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1. SUMMARY

A two and one-half year experimental and theoretical research program on the properties of laminar and turbulent premixed gas flames at microgravity was conducted. Progress during this program is identified and avenues for future studies are discussed.

2. RESULTS

2.1 Overview

The work of the Principal Investigator (PI) supported by NASA Grant No. NAG3-1242 has encompassed four topics related to the experimental and theoretical study of combustion limits in premixed flames at microgravity, as discussed in the following sections. These topics include:

- Radiation effects on premixed gas flames
- Flame structure and stability at low Lewis number
- Flame propagation and extinction in cylindrical tubes
- Experimental simulation of combustion processes using autocatalytic chemical reactions

This work has resulted in the following reviewed publications or submissions to reviewed journals thus far:

Haslam, B. D., Ronney, P. D., "Fractal Properties of Propagating Fronts in a Strongly Stirred Fluid," submitted to Physics of Fluids.

Zhu, J. Y., Ronney, P. D., "Simulation of Front Propagation at Large Non-dimensional Flow Disturbance Intensities," submitted to Combustion Science and Technology (1993).

Lozinski, D., Buckmaster, J. D., Ronney, P. D., "Absolute Flammability Limits and Flame Balls in Optically Thick Mixtures," to appear in Combustion and Flame (1994).

Ronney, P. D., "Laser versus Conventional Ignition of Flames," to appear in Optical Engineering (1993).

Abbud-Madrid, A., Ronney, P. D., "Premixed Flame Propagation in an Optically-Thick Gas," to appear in AIAA Journal (1993).

Ronney, P. D., Whaling, K. N., Abbud-Madrid, A., Gatto, J. L., Pisowicz, V. L., "Stationary Premixed Flames in Spherical and Cylindrical Geometries," to appear in AIAA Journal (1993).

Sloane, T. M., Ronney, P. D., "A Comparison of Ignition Phenomena Modeled with Detailed and Simplified Kinetics," Combustion Science and Technology 88, 1-13 (1993).

Buckmaster, J. B., Gessman, R., Ronney, P. D., "The Three-Dimensional Dynamics of Flame Balls," Twenty-Fourth International Symposium on Combustion, Combustion Institute, Pittsburgh, 1992, pp. 53-59.

Shy, S. S., Ronney, P. D., Buckley S. G., Yakhot, V., "Experimental Simulation of Premixed Turbulent Combustion Using Aqueous Autocatalytic Reactions," Twenty-Fourth International Symposium on Combustion, Combustion Institute, Pittsburgh, 1992, pp. 543-551.

Chen, R. H., Mitchell, G. B., Ronney, P. D., "Diffusive-Thermal Instability and Flame Extinction in Non-Premixed Combustion," Twenty-Fourth International Symposium on Combustion, Combustion Institute, Pittsburgh, 1992, pp. 213-221.

Ronney, P. D., Yakhot, V., "Flame Broadening Effects on Premixed Turbulent Flame Speed," Combustion Science and Technology 86, 31-43 (1992).

Buckmaster, J. D., Joulin, G., Ronney, P. D., "Structure and Stability of Non-adiabatic Flame Balls: II. Effects of Far-Field Losses," Combustion and Flame 84, 411-422 (1991).

The following work, not yet published in refereed journals, has been presented:

Wang, Q., Ronney, P. D., "Mechanisms of Flame Propagation Limits in Vertical Tubes," Spring Technical Meeting, Combustion Institute, Eastern/Central States Section, March 15-17, 1993, New Orleans, LA (to be presented).

Buckmaster, J. D., Ronney, P. D., Smooke, M. "Flame Balls: Past, Present, and Future," AIAA Paper No. 93-0712, 1993.

Haslam, B. D. and Ronney, P. D., "Experimental Simulation of Premixed Turbulent Combustion in Flows with a Broad Range of Temporal and Spatial Scales," Fall Technical Meeting, Western States Section, Combustion Institute, Oct. 12-13, 1992, Berkeley, CA.

Buckmaster, J. D., Lee, C. J., Joulin, G., Ronney, P. D., "Modeling of Microgravity Ignition Experiments," in: Recent Advances in Combustion Modeling (B.

Larrouturou, ed.), Series in Advances in Mathematics for Applied Sciences, Vol. 6, pp. 1-18, World Scientific Press, Teaneck, NJ, 1991.

"Effects of Ambient Atmosphere on Flame Spreading and Extinction," Workshop on Spacecraft Fire Safety Risk Analysis Assessment, Oct. 31 - Nov. 1, 1991, Los Angeles, CA (Invited presentation).

"New Premixed Gas Combustion Phenomena," Gordon Research Conference on Gravitational Effects in Physico-Chemical Systems, June 16-21, 1991, Plymouth, NH. (Invited Presentation).

Ronney, P. D., "An Investigator's Suggestions for Effective use of the NASA-JSC Reduced Gravity Program KC-135A Aircraft," in: JSC Reduced Gravity Program User's Guide, NASA JSC-22803, July 1991.

2.2 Narrative descriptions of research results

2.2.1 Radiation effects on premixed gas flames

Prior work by the PI had demonstrated the importance of gas radiation in the propagation and extinguishment of premixed gas flames. However, gas radiation is a weak effect and is only a significant influence for very slowly burning flames. These flames can only be observed at μg due to buoyancy effects at one-g. Furthermore, it is not clear whether radiation is a fundamental limit because in very large systems (say, tens of meters) emitted radiation could be reabsorbed within the gas and thus not lost from the system. In this case the propagation rate could actually increase because radiation would augment the usual transport by conduction of heat from the flame front to the unburned gas.

To test these hypotheses, we examined radiation effects by studying flames at μg in gas mixtures to which small quantities of inert, radiant particles have been added. Solid particles emit and absorb radiation across a broad spectrum, unlike gas molecules which are active only in narrow spectral bands. Thus, appreciably more radiation and absorption can be expected in particle-laden gases. The experiments showed that at small particle loadings, burning rates are reduced, peak pressures in constant-volume combustion are lower and thermal decay rates in the burned gases are increased. This indicates that the significance of radiative loss is enhanced by the addition of particles to the gas. With sufficient seeding, the burning rates are practically the same as those found in particle-free mixtures, the peak pressures are comparable and the thermal decay rate is smaller than particle-free mixtures. All of these observations are consistent with the hypothesis that at sufficiently high particle loadings, radiation is reabsorbed within the combustible medium, and thus may not constitute a fundamental limit in very large systems. In essence, the presence of particles has made the system "larger" in that the ratio of the system size to mean absorption length of radiation is increased.

The aforementioned experiments employed the NASA-Lewis 2.2 second drop tower. μg was necessary because only slow burning mixtures are affected by the relatively weak process of radiation, thus at one-g buoyancy-driven transport dominates the radiative effects of interest. Some tests require additional duration of μg, however. To facilitate this, the PI initiated a cooperative program with Prof. John H. S. Lee of McGill University in Montreal, Canada. Prof. Lee's work, funded by the Canadian Space Agency, involves burning combustible dusts in air in a low-g environment. This apparatus had previously flown 3 campaigns on the KC-135 in Houston and one on the French Caravelle aircraft. This apparatus was suitable for studying inert dusts in combustible atmospheres. A set of low-g experiments was performed on the KC-135 aircraft during the week of March 8, 1993. The effect of inert and combustible particles on the propagation rates and flammability limits of gaseous methane-air mixtures was studied using flight hardware from both the PI's and Prof. Lee's laboratories. It was found that with care, it was possible to produce quite uniform and optically thick dispersions of particles in the combustion chamber. Preliminary results indicate that the addition of particles did not seem to enhance the flammability of the methane-air mixtures as much as anticipated based on theory and the results of the drop tower experiments, which appeared to be due to the lower quality of low-g in the KC135 as compared to the drop tower.

In collaboration with Prof. John D. Buckmaster of the University of Illinois, a model of the effects of reabsorption of emitted radiation on propagating flames and flame balls (described below) was developed. The model demonstrated that when reabsorption of emitted radiation is considered, there are additional branches of flame ball solutions not present without reabsorption. The solutions having smaller radii correspond to conduction-dominated flame balls and the larger solutions correspond to radiation-dominated flame balls. The large, radiation-dominated flame balls are unstable to radial disturbance and thus evolve to propagating planar flames or collapse to smaller flame balls (or to extinguishment, depending on the composition.) All flammability limits are eliminated because in the configuration of the problem, there is no loss mechanism. This is in marked contrast to analyses of flame balls or propagating flames under optically-thin conditions, because in this case radiation is always a loss process. This suggests that optically-thick flame balls could be observed only as transient structures in experiments. These theoretical results were compared with experimental observations obtained in the KC135 aircraft and drop towers. The possibility that the "sudden propagation" seen in SF6-diluted mixtures is a manifestation of optically-thick effects is discussed. These results serve as further motivation for space experiments employing these mixtures. Four of these tests are included in the test matrix for the PI's proposed SOFBALL flight experiments.

2.2.2 Flame structure and stability at low Lewis number

The PI's μg experiments have shown that the tendency of flames in mixtures with low Lewis numbers, e.g. hydrogen-air, to break up into cells affects the near-limit behavior dramatically. 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 had been predicted previously but were predicted to be unstable. Experiments supported by Grant

NAG3-1242 showed that such phenomena seem to occur in all mixtures with sufficiently low Lewis number and near flammability limits, independent of the chemical mechanism. Experiments on the KC-135A aircraft showed that these structures are stable for at least the 20 seconds of low-g available. By comparison with analytical models, it was concluded that radiation from the combustion products, along with diffusive-thermal effects in low-Le mixtures, is probably the stabilization mechanism which allows flame balls to exist at µg.

At one-g, these flames are entirely buoyancy-dominated. Consequently, the flammability limits are much leaner at µg than one-g, for example 3.35% vs. 4.0% H_2 for H_2-air mixtures. The µg result is in very good agreement with a computational study of flame balls at µg employing detailed chemistry and transport models in which a limit of 3.5% H_2 was predicted. The close agreement of model and experiment based on first-principle modeling is unprecedented in the study of flammability limits of premixed gases. The results are also relevant to spacecraft fire safety applications because of the many potential sources of hydrogen leaks in manned spacecraft.

The g-jitter in the KC-135A experiments ($\approx 0.02g_o$) caused substantial motion of flame balls. The drift velocity was found to be proportional to $g^{1/2}$, consistent with bubble theory. This drift also led to the formation of two types of quasi-cylindrical flame structures which we have termed "flame strings." These strings are unstable, in that they eventually break into flame balls, but live much longer than would be expected based on thermal diffusion time scales alone. Consequently, they are almost certainly being supported by chemical reaction. Their existence is curious, because no steady solution is possible for cylindrical flames (unlike spherical flames). It seems then that they would evolve radially given sufficient time, but the axial instability which leads to their breakup manifests itself before radial evolution occurs. Recent theory supports this suggestion.

Another type of flame string, not directly related to that discussed above, has been observed in H_2-O_2-SF_6 and CH_4-O_2-SF_6 mixtures, particularly at high pressures. These strings are different in that they are uncorrelated with buoyancy effects and seem to form much faster than any conventional diffusional or hydrodynamic timescale would allow. We conjecture that these are a result of reabsorption of emitted radiation, which is most likely to occur in SF6-diluted mixtures and at high pressures because the Planck mean absorption length (L_p) is much shorter in SF_6 than CO_2 or H_2O.

2.2.3 Flame propagation and extinction in cylindrical tubes

The standard apparatus for measuring flammability limits of premixed gases at earth gravity is the Standard Flammability Limit Tube, or SFLT. This is a tube 5 cm diameter and 200 cm long which is filled with combustible gas and ignited at one end. The mixture is defined to be flammable if it supports propagation throughout the tube. Despite many years of study, the mechanisms of flame extinction in the SFLT is not well understood. The relative importance of buoyancy, flame stretch, heat loss to the tube wall, radiation loss, etc. have not been assessed.

Most theories of flammability limits predict the flame propagation rate at the limit ($S_{b,lim}$). Hence, measurement of $S_{b,lim}$ enables comparison with theories. We have conducted such experiments at earth gravity in tubes of varying diameter, at varying pressures and with mixtures having varying fuels, inerts, and Lewis numbers (Le).

We have shown that many different types of limits can exist in the standard flammability limit apparatus depending on the Lewis and Grashof numbers of the system. Two especially interesting phenomena observed were cases where the upward flammability limits may be narrower than the downward limits, and cases where the burned gases were turbulent and exhibited distributed-like modes of combustion. We have found that the characteristics of the limits can be described in terms of the effect of the Grashof number $Gr \equiv gd^3/\alpha^2$ on the limit Peclet number $Pe \equiv S_{b,lim}d/\alpha$, where g is gravity, d the tube diameter and α the thermal diffusivity at room temperature. For both upward and downward propagation, at low Gr the results are consistent with theoretical predictions for flame extinguishment by conduction loss to the tube walls, namely $Pe \approx$ constant ≈ 40. At higher Gr, results are consistent with buoyancy-induced extinction mechanisms, in the upward propagating case $Pe \sim Gr^{1/2}$ and in the downward case $Pe \sim Gr^{1/3}$. Because of the difference in extinction mechanisms, we have found that the flammability limit can actually be wider for downward propagation than upward propagation (though for most commonly-studied values of Gr and Le, this is not the case).

In the upward case, at $Gr > 2.0 \times 10^8$ (corresponding to a pipe-flow Reynolds number of 2000) turbulent flow is exhibited; the flame behavior can be either flamelet-like or distributed-like depending on Gr and Le. Downward turbulent propagation at the limit is also possible, but would require $Gr \approx 40 \times 10^9$ according to our scaling analysis, and was beyond the limits of our apparatus.

2.2.4 Experimental simulation of premixed flames using autocatalytic chemical reactions

The PI has introduced the use of aqueous autocatalytic propagating chemical fronts (in particular, the arsenous acid-iodate system) for the experimental simulation of premixed combustion in nonuniform and unsteady flows. These fronts more nearly match the assumptions made by most relevant theoretical models that do gaseous flames. These assumptions include that of constant density. The role of density change (due to heat release) in gaseous premixed flame propagation has been a continuing source of controversy, especially in turbulent flows. It has not even been established whether density changes result in an increase or decrease in the turbulent flame propagation rate (S_T). Consequently, it is desirable to compare front propagation in gaseous flames and aqueous autocatalytic fronts to study effects of density change.

We have studied these autocatalytic propagating fronts in a Taylor-Couette (TC) flow, i.e. in the annulus between two rotating concentric cylinders. This flow was chosen because (1) the flow is homogeneous in the direction of front propagation (axially), so that a quasi-steadily propagating front can be studied, and

(2) well-characterized disturbances are generated even at very low Reynolds numbers (and thus low u'). When only the inner cylinder is rotated, pairs of counter-rotating toroidal vortex pair (Taylor vortices) fill the annulus. The measured effect of u'/S_L on S_T/S_L in the Taylor-vortex regime is in good agreement with the predictions of a theoretical model. When the outer cylinder is rotated in the opposite direction, TC flow exhibits a "featureless turbulence" flow. When the "liquid flame" is propagated through this flow, the propagation rate is higher than in the single-scale flow, and the results are in astonishingly good agreement with Yakhot's Renormalization Group model of the propagation velocity of premixed turbulent flames. The same is true when the fronts are propagated in a chaotic Capillary Wave (CW) flow (which could also be employed at μg because the waves are produced by surface tension rather than gravity)

The fractal dimensions of the aqueous fronts in CW flows were measured at values u'/S_L which are at least an order of magnitude higher than any result which can be obtained in gaseous flames. The fractal plots (measured perimeter as a function of the measurement length scale) were very linear, indicating that indeed these surfaces were fractals. Surprisingly, the fractal dimension was found to be close to 7/3, which agrees with a number of previous theories and experimental studies in gaseous flames in 3-d turbulence. It is interesting that this similarity exists because the velocity spectra of the CW flows are very different from 3-d turbulence and according to existing theories the fractal dimension should be different.

The PI has also begun a numerical study of propagating fronts at very high u'/S_L in conjunction with Prof. Jingyi Zhu of the University of Utah. The effect of u' on S_T was calculated in a simulated Taylor-Couette flow at values of u'/S_L up to 50, which is much higher than any previous simulation technique could reach. The results were found to be in very good agreement with the experiments and theoretical predictions of our previous work supported by this grant. Also, the model predicts steady propagation rates after an initial transient, which in itself is not surprising physically since our experiments employing liquid-phase autocatalytic propagating fronts have demonstrated this even at very high non-dimensional disturbance intensities, but to date there is no mathematical proof of the existence of steady solutions for the G-equation.

3. POSSIBLE CONTINUATIONS AND EXTENSIONS

3.1 Radiation effects on premixed gas flames

Understanding the initial results on the effects of particulate radiation on premixed flames is limited by the relatively simple diagnostics employed in that work. In particular, the optical properties of the dust suspensions employed have not been measured in detail. Therefore, future work could focus on methods of quantifying the particle density, scattering and attenuation properties using now-standard laser light scattering and attenuation measurements. These results could also be used to determine the albedo (scattering to attenuation ratio) of the dust suspension, which is an important parameter in some theories of particle-laden gas flames. Once this has been done, experiments could be performed for a variety of mixtures and particle loadings to compare with theory and to determine if flammability limits can be entirely suppressed when radiation losses are eliminated.

To facilitate the interpretation of these data, radiometers could be incorporated into the drop tower and KC135 experiment packages.

In these tests, one could also explore the possibility that since the presence of particles will influence transport of thermal energy (see above) but should not influence mass transport, the radiation-influenced Lewis number may be higher than the particle-free value, thereby inhibiting the cellular instability. Some evidence of this was observed in the PI's initial experiments.

3.2 Flame structure and stability at low Lewis number

It is desirable to further study the effects of optical thickness outlined in section 2.2.1 as it applies to flame balls and flame strings. It is not feasible to do this using particle-laden gases because this appears to increase the effective Lewis number of the system and may suppress the cellular instabilities. The next-best way to provide short absorption lengths is via the use of SF_6-diluted mixtures at high pressures. A new combustion chamber which is considerably larger than the existing one and which is rated to higher pressures has been built by the PI. This chamber also has more viewing ports for multiple camera views and advanced diagnostics. This facility could be used to test H_2-O_2-SF_6 and CH_4-O_2-SF_6 mixtures at high pressure; the Planck absorption length can less than 0.1 cm under these test conditions (3 atm). These mixtures could be used to determine the types of instabilities which can be observed at μg in these radiation-dominated flame structures.

3.3 Flame propagation and extinction in cylindrical tubes

Our scaling analyses indicate that radiation losses are very likely insignificant at earth gravity for any experimentally realizable conditions; either conduction losses or buoyancy effects will dominate. Of course, radiation can be the dominant extinction mechanism in large tubes at μg. A study of the flame extinction mechanisms in large tubes could be useful in this context. Also, scaling estimates suggest that μg experiments may provide the possibility to observe the high Lewis number pulsating instability of freely propagating premixed flames; for several reasons, these are difficult if not impossible to observe at earth gravity. Hence, this conjectured instability has never received a conclusive experimental demonstration. It may be possible to search for this instability in drop tower experiments.

3.4 Experimental simulation of premixed flames using autocatalytic chemical reactions

A limitation on the utility of aqueous fronts is that even the small fractional density change across the aqueous front leads to significant buoyancy influences at one-g (g_o) because of their very low S_L. Only when $u' >> S_L$ is this limitation unimportant. Gaseous flames with $u' >> S_L$ cannot be observed because of quenching which results from the hydrodynamic strain at high $U \equiv u'/S_L$; this makes it impossible to compare the results of aqueous and gaseous front experiments at the same U, and thereby assess the role of density changes. Quenching is not a problem in the aqueous fronts because of their high Schmidt number (Sc) (ratio of kinematic viscosity (ν) to mass diffusivity (D)) which reduces the effect of hydrodynamic strain. High U is also inaccessible to computational studies because of numerical difficulties, especially when density changes are included. This discussion indicates the need for studying aqueous fronts at μg to eliminate buoyancy influences, enabling the study of front propagation at low U and thereby allowing comparison of front propagation in aqueous and gaseous fronts at the same u'/S_L.

An ideal flow for studying the interaction of propagating fronts with flow disturbances, and one which suffers from these buoyancy influences, is the Taylor-Couette flow in the annulus between two rotating concentric cylinders. When only the inner cylinder is rotated, pairs of counter-rotating toroidal vortex pairs (Taylor vortices) fill the annulus. To obtain these vortices, the Reynolds number $Re \equiv \omega d r_i/\nu$, where ω is the angular rotation rate of the inner cylinder, d the cylinder gap and r_i is the inner cylinder radius, must be larger than about 75. Using known properties of the Taylor vortex flow and the effect of buoyancy on the autocatalytic fronts, along with representative values for the relevant parameters, we find that to avoid buoyant convection, we require $d < 0.1$ cm at $g = g_o$ and $d < 2.0$ cm at $g = 10^{-4}g_o$. (The latter g is a typical figure in space flight experiments.)

Our scaling analysis has shown that in a space experiment, it is possible to study aqueous fronts with $U \approx 7$ at $g = 10^{-4}g_o$ (a typical figure for space experiments) without buoyant convection, whereas $U \approx 140$ is the lowest possible U at $g = g_o$ without buoyant convection. It is possible to study $U = 7$ in gas combustion without quenching, whereas $U = 140$ is not possible. Thus, space experiments would enable us to study the aqueous fronts at values of U accessible to gas combustion experiments and numerical simulations, enabling us to create a "bridge" between studies of fronts with and without substantial density changes.

While the time scales for the experiments at $U = 7$ are sufficiently slow that space experiments would be required, in the KC135 aircraft with 20 seconds of low-g it is still possible to study considerably lower U (≈ 20) than is possible at one-g. Consequently, it would be useful to perform these tests on the KC135 aircraft.

The fronts can be imaged by the pH indicators since their thresholds are intermediate between the reactant pH and product pH. The important quantitative results, namely the front propagation rate and shape, are readily obtained in this way.

It would also be useful to build a counter-flowing twin jet apparatus for measuring the response of flat liquid flames to straining effects, in a manner similar to that which is commonly done for gaseous flames. The purpose of this is to determine if the models for the influence of strain on flames (e.g. due to turbulence) which were developed for gas combustion are also relevant to liquid flames.

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16. Abstract

A two and one-half year experimental and theoretical research program on the properties of laminar and turbulent premixed gas flames at microgravity was conducted. Progress during this program is identified and avenues for future studies are discussed.

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