Droplet Combustion in Microgravity

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Burning of petroleum based liquid fuels form harmful products that contribute to global climate change. Yet, the demand for such fuels is expected to continue into the next decade. This white paper outlines a plan for experimental and numerical research that uses an isolated droplet burning in microgravity to access the chemical physics pertinent to global climate change. The areas proposed for study include biofuels, numerical modeling, burning at elevated pressures, transitions to cool flames, partial gravity effects, and development of new diagnostics for microgravity platforms.

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1. Importance of Microgravity Droplet Combustion

Research in droplet combustion in microgravity began over sixty five years ago. The motivation was the recognition that the one-dimensional gas transport around a stationary and burning droplet, promoted by the microgravity environment, provided access to the fundamental combustion physics that control burning of liquid fuels. Today, science has moved well past this early focus on fundamentals to more practical considerations of controlling the impact of combustion of liquid fuels on global climate change.

In the U.S. today over 90% of transportation vehicles are powered by petroleum-based liquid fuels (PBLF)[1]. The emissions they produce are having a deleterious effect on global climate change. In 2020 almost a third of the total CO_2 emissions was due to PBLF emissions from the transportation sector. Strategies that reduce harmful emissions are necessary. Alternatives are being pursued with projections to supplant the internal combustion engine as the prime mover for transportation systems by electrification of the vehicle fleet over the next two decades. By the end of 2020 fewer than 1% of electric vehicles were on the road. The Biden administration has recognized the current state by proposing that cars have an average of 51 miles per gallon by 2026 while alternatives are being pursued. Droplet combustion in microgravity is well positioned to play a key role in achieving this goal.

Liquid fuel is introduced into combustion engines in the form of a spray which sets the initial conditions for operation. However, optimizing engine efficiency through the lens of spray flames is currently too difficult because of turbulence, unsteady gas and liquid transport, fuel evaporation, radiation, inter-droplet interactions, preferential vaporization and phase equilibrium of multicomponent fuels including surrogates, radiation, and the often large number of chemical reactions needed to predict formation of particulate matter and greenhouse gases. The numerical tools for detailed modeling that include these processes are still being developed. The problem can be approached as a progression from the 'simple' to the 'complex'. The isolated and stationary droplet burning in microgravity represents the baseline. Success with this liquid fuel burning configuration will be a stepping stone to the incorporation of more complex transport dynamics found in the engine environment.

The value of the microgravity environment for droplet burning is illustrated by a number of discoveries over the past few years. The transition of traditional 'hot flame' burning to regimes of 'cool flames' controlled by a completely different set of chemical reactions is an example. The unmasking of the strong role flame radiation plays in droplet burning is another, snd the use of the microgravity droplet flame to evaluate performance of new biofuels synthesized in laboratory (bench) experiments has recently been demonstrated. Understanding liquid fuel combustion from the perspective of the microgravity droplet flame can also serve as a bridge to developing numerical tools to simulate burning in more complex environments such as in an engine. Doing so can yield capabilties to develop strategies to control harmful emissions and promote cleaner and more efficient operation of transportation systems.

2. Brief History of NASA's Interests in Microgravity Droplet Combustion

For over four decades NASA has been a world leader in supporting microgravity droplet combustion experimentation. Early designs of the Multiuser Droplet Combustion Apparatus (MDCA) for droplet burning experiments on the Space Shuttle, and later modifications of it for the International Space Station (ISS) as part of the Flame Extinguishment Experiments (FLEX2),

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[1] The bibliography on p. 6 is not meant to be a complete literature review. Rather, the citations are drawn from different sources to inform the narrative. Citations are not embedded in the text.

were first described in the 1980s. The data obtained from the MDCA has been essential for validating detailed numerical models of burning, discovering the role of radiation on the effect of initial droplet diameter on fuel burning rate and transitions to cool flame burning, and understanding the role of fuel composition on burning of miscible mixtures of petroleum fuel surrogates, and soot formation.

While NASA has been the leader in advancing microgravity droplet combustion science, the value of the microgravity droplet burning configuration is recognized internationally for its importance. For example, droplet combustion experimentation has long been a part of Japan's portfolio of microgravity research projects, the European Space Agency has had programs in microgravity droplet burning including the first studies on the subject, and Italy has been one of the leaders in numerically modeling droplet burning. China is most recently poised to emerge as a prominent international partner in microgravity science with the Tiangong Space Station (TSS) becoming operational and recently manned by astronauts. China is making significant investments in microgravity research with droplet combustion figuring prominently in its program, and is developing a new drop tower in support of TSS research programs and its national initiative on microgravity. It is hoped that NASA maintains its leadership in microgravity droplet combustion science to avoid falling behind the international community. The next sections outline several areas for further research where NASA can accomplish this goal while at the same time strengthening the link of the microgravity environment to terrestrially-based combustion technologies.

3. Opportunities for Droplet Combustion Research in Microgravity

Cool Flames

Cool flames have been a remarkable discovery in droplet combustion studies in microgravity. Originally inferred from measurements of gas temperature around droplets burning in microgravity, the definitive documentation was achieved from experiments carried out as part of the FLEX2 program on the ISS and supporting detailed numerical modeling to understand the role that radiation plays on flame extinction mechanisms.

Cool flames in droplet burning have significant fundamental and practical importance. From a fundamental perspective, droplet burning data have provided the capability to validate detailed numerical models that incorporate cool flame chemistry. From a practical perspective, identifying conditions from droplet burning studies that promote transitions to cool flame burning can carry over to developing strategies to promote this burning mode in combustion engines on Earth. The role that ambient pressure, ambient gas, and liquid composition have in developing cool flames is not well understood in these transitions and more work is needed in these areas.

Biofuels

Over the next decade the supply of petroleum reserves will be reduced but the demand for them will continue. Biofuels have potential to offset this shortfall as additives to petroleum fuels (cf, E10 as mixtures of 10% ethanol and 90% gasoline). The ability to 'grow' feedstocks for producing biofuels is attractive to reduce fuel consumption and emissions of conventional fuels since biofuels will typically be oxygenated compounds. Use of biological strains for synthesizing biofuels (e.g., *clostridium* for synthesizing butyl acetate (BA) as an alternative to Fischer esterification to produce BA) offers promise to provide new sources for biofuel production.

The microgravity droplet configuration can play an important role in testing and evaluating biofuel combustion in three ways. Firstly, synthesizing biofuels by new processes will proceed on a pre-production scale to demonstrate the synthesis process. The yields will typically be too small - on the order of milliliters - for engine testing where tens of liters at a time are required. By contrast, droplets burning in microgravity can typically have volumes on the order of 65 *nano*liters (for a 0.5 mm diameter droplet) and so are well suited to evaluate the performance of biofuels in limited supply produced by new processes. Secondly, new and untested synthesis processes can also yield miscible by-products that render the biofuel a miscible mixture. In this case the role of the undesired 'additives' on burning is easily evaluated in microgravity burning experiments. Finally, the biofuel droplet burning data for the spherically symmetric burning case will themselves be essential for validating numerical simulations of biofuel combustion.

High Pressure

High pressure is a ubiquitous state in terrestrial propulsion systems, for example gasoline and diesel engines. Ambient pressure influences many aspects of droplet burning including flame dynamics, fuel evaporation rate, droplet heat-up period, ignition delay time, soot formation, flame extinction processes, radiation and combustion kinetic mechanisms. In certain cases, the long time of the space-based environment will be necessary to access some of these aspects of droplet burning at high pressures, such as transitions of burning into supercritical conditions.

Combustion at elevated pressures brings with it many experimental and modeling challenges. The experimental challenges of high pressure burning include the following: obtaining high quality data in the presence of increassinglyoptically thick soot clouds as pressure is increased; developing an operational procedure for forming, deploying and auto-igniting droplets in a high pressure chamber without initially disturbing the ambient gas surrounding the droplet; potential for dissolving ambient gases into the liquid fuel as pressures approach the critical state; and disappearance of surface tension that leads to blurring the distinction between phases as pressure is increased. Many of these challenges can be addressed in a well-developed program of ground-based experimentation. Added to these concerns are challenges in numerical simulation of the 1-D droplet flame. The transport properties needed for modeling high pressure burning do not generally exist. The problem of developing mixing rules and using equations of state to determine phase equilibrium conditions is complicated by the lack of data for validation. And deviations from ideal solution behavior for surrogates can be significant.

Multicomponent and Surrogate Fuel Systems

PBLFs consist of hundreds of miscible organic liquids with wide ranges of boiling points, sooting propensities, kinetic mechanisms and transport properties. Mixture effects in droplet burning are therefore essential to understand. Topics of interest include preferential vaporization and internal diffusional processes inside surrogate droplets, conditions leading to confinement of volatile components inside the droplet to create the potential for internal superheating and microexplosion phenomenon, the kinetic mechanism for combustion of the mixture, and development of property correlations noted previously and experimental databases for thermal and transport properties with appropriate mixing rules.

Multicomponent mixtures, even if comprised of as few as four or five miscible constituents, can create significant numerical challenges. Transport properties, mixture bubble points and critical state values will not generally and must be measured and simulated. As gas pressure increases to near the critical state, ambient gases dissolution can be important but has not been previously studied in the microgravity droplet burning configuration. Simple mixing rule assumptions such as critical states being linearly dependent on component volume fraction or ideal solution behavior may not be valid near a fuel's critical state. Moreover, preferential vaporization of components in a surrogate mixture will leave behind the heavier and more soot-producing fuels to later in the burning history. These concerns must be addressed to develop high accuracy simulation capabilities to predict the effect of pressure on burning.

Numerical Modeling

Direct numerical simulations of microgravity droplet flames is a core necessity to extract fundamental value from experimental data. Simulations of microgravity droplet burning have provided essential information to interpret experimental results from ISS experiments. Current modeling capabilities of microgravity droplet burning data have also opened up an as yet unexplored use of this modeling capability, namely to validate detailed combustion kinetic mechanisms of new fuel systems. This has been made possible by using microgravity droplet burning data from ISS experiments to inform the efficacy of detailed combustion kinetic mechanisms in numerical simulations of droplet burning. Traditionally, mechanism validation of is carried out on gaseous fuel systems - the liquid is pre-vaporized. In so doing, the resulting kinetic mechanism remains untested for its ability to determine the relationship between droplet evaporation and the chemistry of ignition which are known to be linked. For new mixtures and environmental conditions of burning (e.g., high pressures) detailed modeling will be important in the kinetic mechanism validation process.

Computational challenges for microgravity droplet burning are significant. Included are the need develop the following: efficient solutions to the governing equations; methodologies to reduce the large number of reactions of a comprehensive kinetic mechanism from tens of thousands of reactions to a few thousand; the ability to predict and validate transport and thermodynamic properties including mixing rules for multicomponent blends; predictions of bubble and dew points as the critical state of surrogate mixtures is approached; methodologies to deal with the prospect of incorporating gas dissolution effects. The droplet burning problem is so complex, even with the simplification of gas transport afforded by the microgravity environment, that efficient numerical methods and high-power computer platforms will be needed to enable solutions to the governing equations. Some of these challenges can benefit from considering reduced kinetic models that enable solutions by alternative methods (e.g., activation energy asymptotics) which can be useful for interpreting results from detailed kinetic mechanisms.

Particulate (Soot) Formation

PBLFs in general form soot when they burn. The carbon black produced is largely responsible for global warming effects. The small size of soot precursor particles also makes them prone to being inhaled into the respiratory tract creating health concerns. In the microgravity environment for droplet combustion, soot formation is easily tracked as particles collect in a shell-like structure by the forces acting on the aggregates. This configuration is attractive for modeling. On the other hand, a more random and chaotic soot pattern arises for burning in a non-symmetric transport field such as in a spray flame or even a monodispersed stream of droplets.

The presence of soot formed during microgravity droplet combustion was originally considered as more of a curiosity than as a problem of significance to global climate change and worthy of further work. Today, that has changed. The ability to predict formation of the soot shell is just now emerging and can provide a view into the soot volume fraction and its control. More work in this area is needed including developing sub-models for soot formation to simulate the soot volume fraction in comprehensive simulations which is still in the early stages.

Partial Gravity Effects

With missions being planned to the Moon, Mars, and beyond, an understanding of how liquid fuels burn at partial gravity levels can be important. Many studies have reported droplet burning data at normal gravity ($g/g_o=1$ where $g_o=9.8$ m/s^2), at $g/g_o\sim10^{-2}$ (in aircraft undergoing parabolic flight trajectories or drop towers without a drag shield), at $g/g_o\sim10^{-4}$ (drop towers with a drag shield) and at $g/g_o \sim 10^{-6}$ (in the ISS). However, for the environment of the Moon $g/g_o\sim0.16$ and on Mars $g/g_o\sim0.38$. No droplet combustion studies are known at these intermediate gravitational levels. Simulating droplet burning at partial gravity will be beneficial, but data to validate the simulations and the inputs to them will need to be developed. Ground-based facilities that provide for a well-controlled free fall to create a partial gravitational acceleration in the moving frame of reference are required to provide such data.

Diagnostics and Facility Improvements for Microgravity Droplet Combustion Experimentation

Since the earliest studies of microgravity droplet combustion experimentation, video imaging has been the mainstay to provide measurements of the droplet, flame and soot shell diameter. The data themselves are extracted from video images by computer-based image analysis algorithms. However, numerical modeling provides a wealth of additional data that can be modeled, but which so far has been unable to be measured. Such variables as the concentration and distribution of combustion gas products and soot volume fraction and combustion gas temperature are useful because they are all related to global climate change. Gas temperature in particular has been measured from droplets supported by thermocouples, and the effect of the support needs to be evaluated. These variables are routinely measured in combustion configurations in the laboratory (normal gravity) frame often with non-intrusive diagnostics. However, beyond radiometers for measuring radiant emissions from flames, the microgravity environment does not lend itself to incorporating delicate instrumentatoin for measurement (in drop towers or the ISS). For drop towers, the shock of the impact in drop towers is a concern, and in the ISS the need for frequent hands-on adjustments for optical diagnostics by ISS crew is impractical.

Development of small-scale solid state micro gas sensors, or other sensing devices which would overcome these challenges would be beneficial to enhance the range of data obtained. Such capabilities would have applications well beyond the microgravity environment.

In addition to concerns with diagnostics, an important virtue of the ISS is the unlimited observation times it provides for microgravity. This feature has been especially beneficial in the discovery of transitions to cool flame regimes of burning and radiative extinction processes that require long times to develop. Several options exist in ground-based drop towers to extend microgravity observation times to greater than 10s. Developing them along with new diagnostic capabilities can be beneficial to providing a cost-effective database for microgravity droplet burning.

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