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Advanced Propellants for Space Propulsion – A Task within the DLR Interdisciplinary Project "Future Fuels"

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Abstract

Within the sub-project “Advanced Rocket Propellants” (TP5) of the DLR interdisciplinary project "Future Fuels" three promising advanced green propellant systems are under investigation with regard to applicability and efficiency in rocket engines. The work aims at different mission scenarios and tasks, whereas performance, low costs, environmental friendliness, and safe handling characteristics are essential. The objective of this sub-project is to show and elaborate the potential of these propellant systems, to develop and to understand combustor processes with the task to advance the technology development in direction to commercial development and usage and to provide this technology to industry. This paper gives an overview of the planned work together with first obtained results.

1. Introduction

The development process of rocket propulsion systems is no longer affected only by the demand for better performance properties like higher thrust, enhanced specific impulse and/or increased velocity gain. Instead, requirements are coming more and more into focus, which have been rated up to now as secondary [1,2]. These include amongst others a free and versatile thrust variation capability, simple handling and storage characteristics, low toxicity and health hazard risks both for propellant and exhaust flow species, improved safety in handling and use, environmental friendliness, reusability, and strategies for upgrading and decommissioning under the above mentioned aspects. Furthermore mission scenarios are getting more and more complex and existing propulsions systems with conventional propellants are not able to fulfil all of the envisioned demands of contemplated missions.

Considerable efforts are currently undertaken worldwide to develop greener propellants, fuels and propulsion systems (see e.g. Refs. [3-6]). For hydrazine replacement, energetic ionic liquids based on ADN and HAN are most promising. First satellites are in orbit with small thrusters using advanced green propellants based on ADN [7-9]. Nevertheless there is still a long way to go before the majority of propulsion systems using highly toxic and aggressive propellants may be replaced on all thrust levels and for all mission demands by greener ones.

The search for greener propulsion systems is not only a task for the space propulsion sector. All aspects of human energy use, transfer and consumption on planet earth request the development of a greener and more sustainable energy production and use. Therefore the German Aerospace Center (Deutsches Zentrum für Luft- und Raumfahrt; DLR) has started the four year interdisciplinary research project “Future Fuels” in 2018, where institutes of different subject areas and specializations are working in close collaboration on novel fuels and propellants for various terrestrial and space applications.

Within the sub-project 5 (TP5) “Advanced Rocket Propellants” three promising advanced green propellant systems are under investigation with regard to applicability and efficiency in rocket engines. This manuscript gives an overview on the ongoing and planned work within this project and also on first obtained results.

DOI: 10.13009/EUCASS2019-276

2. Overview DLR Interdisciplinary Project "Future Fuels"

Advanced and synthetic fuels are a decisive building block for the successful implementation of the German Energiewende (Energy Transition) as they allow storing energy in a validated simple, flexible, efficient, and sustainable way. They are also critical guarantors of future mobility in a vast range: as fuels for road vehicles, trains, and ships as well as for aircraft and rockets. The challenges are manifold, but climate-neutral production and fuel design for optimized properties are key aspects in current research. Eleven institutes of the German Aerospace Center (Deutsches Zentrum für Luft- und Raumfahrt; DLR) are researching the development and application of advanced and synthetic fuels in DLR's interdisciplinary "Future Fuels" project [10].
Carbon and hydrogen (and sometimes oxygen) are the basic elemental building blocks of almost all fuels. Chemical processes and renewable electrical energy are used to produce liquid hydrocarbons from them, thus serving as the basis for application-specific future fuels. Synthetic fuels have a triple advantage: they can be easily integrated into our existing energy and mobility infrastructures, they can be optimized for the respective applications, and they allow optimizing their chemical properties in a way that no or at least significantly fewer pollutants such as soot particles and nitrogen oxides are formed.
The scientists and engineers within the project are investigating how synthetic fuels can be produced using solar energy and electrolysis processes (Solar Fuels) and are developing concepts for the re-conversion of these fuels into electricity. They are working on emission-optimized fuels for transport and aviation (Designer Fuels), including their flight testing, and advanced Green Propellants for space applications, for example, to replace the highly toxic hydrazine. System analyses and technology assessments are carried out that take a holistic view of future fuels and include factors such as cost-effectiveness, performance, security of supply, and social acceptance.
A detailed overview on the DLR project “Future Fuel” can be found in Ref. [10]. Figure 1 shows the key elements of the encompassing DLR approach.

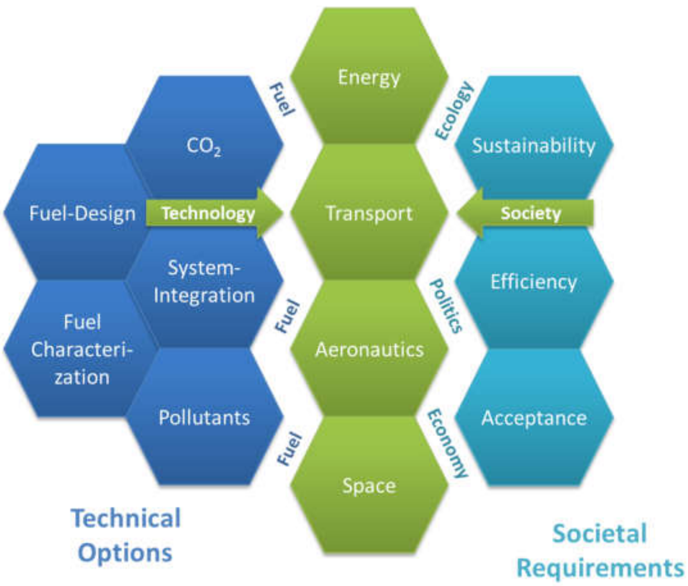

Figure 1: The holistic approach of the DLR project “Future Fuels”.

3. Overview sub-project (TP5) “Advanced Rocket Propellants”

Within the sub-project “Advanced Rocket Propellants” (TP5) three promising advanced green propellant systems are under investigation with regard to applicability and efficiency in rocket engines. Here, the DLR institutes of Space Propulsion, of Combustion Technology and of Structures and Design are working together in a close collaboration. The work with three different propellant system candidates aims on different mission scenarios and tasks, whereas performance, low costs, environmentally friendliness, and safe handling characteristics are essential. The objective of this sub-project is to show and elaborate the potential of these propellant systems, to develop and to understand combustor processes with the focus on the technology development in direction to commercial development and usage and to provide this technology to industry.

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The three selected promising propellant system candidates for future space propulsion applications within this sub-project are:

- The cryogenic bipropellant combination liquid methane / liquid oxygen (LCH4/LOX).
- Liquid monopropellants consisting of hydrocarbons and nitrous oxide (HyNOx).
- Green gelled propellants.

This manuscript gives a detailed overview on the ongoing and planned work within this project and also on first obtained results. LCH4/LOX is investigated in the main work package HAP 5.1, HyNOx in HAP 5.2 and gelled propellants in HAP 5.3.

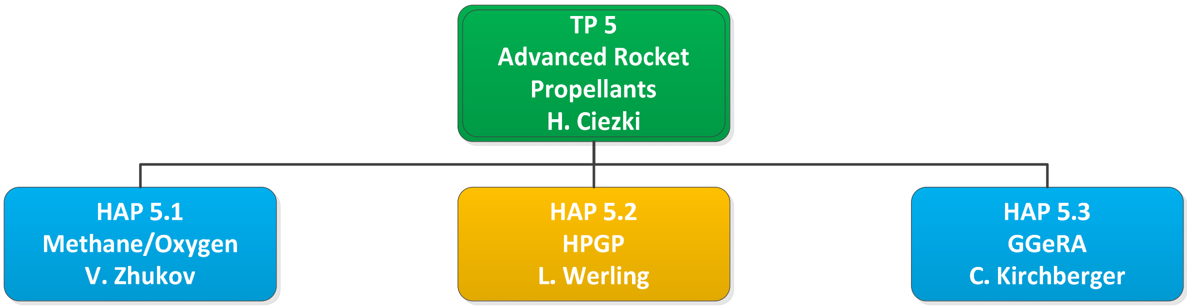

Figure 2: Structure of sub-project TP5.

4. Cryogenic bipropellant combination LCH4/LOX

Liquid methane (LCH4) / liquid oxygen (LOX) is the most promising rocket propellant combination for the development of the next generation of launchers [11]. The great interest in methane stems from the possibility to build cost-effective space transportation systems for a wide range of applications ranging from space tourism [12] to Mars missions [13, 14]. The possibilities for the cost reduction offered by methane are due to its unique properties: the high energy value of the low molecular weight of the combustion products, the small difference in temperature and density between methane and oxygen in the liquid states, and the acceptable cooling performance at low thermal loads on the liner [11]. At the moment, the developments of methane rocket engines are carried out by all large space countries: USA, Russia, China, France, Germany, and Italy. Here, the non-toxicity of methane also plays a role, and it is called often a „green propellant“ in comparison to propellants based on nitrogen oxides and hydrazine.
Nowadays, rocket engines are being designed with the use of CAD and CAE systems through the extensive use of numerical simulations. Computational fluid dynamics (CFD) simulations of the flow in combustion chambers are of high relevance in the development process of future rocket engines. They can help to speed up the development process while decreasing the cost. In order to fulfil those goals the numerical simulations need to be reliable and produce precise and accurate predictions for the flow conditions within the combustion chamber and the thermal loads on the chamber walls, while being reasonably cost effective in terms of computational effort. Projects like Prometheus from ESA or LUMEN from the German Aerospace Center (DLR) show the increasing interest for methane as a rocket propellant. Methane offers better properties over hydrogen in respect to costs and the overall performance from a system point of view. Using Methane as a rocket propellant the tank size can be decreased, there are lower requirements for cooling, the reusability is increased and methane offers and overall better availability. However compared to hydrogen there is still a lack of knowledge about the fuel/oxidizer combination CH_4/O_2. In order to increase the confidence into the numerical models the simulation results need to be validated against experimental data. Due to the extreme conditions and the complex physical processes in rocket combustion chambers, the availability of experimental data is limited and often only measurements on the chamber wall are available (temperature, wall heat flux, and/or pressure).
In the framework of the project “Future Fuels”, hot fire test runs of a single-injector methane rocket combustor will be carried out and the methane rocket combustor will be modelled using the in-house DLR CFD code TAU. The goal of the CFD simulations is to validate the developed numerical model. The single-injector combustion chamber, which will be used in the experiments, is shown in Figures 3 and 4. The chamber is instrumented with pressure transducers and thermocouples and has an optical access, which allows carrying out the comprehensive validation of numerical models. The validated model will be used in the future R&D of methane rocket engines at the Institute of Space Propulsion.

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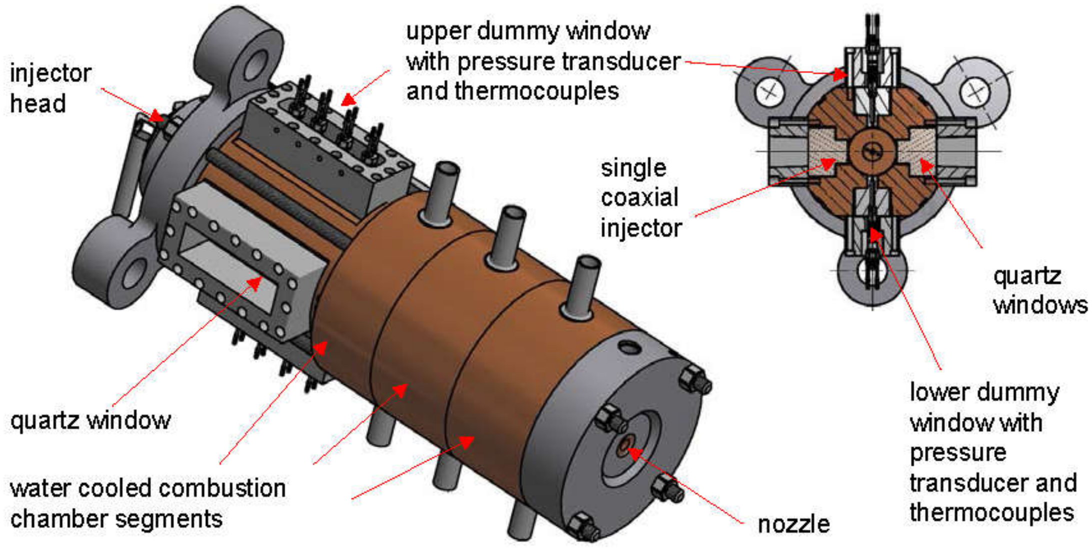

Figure 3: Single injector combustion chamber with optical access (BKC) (figure courtesy of Dmitry Suslov) [15].

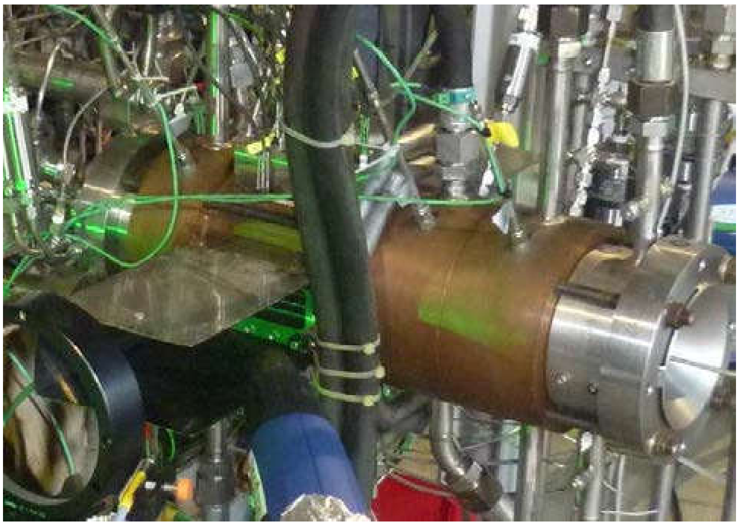

Figure 4: Combustion chamber BKC at DLR test bench P8 (photo courtesy of Dmitry Suslov) [15].

5. Liquid monopropellants based on hydrocarbons and nitrous oxide (HyNOx)

To this day hydrazine (N_2H_4) is the commonly used monopropellant to propel satellites, planetary probes or landers. Additionally, hydrazine is used in bipropellant systems as a fuel in combination with dinitrogen tetroxide (N_2O_4) [16]. Advantages of hydrazine as monopropellant are its sufficient I_{sp} (up to 240 s), the long term storability, it can easily be decomposed via catalyst and an explosion of the propellant is very unlikely. Used in bipropellant systems, N_2H_4 reacts hypergolic with N_2O_4, which makes an ignition system for upper stages redundant [17].
One the opposite, hydrazine is highly toxic and carcinogenic. Due to the high toxicity the fuelling process of a spacecraft is a complicated process with high safety measures. Along with the high safety precautions, high costs for the overall fuelling and transportation process occur. As a consequence of hydrazine's high toxicity it was added to the candidate list of substances of very high concern (SVHC) in the context of EU's REACH (Europe's Registration Evaluation Authorization and Restriction of Chemicals) regulation [18]. Due to this the use of hydrazine could be restricted or even forbidden in future which also may have impact to space propulsion applications.
Caused by the mentioned economic and political reasons several alternatives for hydrazine are currently under investigation. Among those "green propellants" several substances seem to be promising to fulfil the needs for a

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future replacement of hydrazine. The most prospective and developed candidate for low thrust systems is LMP-103S, based on ADN (ammonium dinitramide) and developed by the Swedish company ECAPS [19-23].
Nevertheless other possibly prospective propellants exist. Concerning lower costs and negligible toxicity, highly concentrated hydrogen peroxide (H_2O_2) might be a suitable alternative. The main advantages of H_2O_2 are beside the aforementioned negligible toxicity, easy ignitability via catalyst, relatively low decomposition temperatures (up to 1230 K) and a high density (1.44 kg/l) [24,25]. The drawbacks of H_2O_2 are a lower I_{sp} than hydrazine (up to 185 s, depending on concentration), the incompatibility with several materials (e.g. copper, iron, magnesium alloys, titanium) [26-28] and decomposition/detonation hazards [29-30].
In the USA, in China, in Russia and in Japan HAN (hydroxylammonium nitrate) based propellants are intensively studied [31–35]. Those propellants offer low toxicity due to negligible vapour pressures and higher performance than conventional hydrazine. Drawbacks of HAN based propellants are high combustion temperatures, which are challenging for combustion chamber and catalyst material and the danger of explosion of the propellant [33].
Furthermore the so called "water propulsion" is under investigation for satellite propulsion systems [36,37]. Here the satellite is fuelled with water which then is decomposed into hydrogen and oxygen via an electrolyser in orbit. The gaseous hydrogen and oxygen are stored in separate tanks and used for thrusters at a small mixture ratio or with a suitable film cooling to avoid overheating of the combustion chamber or catalyst. If the H_2 and O_2 are combusted at a rich mixture ratio, the excess oxygen could be used in cold gas thrusters or as oxygen supply for manned space stations. Drawbacks of water propulsion systems are the complex propulsion system, limitations on low thrust levels or burn times due to the production rate of H_2 and O_2 by the catalyst and the limited amount of gaseous H_2 and O_2 which have to be stored in separate tanks.
Another class of prospective, low cost and high performance propellants are mixtures of nitrous oxide (N_2O) and fuels, also known as nitrous oxide fuel blends [38-41]. Here the nitrous oxide and a fuel are stored pre-mixed as a monopropellant in one tank. These mixtures offer a performance similar to bipropellants ($I_{sp} \geq 300$ s), while only one tank and one feeding system are needed. In addition to this simplified fluid system, self-pressurization of the propellant tank is possible due to the high vapour pressure of nitrous oxide. Further advantages of those propellants are the non-toxic and very cheap constituents.
In contrast to those benefits, the main challenges regarding N_2O/fuel propellants are high combustion temperatures and the danger of a flame flashback across the injection system upstream into the tank structure. To handle the high combustion temperatures, an active cooling system is needed, which increases the complexity of the thruster. To avoid flame flashback suitable flashback arresters have to be designed, tested and qualified to be used in a propulsion system which is a vital aim of work package 5.2.

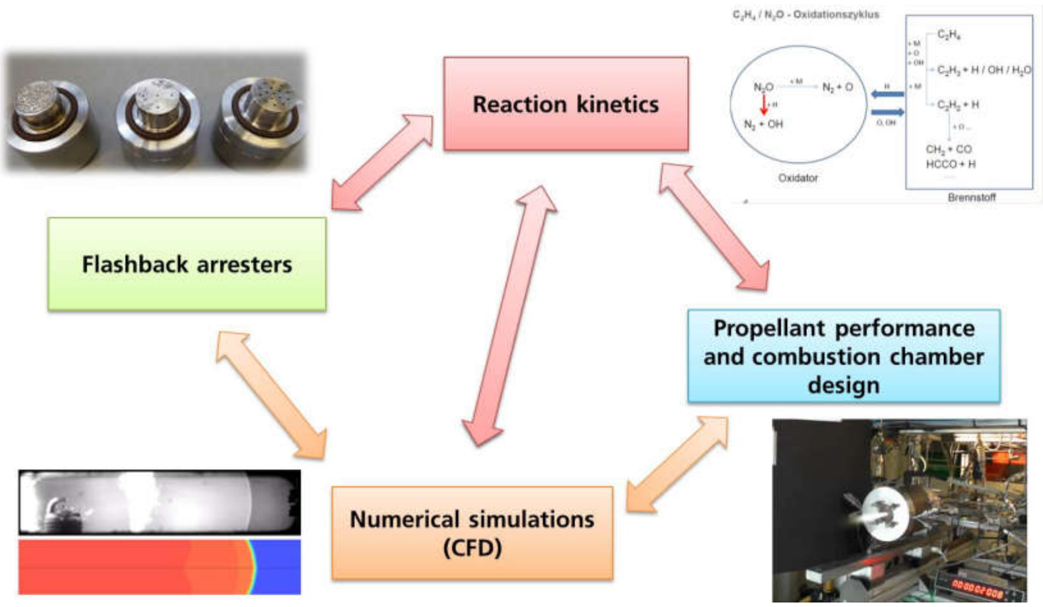

Figure 5: Working areas and interconnection in the HPGP main work package 5.2.

To solve the above mentioned challenges, DLR selected a mixture consisting of N_2O and C_2H_4/C_2H_6 called “HyNOx” (hydrocarbons mixed with nitrous oxide) for further investigation in the Future Fuels project. Tasks in the project are split in between DLR Institute of Space Propulsion in Lampoldshausen and the DLR Institute of Combustion Technology in Stuttgart.

DOI: 10.13009/EUCASS2019-276

Figure 5 shows the overall work logic of the "High Performance Green Propellant (HPGP)" main work package. A strong interconnection between the work packages and tasks is needed to solve the above mentioned challenges. Activities regarding a mixture of N_2O and C_2H_4 started during a precursor project (conducted in the years 2015-2018) and will be continued in this interdisciplinary Future Fuels project. Furthermore the gained knowledge with the N_2O/C_2H_4 propellant is used to expand the investigations on a N_2O/C_2H_6 mixture in the scope of the interdisciplinary project.
The final aim of the HPGP main work package is to design, develop and test a 22 N TRL 4 thruster under vacuum conditions at the M11 test bench [42,43] in Lampoldshausen. To achieve this aim several tasks need to be completed: By using numerical simulations the flame propagation and flashback behaviour of the propellant will be analysed, investigations on a regenerative cooling system will be used to design the cooling channels of the thruster, further investigations on flashback arresters help to assure a proper function during all operation modes, different ignition methods will be tested and the performance of the propellant (characteristic velocity c^*, specific impulse I_{sp}) will be evaluated in an experimental combustion chamber.

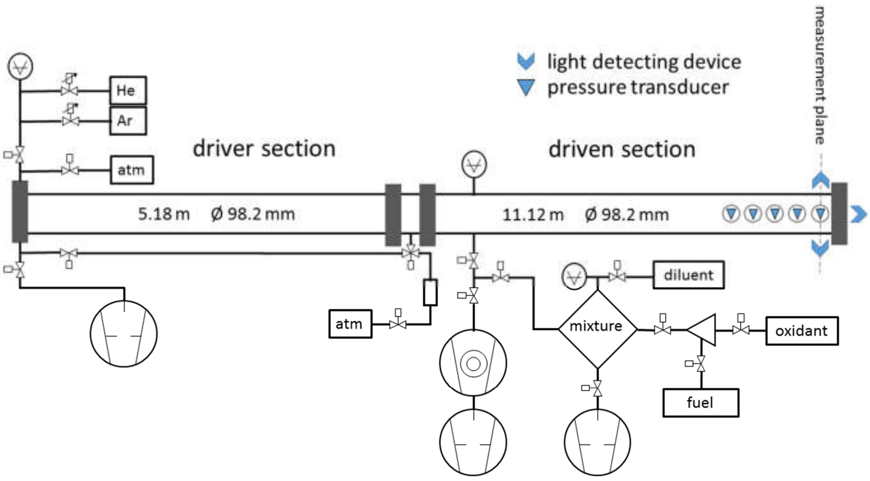

Figure 6: Sketch of DLR's Ø 98.2 mm shock tube used for ignition delay time measurements. The measurement plane is located 10 mm in front of the end plate.

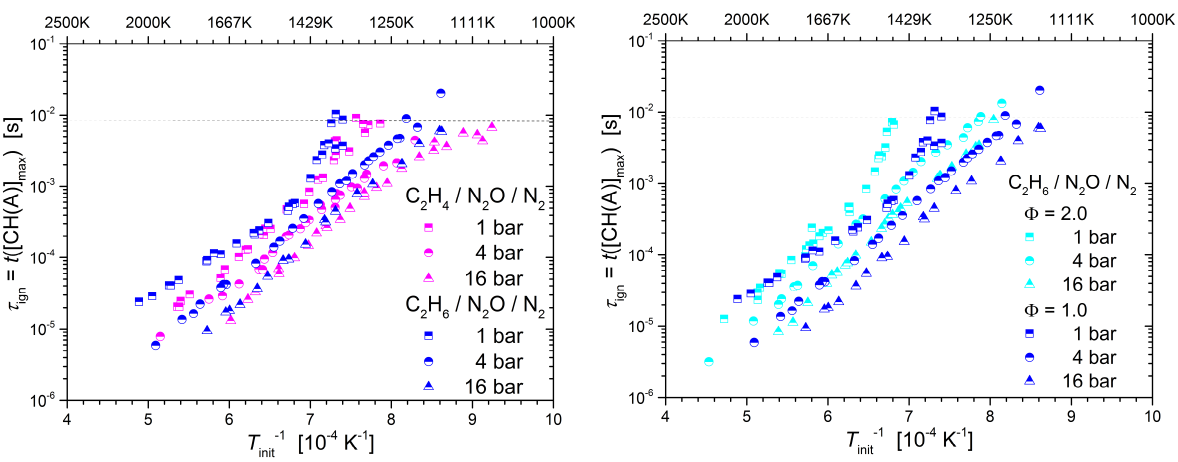

Figure 7: Ignition delay times of stoichiometric nitrous oxide / ethene and ethane – mixtures diluted 1:5 with nitrogen at initial pressures of 1, 4 and 16 bar.

Starting with the precursor project, different aims were successfully achieved. Via extensive flame speed measurements and ignition delay time experiments in shock tubes (see Figs. 6 and 7**Fehler! Verweisquelle konnte nicht gefunden werden.**), chemical kinetic reaction mechanisms for the N_2O/C_2H_4 and the N_2O/C_2H_6 mixtures, resp., were validated, optimised, and with respect to the N_2O/C_2H_4 – reactive system, a reduced mechanism has been

DOI: 10.13009/EUCASS2019-276

provided [29,30]. Subsequently this mechanism was used in numerical simulations to analyse the flame propagation mechanisms [46] and to compare the flame behaviour to results of experimental investigations. Furthermore, the chemical-kinetic reaction mechanisms were used to calculate quenching diameters and critical Péclet numbers for quenching of the N_2O/hydrocarbon flame.
In more than 500 combustion tests, the performance of a N_2O/C_2H_4 and a N_2O/C_2H_6 propellant in an experimental combustion chamber was evaluated. Here the performance (c* and c* efficiency) of the propellant depending on the mixture ratio, the characteristic chamber length (L*) and the chamber pressure was derived [39,47,48]. Furthermore heat loads on the combustor were analysed to generate a valid data set for a future regenerative cooling system [49].
In addition a specific ignition and flashback test setup was used to test porous materials and capillaries as flashback arresters [50,51]. The results of those experiments provided data to design suitable flame barriers for the experimental thruster and served as reference experiments for numerical simulations.
Figure 8shows a hot run of the experimental combustor at the test bench M11.

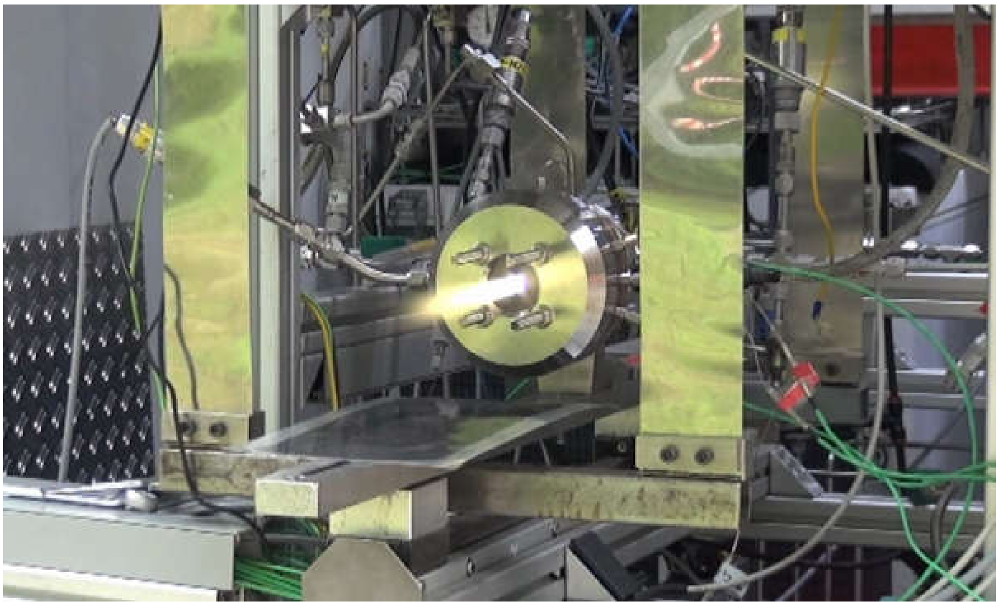

Figure 8: Hot run of experimental combustion chamber with a N_2O/hydrocarbon propellant.

During the project, a N_2O/C_2H_6 propellant will also be used non-premixed in a conventional bipropellant system. To conduct the experiments a bipropellant thruster will be designed and tested.
Furthermore throughout the project a detailed system study regarding the different classes of green propellants will be conducted. Thus the operation range of different propellants and propulsion systems will be shown and the best solution for a given mission or task can be selected.

6. Green gelled propellants (GGeRA)

Gel propellants offer the possibility to build throttleable propulsion systems with easy handling and storage characteristics. At the German Aerospace Center (DLR) at Lampoldshausen test site basic research and technology development is performed on production, rheological properties, flow behaviour, spray characteristics and combustion behaviour of gelled propellants. In context of the interdisciplinary project “Future Fuels”, novel hypergolic and green gelled propellants and associated technologies are investigated.
In recent years a growing interest in gelled propellants for propulsion applications is observable worldwide. First work on gel propulsion was performed in the USA already in the 1960s, followed by contributions in Israel since about mid of the 1990s. In Germany, basic research on green gel propulsion began at the German Aerospace Center (DLR) in Lampoldshausen in 1999 and hereafter at Bayern-Chemie and Fraunhofer-Institute for Chemical Technology (ICT) from the early 2000s on. Lately, new research was also published in e.g. China, South Korea, India and Japan. General information about gel propulsion and a summary of the status of worldwide activities in the year of publication is given e.g. in the overview reports in Refs. [52] and [53].
Gelled propellants and their specific properties are of interest for both rocket and ramjet applications due to the simplicity of implementing a highly variable “on-demand” thrust control, easy handling, possibility to tailor the

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propellant by addition of particles or even energetic materials and an improved operational safety [54]. The beneficial combination of both performance flexibility and storage characteristics merges major advantages of liquid and solid propulsion systems and is caused by the non-Newtonian flow behaviour of gels. Usually, a net-like structure is formed by the gelling agent wherein a liquid – a monopropellant, a fuel or an oxidizer – is embedded. The gelled fluid behaves like a solid at rest, but once the colloidal network is destroyed by a sufficiently high shear stress, the gel is liquefied. At very high shear rates, typically reached during the propellant injection processes, the properties of the gelled liquid become very similar to the properties of the pure base liquid itself.

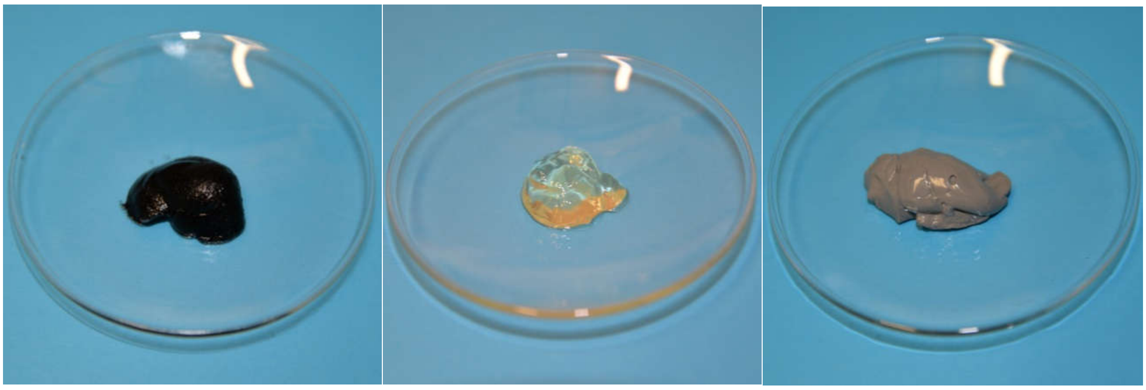

Figure 9: Examples of different gel propellant samples.

At the German Aerospace Center mainly gelled monopropellants were investigated in the former years. These gelled monopropellants consist of two or more components, which are the fuel/propellant blend, a gelling agent and partly also additives. Gelling agents like organic gelators (gelatine, agar, etc.) and inorganic particles (Aerosil, Cabosil, etc.) have been tested [55, 56] although latest work focused on Carbon-based and commercial urea-based gellants. Also first-hand experiences with the addition of metal particles (e.g. μ- and Nano-Al) were gained [57].
Recently, bipropellants and especially green hypergolic systems are moved into focus. Even though a bipropellant system is more complex than a monopropellant system, it bears many advantages. Firstly, a bipropellant system is inherently safer; hence oxidizer and fuel are neither premixed nor are contained in one molecule. Secondly, bipropellant rockets feature higher specific and density impulses than monopropellants. And lastly, a hypergolic propellant system does not require an external ignition source and can be easily reignited multiple times.
For the selection of suitable gel bipropellant candidates, reasonable performance, production, handling and treatment properties must exist or must be developed. Thereby, an important element of the propellant development in Germany is the avoidance of substances – as far as possible – that exhibit any danger to the staff in case of an accident or destruction, and the same holds for the exhaust gases. This means amongst others that the propellants should not be toxic and the sensitivity to shock and friction should be as low as possible.
In order to identify possible fuels and oxidizers for a green easy to handle storable hypergolic propellant system a set of criteria based on GHS hazard sentences was created. Additionally, the bipropellant system is required to have better specific and density specific impulses than the best existing gel monopropellant system. A detailed description of the investigation is given by Kurilov in Ref. [58]. As a first step of a screening process, two potential oxidizers were identified, i.e. hydrogen peroxide and white fuming nitric acid (WFNA). Because of its higher performance and environmentally benign exhaust products, hydrogen peroxide is preferred as oxidizer. Mixtures based on hydrogen peroxide, ammonium dinitramide (ADN) and ammonium nitrate (AN) were evaluated promising at laboratory scale before [59]. A review showed that alkaline fluids mixable with hydrogen peroxide are favourable as fuels, because their properties support its decomposition thus supporting hypergolic ignition reactions. As a result, the group of liquid methylated diamines was identified as promising high performance fuels candidates. However, because of their toxicity, scarcity and high price not all substances of that group will be investigated. Additionally, since hydrazine is the only known rocket fuel which “as is” hypergolically reacts with hydrogen peroxide, a reaction catalyst, such as a transition metal compound or a strong reducing agent, is needed to be incorporated into the fuel gel.
To quickly test a large number of fuel/catalyst combinations, a simple drop test experiment was implemented (see Figure 10). In this setup hydrogen peroxide is dropped from a height of 0.1 m into a vial with fuel gel. By means of filming the reaction with a high speed video camera it is possible to measure ignition delay times (Figure 11). With this setup various combinations of methylated diamine gels and hypergolic catalysts were tested [58]. As a result, Kurilov identified N,N,N′,N′-Tetramethylethylendiamine (TMEDA) hypergolically activated with copper-(II)-chloride as a very promising fast igniting fuel. Short ignition delay times (< 15 ms) and a good performance, which is

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comparable to the performance of liquid MMH/NTO, are actually feasible. Additionally, handling is easy and the potential fuel is available in high quantities to a fair (retail) price.

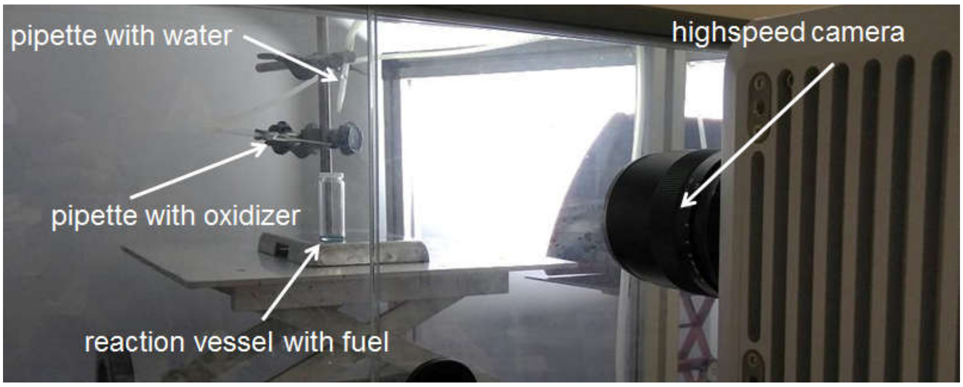

Figure 10: Test setup for drop tests (courtesy of M. Kurilov).

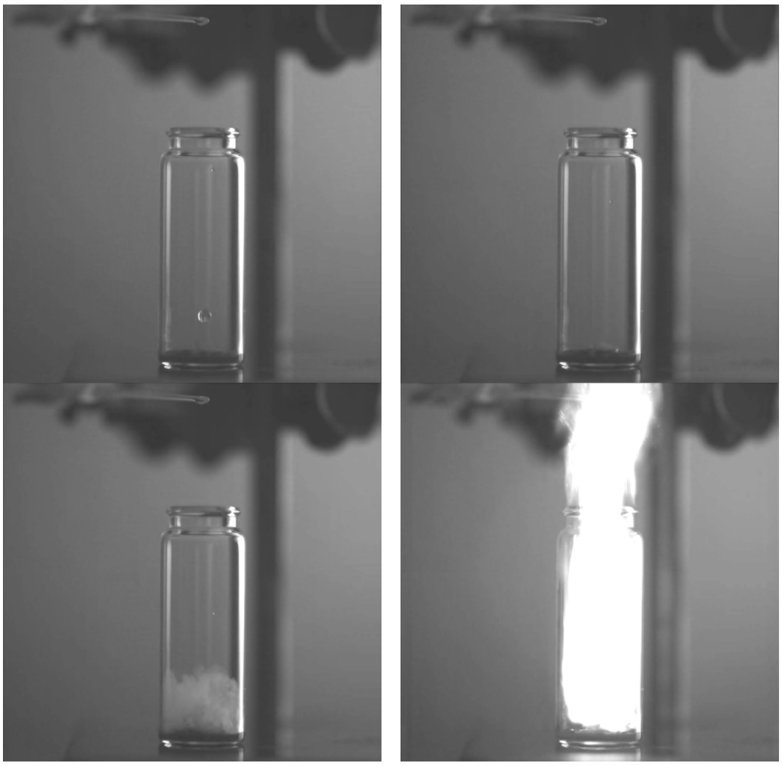

Figure 11: Hypergolic reaction (sequence from high speed video recording, courtesy of M. Kurilov).

Since with an addition of only 1 wt.-% of the ignition catalyst the goal aspired by Kurilov [58] of an ignition delay time of less than 10 ms could yet not be achieved, further pre-tests are carried out both on content und type of the catalyst. In parallel, a dedicated bipropellant model combustor setup has been designed and manufactured. First ignition and hot fire test a planned near-term in order to verify the test results achieved in laboratory.

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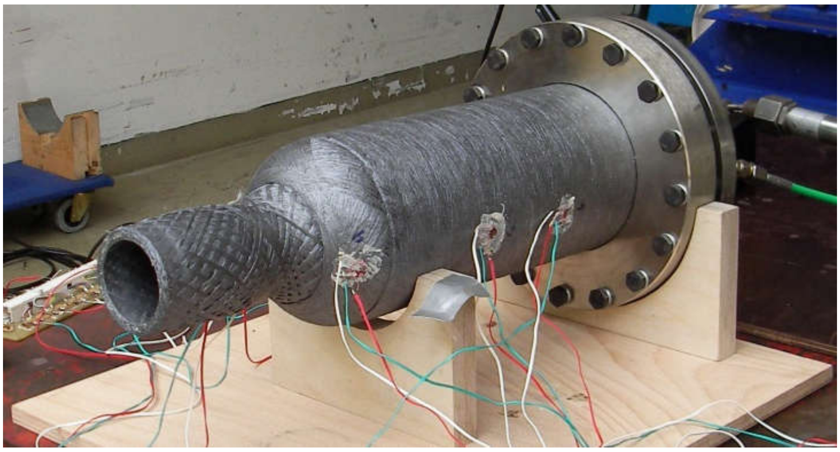

Figure 12: CMC combustion chamber and nozzle for monopropellant gel.

The compatibility of combustion chamber materials with the reactive gases and higher combustion temperatures of a bipropellant system have to be verified for the longer operation times desired. CMC-based combustion chamber designs have been successfully deployed for monopropellant gels (see Figure 12, [60]). In the context of project “Future Fuels”, the thermal management, the overall combustion chamber design including novel cooling approaches and the utilization of new and advanced materials and manufacturing methods e.g. additive layer manufacturing (ALM) and ultra-high temperature ceramic matrix composites (UHTCMC) is investigated. Because of the modularity of the combustion chamber setup, tests are envisaged as soon as first test specimen will become available.

7. Summary and conclusion

Three promising advanced green propellant systems are investigated with regard to applicability and efficiency in rocket engines within the sub-project “Advanced Rocket Propellants” (TP5) of the DLR interdisciplinary project "Future Fuels", which was started in 2018 and will last 4 years. First results have been obtained and are presented briefly in this overview paper. They are encouraging and show that these candidates are indeed interesting for the application in future propulsion systems. Nevertheless there are still important tasks to solve on the way to first applications.

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Nomenclature

c^*	characteristic velocity, [m/s]
I_{sp}	(weight) specific impulse, [s]
L^*	characteristic chamber length, [m]

DOI: 10.13009/EUCASS2019-276

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