to as the Santoro burner [6]. The actual burner used in this investigation is a modified Santoro burner developed to investigate acoustically forced, time-varying diffusion flames [7]. A drawing of the burner is shown in our HOTWC 2000 paper [3]. It consists of an open **13.7** cm length of tubing with a **1.27** cm outer diameter that serves as the central fuel tube. The tube lip thickness is 0.065 cm, yielding an inner diameter of **1.14** cm. One end of the tube is attached to a plenum containing a loud speaker. The speaker, used to force the fuel flow acoustically, was not employed during this study. The open end of the fuel tube extends **0.4** cm above a surrounding **10.2** cm diameter ceramic honeycomb formed from 0.15 cm square cells and having an overall length of **2.5** cm. After passing through a chamber filled with glass beads and several screens, a flow of air and agent enters the honeycomb to form the surrounding coflow for the flame.

Fuel and the air/agent mixture were delivered to the burner using a system of thermal mass flow controllers (MFC). The flow-control system is a modification of a system developed at NIST for a previous investigation as described in detail by Pitts et al. [8]. The flow system was software controlled by a desktop 486-DX personal computer. The volume flow rates required to generate precise mixtures of air and agent were specified by the software. The air and agent streams were combined upstream of the burner in a mixing chamber designed to ensure a homogenous mixture. Pneumatic shut-off valves prevented unwanted backflow of gases and allowed for easy calibration of each MFC. Modifications for the current study included the addition of a 30 SLM (standard liter per minute) mass-flow controller for the air flow and movement of the **10** SLM and **2** SLM mass-flow controllers to the agent and fuel streams, respectively. Gas supplies for the fuel and agents were bottled gases with nominal purities of 99.0%. Air was supplied from an in-house high-pressure line, and its relative humidity was a consistent **4.5%**.

DETAILED CHEMICAL-KINETIC MODELING

The approach used to model the extinguishment of diffusion flames by thermal agents is discussed in detail elsewhere [2, 9]. The code OPPDIF [10] developed by Sandia National Laboratories and now available from Reaction Design of San Diego, was used for the calculations. OPPDIF is built on a number of general-purpose subroutines, collectively known as CHEMKIN-III [11], which handle many of the tasks associated with the calculation. Data describing the reaction mechanism and thermodynamic and

* Certain commercial equipment, instruments, or material are identified in this paper to adequately specify the experimental procedure. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials or equipment is necessarily the best available for the purpose.

transport properties are either incorporated in OPPDIF or accessed as databases. OPPDIF solves the pseudo-one-dimensional equations describing an axisymmetric opposed-flow laminar diffusion flame. These codes have previously been used to investigate the extinguishment of methane flames as the result of adding inert agents to the air counterflow [2, 9]. The required chemical mechanism, thermodynamic data, and transport data were taken from the GRIMech 1.2 mechanism developed with support from the Gas Research Institute [12]. GRI-Mech 1.2 consists of 32 chemical species undergoing 177 reactions. One- and two-carbon species are included. Thermodynamic and transport data are provided as separate files. The mechanism was created by starting with appropriate estimates for the rate constants and then optimizing the mechanism to provide the best agreement with experimental measurements in such systems as premixed flames, shock tube studies, and flow reactor investigations.

Predictions of extinguishing volume fractions for various thermal agents were obtained by using an experimental value for one gas, N₂, to determine the maximum calculated flame temperature with the required amount of N₂ added and assuming that the other agents would induce flame extinguishment when the maximum flame temperature was reduced to the same value. This procedure yielded good agreement with the limited data available for the extinguishment of methane flames by thermal agents [2, 9]. The calculated extinguishing concentrations were also shown to be in close agreement with cup-burner results using heptane as fuel.

For the current study, it was desired to extend the OPPDIF estimates for extinguishing concentrations to propane flames. Detailed reaction mechanisms for propane combustion have not been as extensively developed as for methane, and no widely accepted version comparable to GRI-Mech is available. A mechanism assembled at NIST by Babushok and Tsang was used [13]. This mechanism was created by adding a block of reactions for C3-C4 compounds taken from Marinov et al. [14, 15] to a C1 and C2 mechanism previously developed at NIST [16]. It consists of 83 species undergoing 509 reactions.

In general, it was found that calculations for the methane flames required considerably less time and were much more computationally stable than those for propane flames. For this reason, the propane computations were generally limited to conditions directly applicable to the current investigation.

EXPERIMENTAL RESULTS

Experimental results for the volume fractions of thermal agents required to extinguish methane and propane Tsuji-burner flames in the DLAFSS are included in Table 1. Each measurement was repeated a minimum of three times, and the uncertainty limits refer to measured standard deviations. The value included in Table 1 for extinguishment of the propane flame by CO, differs slightly from that reported earlier due to a change in the way the agent volume fraction is estimated [3].

TABLE 1. SUPPRESSION RESULTS USING THE DLAFSS

Agent	Methane		Propane	
	Agent Vol. Fraction at Blowoff	$aT = 2V_o R$ (s ⁻¹)	Agent Vol. Fraction at Blowoff	$aT = 2V_o R$ (s ⁻¹)
Carbon Dioxide	0.240 ± 0.003* (n = 4)	39 ± 1	0.257 ± 0.008 (n = 3)	45 ± 2
Nitrogen	0.332 ± 0.004 (n = 5)	52	0.390 ± 0.004 (n = 5)	56
Helium	0.207 ± 0.003 (n = 5)	94	0.224 ± 0.003 (n = 5)	95* 1
Argon	0.468 ± 0.002 (n = 4)	44* 1	0.550 ± 0.005 (n = 5)	44

*Mean ± standard deviation

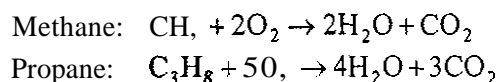
No. of samples = n

The table also lists values for the global strain rate, a_T , and its definition, which is often used as a characteristic strain rate for Tsuji-type flames. V_O refers to the nominal velocity of the oxidizer and R is the radius for the cylindrical burner. Note that for He the values for a_T are almost a factor of two higher than those obtained using N₂, CO₂, and Ar. Recall that the blower in the DLAFFS is set to operate at the lowest possible revolution rate. Even though the blower speed is nominally constant, when the density downstream of the blower is significantly reduced, it generates a higher volume flow rate. Due to the higher value of a_T for He, it is to be expected that the observed extinction concentration will be somewhat lower than would be measured for a purely buoyancy-dominated flame.

Even though the Santoro burner is a fairly simple burner configuration, quite complex flame behaviors are observed during extinguishment experiments using thermal agents. As discussed in an earlier HOTWC paper, when a thermal agent was added to the oxidizer stream surrounding a methane flame, an agent volume fraction was reached for which a significant up and down motion of the flame base developed [3]. The methane and oxidizer flow velocities were both nominally 7 cm/s. It was shown that the results were relatively insensitive to the flow velocities over a substantial range [3]. Similar instabilities have been reported in the literature for diffusion flames near extinguishment [17, 18]. When N₂, Ar, and CO₂ were used as diluents, additional small increases in agent volume fraction resulted in flame lift off. For N₂, the flame continued to move away from the fuel tube with increasing N₂ volume fraction and assumed a shape typical of a triple flame [19, 20, 21, 22]. Eventually a N₂ volume fraction was reached where the flame simply floated away, i.e., blew off. When CO₂ and Ar were used as diluents, the lifted flame at first moved downstream with increasing volume fraction, but then stabilized at a height well above the fuel exit. This behavior was attributed to the entrainment of air from regions outside of the coflow reaching the burning regions and stabilizing the flame.

The methane flame behavior was somewhat different when He was used as the diluent. The flame developed an initial instability similar to that observed with the other agents. However, after a few fluctuations, the flame immediately extinguished. Since the only common behavior for all four agents was the onset of the instability, its occurrence was adopted as the characteristic behavior for methane flame extinguishment. As will be seen shortly, the agent concentrations necessary to induce flame lift-off and blow off were only slightly higher than those necessary to induce the flame fluctuations.

When the baseline fuel and oxidizer volume flow rates for the methane case were used for propane, the resulting flame length was much greater than observed for methane. This observation is easily understood by considering the flame stoichiometries for the two fuels,



which indicate that combustion of a given volume of propane requires 2.5 times more oxidizer than the same volume of methane. Since the entrainment rate of oxidizer into laminar diffusion flames is roughly independent of the fuel flow rate, a longer flame length is required for the complete combustion of propane. To have roughly comparable flame lengths for the two fuels, the nominal fuel velocity for propane was reduced to 40% of that used for methane (i.e., a nominal velocity of 2.8 cm/s, volume flow rate of 0.171 SLM). With the oxidizer nominal velocity maintained at 7 cm/s, the propane flame length was comparable to that for the baseline methane flame. These flow velocities were adopted as the baseline conditions for the propane flame.

Similar to the baseline methane flame, the propane flame was attached to the fuel tube prior to the addition of N₂ to the coflow. As the N₂ volume flow rate was increased, a blue region at the base of the flame occupied a larger volume, as was also observed for the methane flame. However, unlike the methane flame, the base of the propane flame broadened substantially as it moved away from the burner tube with increasing N₂ addition. When the N₂ volume fraction reached 26.5%, the flame base detached from the

fuel tube and stabilized approximately 5 mm downstream. By the time the N_2 volume fraction was increased to 27.4%, the flame base was approximately 10 mm above the fuel tube, and began to oscillate with a combined vertical and radial motion. Further downstream the flame formed a necked-down region. With only an additional increase of 0.1% N_2 volume fraction, the flame suddenly extinguished by collapsing at the necked-down region. A sketch of the nearly extinguished flame is shown in Figure 1. The extinguishment of the propane flame following lift off stands in sharp contrast to the methane flame, which formed a stable lifted flame following detachment from the burner.

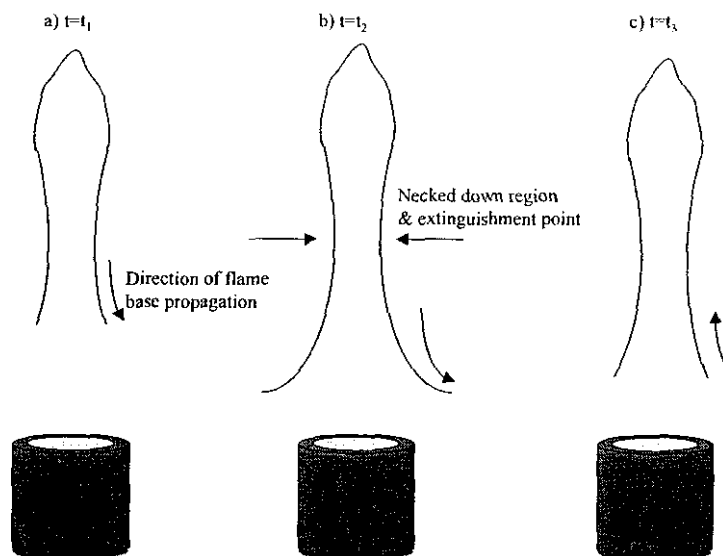


Figure 1. A sketch of the time evolution of a nearly extinguished propane flame is shown for the case of N_2 dilution of the co-flow air, $t_1 < t_2 < t_3$.

Unlike the baseline methane flame, the general response of the propane flame to dilution of the coflow air by CO_2 , Ar , and He was the same as described above for N_2 dilution. Since an easily characterized extinguishment was observed for each agent using propane, this event was designated as the characteristic flame extinguishment behavior.

The results of extinguishment studies for baseline methane and propane diffusion flames on a Santoro burner are summarized (Table 2). Agent volume fractions corresponding to the onset of flame fluctuation and flame extinguishment (if observed) are included. Uncertainties represent the standard deviation, s , for six repeats of each measurement. Blowout was not observed for the methane flame with CO_2 and Ar . The volume fraction of agent required to induce oscillation is taken as the characteristic extinguishing concentration for the methane flame, while the actual concentration at flame extinguishment is used for the propane flame. Note that the small differences observed between the two values for a given agent and fuel indicate that only small uncertainties are introduced by the use of the different definitions for extinguishment.

DETAILED CHEMICAL-KINETIC MODELING RESULTS

The same approach, based on detailed chemical-kinetic modeling of opposed-jet diffusion flames, was used to estimate thermal agent extinguishing volume fractions for propane flames as was used previously for methane flames [2, 9]. Figure 2 shows plots of maximum flame temperature versus fuel and oxidizer velocity for a series of propane flames burning in air diluted with N_2 . To determine an appropriate maximum flame temperature for the extinguishing condition, it is necessary to compare the calculations with an experimental extinguishing volume fraction. As discussed previously with regard to methane flames,

TABLE 2. MEASURED AGENT VOLUME FRACTIONS AT ONSET OF FLAME-BASE INSTABILITY (OSC) AND FOR FLAME EXTINGUISHMENT (EXT) OF BASELINE METHANE AND PROPANE DIFFUSION COFLOW FLAMES.

	Methane		Propane	
	osc	Ext	Osc	Ext
Carbon Dioxide	0.120 (s=±0.001)	-	0.169 (s=±0.004)	0.176 (s=±0.001)
Nitrogen	0.202 (s=±0.002)	0.218 (s=±0.002)	0.276 (s=±0.005)	0.280 (s=±0.008)
Helium	0.200 (s=±0.001)	0.200 (s=±0.001)	0.271 (s=±0.003)	0.273 (s=±0.002)
Argon	0.290 (s=±0.001)		0.356 (s=±0.002)	0.368 (s=±0.003)

s = Standard deviation; No. of samples = 6

extinguishing volume fractions are found to depend on the type of diffusion-flame burner employed, with the opposed-flow hemispherical and cylindrical porous burners requiring the highest values. The measurement reported by Simmons and Wolfhard was used to define the appropriate extinguishing volume fraction, 39.5%, for N₂ [23]. This value can be compared with the value used for methane, 33% [2, 9]. Apparently, the propane flame is harder to extinguish. From Figure 2 it can be seen that the maximum flame temperature at extinction for the propane flame burning in air diluted with 39.5% N₂ is 3 1433 K. This is more than 100 K lower than the corresponding methane value of 1550 K.

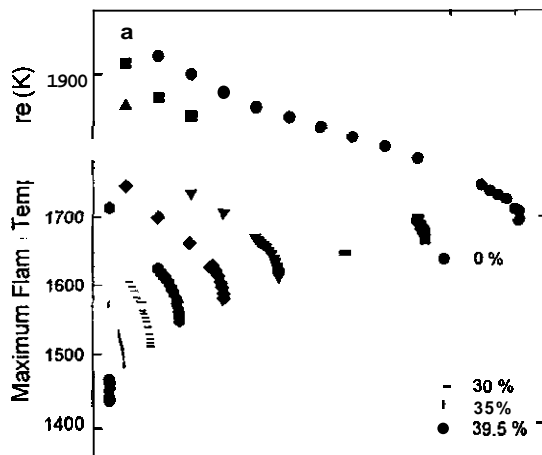


Figure 2. Maximum calculated flame temperatures are plotted against propane and oxidizer exit velocity magnitudes for a series of opposed-jet diffusion flames burning in air diluted with N₂.

Extinguishing volume fractions for propane flames burning in air diluted with CO₂, Ar, and He were estimated by determining the concentrations required to induce extinction with a maximum flame temperature of 1433 K. The results are summarized in Table 3 along with the corresponding values for methane reported previously [2, 9].

TABLE 3. CALCULATED EXTINGUISHING VOLUME FRACTIONS FOR METHANE AND PROPANE OPPOSED-FLOW DIFFUSION FLAMES FOR INDICATED THERMAL AGENTS

Thermal Agent	Methane Flame	Propane Flame
Nitrogen	33%	39.5%
Carbon Dioxide	22%	28.4%
Argon	43%	50.2%
Helium	33%	39.0%

COMPARISON OF EXPERIMENTAL AND MODELING RESULTS

It is interesting to compare various experimental measurements of extinguishing concentrations for diffusion flames with predictions based on the detailed chemical-kinetic modeling. Figure 3 includes experimental results from the current work (solid symbols) and literature results for various types of methane diffusion flames* [23, 24, 25, 26] plotted against reference volume fractions based on the modeling results for methane flames. The symbols represent different types of diffusion flames as follow: circle, porous burner in counterflow; square, opposed jet; and triangle, coflow. The OPPDIF results are indicated by pluses and are connected with a straight line. Note that the results for He have been offset by 0.01 volume fraction for clarity.

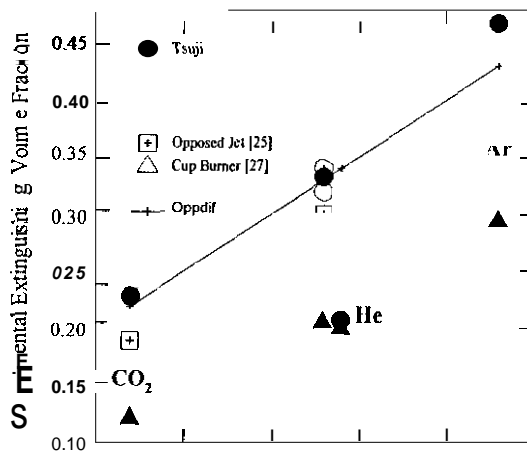


Figure 3. Experimental values of extinguishing volume fractions for N₂, CO₂, Ar, and He are plotted against the corresponding OPPDIF predictions for a number of methane-air diffusion flames. The reference volume fraction for helium is offset by +0.01.

* Hamins, A., Building and Fire Research Laboratory, NIST, personal communication, 2001.

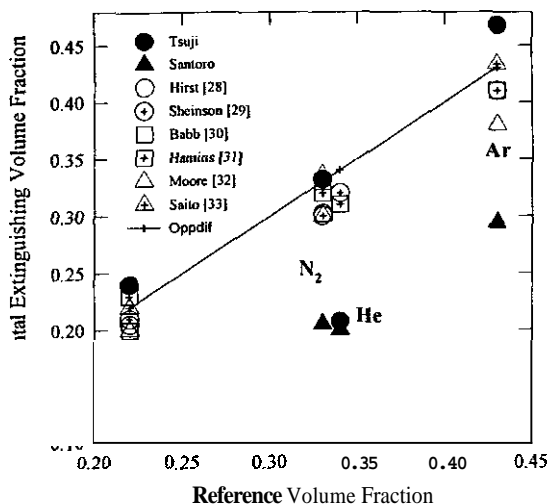
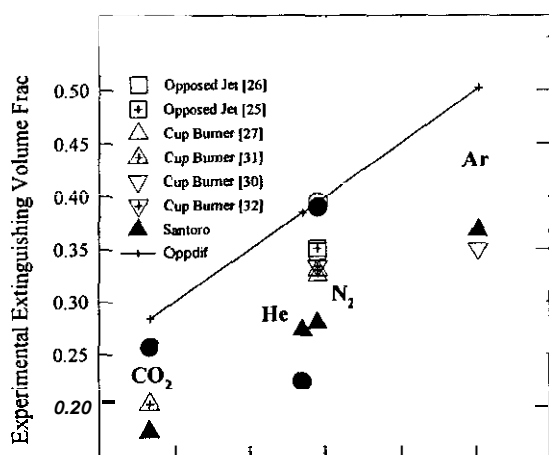


Figure 4. Experimental measurements of thermal agent extinguishing volume fractions for methane flames are plotted against reference volume fractions and compared with heptane cup-burner results.



triangle, coflow. The OPPDIF results are indicated by pluses and are connected with a straight line. Note that unlike the results for methane shown in Figure 3, the values for He have not been offset.

It can be seen that the N_2 extinguishing volume fraction recorded during the current experiment using the Tsuji burner is in very good agreement with the hemispherical porous burner result of Simmons and Wolfhard [23]. The Tsuji burner results for CO_2 and Ar fall close to the predictions based on the detailed chemical-kinetic modeling. As was also found for the methane results, the measured extinguishing volume fraction for He is much less than predicted and appears to be unreasonably low. Again, this is probably due to the high strain rates present during the DLAFSS experiments for this agent.

The results for the Santoro burner once again fall well below those for the Tsuji burner. As was also true for the methane flames, the extinguishing volume fractions for the Santoro burner lie on a straight line when plotted in terms of the reference volume fraction, i.e., the predicted extinguishing volume fractions based on the detailed chemical-kinetic modeling. Significantly, both the experiment and model predict that a slightly lower volume fraction of He is needed for flame extinguishment than is required for N_2 .

The required extinguishing N_2 concentrations for the various types of diffusion flame burners included in Figure 5 depend on burner type in the same way as found for the methane flames. The highest concentra-

tions are required by the porous burners in counterflows, followed by the opposed-jet flames, and finally the coflow flames. Once again, it is found that the Santoro burner is much easier to extinguish than the corresponding cup-burner flame. Literature values for the other agents are limited and do not provide additional insight into the extinguishment behavior.

Comparison of Figures 3 and 5 indicates that higher concentrations of the thermal agents are required to extinguish propane flames than required for methane flames. This can be seen more clearly in Figure 6 where the measured extinguishing volume fractions for the Tsuji and Santoro burners for the two fuels are directly compared. The data are plotted as functions of the predicted extinguishing concentrations for the methane flame taken from the detailed chemical-kinetic modeling results. The reference volume fraction for He has been offset by 0.01 from its true value. Once again excluding the DLAFFS results for He, the results shown in the figure confirm the two major trends identified during the current work. Higher concentrations of a given thermal agent are required to extinguish propane flames than are required to extinguish methane flames, and considerably less of an agent is needed to extinguish a flame burning on the Santoro burner than for the Tsuji burner used in the DLAFFS.

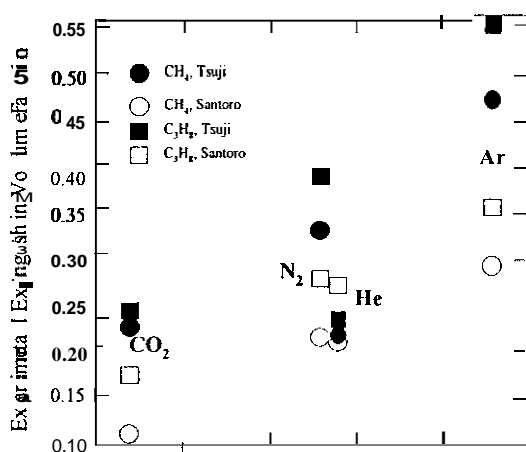


Figure 6. Extinguishing volume fractions for N₂, CO₂, Ar, and He for methane and propane flames burning on the Tsuji and Santoro burners are plotted as a function of predicted concentrations for extinguishment of methane flames based on detailed chemical-kinetic modeling. The reference volume fraction for He is offset by 0.01

DISCUSSION

The experimental measurements reported here show clearly that higher concentrations of thermal agent are required to extinguish propane flames than methane flames. This observation is consistent with the discussion of Macek who has provided an enlightening discussion of flammability limits for straight-chain hydrocarbons [33]. This author used the calculated adiabatic flame temperatures for mixtures of fuel and air with N₂ added to a level corresponding to the lower oxygen index (LOI) as a measure of flammability for fuels having 1 to 10 carbons. The LOI is defined as the minimum oxygen concentration in N₂-diluted air that will support diffusion-flame combustion. The experimental data used for the LOI were taken from Simmons and Wolfhard [23]. This analysis indicated that methane flames are considerably less flammable than propane flames. Interestingly, the analysis also indicated that methane and heptane flames have comparable flammability, which is consistent with the results shown in Figure 4.

The greater stability of propane flames is reflected by the lower maximum flame temperature at extinguishment calculated for this fuel (1433 K) as compared to that for methane (1550 K). Macek calculated adiabatic flame temperatures for LOI conditions of 1780 K and 1720 K for methane and propane, respectively. These values are considerably higher than the maximum calculated diffusion flame temperatures, reflecting the effects of heat losses from the combustion zone of the diffusion flame. Even so, it appears as if both measures provide an indication of the relative flame stability for the two fuels.

The experimental results reported here as well as literature results indicate that varying volume fractions of thermal agents are required to extinguish different types of diffusion flame burners with the general order being porous burner in counterflow flame > opposed-jet flame > coflow flames. It is also clear that significantly lower concentrations are required for the Santoro burner than for a cup burner, even though both use coflow flames, and the fuel and oxidizer velocities are comparable. For both types of burner the results have been shown to be relatively insensitive to flow velocities [3, 25]. The reasons for these differences in extinguishing behavior are currently unknown, but it should be noted that previous work has indicated that the amount of an agent required to extinguish cup-burner flames decreases with the diameter of the fuel cup [34, 35]. The fuel tube diameter of the Santoro burner is less than half of that for a standard cup burner.

The opposed-flow diffusion flame calculations along with the assumption of a well-defined maximum flame temperature at extinguishment provide predictions for thermal agent extinguishing volume fractions that agree very well with the observed experimental behaviors. This is particularly true with regard to the effectiveness of He. Traditional analyses for predicting the effectiveness of a thermal agent have used such properties as adiabatic flame temperature or heat capacity as dependent variables. Since He and Ar are both monatomic ideal gases, such approaches predict that the effectiveness of these two thermal agents are the same. On the other hand, the use of the diffusion flame calculations captures the true behavior in which the extinguishing ability of He is significantly greater than that for Ar and actually compares very favorably with that for N₂. Other workers have noted the enhanced effectiveness of He and have attributed it to the high thermal diffusivity of He [28, 36, 37]. This effect is automatically incorporated into the opposed-jet diffusion flame calculations.

Even though there is a substantial dependence on fuel and burner type for the amount of an agent required for flame extinguishment, both the experiments and calculations indicate that the relative effectiveness of a thermal agent has little or no dependence on these properties. Similar conclusions concerning fuel effects have been discussed recently in the literature for wider classes of fire extinguishing agents [38, 39]. The current findings suggest that the relative effectiveness of various firefighting agents can be assessed using generic diffusion flame burners, but that care should be exercised when determining extinguishing concentrations for design purposes.

ACKNOWLEDGMENTS

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