

Investigations on Rheology, Spray and Combustion Processes of Gelled Propellants at DLR Lampoldshausen

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Abstract

Gelled propellants and their specific properties are of interest for both rocket and ramjet applications due to the capability for implementation a freely adjustable thrust control in combination with easy handling and high operational safety. The combination of performance flexibility and storage characteristics merges the major advantages of liquid and solid propulsion systems. At the Institute of Space Propulsion, German Aerospace Center (DLR), basic research on the development, the rheology, the flow behaviour, the spray properties and the combustion characteristics of gels is performed. This paper provides a general overview of the activities and presents recent achievements.

1. Introduction

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. [1] and [2].

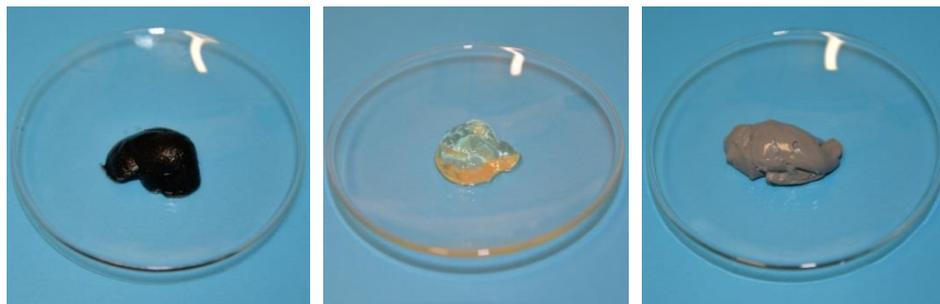


Figure 1: Examples of different gel propellant samples.

Gel propellants offer the possibility to build throttleable propulsion systems with easy handling, good storage characteristics and an improved operational safety [3]. Furthermore, gelling enables the designer to tailor the propellant with respect to the mission requirements by adding e.g. particles or energetic materials without the risk of sedimentation or separation. The beneficial combination of both performance flexibility and storability merges major advantages of liquid and solid propulsion systems and is caused by the non-Newtonian flow behaviour of gels. The gelling agent typically forms a net-like structure wherein a liquid – a monopropellant, a fuel or an oxidizer – is embedded. Therefore, 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 usually become very similar to the properties of the pure base liquid itself.

At the Institute of Space Propulsion of the German Aerospace Center (DLR) basic research and technology development is performed on production, rheological properties, flow behaviour, spray characteristics and combustion behaviour of gelled propellants. Here, the main goal is to obtain detailed knowledge of the processes within combustion chambers, injectors and feed lines, as well as compositions and production processes. This shall enable the realization of reliable design processes for future gel rocket motors with high power densities and stable operating envelopes at reduced costs and a lower demand for testing. The investigations are carried out within the frame of institutional funding e.g. in context of the interdisciplinary project “Future Fuels” or the Defence & Security Research at DLR, but are as well partly covered by contractual research. This paper gives an overview on activities, which have been conducted and published in the last few years. For more detailed information an interested reader is encouraged to consult the references given in the text.

2. Infrastructure

For the research purposes on advanced and novel rocket propellants and high-speed air-breathing propulsion the Institute of Space Propulsion, Propellants Department, operates the test complex M11, the physico-chemical laboratory at M3 and the and production facility G49, which are located at the DLR site in Lampoldshausen. These facilities provide the necessary infrastructure and staff for the research efforts conducted on gelled propellants.

2.1 Test Facility M11

The first part of the test complex M11 with two test cells and a small office and laboratory building was already built in 1966. Since then, research and testing in the field of rocket and ram-/scramjet propulsion were carried out here. In order to be able to face new challenges, the facility has been enlarged and refurbished several times. Current work is focused on research in the field of advanced green propellants, gelled propellants, hybrid propulsion and scramjet propulsion. Thereby, test activities both for DLR-internally and externally funded projects as well as contract R&D work for partners from industry, other research organizations, universities and agencies are performed.



Figure 2: Test complex M11 at DLR site Lampoldshausen

The test complex M11 (Figure 2) consists of two test cells with two test positions each (M11.1 to M11.4) and the research and student test field (M11.5). Each test position at the M11 is equipped with newest data acquisition and control systems and enables a specific kind of test conditions while ensuring high flexibility. The test facility provides 20 MPa gas supply systems for H₂, O₂, N₂ and pressurized air. All valves and pressure regulators of the test bench are controlled by a redundant Siemens PLC. Additionally, each test position features a versatile measurement system which can easily be configured to meet very specific test requirements. This offers the possibility to conduct combustion, flow and spray tests with a wide range of different rocket propellants. Furthermore, the simulation of relevant ram-/scramjet combustor inflow conditions or generally the testing of various propulsion hardware components and materials is possible.

The test position M11.4 is exclusively used for investigations on the combustor processes i.e. injection, atomization, evaporation and combustion of gelled propellants. The gelled propellants are stored in easily exchangeable piston-type accumulators, so-called cartridges. Using hydraulic drives with a continuously adjustable control, the gels are fed with exact and freely selectable mass flow rates through the feed pipes to the injectors. For a reliable and smooth ignition a H₂/O₂ gas torch igniter is normally used. Additionally, the start of combustion may be boosted by an auxiliary oxygen injection. A 100 l / 1 MPa water tank is available for emergency cooling and purging. The test position is also equipped with a high pressure water cooling system providing up to 1.2 m³ H₂O at up to 20 MPa. A comprehensive measurement instrumentation including thrust, pressures and temperatures as well as optical diagnostics is present. The DAQ system allows currently the simultaneous acquisition of up to 160 channels. It also features some unique capabilities e.g. an integrated test database and redline monitoring.

2.2 Physico-Chemical Laboratory at M3 and Production Facility G49

The physico-chemical laboratory at M3 (Figure 3) is of central importance concerning the provision of specific lab services and in its function as a research establishment. The emphasis is on chemical analysis, development and prequalification of new advanced propellants or fuels and oxidizers. For this purpose, the M3 laboratory wing is equipped with a large number of different analytical instruments, ranging e.g. from trace metal analytics to comprehensive surface analysis. This opens up comprehensive new perspectives for the fundamental understanding of the propellants' chemistry in the solid, liquid or gaseous state as well as its physical properties at different loads.



Figure 3: Part of the physico-chemical laboratory at M3

For preparing of propellants and handling of substances, which could be subject of the German Explosives Act, the laboratory staff also operates adjacent to test complex M11 the so-called production facility G49. The main task is to provide experimental liquids and gels for spray and hot fire testing of internal and external customers. The amounts of propellants, which can be handled here, are usually on the lab-scale and may come up to a 2.5 kg TNT equivalent. Two dissolver apparatuses are available for the production of gelled or particle-loaded fuels and oxidizers. They are especially suitable for the production of particulate gels because they are capable to generate high shear rates necessary to disperse e.g. metal particles.

3. Key Topics of Research on Gel Propellants at German Aerospace Center (DLR)

3.1 Monopropellant Gels

For successful deployment of new propellants, reasonable performance, production, handling and treatment properties must exist or must be developed. An important element of the propellant development in Germany is as far as possible the avoidance of harmful substances. This means for example that neither the propellants nor exhaust gases shall be carcinogenic, mutagenic or toxic for reproduction. Otherwise, toxicity shall be as low as possible. So, need for special procedures and personal protective equipment for manufacturing and operating may be reduced to a minimum.



Figure 4: Hot fire test with Aerosil-based gel (left) and a Carbon-based gel (right)

At the German Aerospace Center (DLR) 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 (i.e. Aerosil, Cabosil) have been tested [4][5] although the focus was later set on Carbon-based particulate and commercial urea-

based gellants (cf. Figure 4). These so-called “2nd generation” gel propellants feature high performance and good delivery characteristics with less than 8% of admixtures. Also first-hand experiences with the addition of metal particles (e.g. μ - and Nano-Al) were gained [6].

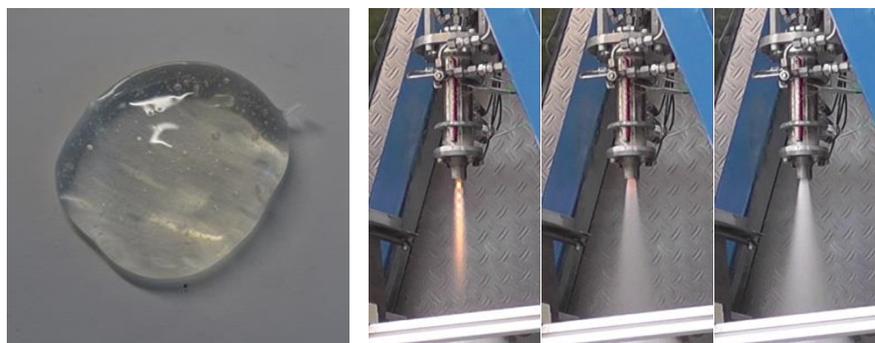


Figure 5: Test sample of a particle free gelled monopropellant (left) and sequence from an ignition test (right)

Most recent research on monopropellant gel addresses requirements for special applications. For the purpose of optical investigations on injection, atomization and flame structure of gelled propellants new particle free gel compositions are in development (Figure 5). Such a gel may also be useful for applications like gas generators and reaction control systems. In contrast, for the use in booster and sustainer stages also metallized “high-performance” propellant gels were tested lately. Another focus of current work is the development of monopropellant gels with high insensitivity and/ or extended operational envelope. Here, great progress was made by use of ionic liquids.

3.2 Bipropellant Gels

At German Aerospace Center (DLR) bipropellants and especially green hypergolic systems moved again into focus recently. 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 is the energy released through an exothermic chemical decomposition of a single 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 therefore be easily reignited multiple times.

In order to identify possible fuels and oxidizers for an environmental-friendly “green”, easy to handle and storable hypergolic propellant system, a set of criteria based on GHS hazard sentences was created [7]. Additionally, the bipropellant system is required to have appropriate specific and density specific impulses. As a first step of the screening process, two potential oxidizers were identified, i.e. hydrogen peroxide (H_2O_2) and white fuming nitric acid (WFNA). Preliminary studies also proofed sufficient gelation of both using Aerosil (see Figure 6). Because of its higher performance and environmentally benign exhaust products, hydrogen peroxide is favourable as oxidizer. Mixtures based on hydrogen peroxide, ammonium dinitramide (ADN) and ammonium nitrate (AN) were evaluated promising at laboratory scale before [8].

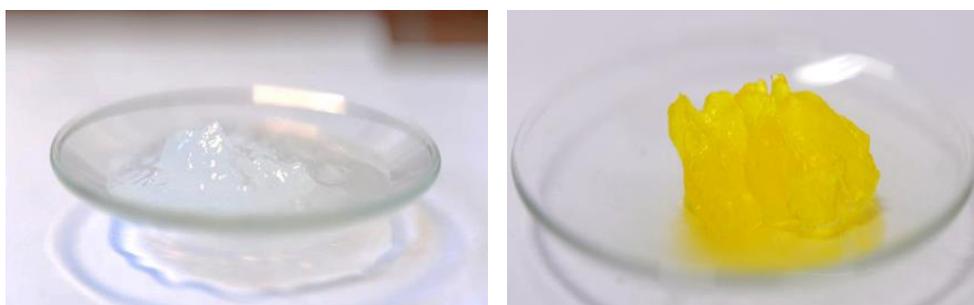


Figure 6: Gelled hydrogen peroxide (left) and gelled nitric acid (right)

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. In order to enable hypergolic reaction with hydrogen peroxide, an additional catalyst, such as a transition metal compound or a strong reducing agent, is needed.

To quickly test a large number of fuel/catalyst combinations, a simple drop test experiment was implemented (cf. Figure 7). In this setup, hydrogen peroxide is dropped from a height of 0.1 m into a vial containing the fuel gel. By filming the reaction with a high speed video camera, the ignition delay times are determined. With this setup various combinations of methylated diamine gels and hypergolic catalysts were tested [7]. As a result, N,N,N',N'-Tetramethylethyldiamine (TMEDA) activated with copper-(II)-chloride was identified as a very promising fast igniting fuel. Ignition delay times in the order of 15 ms and a performance comparable to the one of common monomethyl hydrazine/ nitrogen tetroxide seem to be feasible (Figure 8). Additionally, handling is easy and the potential fuel is available in high quantities to a reasonable price.

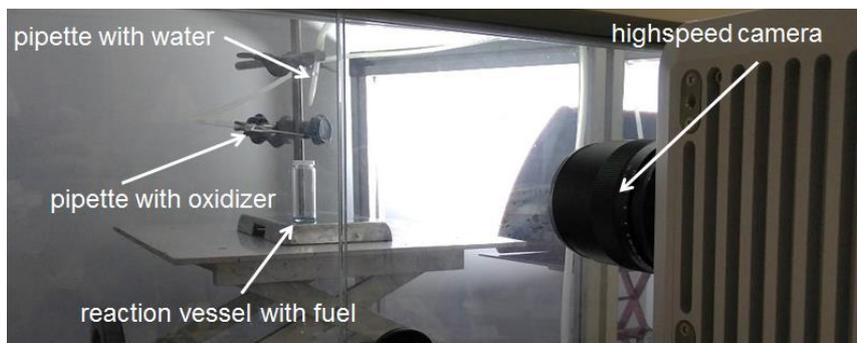


Figure 7: Test setup for drop tests.



Figure 8: Hypergolic reaction of TMEDA gel with hydrogen peroxide (sequence from high speed video recording).

Further tests both on content and type of the catalyst are carried out. In parallel, a dedicated bipropellant model combustor setup has been designed and manufactured. First ignition and hot fire tests are planned near future in order to verify the test results achieved in laboratory.

3.3 Combustion Performance

A set of modular, capacitively cooled combustion chambers with different diameters and lengths is used for assessing the combustion performance of new gel propellants. In the past, a round combustion chamber with an inner diameter of approx. 21 mm (called BK21) was used for comprehensive investigations on the optimum ratio of combustion chamber volume and nozzle throat area V_c/A_t i.e. the optimum characteristic length L^* of the combustor. Therefore, the combustion chamber length was varied from 400 mm down to 80 mm, which is equivalent to a variation of L^* between 7 m and 1 m taking into account different nozzle configurations applied. In comparison, hot fire tests with the larger BK50 with an inner diameter of 50 mm were conducted. With the smaller combustion chamber, for the monopropellant gel under investigation an optimum L^* of 1.5 m for 70 bar combustion chamber pressure and 1.8 m for 50 bar was determined, albeit a minimum pressure of 40 bar was required for a stable and self-sustained combustion. In contrast, hot fire tests with the BK50 showed generally lower combustion efficiency with an optimum characteristic length of approx. 7.5 m but with a more extensive operational pressure range. The enhanced efficiency of the smaller combustion chamber has been attributed to beneficial spray-wall-interactions increasing the heat exchange within the evaporation-driven combustion process (cf. [10][11]).

The assumption of favourable spray-wall-interactions was supported by tests using the newly designed and built combustion chambers BK20, BK30 and BK40 with 20 mm, 30 mm and 40 mm inner diameter, respectively (Figure 9). By their handy and versatile design featuring e.g. additional pressure sensors and wall temperature

measurements these combustion chambers are very valuable for characterization of combustion and performance. Actually, BK40 became a workhorse and reference configuration for the comparison of different gel formulation at DLR. It also serves as the base for derivatives e.g. for the investigations on gelled bipropellants (BK40ZS)

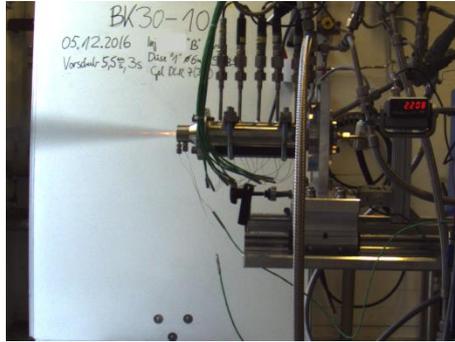


Figure 9: Hot fire test using combustion chamber BK30.

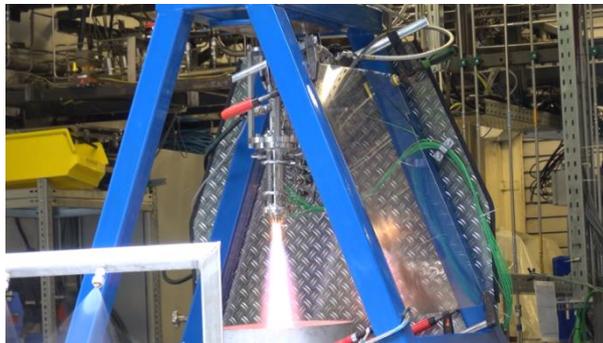


Figure 10: Combustion chamber BK40 is fired at vertical test stand.

In order to investigate the behaviour and performance of metallized and other particle-loaded propellants under application-relevant conditions, a vertical test stand (VTS) was erected at M11.4. A hot fire test at the VTS with an Aluminium-loaded propellant is shown in Figure 10. The stand enables testing of versatile propellants reducing the risk of depositions in the combustion chamber and the risk of a clogging of the nozzle. This configuration has significantly enhanced the test capabilities for gelled propellants in Lampoldshausen.

3.4 Optical Analyses

Two different model combustion chamber designs with windows are available and allow access for optical diagnostic tools allowing enabling the visualization and investigation of the combustor processes. Preliminary tests with a round chamber demonstrated the feasibility and the benefits of a visual examination of the gel injection and combustion using e.g. shadowgraph technique [10]. Taking into account experiences from these tests regarding e.g. field of view and fouling of the windows, a modular rectangular chamber was designed, manufactured and successfully deployed [12][13].

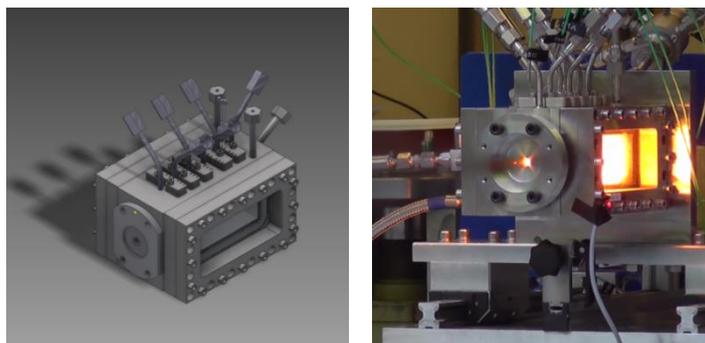


Figure 11: Rectangular gel rocket combustion chamber with optical access (left: sketch; right: hot fire test).

Table 1: Main characteristics of rectangular gel rocket combustion chamber with optical access

| Dimensions | | | |
|---------------------------|--------------|-------|-------------|
| Length | L | [mm] | 120 |
| Width | W | [mm] | 80 |
| Height | H | [mm] | 50 |
| Throat Diameter | d_t | [mm] | 6; 8; 10 |
| Pressure | | | |
| nominal | $p_{c,nom}$ | [MPa] | 5.0 |
| maximum | $p_{c,max}$ | [MPa] | 8.0 |
| Characteristic Length | L^* | [m] | 17; 9.5; 6 |
| Chamber Contraction Ratio | ϵ_c | [-] | 140; 80; 51 |

The rectangular combustor with optical access as shown in action right-hand-side in Figure 11 is made of stainless steel and features both an interchangeable nozzle and a changeable single injector. Main design data is provided in Table 1. For the window, sapphire was chosen due to its superior mechanical and thermal properties. The sapphire window is preinstalled in a metallic frame simplifying the handling at the test facility and reducing changeover times in between tests. The design is also very versatile with respect to test configurations since the sapphire can easily be replaced by dummy elements, dedicated measurement segments or material samples. With the exception of a nitrogen purge / film cooling system for reducing the deposition of soot on the window and to limit the heat loads during start-up, no active cooling is foreseen. Therefore, the test time is limited to a few seconds.

Optical investigations carried out so far comprise e.g. shadowgraph and true image video recordings using either a general purpose industrial camera or a high speed black-and-white camera. Different filters were applied where appropriate. Snapshots out of the recorded high speed videos from the ignition of the gel propellant (boosted with gaseous oxygen) are given in Figure 12. The image indicates the zone, where the auxiliary oxygen and the gelled propellant react during the start-up phase. Otherwise, analysis of the flame pattern has been hampered by recirculation zones and radiation from soot coming from the carbon-based monopropellant gel used for these tests. Here, new and more significant results are expected once the new particle free propellants are fully characterized and available in sufficient quantity.

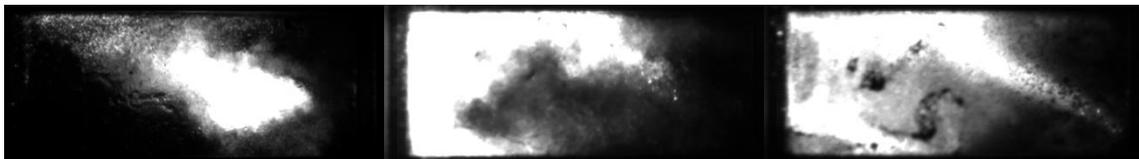


Figure 12: Gel ignition, sequence from high speed camera recordings (flow direction from right to left).

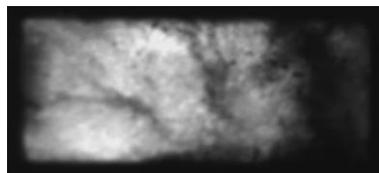


Figure 13: Steady-state operation; radiation from soot and recirculation hamper analysis (flow direction from right to left).

Detailed investigations were conducted on the spray behaviour of gels using optical measurements in the past. The atomization of the propellant into small droplets and therewith a high free surface is favourable and mandatory for a quick conversion of the propellant within the small combustor. However, some gels cannot be sprayed to small droplets but large threads are formed instead. As shown by Negri [14], this behaviour is caused by viscoelastic effects. While these investigations on the spray behaviour have mostly been performed at ambient or low pressure only, tests on spray formation and combustion also at high pressures are pursued. Preliminary studies have been conducted using the rectangular combustion chamber with optical access. Since data quality will suffer from spray mist or droplets interacting with the windows, a dedicated pressure vessel with a large diameter is currently designed in-house maximizing the distance between injector and pressure window.

3.5 Rheology and Pressure Loss

From a rheological point of view a gel can be defined as a viscoelastic shear-thinning fluid with a storage modulus higher than its loss modulus if exposed to oscillatory conditions. From an application point of view with respect to rocket and ramjet propulsion, the gelled propellant should behave similar to a solid under storage conditions and similar to a liquid when a sufficiently high shear stress is applied. The gel should be easily sprayable to small droplets and necessary feeding pressures should be low to moderate only.

The flow behaviour of non-Newtonian fluids shows several differences to Newtonian fluids. Newtonian fluids have a parabolic velocity profile for flows through tubes of constant diameter under laminar, fully developed, incompressible and steady state conditions. In contrast, shear-thinning fluids show a “broader” velocity profile with lower velocities at the centreline but steeper gradients near the tube wall. A flow region with a constant velocity in a distinct area of the tube cross section around the tube centreline may occur when the shear stresses τ are being lower than the yield stress τ_0 of the fluid. The size of this so called plug flow region can be described by the ratio of the yield stress to the stress at the wall τ_0/τ_W . This distinct flow behavior has to be considered for the design of feed pipes, injectors and valves and also affects the heat transfer and heat transport properties. Therefore, for the development of gel propulsion systems a very detailed knowledge of non-Newtonian rheological and physical properties and of the flow and spray behaviour of gels is an essential base.

The three most relevant aspects of the rheology of a gel (the shear thinning behaviour, the upper Newtonian plateau and the yield stress) are described using the so called Herschel-Bulkley Extended (HBE) equation, as depicted in Eq. 1, which was evolved and verified at DLR [15][16][17].

$$\eta_{HBE} = \frac{\tau_0}{\dot{\gamma}} + K\dot{\gamma}^{n-1} + \eta_\infty \quad (1)$$

This equation describes the dependence of the dynamic shear viscosity η on the shear rate $\dot{\gamma}$ with a satisfying accuracy in the whole propulsion relevant shear rate range.

In a dedicated experimental setup (see Figure 14 left) the behaviour of gels while flowing through an injector-like geometry has been examined [18][19][20]. The aim of this work is to understand pressure losses and the specific flow behaviour of gels flowing through a constriction within a tube. The test sections (cf. Figure 14 right) feature a reduction of diameter from $d_1=12$ mm to a diameter d_2 between 4 mm and 8 mm at an angle of 118° . This simple geometry was chosen because it is relevant for the feeding system of rocket engines.

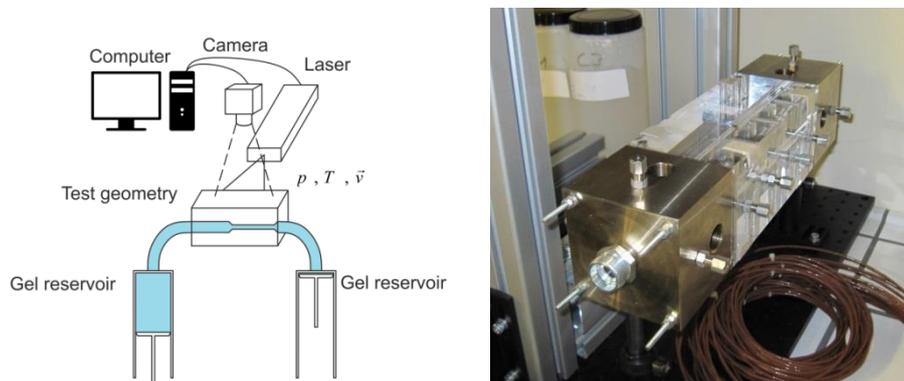


Figure 14: Sketch of test setup for investigation of pressure loss and flow behaviour (left). Measurement section made of acrylic glass (right).

A Cellosize based gel (Gel C), an Aerosil based gel (Gel A), a Thixo-2 based gel (Gel T) and – for reference – a Newtonian fluid have been investigated. As expected, the pressure losses follow predominantly a quadratic equation as it is known for Newtonian fluids from literature [21]. However, for certain load points the pressure loss of the gelled fluid may even be lower than the pressure loss for the corresponding Newtonian fluid. A correlation was found for the pressure loss coefficient ζ with respect to the generalized Reynolds number determined by means of the Herschel-Bulkley Extended (HBE) equation (Figure 15). This enables the prediction of pressure losses from flow checks using substitutes.

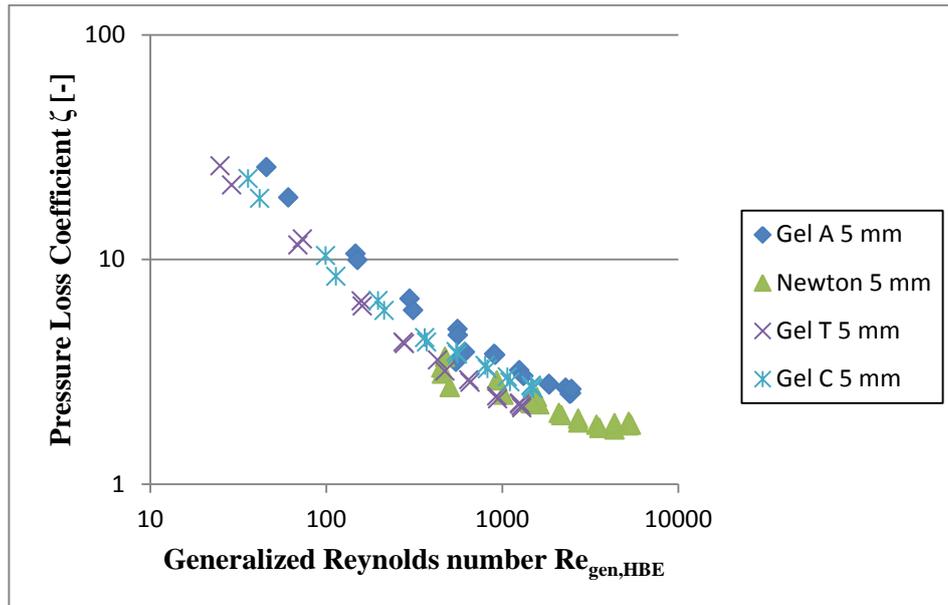


Figure 15: Pressure loss coefficient ζ with respect to generalized Reynolds number $Re_{gen,HBE}$ in pipe for target diameter $d_2=5$ mm.

From optical investigations by means of particle image velocimetry (PIV) three different flow regimes have been identified. As expected, the Newtonian fluid shows recirculation zones (“vena contracta”) after the contraction which enlarge when velocity is increased. Some gels show a very similar behaviour but with usually smaller recirculation areas. A second regime is characterized by oscillating streamlines i.e. an unstable flow behaviour. Since these instabilities may trigger malicious viscoelastic effects in the injector, this flow regime shall be avoided. Finally, the third flow regime is denoted by a highly viscous fluid in the centre of the pipe (“plug”) but a low viscous fluid near the wall. Thus, the flow near the wall may pass the contraction easier resulting in higher velocities in the outer flow upstream the constriction.

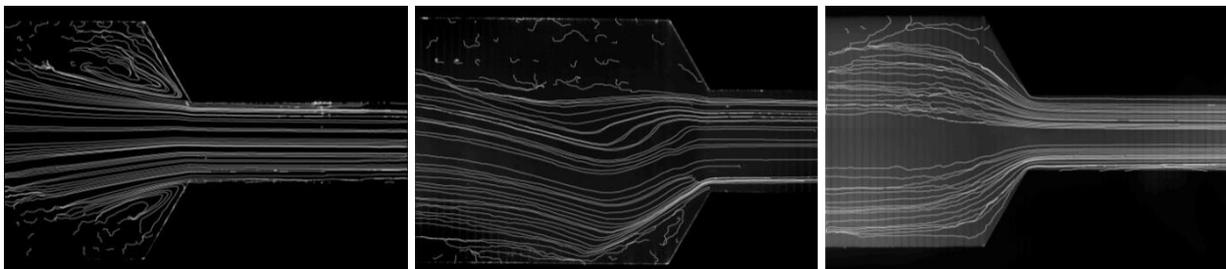


Figure 2: Flow regimes: Gel flow with recirculation areas (left), oscillating flow (middle), gel flow without recirculation areas but high speed near the wall (right)

Extensive testing has been performed and the data is currently evaluated. The obtained information will help to design and optimize injector flow paths as well as control valves for future gel propellant rocket motors.

4. Conclusion

Gel propellants offer the possibility to realize propulsion systems, which are easy to throttle but have simple handling and storage characteristics. At the Propellants Department of the Institute of Space Propulsion, German Aerospace Center (DLR), in Lampoldshausen basic research on the development, the rheology, the flow behaviour, the spray and the combustion behaviour of gel propellants is performed. The aim of the activities is the maturation of the gel propulsion technology as well as the improvement of the performance, safety and storage characteristics with the scope to various applications and gel motor types.

Acknowledgments

Parts of the presented work are embedded in the Defence & Security Research at DLR conducted in the context of the DLR project on “Innovative Technologies and Methods for Missiles” (ITEM-FK). Novel hypergolic and green gelled propellants are investigated in the frame of the interdisciplinary project “Future Fuels”. The authors would like to thank their colleagues from the various DLR institutes for the exchange and good collaboration.

The financial support by Bayern-Chemie for the presented work on gel flow behaviour is kindly acknowledged.

The authors also thank the colleagues of Fraunhofer ICT and the DLR physico-chemical laboratory for producing the gel propellants and performing all necessary chemical analyses as well as the DLR M11 test complex team for supporting and conducting the hot fire tests.

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