# A Contribution to the Characterization of Rheological and Flow Behavior of Gel Fuels with Regard to Propulsion Relevant Conditions

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## **Abstract**

A detailed knowledge about the rheological and the flow behavior of gel fuels and propellants is necessary for the characterization of their spray properties, which is a base for the development of efficient ramjet and rocket combustor processes. Gelled fuels behave as non-Newtonian fluids and show decreasing shear viscosity values with increasing shear rates. The present publication gives detailed information about relevant characteristics of selected gel fuels. The results include the description of the shear viscosity dependence by an extended version of the Herschel-Bulkley equation, the determination of elongational viscosities and the characterization of the flow behavior of gels in tubes of constant diameter by a generalized Reynolds number and a critical Reynolds number. The spray behavior of various droplet forming gels is characterized by a regime diagram with generalized Reynolds and Weber numbers.

# 1. Introduction

Gelled fuels, propellants or propellant combinations are of increasing interest for rocket and ramjet propulsion systems especially in the last two decades, because of their safety and performance benefits. Their non-Newtonian flow behavior, which can be shown for example by the shear-rate dependency of the shear viscosity, offers the possibility to build engines, which both can be throttled similar to engines with liquid fuels and which have similar simple handling and storage characteristics like engines with solid fuels.<sup>1-3</sup>

The addition of gelling agents to conventional liquid fuels, which commonly behave as Newtonian fluids, and the conduction of a gellation process changes the rheological properties of these liquids dramatically. Without any applied shear stress gels are more or less incapable of flow, because of their very high dynamic shear viscosity values and their often existing distinct yield stress. Applying high shear rates during the injection process, however, it is possible to reach relative low viscosity values and possibly even liquefaction in the area near the injector exit plane. Thus an atomization process, which shows similarities to the atomization of conventional liquid fuels in a large range, is possible for distinct injection conditions and set-ups as previous experiments have shown. But it has to be mentioned that this process is more difficult to conduct than in the case of pure (and ungelled) liquid fuels.<sup>1,3-6</sup>

For a detailed description of flow and spray characteristics, which is essential for the development of an effective combustion process within a limited combustor length, a better understanding of the basic processes of the flow behavior is necessary, because these processes are strongly correlated to the rheological properties of the used gelled fuels and oxidizers. For the characterization of flow and spray processes dimensionless numbers are commonly used. For example the Reynolds number, which can be interpreted as the ratio of the inertial forces to the viscous forces, is written for Newtonian fluids as  $Re = \rho u L / \eta$ 

( $\rho$  density, u velocity, L characteristic length,  $\eta$  dynamic shear viscosity). For non-Newtonian fluids this formula cannot be used because of its constant viscosity value  $\eta$ . Furthermore not all gels can be sprayed to small droplets as can be seen for example later in Fig. 7. This behavior cannot be explained only by the influence of shear viscosity and surface tension. Furthermore in tapered geometries, which exist in feeding lines and injectors, effects occur, which can be correlated to the extensional viscosity. Thus pressure losses for distinct gels and distinct flow conditions depend not only on shear viscosity influenced processes.

In the last years significant progress has been made in basic research as well as in work on propulsion system development and demonstration. This can be seen both in various publications (see e.g. Refs. [1, 6-12]) and in the conduction of complete sessions, which are dedicated to the gel propulsion theme, at international conferences (e.g. EUCASS 2006, AIAA Joint Propulsion Conference 2008).

The present publication shows relevant steps on the way to characterize gel spray properties, which are results of the basic research activities at DLR - Institute of Space Propulsion. The paper includes various aspects of rheology, flow behavior and spray characterization. Finally a Regime diagram will be given for droplet producing gels. In this context it has to be mentioned that some of the used characterization tools in the present publication were presented in previous publications, but the here obtained results are goal-oriented to the here presented gels with its different gellants.

# 2. Experimental Setup

# 2.1 Gel production

A Getzmann dissolver stirrer apparatus was used for the production of gelled fuels. In the present publication 4 different gel test fuels TF1 to TF4 were mainly used, whose composition is presented in Table 1. Jet A-1 (kerosene), paraffin and ethanol were chosen as the basic liquids to be gelled. One inorganic and two organic gellants were used for this investigation. Aerosil-200 is hydrophilic fumed silica from Evonic Industries, which consists of particles of about 12 nm average diameter. Thixatrol ST is a castor oil derivative from Rheox, which was used together with 5-Methyl-2-Hexanone (Miak) for the vehicle/solvent mixture for the gellation process. Methocel-311 consists of hydroxypropylmethyl cellulose and was delivered from Dow Chemical. All gels were produced under vacuum condition to avoid having air bubbles within the gel.

	Fuel	Gellant	Additive
TF1	85 % paraffin	7.5 % Thixatrol ST	7.5 % Miak
TF2	96 % paraffin	4.0 % Aerosil-200	-
TF3	85 % Jet A-1	7.5 % Thixatrol ST	7.5 % Miak
TF4	96.5 % ethanol	3.5 % Methocel-311	-

Table 1: Composition in wt.-% of investigated gel test fuels.

#### 2.2 Rheometrical equipment

For the determination of the rheological properties two different rheometers were used. In the shear rate range up to approximately  $\dot{\gamma}=10^4~\text{s}^{-1}$  the shear viscosity was determined with a Thermo Haake Rheo-Stress 1 rotational rheometer, whereas a plate and cone geometry with a diameter of 35 mm and a cone angle of 2° was chosen for the measurements. Due to centrifugal effects in the rotating cone-and-plate set-up a Rosand RH2000 capillary rheometer was used to determine the viscosity characteristic at higher shear rates. The measurements were conducted up to approximately  $\dot{\gamma}=10^6~\text{s}^{-1}$  for TF1 - TF3 and up to  $2\cdot10^5~\text{s}^{-1}$  for TF4. Bagley correction was applied to the measured data as well as the Weissenberg-Rabinowitsch

correction. Wall slip effects were determined to be negligible. A detailed description of the used corrections is given in Refs. [13, 14].

# 2.3 Experimental setup for spray investigations

Figure 1 shows a sketch of the experimental set-up for the spray investigations. The gelled fuel to be investigated is stored in a cartridge. It is fed to the injector unit by moving a piston inside the cartridge with a remote controlled hydraulic driving unit so that the gel is pushed with a pre-selected volume flow rate through the pipe, which connects the cartridge with the injector unit. The pressure inside the cartridge and in the injector unit is monitored by pressure gauges.

For the spray investigation presented here, a doublet like-on-like impinging jet atomizer set-up was chosen. This injector type is often used in liquid rocket engines operated with storable Newtonian fuels due to its simplicity and its good atomization and mixing characteristics. The modular injector set-up allows easy variation of impingement angle, impingement distance and injector exit diameter. All experiments presented in this publication were conducted with the same injector exit diameter D = 0.7 mm, a distance of 10 mm between the injector exit plane and the impingement point, and an impingement angle between the two jets of  $2\theta = 90^\circ$ . The shadowgraph technique was applied for the visualization of the spray behavior together with Xenon flash lights with a flash duration of 150 ns (FWHM) and CCD cameras with a maximum resolution of 1024x1024 pixels. The two used shadowgraph systems were perpendicular oriented so that the atomization behavior could be observed from two sides. Further information about the set-up and the injector design is given e.g. in Refs. [10, 16].

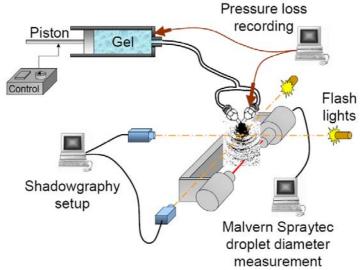


Figure 1: Experimental setup for spray investigations

#### 3. Results and discussion

#### 3.1 Rheological characteristics

Figure 2 shows the shear viscosity dependence upon the shear rate of the four investgated gel test fuels TF1 - TF4 in log-log diagrams. It can be seen that at low and medium shear rates  $\dot{\gamma}$  decreasing viscosity values  $\eta$  occur with increasing shear rates. At high shear rates, however, the slope flattens up to a constant value  $\eta_{\infty}$ , which is called upper Newtonian plateau. The dashed lines of constant viscosity in the diagrams

indicate the viscosity of the ungelled basic liquids, which behave as Newtonian fluids. In most cases  $\eta_{\infty}$  is near the viscosity value of the ungelled basic liquids. Only the ethanol/Methocel gel (TF4) shows a distinct difference. It has to be mentioned in this context that shear rates at about 0.01 s<sup>-1</sup> are significant for storage characteristics. Shear rates higher than  $10^3$  s<sup>-1</sup>, however, occur for example in injector units and are relevant for the spray behavior.

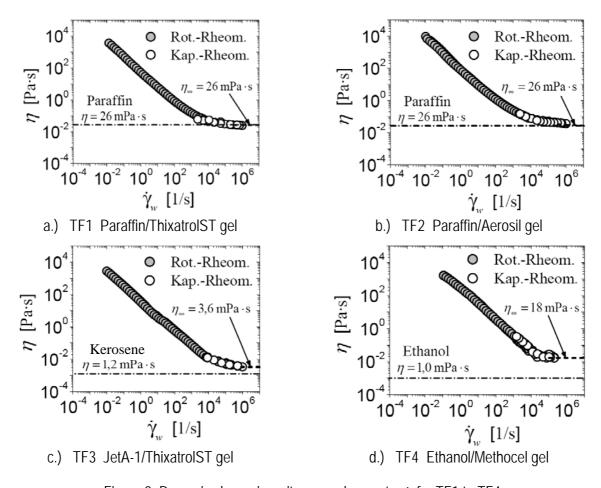


Figure 2: Dynamic shear viscosity  $\eta$  vs. shear rate  $\dot{\gamma}$  for TF1 to TF4

		TF1	TF2	TF3	TF4
		Paraffin/Thixatrol	Paraffin/Aerosil	JetA-1/Thixatrol	Ethanol/Methocel
$ au_0$	[Pa]	45	83	33	360
Κ	[Pa·sn]	5.07	2.54	11.76	1.19
n	[-]	0.38	0.57	0.19	0.16
$\eta_{\scriptscriptstyle \infty}$	[Pa⋅s]	0.026	0.026	0.0036	0.018
Re <sub>crit HBE</sub>	[ - ]	2315	2355	2415	2319

Table 2: HBE parameters of the four investigated gel test fuels TF1 – TF4. Additionally the critical HBE Reynolds number is given for injector orifice diameters of D = 0.7 mm.

The viscosity characteristics of the here and in other publications presented gels can theoretically be approached over the entire propulsion relevant shear rate range of  $10^{-2}$  s<sup>-1</sup> <  $\dot{\gamma}$  <  $10^6$  s<sup>-1</sup> with the Herschel-

Bulkley-Extended equation (HBE). This equation is presented in Eq. (1) and was introduced by Madlener and Ciezki<sup>17,18</sup> as an extended version of the well-known Herschel-Bulkley law, which considers only the power-law range and the yield stress  $\tau_0$ . The HBE equation, however, considers additionally the viscosity characteristic in the high shear rate range with its constant viscosity parameter  $\eta_{\infty}$ . Table 2 presents the HBE parameters, which were determined for the four test gel fuels.

$$\eta = \frac{\tau_0}{\dot{\gamma}} + \mathbf{K} \cdot \dot{\gamma}^{n-1} + \eta_{\infty} \tag{1}$$

Furthermore gels show partly a distinct resistance to elongational flows, which occur in tapered geometries like in injectors. From capillary rheometer measurements also extensional (or elongational) viscosities  $\eta_E$  in dependence upon the strain rate  $\dot{\varepsilon}$  were determined making use of the method of Cogswell.<sup>19</sup> It can be seen in Fig. 3 that the kerosene (Jet A-1) and the paraffin based gels show decreasing values with increasing strain rate in the low and medium strain rate range. Furthermore a distinct minimum can be seen for these gels. The Ethanol/Methocel gel, however, shows significantly higher  $\eta_E$  values. This different behavior will later be discussed in connection with the spray investigation.

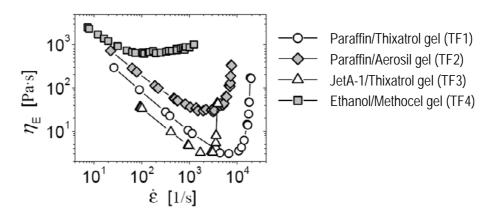


Figure 3: Extensional viscosity  $\eta_E$  vs. strain rate  $\dot{\varepsilon}$  for various gelled fluids

#### 3.2 Flow characteristics

Velocity profiles of laminar (HBE) gel flows have jet been determined numerically in previous publications, see e.g. Ref. [17]. To determine the area of validity of these laminar calculations as well as to determine a basis for the classification of spray regimes for gel sprays a generalized Reynolds number and a critical Reynolds number are necessary. Equation (2) shows the analytically determined generalized Reynolds number for fully developed flows in pipes of constant diameter D, which has yet been presented at the previous EUCASS2007 conference, see Ref. [20].

$$Re_{gen\,HBE} = \frac{\rho \cdot \overline{u}^{2-n} \cdot D^{n}}{\frac{\tau_{0}}{8} \left(\frac{D}{\overline{u}}\right)^{n} + K \left(\frac{3m+1}{4m}\right)^{n} 8^{n-1} + \eta_{\infty} \frac{3m+1}{4m} \left(\frac{D}{\overline{u}}\right)^{n-1}}$$

$$with \quad m = \frac{n \cdot K \left(\frac{8\overline{u}}{D}\right)^{n} + \eta_{\infty} \left(\frac{8\overline{u}}{D}\right)}{\tau_{0} + K \left(\frac{8\overline{u}}{D}\right)^{n} + \eta_{\infty} \left(\frac{8\overline{u}}{D}\right)}$$
(2)

The critical Reynolds number  $Re_{crit}$ , however, which defines the laminar/turbulent transition point, cannot be determined analytically as it is possible for power law fluids, conducted by Metzner and Reed. Thus an iterative method was used to determine critical Reynolds numbers, which is based on the stability parameter method of Ryan and Johnson. Figure 4 shows the dependence of the critical Reynolds number  $Re_{crit}$  on the exponential factor n in the HBE equation for a constant pre-exponential factor K and further parameters of the HBE equation. It can be seen that the critical Reynolds number  $Re_{crit}$  HBE shows a maximum at medium exponential factors n. For n = 1, which represents a Newtonian fluid, the well known value of 2300 occurs. At low n a limiting value above the Newtonian value can be seen. In comparison to those results the critical Reynolds number for power-law fluids  $Re_{crit}$  HB show both non-realistic low values for low exponents. This is due to the fact that for low exponents n both, the power-law and the Herschel-Bulkley law, calculate with unrealistic low viscosity values in the high shear rate range. A more detailed discussion and a comparison with experimental results are presented in Refs. [13, 14]. The critical generalized Reynolds number for injector orifice diameters of D = 0.7 mm is given additionally in Table 2.

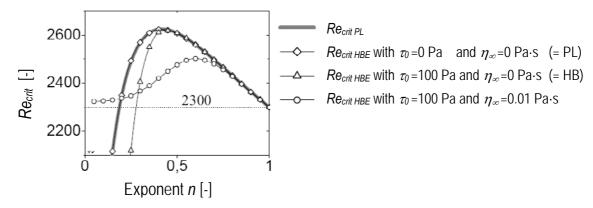


Figure 4: Calculated critical Reynolds number  $Re_{crit}$  vs. the exponential factor n of the HBE equation.  $K = \text{const.} = 10 \text{ Pa} \cdot \text{s}^n$  [14]

# 3.3 Spray characteristics

Figure 5 shows typical shadowgraph images of the break-up process of the kerosene/Thixatrol gel (TF3) in dependence upon the generalized HBE Reynolds number. The left images of each pair of shadowgraph images provide the view perpendicular to the plane spanned by the two gel jets, while the parallel view to the liquid plane is shown on the right images. It can be seen that at the impingement point of the two gel jets a thin sheet is formed, which is perpendicular oriented to the plane with the two jets. Comparing the images it is obvious that the breakup process changes with  $Re_{gen\ HBE}$ . At low and medium  $Re_{gen\ HBE}$  (Fig. 5a-e) ligaments are separated from the sheet, which decay downstream to droplets. At the highest  $Re_{gen\ HBE}$  (Fig. 5f) a direct decay to droplets is visible, which are separating in a periodic manner. These different breakup modes were also found for Newtonian fluids and thus the naming could partly be related to Newtonian breakup modes, see e.g. Refs. [23-25].

The break-up behavior of the paraffin/ThixatrolST gel TF1 is presented in Fig. 6. The gel shows similar break-up modes with its decay to droplets like TF3. The same break-up modes occur at different injection velocities but at similar values for the generalized Reynolds number. Also the paraffin/Aerosil gel TF2, see Ref. [14], shows similar breakup modes with the decay to droplets. The ethanol/Methocel gel (TF4), however, shows a completely different behavior. It can be seen on the images of Fig. 7 that the sheet is strongly bended. At the lower  $Re_{gen\,HBE}$  a breakup mode, which is called "closed rim", can be seen. At the higher  $Re_{gen\,HBE}$  the sheet breaks up in fibers, which do not decay to droplets. This fiber-producing effect was also observed earlier with other gels and also with an air blast atomizer; see e.g. Refs. [26, 27].

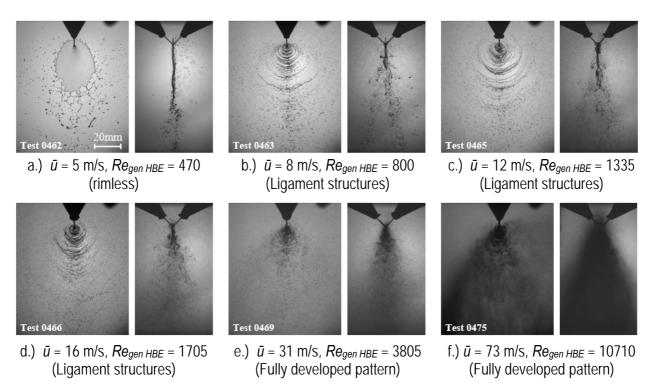


Figure 5: Break-up process with droplet formation of the kerosene/Thixatrol gel (TF3) in dependence of the generalized HBE Reynolds number

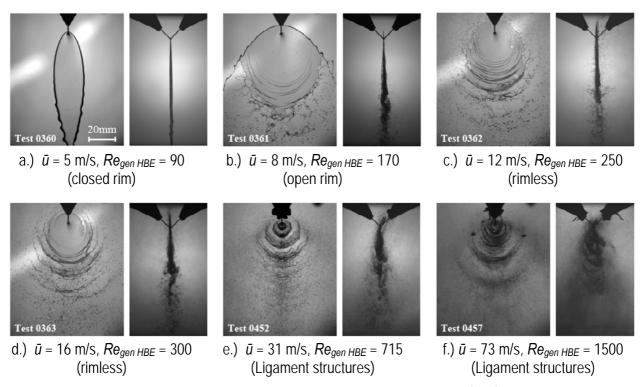


Figure 6: Break-up process with droplet formation of the paraffin/Thixatrol gel (TF1) in dependence of the generalized HBE Reynolds number

It is assumed that the reason for this difference between droplet and fiber formation is the higher elongational viscosity of the fiber forming ethanol/Methocel gel. Due to the long-chain\_cellulose molecules, which are winding around each other and which hinder a total separation of the molecules in an elongational flow, the decay to droplets seems not to be possible. Further investigations are necessary to get a better understanding and to verify this assumption.

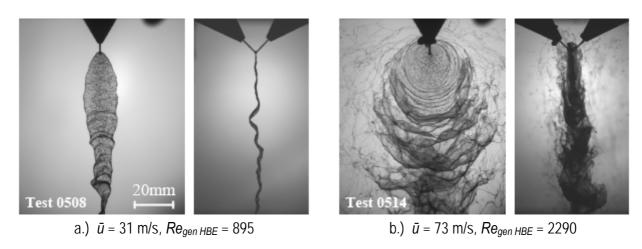


Figure 7: Break-up process with fiber formation of the ethanol/Methocel gel (TF4) for two different generalized HBE Reynolds number

Figure 8 presents the calculated generalized HBE Reynolds number  $Re_{gen\,HBE}$  in dependence upon the jet exit velocity for the four investigated test gel fuels and their basic fluids. The calculated critical Reynolds numbers for all Newtonian fluids and gels are in the range 2300 till 2415. The position of this range in the diagram is approached by the dotted line. This diagram serves as the base for the regime diagram presented in Fig. 9. Here the position of the different breakup regimes in dependence of the generalized Reynolds number  $Re_{gen\,HBE}$  and the Weber number  $We = \rho \cdot u^2 \cdot L / \sigma$  for the droplet producing gels TF1 – TF3 and their basic fluids is presented. For the determination of Weber numbers constant surface tensions  $\sigma$  were assumed for all investigated gels, which were set to be equal to these of their basic liquids, see Ref. [14]. The closed rim mode occurs at low Weber and  $Re_{gen\,HBE}$ , followed by the periodic drop mode at higher We and  $Re_{gen\,HBE}$ , etc. It can be seen furthermore that for the fully developed pattern with its direct decay of the sheet to droplets the gel flow in the injector orifices show turbulent conditions for all conducted experiments. The ligament-structure mode shows both laminar and turbulent flow conditions, while all other breakup modes have definitely laminar injector flow conditions for the investigated gels and fluids.

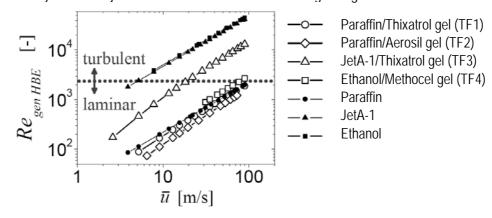


Figure 8: Generalized HBE Reynolds number vs. the jet exit velocity  $\bar{u}$  for the 4 gel test fuels and their basic liquids

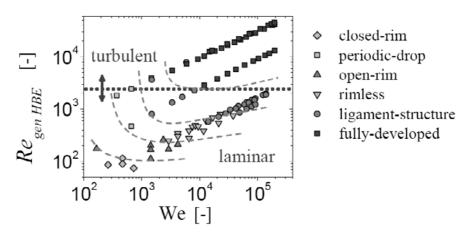


Figure 9: Regime diagram for investigated droplet producing gel test fuels and their basic liquids

# 4. Summary and conclusions

Gelled fuels are shear thinning non-Newtonian fluids, which rheological behavior concerning applied shear stresses can be described by an extended version of the Herschel-Bulkley equation. For the characterization of the spray behavior generalized Reynolds numbers and critical Reynolds numbers can be used. The spray behavior of three investigated gels with droplet formation characteristic shows different break-up modes at different generalized HBE Reynolds numbers. These break-up modes can be arranged in a regime diagram with the generalized HBE Reynolds number and the Weber number. With the ethanol/Methocel gel, however, long fibers were produce instead of droplets. It is assumed that the higher elongational viscosity of this gel in comparison to the other gels is responsible for this behavior.

# Acknowledgements

impingement half angle, -°

density, kg/m<sup>3</sup>

 $\theta$ 

ρ

The help of A. Feinauer during the running of the spray tests is kindly acknowledged.

# **Nomenclature**

d D K L	tube diameter, m injector orifice diameter, m pre-exponential factor, Pa s <sup>n</sup> characteristic length, m	$\eta$ $\eta$ E $\eta_{\infty}$	dynamic shear viscosity, Pa·s elongational viscosity, Pa·s viscosity of upper Newtonian plateau, Pa·s
n Re u ū We	exponential factor, - Reynolds number, - velocity, m/s average jet exit velocity, m/s Weber number, -		surface tension, N/m shear stress, Pa yield stress, Pa ripts and abbreviations
$\frac{\textit{Greek}}{\dot{\gamma}}$	shear rate, 1/s strain rate, 1/s	crit gen HB HBE	critical generalized Herschel-Bulkley Herschel-Bulkley-Extended

PL

TFx

power law

test gel fuel x

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