MECHANISMS OF COMBUSTION LIMITS IN PREMIXED GAS FLAMES AT MICROGRAVITY
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SUMMARY

A three-year experimental and theoretical research program on the mechanisms of combustion limits of premixed gas flames at microgravity was conducted. Progress during this program is identified and avenues for future studies are discussed.

Introduction

The work of the Principal Investigator (PI) has encompassed two major topics related to the experimental and theoretical study of combustion limits in premixed flames at microgravity. These topics are as follows:

- Flammability limits and flame extinction processes at $\mu g$
- Stability limits of freely propagating flames at $\mu g$

Progress on these specific topics will be discussed in following sections.

The work supported by NASA has resulted in nine reviewed publications or submissions to reviewed journals thus far:


The following papers have been or will be presented at technical conferences (most will be submitted for publication in reviewed journals):

Ronney, P. D., "A Review of Flame Ignition," Spring Technical Meeting, Combustion Institute, Western States Section, April 30 – May 2, 1990, Banff, Alberta, Canada

Whaling, K. N., Abbud-Madrid, A., Ronney, P. D., "Effect of Buoyancy on the Structure and Stability of Flames with Low Lewis Number," Fall technical Meeting, Combustion Institute, Western States Section, October 15-16, 1990, La Jolla, CA (to be presented.)


Ronney, P. D., Padmanabhan, K. T., Hardy, T. E., "Effect of Catalyst Type on the Burning Rate of Carbon Monoxide Flames," Fall Technical Meeting, Combustion Institute, Western States Section, October 17-18, 1988, Dana Point, California.


The work of the following graduate students have been supported in whole or in part by NASA funds:

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SUMMARY OF ACCOMPLISHMENTS AND FUTURE DIRECTIONS

Flammability limits and flame extinction processes at μg

Experimental

In previous studies of freely propagating premixed gas flames at microgravity, it has been found that flames in mixtures having Lewis number (Le) slightly less than a critical value near unity exhibit a flammability limit and an unstable mode of flame propagation in sub-limit mixtures, termed Self Extinguishing Flames or SEFs, unlike those found at normal gravity. SEFs are unstable spherically expanding flames in mixtures which are apparently outside the planar flammability limit; the flammability limit appears to be caused by radiat heat losses from the burned gases. SEFs are characterized by an energy release before extinguishment which is often orders of magnitude greater than the ignition source energy, a flame kernel radius which increases in proportion to the square root of the time lapse from ignition, and sudden extinguishment at a finite propagation rate.

In early work performed under this grant (Combustion Symposium paper), it was found that SEFs are probably a result of the greater resistance of curved flames fronts (as opposed to planar fronts) in mixtures with Le < 1 to extinguishment by heat losses; as the flame front expands beyond a critical radius of curvature, sudden extinguishment may occur. Fuel chemistry was previously (CST paper) found to be a minor factor in SEFs. These conclusions were obtained in the following way. Several models which relate flammability limits to volumetrically distributed heat losses such as gas radiation are available. Probably the most important prediction of these models which is easily tested experimentally is the burning velocity at the flammability limit.
(Su\_lim). Similar results in slightly different forms are obtained from these models but all give the following expression for Su\_lim:

\[
Su\_lim = \frac{1}{\rho_u C_p T_b} \left( \frac{CLb k_b E_a}{R_g} \right)^{1/2}
\]

where \( \rho_u \) is the unburned gas density, \( C_p \) is the specific heat at constant pressure, \( T_b \) is the adiabatic flame temperature, \( L_b \) is the heat loss per unit volume per unit time from the burned gases, \( k_b \) is the thermal conductivity of the burned gases, \( E_a \) represents the activation energy, \( R_g \) is the gas constant and \( C \) is a constant of order unity which reflects the effects of temperature on heat loss. Williams shows that \( C=1.2 \) is appropriate for limits caused by radiant heat loss. Another prediction of the theories is a "response curve" of burning velocity (Su) versus fuel concentration or adiabatic flame temperature. Of special importance is the prediction that the slope of this curve approaches an infinite value at the flammability limit.

These early tests concentrated on the use of different fuels (mainly CH₄, H₂, C₃H₈ and NH₃) in air, i.e. with nitrogen as the diluent. In more recent work (Combustion Symposium Paper) the use of different diluents with the same fuel–oxidizer combination was employed to allow a more versatile way to change Lewis number, radiation characteristics and transport properties, all of which are known to affect flame behavior near the flammability limit region. As a comparison with the behavior predicted by the theories relating flammability limits to radiative heat loss, Figures 1 and 2 show the experimentally observed relationship between burning velocity (Su) and mixture dilution. The previously mentioned behavior of the slope of this curve seems to agree with the experimental findings as the mixture dilution approaches the flammability limit. A comparison of calculated and observed limit burning velocities of all mixtures considered is presented in Table 1. Table 1 shows good agreement between experimentally observed and theoretically calculated burning velocities at the limit in the case of CH₄–O₂–CO₂, H₂–O₂–He and CO–O₂. This results were to be expected from mixtures with Lewis number less than unity, but high enough to avoid flame front wrinkling and cell formation. Nevertheless, wrinkling and splitting of the flame front was observed in the CH₄–O₂–CO₂ flames near the flammability limit; two-lobed flame fronts were observed first (63.15% of CO₂ dilution) followed by a three-lobe (63.20%) and a six-lobe flame front (63.25%) as dilution was increased. Below this dilution, a very well defined SEF region was observed (63.30%-63.50%) below which no ignition could be observed.

Lean C₃H₈–O₂–CO₂ mixtures exhibit considerably higher values of Su\_lim than those predicted by the models based on extinction caused exclusively by radiant heat losses. It is believed that in this case an ignition limit rather than a flammability limit is observed. This is because a high Le and a high thermal diffusivity leads to a very high minimum ignition energy. This high minimum ignition energy in turn causes an ignition limit to be reached before the radiant flammability limits are reached, thus causing higher values in limit burning velocity. As a consequence no SEF behavior is observed.

Experiments were also conducted with rich C₃H₈–O₂–CO₂ but no clearly defined limit was observed due to the unusual behavior of these mixtures; the simultaneous appearance of small sooting cells, medium sized slowly propagating "flame balls" and fast elongated flamelets made it impractical to assign a burning velocity to such a flame. Even more surprisingly, as these mixtures were
diluted, a flammability limit was reached (as expected), but if the mixture was diluted still further, it became flammable again! (Table 2). No clear explanation has been found for this behavior, which we have termed "dilution-enhanced flammability." Further dilution caused permanent non-flammability.

Rich H₂-O₂-CΟ₂ flames also exhibited unexpected behavior. After ignition a relatively fast spherical flame front traverses the entire chamber and it disappears upon reaching the vessel walls. It is then immediately followed by another flame born on the platinum thermocouples, traveling with a slightly higher velocity and generating a higher temperature than the first one. It also disappears at the vessel walls well before the end of the drop. The dynamic response of the two flame fronts, as well as their pressure and temperature records, are shown in Fig. 3. This behavior was found only very near at the flammability limit. Single flames were present in slightly less diluted mixtures and no ignition was possible in slightly more diluted mixtures. Gas sampling extracted during the free-fall indicate that in the case of double flames, the first flame consumes only two-thirds of the oxygen and produces equilibrium CO (due to the water-gas shift), then the second flame consumes most of the remaining O₂ (see Table 3). These pressure and temperature records are consistent with the gas-sampling results in terms of the observed heat release.

No explanation has yet been found for these "double flames" but it is clearly related to some (as yet unknown) water-gas shift effect. It is also suspected that it may be related to the presence of two reactants with different Lewis number. This effect is described more fully in a manuscript in preparation in conjunction with G.I. Sivashinsky. Depending upon the flame curvature and the heat loss from the combustion products, either reactant can be the abundant one at the flame front. The other reactant will "leak" through the flame front and creates a surplus on the burned gas side. If O₂ (the globally deficient reactant) were to leak through the flame front, after a pressure and temperature rise due to the expansion of the flame kernel in the constant volume vessel another flame could be formed.

In future work, we intend to examine radiation effects by adding small quantities of inert, radiant particles to the mixture. If the amount of particles added is sufficiently small, it will not affect the heat capacity of the mixture significantly, and thus these particles will only affect the radiative (and not chemical, thermodynamic, or transport) properties. This is more systematic our previous technique of using dilution to change the impact of heat loss parameter (primarily because it reduces burning velocity), plus it enables the study of premixed flames in the optically thick regime, a regime that has not received systematic study to date. Additionally, we will study the effects of turbulence on flammability limits at μg. At one-g such experiments are very difficult to perform because buoyancy-induced turbulence (that is the ratio of the RMS velocity fluctuation to S_l) becomes very high. Thus, a systematic study of the effects of turbulence on limits necessitates the use of μg to eliminate buoyancy-induced turbulence. In particular, we wish to determine if turbulence alone can extinguish these flames in the absence of losses.

Theoretical work
It has been found that SEFs occur only in mixtures with Lewis number less than a critical value near unity. Theory shows that radiation extinction limits for planar flames should be independent of Lewis number (Le). It is well known that Lewis number effects in premixed flames are usually a result of the influences of curvature, hence curvature effects are likely to be of importance in these flames. As a result, a theoretical study of the dynamics of nonadiabatic spherical flame fronts was initiated in collaboration with G. I. Sivashinsky. A simple non-dimensional evolution equation for the flame front was derived (SIAM paper):

$$\frac{dS}{dR} + S^2 \ln(S^2) = 2S/R - Q$$

where $S$ is the flame speed, $R$ is the flame radius (which is positive for $Le < 1$ and negative for $Le > 1$), and $Q$ is the heat loss. The previous steady planar theories are identical to this except that the unsteady term ($dS/dR$) and the curvature term $(2S/R)$ are missing. This simple equation shows very similar trends to the experimental results for $Le < 1$, as shown in Figure 4.

Numerical results

Current numerical models of premixed gas flame propagation without external losses do not predict flammability limits; instead they predict that even very dilute mixtures should be flammable although they may burn very slowly. We have employed time-dependent one-dimensional flame propagation models with detailed kinetic and transport processes to which we are adding other processes, e.g. heat losses, in an attempt to explain the results of the aforementioned earth-gravity and low-gravity experiments. This work is an extension of the theoretical work described above to more complex chemical and transport schemes. Computations have been run on CYBER-205, CRAY-XMP, and CONVEX equipment at the NSF-John von Neumann Computing Center, Naval Research Laboratory, and Princeton, respectively.

While "complete" chemical and transport models for methane-air combustion is prohibitively expensive in terms of CPU time, we have identified a suitable reduced reaction mechanism which is appropriate for near-limit and transient conditions. Using this reduced mechanism, it was found, as with the theoretical work, that the most important effects are the interactions of heat loss from spectral radiation from the gases to the environment, the curvature of the flame front, the divergence of the flow field in which the flame propagates, and the Lewis number of the mixture which represents the relative rates of chemical and thermal enthalpy in the flame front (CST paper.) Typical results are shown in Figures 5 and 6. It is significant that SEFs are predicted and the strong effect of ignition source strength, which could not be predicted by the analytical work, is seen. This is because of the scaling assumptions inherent in the analytical results do not cover the range of scales necessary to model how the ignition effects which occur on small scales manifest themselves on the larger scales where SEFs extinguish.

Sub-topic: Technique for the Systematic Study of the Lewis Number Effects in Premixed and Diffusion Flames
As discussed above, Lewis number effects are prevalent in many types of premixed and diffusion flames, particularly when flow or flame front curvature is present. These effects may be studied experimentally by comparison of flames in fuels having different molecular weights, however this technique has limited value because changing fuel type also changes many other flame properties in ways which cannot be assessed a priori. The use of CO-O₂-N₂ mixtures with small concentrations of hydrogen-bearing catalysts such as H₂, CH₄, C₂H₆, and C₃H₈ may enable experimental study of the effects of preferential diffusion without changes in flame chemistry, temperature, or transport properties. This is possible because hydrogen-free CO-O₂ mixtures burn very poorly if at all but a very small concentration of a hydrogen-bearing substance greatly accelerates the reaction.

To exploit such a technique the system must first be "calibrated" by measuring the burning velocities of premixed planar flames with these catalysts. This task has been accomplished; sample results are shown in Figure 7 (Western States Combustion Institute paper). It can be seen that different catalysts have nearly the same influence on the burning rate of a planar flame when normalized by the amount of H atoms in the system. We believe this is because the slowest reaction in the system is

\[ \text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H} \]

which is not a function of the hydrogen-bearing substance. In other words, the above reaction is much slower than the reactions which break down the hydrogen-bearing molecule into OH, and thus the form in which hydrogen is introduced into the system does not affect the overall reaction rate or burning velocity. We are initiating a numerical modelling effort, in parallel with the other modelling described above, to determine if this conjecture can be validated.

The effects of curvature can now be tested by comparing the properties of flames in different mixtures having the same planar burning velocities but varying catalyst diffusivities. This will enable us to study the effects of preferential diffusion in curved premixed flames such as near-limit upward propagating flames in tubes, flames in turbulent flows, and spherical flame kernels emanating from point sources of ignition, and diffusion flames such as gaseous fuel jets and counterflow diffusion flames.

**Stability limits of freely propagating flames at \( \mu \)**

**Experimental**

We have found in previous work that the tendency of flames with low Lewis numbers, e.g. hydrogen-air mixtures, to break up into cells affects the near-limit behavior dramatically (paper to appear in Combustion and Flame.) Very slowly burning mixtures may be observed in mixtures with low Le, and in some cases flame propagation ceases entirely without flame extinction occurring. In these cases stable, stationary spherical flames ("flame balls") seem to exist. Stationary spherical flames have been predicted previously but they are predicted to be unstable. In recent experimental work, it has been found that the same behavior found with nitrogen-diluted mixtures also occurs with CO₂-and
SF$_6$-diluted mixtures. In the latter case, the Lewis number is about 0.1, and the cellular structures observed are quite striking although qualitatively not different from those observed in nitrogen. Very significantly, we have also studied CH$_4$-O$_2$-SF$_6$ mixtures (Le = 0.3) and found very similar results. This indicates that these trends are strictly a result of low Lewis number and not an artifact of H$_2$-O$_2$ chemistry.

For many of the experiments performed in the course of this work, 2.2 second of low-gravity is adequate. However, some of the test, particularly the studies of "flame balls," this time is woefully inadequate. This is because the flame balls are found to evolve on a very slow time scale, much slower than that of "ordinary" flames. This is also predicted by the analyses of flame balls discussed above. Hence, we had been unable to determine if these low Lewis number structures are indeed stationary or are slowly evolving to some other structure. An improved experimental apparatus, based on an IBM-PC compatible single-board computer and data acquisition system, was built and flown on the KC-135A aircraft. Experiments showed that, indeed, the structures appear to be stable for long periods of time (= 15 sec.) Two other advantages accrued due to the use of this facility. First, we were able to use an infrared-sensitive camera which enabled us to study flames which are not doped with a "coloring" agent. The dopant was necessary in prior work because hydrogen flames are nearly transparent in the visible range. The advantage of eliminating the coloring agent is that the chemical and radiant transport properties of the coloring agents are not well known and thus make detailed numerical modelling very difficult. Secondly, the "g-jitter" in the aircraft caused yet another new phenomena to appear: "flame cylinders" resulting from moving "flame balls." While theory shows that steady "flame cylinders" are not possible (unlike flame balls, which are theoretically possible), the structures can persist for relatively long periods of time (steady solutions are only logarithmically divergent). What is observed experimentally is that flame cylinders persist for several seconds then break up in a small fraction of a second into flame balls.

In future work, we intend to study flame balls in undoped H$_2$-air mixtures because of their impact on fire safety considerations. We did not study these previously because we had no suitable way to image them. Also, we hope to study low Lewis number flame structures in particle-laden mixtures to study the effects of optical thickness, as discussed above.

Theoretical

We are interested in determining the stabilizing mechanism for these flames. It was speculated that heat losses cause the apparent stability and theoretical work performed in conjunction with Prof. John Buckmaster (Univ. of Illinois) and Dr. G. Joulin (Univ. of Poitiers, France) has confirmed this speculation, at least for the case of a high activation energy reaction such as that of lean H$_2$-air flames. Model predictions are shown in Figures 8 and 9 (papers to appear in Combustion and Flame.) It is found that in the presence of heat losses, two steady solutions are possible but linear stability analysis shows that the lower branch of solutions is unstable (figure 8). In the presence of far-field losses (unburned gas), oscillatory solutions are also possible. Our model also predicts that for low heat losses, the flame balls are unstable to
three-dimensional disturbances, i.e. they break up into cells (figure 9). This is precisely what is observed in experiments. It is particularly noteworthy that mixtures with low Lewis numbers may exhibit much wider flammability limits in these curved geometries than is possible in planar flame fronts (figure 9).

In future work, we intend to compare measured flame balls diameters, from mixtures without coloring agents, with detailed numerical computations for the $\text{H}_2-\text{O}_2$ system to see if quantitative agreement is obtained. Such a task can be performed with FLAMEID as described above. Additionally, a stability analysis of "flame cylinders" has been initiated to determine if there is a "most unstable" wavelength which could result in the sudden breakup of the flame cylinders into flame balls.
Figure 1
Flame radius vs. time for near-limit and sub-limit spherically expanding flames in mixtures with $Le<1$
Figure 2

Burning velocity vs. dilution for near-limit flames
Figure 3

Dynamic response, temperature, and pressure characteristics of "double flames"
Figure 4
Theoretical dynamic response of spherically expanding flames as a function of heat loss
Figure 5. Computed dynamic response of spherical flames in a near-limit methane-air mixture ($\phi = 0.519$) as a function of ignition energy.
Figure 6. Computed extinguishment radii of near-limit spherical flames in methane-air mixtures at \( \mu_g \).
Figure 7. Burning velocities of CO-O$_2$-N$_2$-catalyst mixtures.
Figure 8
Size and stability boundaries for non-adiabatic flame balls
Figure 9

Stability boundaries of non-adiabatic flame balls vs. a function of Lewis number
<table>
<thead>
<tr>
<th>Mixture</th>
<th>Equiv. ratio (φ)</th>
<th>% Diluent at the limit</th>
<th>Pressure (atm)</th>
<th>( S_u,\text{lim} ) (exp) (cm/sec)</th>
<th>( S_u,\text{lim} ) (theo) (cm/sec)</th>
<th>( E_a ) (kcal/mole)</th>
<th>Estimated Lewis Number</th>
<th>( T_{ad} ) (K)</th>
<th>( T_{max} ) (K) (exp)</th>
<th>( T_{max} ) (K) (theory)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH(_4)-(\text{O}_2)</td>
<td>0.50</td>
<td>63.20% CO(_2)</td>
<td>0.40</td>
<td>2.11</td>
<td>2.91</td>
<td>32.1</td>
<td>0.74</td>
<td>1544</td>
<td>1331</td>
<td>1396</td>
</tr>
<tr>
<td>CH(_4)-(\text{O}_2)</td>
<td>0.50</td>
<td>63.00% CO(_2)</td>
<td>1.00</td>
<td>1.62</td>
<td>1.97</td>
<td>48.9</td>
<td>0.74</td>
<td>1551</td>
<td>1334</td>
<td>1453</td>
</tr>
<tr>
<td>CH(_4)-(\text{O}_2)</td>
<td>0.50</td>
<td>62.80% CO(_2)</td>
<td>2.20</td>
<td>1.25</td>
<td>1.34</td>
<td>52.5</td>
<td>0.74</td>
<td>1558</td>
<td>1348</td>
<td>1466</td>
</tr>
<tr>
<td>(\text{H}_2)-(\text{O}_2)</td>
<td>0.50</td>
<td>83.75% He</td>
<td>1.00</td>
<td>2.93</td>
<td>3.11</td>
<td>41.3</td>
<td>0.97</td>
<td>1177</td>
<td>1143</td>
<td>1110</td>
</tr>
<tr>
<td>(\text{H}_2)-(\text{O}_2)</td>
<td>0.50</td>
<td>82.50% He</td>
<td>2.00</td>
<td>2.38</td>
<td>2.19</td>
<td>52.8</td>
<td>0.97</td>
<td>1240</td>
<td>1212</td>
<td>1182</td>
</tr>
<tr>
<td>CO-(\text{O}_2)</td>
<td>7.16</td>
<td>-</td>
<td>1.00</td>
<td>2.36</td>
<td>1.93</td>
<td>55.8</td>
<td>1.02</td>
<td>1338</td>
<td>1157</td>
<td>1274</td>
</tr>
<tr>
<td>(\text{C}_3\text{H}_8)-(\text{O}_2)</td>
<td>0.50</td>
<td>66.85% CO(_2)</td>
<td>1.00</td>
<td>3.67</td>
<td>2.05</td>
<td>57.2</td>
<td>1.43</td>
<td>1550</td>
<td>1401</td>
<td>1466</td>
</tr>
<tr>
<td>(\text{H}_2)-(\text{O}_2)</td>
<td>2.00</td>
<td>55.00% CO(_2)</td>
<td>1.00</td>
<td>10.5</td>
<td>3.16</td>
<td>52.1</td>
<td>0.83</td>
<td>1279</td>
<td>1164</td>
<td>1216</td>
</tr>
</tbody>
</table>

Table 1. Comparison of experimental and theoretical limit burning velocities and flame temperatures. For mixtures which show a region of asymmetric propagation, the experimental \( S_u,\text{lim} \) is the \( S_u \) of the most dilute mixture which exhibited symmetric propagation. For the \( \text{H}_2\)-\(\text{O}_2\)-CO\(_2\) mixture at 1.0 atm, \( S_u,\text{lim} \) was obtained from the first flame in the double flame mixture.
<table>
<thead>
<tr>
<th>CO₂ dilution</th>
<th>( T_{ad} ) (K)</th>
<th>Observed behavior</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;66.0%</td>
<td>&gt;1460</td>
<td>Normal flames</td>
</tr>
<tr>
<td>66.0%</td>
<td>1460</td>
<td>Normal flame having sooty core surrounded by wrinkled blue flame</td>
</tr>
<tr>
<td>66.4%</td>
<td>1449</td>
<td>Asymmetric flame</td>
</tr>
<tr>
<td>66.8%</td>
<td>1438</td>
<td>SEF-like flame</td>
</tr>
<tr>
<td>68.5%</td>
<td>1390</td>
<td>Initial flame kernel develops into stationary flame balls</td>
</tr>
<tr>
<td>71.0%</td>
<td>1319</td>
<td>Non-ignition</td>
</tr>
<tr>
<td>71.5%</td>
<td>1305</td>
<td>Blue flame ball splits into cells; soot observed during splitting</td>
</tr>
<tr>
<td>72.0%</td>
<td>1290</td>
<td>Two initial cells split into four cells; soot observed during splitting</td>
</tr>
<tr>
<td>73.0%</td>
<td>1261</td>
<td>Regularly dividing cellular structures; soot observed during splitting</td>
</tr>
<tr>
<td>&gt;73.0%</td>
<td>&lt;1261</td>
<td>Non-ignition</td>
</tr>
</tbody>
</table>

Table 2. Observed behavior in rich (\( \phi=2.0 \)) \( \text{C}_3\text{H}_8-\text{O}_2 \) mixtures progressively diluted with CO₂.


<table>
<thead>
<tr>
<th>Mixture</th>
<th>$O_2$ (%)</th>
<th>CO (%)</th>
<th>$CO_2$ (%)</th>
<th>$H_2 + H_2O$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial mixtures</td>
<td>8.9 - 9.0</td>
<td>0.00</td>
<td>54.9 - 55.1</td>
<td>35.7 - 35.9</td>
</tr>
<tr>
<td>Normal Flame (54.9% $CO_2$)</td>
<td>0.8</td>
<td>15.0</td>
<td>44.6</td>
<td>39.6</td>
</tr>
<tr>
<td>Double flame (55.0% $CO_2$)</td>
<td>2.2</td>
<td>13.1</td>
<td>46.3</td>
<td>38.4</td>
</tr>
<tr>
<td>SEF-like flame (55.1% $CO_2$)</td>
<td>2.3</td>
<td>12.7</td>
<td>46.7</td>
<td>38.3</td>
</tr>
<tr>
<td>Adiabatic equilibrium composition (calculated)</td>
<td>&lt;0.01</td>
<td>13.6 - 13.8</td>
<td>46.6 - 46.8</td>
<td>39.5 - 39.7</td>
</tr>
</tbody>
</table>

Table 3. Gas compositions for rich (ϕ=2.0) $H_2$-$O_2$-$CO_2$ mixtures: normal, double and SEF-like flames. Samples for normal and SEF-like flames were taken after flame passage by the probe; double flame samples were taken immediately after the passage of the first flame and before the onset of the second flame. Contributions of $CF_3Br$ to the product gases are neglected.