Preparations of the NO_x Measurements for the Combustion of an n-Decane Droplet Array Under Microgravity Conditions

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Abstract

It is recognized that the burning of droplet arrays represents an idealized condition to study the complex, interacting phenomena of multi-phase flow, thermodynamics, and chemical kinetics. Droplet arrays provide a fundamental foundation upon which descriptions and models of the complex spray combustion can be developed. The major objective of the present study on droplet combustion is to investigate the burning characteristics and exhaust formation mechanisms. This paper presents the preparations for the exhaust sampling and analysis by burning an array of five n-decane droplets on a sounding rocket platform. The primary problem, which has to be solved, is to obtain a representative gas sample from every, specific combustion run. The pursued concept is intrusive as it is necessary to immerse four probes into the combustion chamber. After flame extinction the samples are withdrawn and stored in sampling cylinders for the succeeding analysis. High efforts are put into a reliable and ideal sampling system as well as the preparation of the experiment procedures to ensure high-quality results. This includes experimental testing and numerical studies. The analysis process itself is ground-based and carried out by FT-IR (Fourier Transform Infrared) spectroscopy after the payload retrieval.

1. Introduction

The scientific cooperation and exchange between the Japanese and the European communities, working on combustion under microgravity (µg) conditions, have been strong for many years. By resorting to a successful JAXA droplet combustion experiment, a sophisticated cooperative research project was initiated in 2006, aiming for a flight opportunity on an ESA sounding rocket campaign. In the meantime intensive preparations for the TEXUS #46 campaign are in progress. Mikami et al. report about a new droplet generation technique of the experiment module 1) and the related µg experiments at high temperatures 2) in a droptower. Kikuchi et al. conducted a numerical study on the flame spread of an n-decane ($C_{10}H_{22}$) droplet array in different temperature environments and for different, dimensionless droplet spacing ratios 3). Their work also included a comparison of numerical and experimental results as well as a discussion of the relevant flame spread modes 4).

The cooperation focuses on two effects of the degree of fuel vaporization (Ψ) along a linear array of five n-decane droplets: the flame propagation (JAXA's main focus) and the NO_x production rates (the main focus of ESA's research team, working on the Combustion Properties of Partially Premixed Sprays, CPS) 5). The main experimental parameters are: the degree of vaporization, the equivalence ratio, the temperature, the pressure, and the droplet/spray mode. In order to further develop an understanding of the effect of fuel pre-vaporization in droplet/spray combustion, the experiments must be conducted on a µg facility that allows observation times in excess of 40s, such as the TEXUS sounding rocket system.

Different regimes of droplets need to be investigated to fully develop a model of the NO_x generation in spray combustion. Furthermore, it is important for the interpretation of the experiments to have an equal degree of vaporization for all droplets prior to ignition 6), 7). The developed setup is able to set the required initial conditions for a defined number of five droplets 1), 2), 3).

2. Technical Background

The original experiment module was used successfully in a number of droptower campaigns, but modifications became necessary for the planned sounding rocket flight. Apart from the modified core systems, a completely new Exhaust Gas Sampling (EGS) system will be incorporated into the TEXUS setup (cf. to **Fig. 1**).

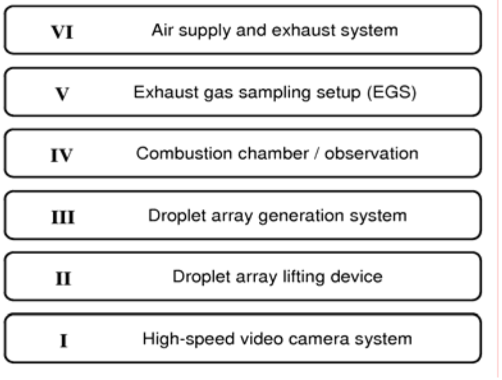

Fig. 1 Overview of the subsystems and platforms of the combustion module.

It will complete the original setup (without EGS) and ensure the highest accuracy for a successive gas analysis on ground. Due to a close collaboration between the Japanese and European work groups, a very promising concept could be tailored, which will satisfy the attainment of the experimental goals and enhance the research activities of both parties.

3. Exhaust Gas Sampling System

Models on the formation of NO_x in partially pre-vaporized/premixed spray flames are frequently based, at least to some extend, on empirical data obtained from well-defined combustion systems [5) 8)]. The reliability of these models therefore depends on the accuracy of the experimental measurements. Considering the investigated droplet combustion, it is necessary to collect exhaust samples by probes from the combustion chamber before the analysis can be performed. Allen [8)] reports on the problems associated with the catalytic reduction of NO_x in metallic probes as well as the NO to NO_2 conversion in non-metallic probes (without affecting the total NO_x content of the sample). Tiné [9)] gives an extensive overview of ideal and practical gas sampling, discusses relevant design aspects, and describes analysis methods. He points out the reliability of the sample composition since sampling procedures can involve errors. Tiné distinguishes between two different kinds [9)]:

- Those which originate from physical causes, and ultimately result in a segregation of some of the species present within the original medium.
- Those which originate from chemical causes and ultimately result in a partial chemical change.

Several modifications and additions consequently had to be implemented in the original combustion experiment in order to be able to perform a gas sampling during a sounding rocket flight. An in-situ measurement of the emissions was not feasible since it would require too many resources. Fig. 1 shows the subsystems of the adapted combustion module as they are arranged bottom-up on standard experiment platforms for TEXUS. Platform V indicates the new, additional EGS setup. It is allocated directly above the combustion chamber. This is done to keep the volume of the piping low, which therefore results in low contamination of the samples. The design meets the following, very strict relation: $V_{piping}/V_{sample} \leq 1\%$. The piping itself has a surface/volume ratio of 1.85 with an inner diameter of ID = 2.16mm.

The four sampling probes of the herein presented experiment are made of thin-walled stainless steel piping, which is coated with hydrogenated amorphous silicon (a-SiH) [10)]. This surface treatment increases the resistance to corrosion, reduces interactions between the steel surface and active compounds, and inhibits coking. Of particular importance for the handling of the whole EGS setup (including sampling cylinders and piping) is to achieve fast pump-down times by a significantly reduced outgassing. An improved moisture uptake (wet-up) and release (dry-down) is essential, too. As confirmed by extensive tests with several EGS components, all three aspects could be improved by at least one order of magnitude due to the utilization of this silicon coating. In agreement with the Japanese party, four symmetrically aligned probes were defined (cp. **Fig. 3**). Their integration into the combustion chamber and their connection to the piping aims at a spatially uniform sampling process.

The EGS setup is arranged on a compact sampling platform and mounted in close proximity to the combustion chamber. **Fig. 2** gives an illustration of the EGS including combustion chamber and gas sampling cylinders with integrated electro-pneumatically operated valves. Furthermore the EGS consists of a late-access port for a turbomolecular or diffusion pump (for the cylinder re-evacuation during the pre-launch activities), a cold cathode gauge to determine the internal vacuum level of the sampling cylinders, and an inlet and outlet shut-off valve. Four sampling cylinders are required to sample the emissions of the altogether four combustion cycles separately (cf. to Fig. 2). Since metal surfaces generally are rather rough and subject to gas adsorption, the cylinders were also coated with amorphous silicon, as well as all piping, the valves, and the fittings in contact with the exhaust gas. Electro-polishing and PTFE-coating were not pursued because of the very strict requirements on the EGS. As shown in Fig. 2, the EGS provides the possibility to evacuate the sampling cylinders prior to lift off. The specified vacuum level is $10^{-5} – 10^{-6}$mbar with an associated leakage rate of $< 10^{-5}$(mbar l)/s.

It is required that the bottom of the combustion chamber remains partially open for an unobstructed operation of the droplet deployment mechanism. After the droplet generation by the fuel dispenser on platform III (below the combustion chamber), this mechanism lifts and inserts the droplet array holder into the combustion chamber. A sealing of the chamber with the deployed droplet array holder would technically not be feasible and

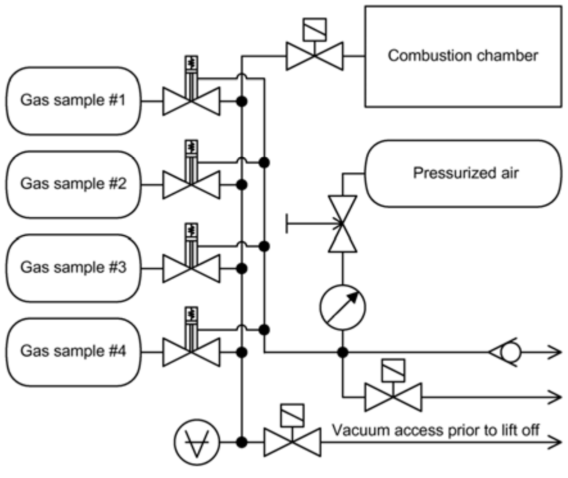

Fig. 2 Concept for exhaust gas sampling.

have a falsifying effect on the flame spread. Therefore, it is even important to have an “open” combustion chamber for an experiment operation under isobaric conditions 1), 2), 5).

4. Combustion and Gas Sampling Process

An essential condition of the investigated combustion process is an equal degree of fuel vaporization (Ψ) for all five droplets. It is perceivable by the vapor distribution formed around each droplet. As the combustion chamber is set at a temperature of 500K (+/–3K) for all four combustion cycles, the time of exposure or “pre-vaporization” prior to ignition is the only parameter determining Ψ. The different amounts of waiting time are therefore essential for the development of a flammable gas layer around the droplets 3) 7) and to diversify the scientific results. After ignition the combustion process will initially be characterized by a partially premixed flame, which propagates from droplet to droplet, followed by a diffusion flame around each droplet until flame extinction 1) 2).

N-decane is employed as fuel. The droplet array is generated at atmospheric conditions (0.110MPa with +/–0.002MPa tolerance) and at an ambient temperature of $300K < T_\infty << 330K$ on platform III. Each droplet is suspended at the intersection of a pair of X-shaped 14µm SiC fibers on the droplet array holder, which imparts lowest residual motion and highest sphericity 1). After the successful generation of the array’s five droplets, the holder is lifted up into the combustion chamber ($\Delta t = 1.5s$).

According to the degree of vaporization a hot-wire igniter (iron-chrome alloy) ignites one end of the droplet array to initiate the combustion process. The linear flame propagation mode has an averaging effect on the overall combustion temperatures. Therefore, the averaged production of thermal NO (Zeldovich mechanism) can be considered as representing the NO quantities of what a single droplet produces as a single link in an infinite droplet array or as in a stationary combustion process.

The density decrease of the gas inside of the combustion chamber, pursuant to the temperature increase during the combustion process, is one of the most critical issues of the experiment. It causes a volume expansion of a factor of approximately 2. This expansion further results in a discharge of some exhaust off the combustion chamber. Since 200ml of exhaust need to be collected for the succeeding gas analysis and the combustion chamber’s volume itself is about 380ml, the losses are significant and need to be considered within the analysis process and the scientific interpretation of the results (cf. to Fig. 3).

The sampling process itself aims to avoid contamination effects and to keep the sampling time to a minimum. After flame extinction the gas sampling will be performed to investigate the NO_x production rates in relation to the main experimental parameters. Fig. 3 illustrates the front and side view of the combustion chamber during the sampling process and a simplified model of the gas flow. By sampling the exhaust through the sampling probes, “fresh air” is entrained into the open combustion chamber.

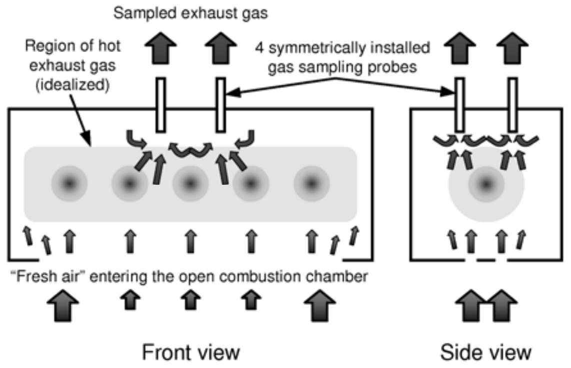

Fig. 3 Sectional views of the combustion chamber, displaying a notion of the gas sampling process.

Choi et al. report a minimum time of 5s to complete the burning of a 1.75mm droplet in a droptower 11). The observation or burning time t_b, needed for extinction, increases with the reduction of the droplet vaporization rate constant K, as shown in Equation (1), where d_i is the initial droplet diameter at the moment of ignition and d_e is the diameter at extinction 12).

$$t_b = \frac{\left(d_i^2 - d_e^2\right)}{K} \tag{1}$$

Extinction occurs at a critical Damköhler number, which is defined by the ratio of the characteristic chemical time at extinction and the characteristic time for the critical diffusive flow process. To guarantee a gas sampling after flame extinction and an identical, succeeding homogenization time of about 2s for all four combustion runs, empirical tests are currently under preparation. They will help to establish a correlation between extinction (in the ideal case: complete droplet burnout) and the temperature readings from inside of the combustion chamber. Having developed this correlation, a simple “if – then” algorithm can be used to trigger the start of the exhaust sampling.

Critical flow through the probe orifice itself will not be achieved in this experiment. However, a quasi-sonic flow “orifice” is expected to develop directly downstream of the physical probe orifice, where the four probes are fitted to one piping joint. Here, the sample undergoes rapid adiabatic expansion and cooling. Due to the transient process of this combustion experiment and a gas sampling after flame extinction, the exhaust cooling rate as well as the decay of the oxygen atom (O) concentration in the post-flame zone are considered to be sufficient to quench the high-activation energy reactions forming NO. However, NO itself can undergo recombination reactions with free radicals 8) 9), as illustrated by Equation (2). Thus, the process of quenching in the sample probe may significantly affect the NO/NO$_2$ ratio in the sample.

$$NO + O \xrightarrow{\text{probe wall}} NO_2 \qquad (2)$$

In addition there is a high probability of aerodynamic quenching in the combustion chamber. This quenching effect is of the same nature as the conversion reaction occuring inside of the probe and is a consequence of the steep temperature gradient in the regions surrounding the droplet array.

5. Numerical Study of the Gas Sampling Process

Since the combustion chamber is not pressure-tight, a gas exchange will occur twice: during the combustion process as well as the succeeding gas sampling (cp. Fig. 3 and **Fig. 4**). The first event is due to the temperature increase and the associated density decrease. The second is due to the evacuated cylinders, imbibing the exhaust from the combustion chamber. Both steps are crucial for the quality of the scientific results and an optimized layout and design needed to be found for the EGS system. Therefore a numerical study of the gas exchange processes was conducted within the design process, considering the “open” combustion chamber.

The numerical model uses the advantage of a twofold symmetric setup. However, symmetry was implemented as boundary condition only in the longitudinal plane of the herein presented results. The plane of symmetry is highlighted in the background of Fig. 4. Therefore, only two sampling probes can be seen entering the combustion chamber from the top.

Fig. 4 shows the temporal evolution of the critical sampling process by highlighting the fresh air from outside of the combustion chamber. The fresh air replaces the sampled exhaust by flowing through the open bottom of the combustion chamber. The light cells indicate 30%v/v and the dark cells 90%v/v of fresh air. After 1.0s the first cells inside of the sampling probes indicate a fresh air content of 30%v/v and after 1.9s the first cells indicate 90%v/v. The tracking of the fresh air is performed by the implementation of an additional variable into the CFD (Computational Fluid Dynamics) routines.

The duration of the exhaust sampling was estimated to be approximately 3s, which could be confirmed experimentally. As this is a rather long process for the small dimensions of the combustion chamber, the location of the sampling probes has a significant influence on the quality (the composition) of the sampled gas. Negative effects on the gas sampling are possible if high amounts of fresh air are sucked into the combustion chamber at unfavorable locations, and direct jets develop between the open bottom of the combustion chamber and the sampling probes. Furthermore, the probes should have neither direct contact with the flame nor be allocated too close to the lateral surfaces of the combustion chamber. As the temporal sequence of Fig. 4 reveals, the probe location could be specified at a favorable location for a spatially and temporally uniform sampling.

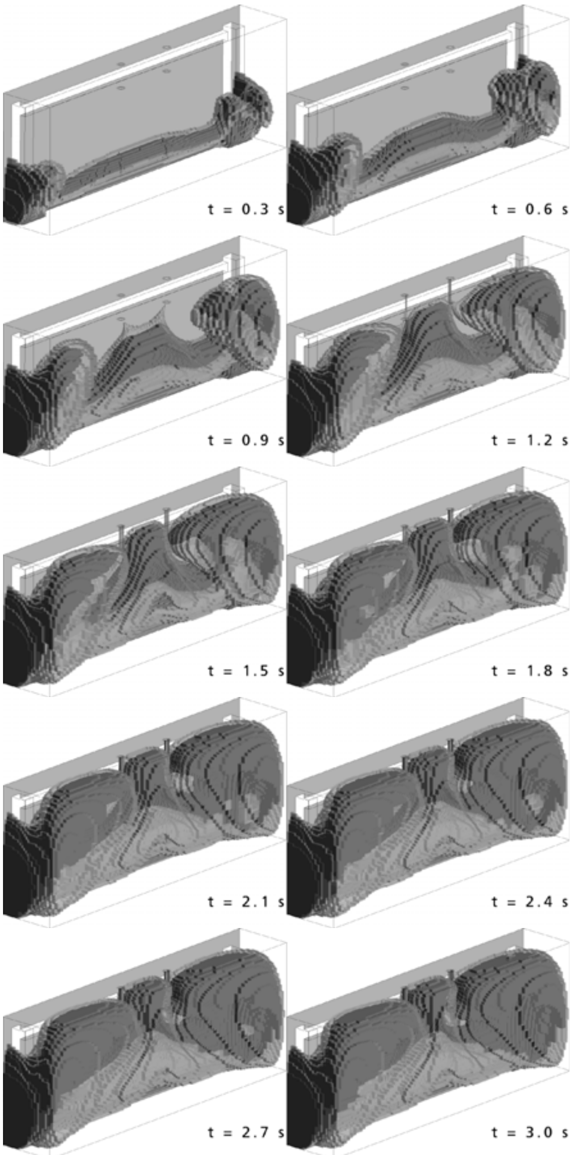

Fig. 4 Entrainment of “fresh air” in the combustion chamber during exhaust gas sampling (symmetric view).

6. Gas Analysis System

An important question for future aero-engine combustors is how partial vaporization influences the NO$_x$ emissions of spray flames [5), 6), 7)]. In order to address this question, the combustion of partially vaporized droplet arrays is studied in this research project under microgravity conditions. To allow a meaningful interpretation of the experimental data, systematic and accurate processes are required. Two main steps characterize the investigations of the NO$_x$ production rates: exhaust gas sampling and analysis.

As a number of methods are available for the gas analysis, the following factors were assessed during the design process [9)]:

- Necessary complexity of the analysis,
- Physical and chemical properties of the gases involved,

- Size of the sample, which can be obtained,
- Time available for analysis, and
- Required accuracy.

The analysis includes measurements of CO_2, CO, NO, NO_2, and N_2O, with the thermal NO being of major interest for technical applications 5), 6), 12). The determination of the gas concentrations is best achieved by the spectroscopic FT-IR method (Fourier Transform Infrared). This method utilizes physical but no chemical properties of the exhaust and the exhaust constituents are not separated during the analytical procedures. FT-IR spectroscopy has analytical and operational advantages, and is capable of analyzing up to 25 species simultaneously 13), 14). The ground-based analysis apparatus requires 200ml of gas, which will be provided by each of the sampling cylinders. As a gas analysis from sampling cylinders has to aim at the highest accuracy and a low contamination impact, an existing FT-IR spectrometer setup was modified for the particularly severe requirements. The reliability of the results depends on the maintenance and calibration of the spectrometer, which is time-consuming in comparison with other methods. The analysis time itself is short.

7. Further Proceedings

The sounding rocket flight itself foresees only four combustion cycles. However, a diversification of the experiments and a multiplication of the scientific output are necessary before a reliable model of the NO_x production of droplet and spray flames can be established. Therefore, the utilization of the experiment's engineering model (EM) in a parabolic flight is currently under discussion and preparatory steps have been initiated. Apart from the microgravity experiments on TEXUS, the parabolic flight experiments will help achieve a more complete understanding of the combustion and the NO_x forming processes. Further variations (apart from the degree of vaporization Ψ) could be the temperature of the combustion chamber, the distance, number, and initial diameter of the droplets, and the fuel itself.

Due to the expansion of the air/exhaust mixture inside of the combustion chamber during the combustion process and the quasi-isobaric conditions, gas will pass out of the combustion chamber entraining combustion products from the central combustion zone. To get a deeper insight into this discharge process and the associated mixing process of burned and unburned gases inside of the combustion chamber, a further numerical study is currently conducted.

8. Conclusions

In order to realize ideal droplet combustion, an array of five n-decane droplets is burned in microgravity without relative velocity to the ambient gas. The droplet array, suspended on SiC fibers, is inserted into a high temperature combustion chamber, where one end of the array is ignited to initiate combustion. The main focus of the herein presented study is to parametrically investigate the effect of pre-vaporization (Ψ) on the NO_x generation rates.

The experiment apparatus is developed in cooperation between the JAXA and ESA combustion work groups and is scheduled for a TEXUS sounding rocket flight in 2009. The Exhaust Gas Sampling (EGS) system is a new module in the already existing, basic Japanese setup and complies with the European focus on the NO_x production rates of partially pre-vaporized/premixed spray flames.

A main issue for the future interpretation of the measurement results is the representativeness of the gas samples. Disturbing phenomena are related to chemical, heat transfer, and aerodynamic effects in and around the probes as well as in the whole sampling system. As these effects are interdependent and difficult to quantify, high effort has been put into the sophisticated design and the operation procedures of the EGS, including the gas analysis. Generally, it is important for the quality of the analysis to install a compact sampling setup and to keep contamination and residual effects, including adsorption, as low as possible. A coating is used for all metal surfaces in contact with the exhaust. It aims to overcome the inherent, undesirable molecular activity of the EGS surfaces, including chemisorption and physisorption of other molecules as well as catalytic reactivity.

To gain insight into the fluid dynamics of the sampling process, a numerical simulation was carried out utilizing a half-model of the combustion chamber with a symmetry boundary condition. The qualitative results are promising for an ideal sampling process and confirmed the pursued design concept.

The exhaust gas analysis is ground-based and will be performed by FT-IR spectroscopy. It is a versatile technique and allows the simultaneous measurement of CO_2, CO, NO, NO_2, and N_2O. The analysis requires 200ml of gas, which will be provided by each of the four sampling cylinders, (one cylinder per combustion run).

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