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Rheology and Stability of Hydrocarbon-Based Gelled Fuels for Airbreathing Applications

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Abstract: Gelled fuels are rheologically complex, non-Newtonian fluids. They combine the benefits of both liquid and solid states, reducing risks of leakage, spilling, and sloshing during storage while maintaining the ability to be sprayed inside a combustion chamber. Additionally, suspending energetic particles, such as metal powders of aluminum and boron, can significantly enhance their energy density compared to conventional liquid fuels. In this study, several kerosene-based and ethanol-based formulations were experimentally investigated, using both organic and inorganic gelling agents. The compositions were optimized in terms of the gellant amount and manufacturing process. Some of the most promising gellants for kerosene include fatty acids, such as Thixcin® R or THIXATROL® ST, and metallic soaps, such as aluminum stearate and zinc stearate. The effects of various co-solvents were assessed, including ketones (methyl isoamyl ketone, methyl ethyl ketone, and acetone) and alcohols (ethanol and octadecanol). Sugar polymers like hydroxypropyl cellulose were tested as gelling agents for ethanol. A preliminary rheological analysis was conducted to characterize their behavior at rest and under shear stress. Finally, a novel approach was introduced to study the stability of the gels under vibration, which was derived from a realistic mission profile of a ramjet. Finally, the ideal gravimetric specific impulse was evaluated through ideal thermochemical computations. The results showed that promising formulations can be found in both kerosene-based and ethanol-based gels. Such compositions are of interest in practical airbreathing applications as they have demonstrated excellent stability under vibration, ideal combustion properties, and pronounced shear-thinning behavior.

Keywords: gelled fuels; kerosene; ethanol; ramjet; rheology; storage; vibrations

Academic Editor: Giacomo Silvagni

Received: 30 September 2024
Revised: 17 December 2024
Accepted: 31 December 2024
Published: 13 January 2025

Citation: Dell'Acqua, S.; Morando, F.; Carlotti, S.; Maggi, F. Rheology and Stability of Hydrocarbon-Based Gelled Fuels for Airbreathing Applications. *Aerospace* **2025**, *12*, 49. https://doi.org/10.3390/aerospace12010049

1. Introduction

1.1. Research Background

Gelled fuels belong to the so-called shear-thinning pseudoplastic fluids, characterized by an apparent viscosity that decreases as the applied shear rate increases. These fuels are produced by incorporating gelling agents, or gellants, which modify the rheological properties of the liquid. As a consequence, gelled fuels behave like solids at rest, but flow as liquids when subject to mechanical forces, such as when they are forced through an injector. This unique behavior provides the potential to combine the advantages of both solid and liquid states, resulting in more reliable and safer storage. The high viscosity of gelled fuels reduces the risks of spillage and leakage. In space applications, these materials are less susceptible to impact, friction, and electrostatic discharges than solid propellants. Accidental ignition, detonation, or explosion can be prevented by storing the fuel and the oxidizer in separate tanks, if both are present. Cracks in gels have no effect, as they are atomized before combustion. As for liquid propellants, they can be used to develop throttleable and reignitable solutions. Moreover, eliminating or significantly reducing sloshing addresses potential stability issues caused by the shift of the center of mass due to the susceptibility to acceleration in a pure liquid state. Airbreathing engines, including several classes of ramjets, use liquid fuels and feature superior specific impulses than rockets. Kerosene derivatives are the most preferred option due to their good combustion performance and storage properties, even though other fuels have been investigated as well [1]. Among the alternatives, ethanol is worth noting. It is characterized by good performance, is environmentally friendly, and can be produced from renewable biomass, similar to biokerosene [2,3]. Finally, by suspending metal particles (e.g., aluminum of boron) inside the fuel in the gelled state, increased energy density can be granted [4]. For clarity purposes, the main advantages and disadvantages of hydrocarbon-gelled fuel with respect to classical liquid or solid propellants are listed in Table 1.

Table 1. Main advantages and disadvantages of gelled propellants.

Advantages	Disadvantages
Strong reduction of leakage, spillage, and sloshing.	Worse atomization and combustion quality.
Possibility to develop throttleable and reignitable engines.	Gravimetric specific impulse reduction from gelling agent addition.
Increased energy density if energetic materials are suspended (e.g., metal powders).	Possible aging inconvenience due to thixotropy.

1.2. Review of Existing Studies

Historically, the exploration of gelled fuels dates back to the late 1930s, when Ray and Bayside [5] patented the first solution of this kind. Research efforts accelerated in the late 1960s when Tarpley [6,7] explored different new formulations based on several gelling agents. Moreover, Tarpley deeply focused on thixotropy, i.e., the time-dependent decrease in viscosity when a material is subject to shear stress, followed by a gradual recovery once in rest conditions [8]. In the 1980s, NASA started investigating gelled propellants [9]. In 1999, TRW (Thompson Ramo Wooldridge Inc.) successfully tested a missile based on gels [10], covering a downrange of 8.0 km and firing the engine five times, for a total of 51 s. Since the 2000s, several formulations have been proposed, mostly stressing kerosene and ethanol as fuels. Concerning kerosene, fumed silica is by far the most used inorganic gellant [11,12]. Over the years, other organic gelling agents have been assessed, such as castor oil derivatives like THIXATROL® ST [13] and Thixcin® R [14], polyamide resin [15], paraffin wax [16], and aluminum stearate [16]. Differently, Jyoti and coworkers evaluated several varieties of cellulose to gel ethanol [17,18]. Gel preparations based on inorganic gellants are relatively simple, but the agent does not participate in the combustion. Rather, organic gelling agents require a more complex preparation but contribute to the reaction and propulsion performance.

A gelled propellant is exposed to a wide range of shear rates throughout its life cycle, with the lowest one being around 10^{-4} s^{-1} during storage, and the highest one being about 10^5–10^6 s^{-1} during droplet vaporization [19,20]. The apparent viscosity, also known as shear viscosity, is the most commonly studied property in this context, especially in relation to the shear-thinning behavior of gels. Several models have been developed to describe the specific rheological behavior of gelled propellants [8,21]. Among these, the power-law (PL) model is the simplest and most adopted one, describing an exponential decrease in viscosity, as the shear rate increases. Another key parameter is the yield stress, which represents the critical shear stress at which a gel begins to flow. According to Padwal and Natan [19], this value generally ranges between 10 and 100 Pa and is significantly lower than the pressure drops typically encountered in injectors. The rheological models are instrumental in studying the atomization quality of gels. Indeed, they are crucial for deriving non-dimensional quantities. For example, the generalized Reynolds number [22] is introduced here. This parameter is a variant of the traditional Reynolds number and takes into account the variation in viscosity with the shear rate. This is a typical feature of non-Newtonian fluids and allows for a more comprehensive understanding of the atomization regimes. A high generalized Reynolds number is typically required to achieve fully-developed atomization of gels characterized by the formation of fine droplets, without patterns or ligaments [23]. The reader should note that the definition of the aforementioned non-dimensional groups is influenced by the specific rheological model employed. An increased difficulty was also observed in the combustion, ignition, and flammability of gelled fuel droplets compared to liquid propellants [24,25], indicating the formation of an impermeable elastic layer, primarily consisting of the gelling agent.

1.3. Research Motivation

Despite extensive research and significant advancements in the field, the development of a fully operational engine based on gelled propellants has not yet been achieved. While many issues related to gels have been explored, numerous aspects still require detailed investigation to overcome key challenges. In particular, the stability of gelled solutions during operations—such as the ability to maintain their properties and avoid degradation within the mission envelope—remains an area that necessitates further studies.

1.4. Study Objectives

The present work focuses on the development of gelled fuels for airbreathing applications, using both established and novel gelling agents having a natural origin. Both kerosene and ethanol have been considered as primary fuels. The obtained substances have been characterized in terms of yield stress and shear-thinning behavior through a preliminary rheological analysis. The stability of gels has been analyzed under flight-like vibration conditions, checking both qualitative and quantitative behavior. Finally, a comparison is proposed between the gravimetric specific impulse of the investigated gels and that of the pure liquid fuels, assessing through thermodynamics the difference introduced by the gelling agent. In the current work, Section 2 illustrates the methodology and materials adopted in the experimentation, as well as the rationale behind the investigation technique. The results of the experimental activity are reported, critically reviewed, and discussed in Section 3. Finally, Section 4 summarizes the findings of the present analysis and outlines potential domains and developments for future research.

2. Methodology and Materials

2.1. Formulations

Kerosene and ethanol were used as the primary ingredients in the gel formulations. JET A-1 by Magigas was selected due to its common use within the European aerospace sector, while ethanol was supplied by Sigma Aldrich (CAS no. 64-17-5).

For kerosene-based gels, fumed silica (Sigma Aldrich, CAS no. 112945-52-5) was chosen as the initial gelling agent due to its long-standing use in this field of research and its remarkable ease of application. Hydrophilic silica surface forms hydrogen bonds between the dispersed particles, entrapping liquid molecules and creating a gel-like structure. Castor oil derivatives, such as THIXATROL® ST (Elementis Specialties, CAS no. 51796-19-1) and Thixcin® R (Elementis Specialties, CAS no. 139-44-6), were other gellants commonly mentioned in the open literature. Unlike silica, these substances create a gel by de-agglomeration and dispersion into kerosene, and subsequent formation of hydrogen bonding to form a three-dimensional structure [26]. This process is enhanced by heating and shear stresses. A polar co-solvent may be required to activate the bonds between kerosene and the gellant. Ethanol, MIAK (methyl isoamyl ketone, FLUKA, CAS no. 110-12-3), acetone (Alfa Aesar, CAS no. 67-64-1), MEK (methyl ethyl ketone, or butanone, Acros Organics, CAS no. 87-93-3), and octadecanol (FLUKA, CAS no. 112-92-5) were adopted for this investigation. Additionally, other organic substances were explored to gel kerosene due to their enhanced combustion efficiency compared to silica and other similar inorganic materials. In particular, paraffin wax (Sigma Aldrich, CAS no. 8002-74-2) was selected, because after melting and mixing, it could trap kerosene and form a gel-like structure, still providing almost the same combustion properties. Metallic salts are mainly used as emulsifiers and also provide good combustion performance. In the present study, copper stearate (abcr, CAS no. 660-60-6), zinc stearate (Acros Organic, CAS no. 557-05-1) and aluminum stearate (FLUKA, CAS no. 637-12-3) were adopted. These substances contain one atom of metal that might participate in the combustion process, releasing energy. In this respect, thermal degradation of Cu and Zn stearates reportedly happens at several hundred degrees Celsius [27,28]. A similar mechanism is expected to occur also for Al stearate. It is worth noting that stearic acid is the base constituent of Thixcin® R and THIXATROL® ST.

The search for gelling agents of ethanol started from the open literature. The cross-linked polyacrylic acid polymer, known as Carbopol®, proved to produce water-based gel simulants in many experiments [12,20]. Several formulations of Carbopol® exist, and, in most cases, the acid compound fully activates when its pH is increased by the addition of other substances. Carbopol® 940 (Natural Group) was chosen for its availability. Additionally, as suggested by Jyoti and co-workers [17,29], cellulose was included in the analysis. Klucel® G (Ashland, CAS no. 9004-64-2) was investigated to avoid the often necessary addition to ethanol-based formulations of water as a co-solvent. Also, hydroxypropyl cellulose (HPC)—typically used in artistic restoration—was tested, since a similar compound (i.e., propyl cellulose) was used with ethanol by Jyoti and Baek [29], without co-solvent addition. Finally, other substances commonly used in food processing were tested, such as agar (by Erbotech), xanthan gum (by PureFood), pectin powder (by Special Ingredients), and sodium alginate (by Special Ingredients, CAS no. 9005-38-3). The complete ingredients list is provided in Table 2.

Table 2. Substances adopted in the formulation of hydrocarbon-based gelled fuels.

Main Substance	Gelling Agent	Co-Solvent
JET A-1	Fumed silica Thixcin® R THIXATROL® ST Paraffin wax Al stearate Zn stearate Cu stearate	Ethanol MIAK Acetone MEK Octadecanol
Ethanol	Carbopol® 940 HPC Agar Xanthan gum Sodium alginate Pectin powder	- - -

The gels were mixed with a three-blade impeller and heated with a PID-controlled heating plate (Kyntel K-MI0102002), monitoring the temperature through a thermocouple sensor (PT1000), dipped inside the gel during the preparation. The target point of the heating plate was set to match the preparation temperature, as discussed in Section 3.1. The initial point was the ambient temperature (i.e., 20 ± 2 °C). The mixing container was covered with aluminum foil during preparation to prevent the evaporation of the most volatile liquid components. A small hole allowed the impeller to enter the vessel.

Thermogravimetric and differential thermal analyses (TG-DTA) were performed to verify the actual composition of the gels after preparation. These tests were conducted in an inert argon atmosphere (50 mL/min) at a heating rate of 10 K/min, employing a Netzsch STA 449 F5 Jupiter instrument. Detailed TG analyses were performed only on kerosene-based gels, as the ethanol-based preparations presented excessive fuel evaporation. The same analysis was accomplished on each ingredient of the gels to quantify the mass variation due to the evaporation of each substance.

2.2. Rheological Characterization

The objective of the rheological analysis was to preliminarily describe the flow properties of the produced gelled propellants, both at rest and under dynamic conditions. The tests were all conducted with a Rheometrics RDA II rheometer, controlled by the embedded software RSI Orchestrator (v. 6.5.3), with a 40-mm parallel-plate geometry. Viscosity measurements were performed under both dynamic and steady conditions, at increasing shear rates. Data analysis used existing rheological models for shear-thinning non-Newtonian fluids. The $\dot{\gamma}$-η curve from steady tests provides a quantification of the shear-thinning property, where $\dot{\gamma}$ is the shear rate and η is the apparent viscosity. The steeper the flow curve, the easier the gel flows in pipes and injectors, once shear stress is applied. The analysis was performed for stress rates in the range of $0.01\,\mathrm{s}^{-1}$ to $100\,\mathrm{s}^{-1}$. According to the adopted PL rheological model, the viscosity can be obtained as in Equation (1), where K is the so-called consistency index and n is the pseudoplasticity index. The consistency index provides an indication of the viscosity level of the substance, while the pseudoplasticity index relates directly to the slope of the flow curve and indicates the strength of shear-thinning behavior. A gel with a low n value is characterized by a more significant reduction in viscosity for the same increase in shear rate.

$$\eta = K\dot{\gamma}^{n-1} \tag{1}$$

Since several phenomena cannot be described by viscosity alone, viscoelastic behavior should be taken into consideration [30]. In dynamic rheological analyses, a sample is subject to harmonically varying small-amplitude deformations in a simple shear field. Monitoring the in-phase and out-of-phase responses of the sample to imposed deformations allows us to characterize its viscoelastic behavior and determine possible structural changes at the molecular level. The viscoelastic properties of the gels were tested by first altering the oscillation frequency f from 0.05 Hz to 50 Hz at fixed strains γ (2 % and 20 %), and then by varying the strain from 0.2 % to 200 % at the fixed frequency of 1 Hz. The strain percentage refers to the gap between the parallel plates. Such dynamic tests provide measurements for the complex viscosity $\eta^\star$ and the complex modulus of rigidity $G^\star$, as well as for their real parts η' and G', and imaginary parts η'' and G'', respectively. It should be noted that η' and G'' represent the viscous contributions, while η'' and G' are associated with the elastic components of the two variables. In this respect, G' is also called storage modulus, while G'' is known as loss modulus. An exponentially decreasing f-$|\eta^\star|$ profile—similar to the aforementioned $\dot{\gamma}$-η diagram—was obtained by Li et al. [31]. However, Rathinaraj et al. [32] observed a substantial difference between the steady response (i.e., $\dot{\gamma}$-η profile) and the dynamic behavior of gelled propellants for medium-to-high shear rate values. Hence, the slope of the f-$|\eta^{\star}|$ curve cannot exactly correspond to the exponent in Equation (1), even though the two values are quite similar. Yet, even the dynamic analysis can provide substantial information on the shear-thinning behavior of different gelled propellants. In fact, the analysis of $G^{\star}$ components allows for determining if elastic or viscous effects prevail in the substance by analyzing the profiles of G' and G'' as functions of the strain (γ) or the shear stress (τ). The storage modulus is typically larger than the loss modulus at low strains, as the elastic property dominates. The two curves then intersect, and the viscous behavior overcomes the elastic one at high strains. The intersection point, the so-called flow point γ_{flow} [33], identifies the limit which discriminates the interval where solid-like behavior prevails from the one where liquid-like is dominant. Finally, a τ-G' curve can be obtained. The storage modulus G' generally features a plateau at low stresses and the change in slope of such a curve represents the yield stress τ_{yield} characteristic of the gel [33]. Before the yield point, the material features elastic deformations and will return to its original shape, if the applied stress is removed. Once the yield point is reached, part of the deformation will be permanent and non-reversible. Thixotropy plays a role in the definition of yield stress, as the behavior of the gel also depends on the time during which the shear stress is applied. For this reason, a distinct yield stress could be hardly identified.

2.3. Stability

Gelled propellants must maintain their characteristics during their whole lifetime to remain suitable for use. This primarily means that the separation of liquid from gel phase and, more in general, degradation of the material must be minimal during both storage and operation. During flight, part of the vibrations experienced by the vehicle components might be transmitted to the tank walls and, hence, the fuel. Applying shear stresses to the gel could lead to a significant reduction in viscosity, due to the shear-thinning characteristic previously discussed. This could be desirable from the injection viewpoint, as the overpressure required for complete atomization would be reduced, but it could also cause the deposition of eventual solid particles suspended inside the gel (e.g., metal powders). Moreover, sloshing behavior increases if viscosity lowers in an unexpected fashion, losing one of the advantages derived from using a gelled fuel.

A specific experimental setup was implemented. A pre-defined vibration profile was imposed on a set of vials containing gel fuel. Six glass test tubes with an internal diameter of 12 mm and filled halfway with the gels were attached to a modal shaker through a rigid support using a threaded connection. The shaker (Brüel & Kjær Modal Exciter Type 4824) was capable of reaching frequencies of up to 5000 Hz, accelerations of 44 g, and a displacement of 25.4 mm (i.e., 1 inch). The periodic oscillation was generated by a Protek Sweep Function Generator 8205A, capable of producing rectangular, triangular, and harmonic waves at a wide range of frequencies and amplitudes. The signal was amplified by a Brüel & Kjær Power Amplifier Type 2732 and inspected through a Tektronix TDS 224 oscilloscope. The experimental setup is outlined in Figures 1 and 2. Glass transparency allowed for the visual inspection of gel behavior under vibration.

Different frequencies and amplitudes were studied, assuming that the gel could be used in a ramjet system. In this respect, different phases of the flight were considered. First, a frequency of 2.28 Hz was found to be typical of an aircraft at takeoff [34]. This condition simulates the situation of a missile carried by a mother aircraft, as a payload. The wave amplitude was assumed as the maximum allowed by the modal exciter (i.e., 1 inch), in order to stress the gels as much as possible. This condition was simulated for 5 min with a triangular wave and for another 5 min with a rectangular shape. Subsequently, the frequency was raised to 70 Hz. This choice was justified by the interest in the buffeting phenomenon, which is one of the most problematic issues encountered by an aircraft during the transonic phase [35]. This effect was reproduced through triangular waves for about 30 min. The amplitude was set at about 5 mm, which was dictated by modal shaker limitations, in particular by the acceleration limit.

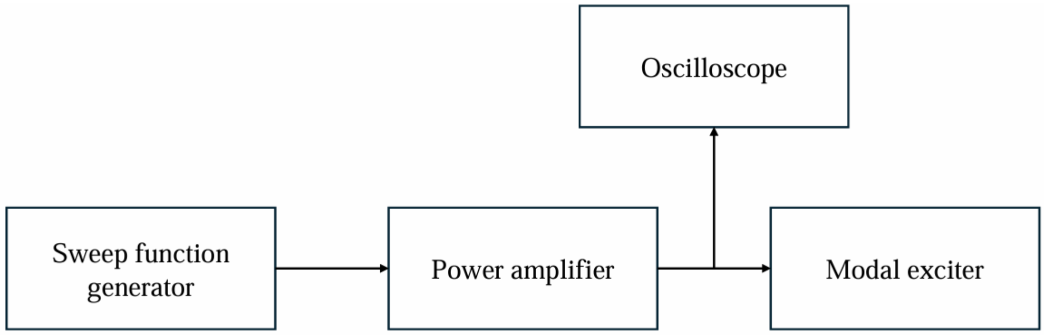


Figure 1. Conceptual scheme of the experimental setup implemented for the vibration assessment of gelled fuels.

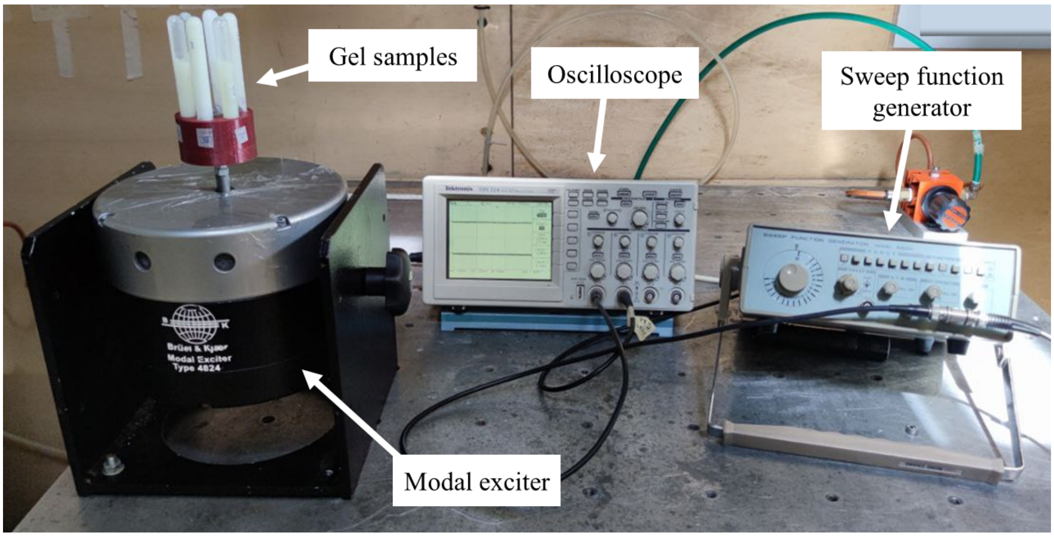


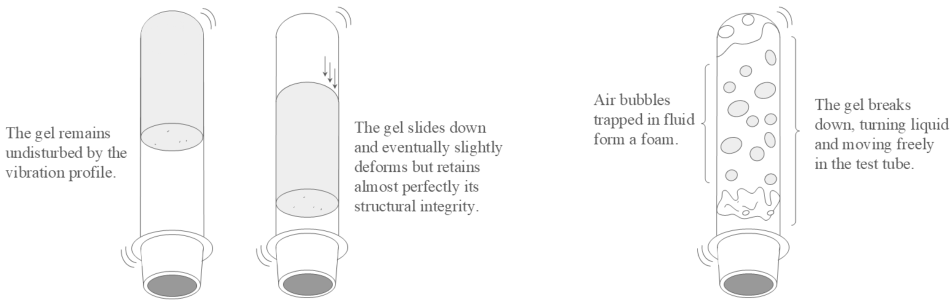
Figure 2. Picture of the experimental setup for the vibration tests. The power amplifier is not shown in the figure.

The reader should be aware that such a test matrix represents a preliminary attempt to characterize the storability of gels in a new way, simulating their behavior under flight-like dynamic conditions. It is important to underline that the open literature on ramjets does not report dedicated input data, such as precise vibration profiles or shapes of the waveform in captive flight conditions or after release. The current test matrix is quite limited, although it is representative of some important documented conditions and has been established on a limited selection of references. Based on further documentation, tests can be expanded for altitude and velocity profiles and should include more accurate details on vibration range, amplitude, and waveform shape.

The response to vibration tests was qualitatively evaluated, according to two criteria: (A) the tendency to liquefy when subject to oscillations, and (B) the recovery of the original compactness up to one hour after the test. The qualitative behavior based on criterion A is schematically highlighted in Figure 3. A grade from 1 to 3 was assigned for each of the two parameters, and the overall oscillation response evaluation was provided by the sum of the two sub-grades. Table 3 summarizes the rating methodology.

Table 3. Rating methodology to evaluate the response to oscillatory loads.

Liquefaction During the Test	**Grade A**
Gel barely moved in the test tube	3
Gel moved along the test tube but did not completely liquefy	2
Gel completely liquefied	1
Recovery after the test	**Grade B**
No change in the gel appearance and compactness	3
Softer gel, but completely recovered after a few minutes	2
Gel never fully recovered its original configuration	1



(**a**) Grade A3. (**b**) Grade A1.

Figure 3. Qualitative representation of gel liquefaction behavior under vibration, describing the best (**a**) and worst (**b**) performances. Grade A2 is associated with an intermediate performance.

2.4. Ideal Performance

The ideal gravimetric specific impulse was computed for a reference ramjet mission profile with an altitude of 15 km and a Mach number equal to 3.5. For this flight condition, the total temperature was 748 K, and the total pressure was 9.24 bar. According to pressure recovery estimation from the military specification MIL-E-5007D [36], the total pressure at the ram burner was assumed equal to 6.85 bar. The gravimetric specific impulse I_{sp} was computed according to Equation (2), which is valid for a single phase and calorically perfect gaseous mixture in an airbreathing device, under the additional assumptions of steady-state isentropic flow and frozen chemical composition during nozzle expansion. Chemical equilibrium and stagnation flow hypotheses were assumed in the combustion chamber. An optimal nozzle was considered.

$$I_{sp} = \frac{1}{g_0}\sqrt{\frac{2k}{k-1}R\frac{T_c}{M_{mol}}\left[1-\left(\frac{p_e}{p_c}\right)^{\frac{k-1}{k}}\right]}(1+\phi) - \frac{v_\infty \phi}{g_0} \quad (2)$$

In Equation (2) g_0 represents the gravitational acceleration of 9.81 m/s^2, k is the specific heat ratio of the expanding mixture, R denotes the universal gas constant, T_c denotes the adiabatic flame temperature, M_{mol} denotes the average molar mass of the gaseous combustion products, p_e denotes the pressure at nozzle exit (assumed as optimized at the considered altitude), p_c denotes the combustion chamber pressure, v_∞ denotes the relative air velocity, and ϕ denotes the air-to-fuel mass ratio. It should be noted that the computed I_{sp} does not take into account combustion inefficiencies, pressure losses in the flow, and other non-isentropic processes. However, this limitation does not impair the scope of the work, which consists of a relative grading among the tested fuels.

The combustion chamber temperature and the composition of the combustion products were obtained by employing the NASA CEA [37], assigning the enthalpy and pressure of the reactants. Performance was evaluated for air-to-propellant mass ratios from 14 to 17 for kerosene-based compounds, and from 6 to 16 for mixtures with both JET A-1 and ethanol. The chemical composition and standard enthalpy of formation Δh_f^0 at 298.15 K for the various substances described in Section 2.1 are summarized in Table 4.

Table 4. Chemical composition and enthalpy of formation of the gel components.

Substance	Chemical Formula	Enthalpy of Formation
JET A-1	$C_{12}H_{23}$	−303.4 kJ/mol
Fumed silica	SiO_2	−910 kJ/mol
Thixcin® R	$C_{57}H_{110}O_9$	−2344 kJ/mol
12-hydroxy octadecanamide	$C_{38}H_{76}N_2O_4$	−1300 kJ/mol
Ethanol	C_2H_6O	−278 kJ/mol
MIAK	$C_7H_{14}O$	−322 kJ/mol
Al stearate	$C_{54}H_{105}O_6Al$	−2736 kJ/mol
Zn stearate	$C_{36}H_{70}O_4Zn$	−1824 kJ/mol
Paraffin wax	$C_{50}H_{102}$	−1437 kJ/mol
Octadecanol	$C_{18}H_{38}O$	−690 kJ/mol
HPC	N.Av.	N.Av.

The chemical formula and enthalpy of formation for JET A-1 and ethanol were retrieved from the CEA database [37]. Properties of other elements were mostly retrieved from the NIST database [38]. On the contrary, paraffin wax was assumed to have a chemical composition equal to $C_{50}H_{102}$ [39], while the enthalpy of formation was computed through Equation (3) by Karabeyoglu et al. [40] as a function of the number of carbon atoms (n_C):

$$\Delta h_f^0 = -6.713 n_C - 7.846 \quad [\mathrm{kcal/mol}] \tag{3}$$

THIXATROL® ST was assumed to be a mixture of Thixcin® R and 12-hydroxy octadecanamide, with compositions of 82.5%/17.5%, as the amide content was reported to be between 10 % to 25 % by weight [41]. Thixcin® R properties were assumed to be equal to those of tristearin (i.e., three chains of octadecanoic or stearic acid), which is the base chemical for Thixcin® R production. In a similar fashion, three times the value of octadecanoic acid (i.e., −912 kJ/mol [38]) was used for aluminum stearate, as it is composed of three stearate chains bonded to an aluminum atom. Similarly, the enthalpy of formation for zinc stearate was estimated based on its di-stearate chemical composition (i.e., two chains of octadecanoic acid and a Zn atom). Thermodynamic properties of HPC were not available in the open literature. It should be noted that this ingredient was excluded from ideal performance computations because its weight fraction was less than 5%. It is the authors' opinion that the small amount and the nature of HPC are unlikely to significantly affect the thermodynamics of the gel fuel combustion process. This assumption was preferred over any other options since—due to a lack of data—effective uncertainty management was not possible.

3. Results and Discussion

3.1. Formulations

Several gel formulations were developed using kerosene and ethanol as main components. The preparation procedure was optimized for each formulation to identify the minimum percentage of a gelling agent needed to produce a stable gel. The main criterion for evaluating the quality of the gels was the test tube inversion method, complemented by a visual inspection for any macroscopic separation of the liquid phase observed up to a few days after production [13].

Fumed silica was confirmed to be the simplest gelling agent, with a minimum concentration of 7% by weight to form a stable gel. Indeed, at lower fractions, the gels were too soft and did not pass the test tube inversion method, while a higher percentage led to the formation of solid lumps. Even though kerosene and silica could be mixed by hand for a few minutes, the three-blade impeller ensured optimal dispersion of gellant particles inside the liquid fuel. JET A-1 gels containing 7% by weight fumed silica showed a tendency to degrade over time and when heated. These compounds dried out within a few months of storage, likely due to kerosene evaporation, even though the samples were stored in a dedicated container. Indeed, TG analysis confirmed the increase in silica content to 15% by weight after 4 months. Moreover, the gel was observed to separate under mild heating (e.g., 35 °C). Although this separation was easily reversible by re-mixing the liquid, it underscores the challenges associated with storing such a gel.

Among organic agents, THIXATROL® ST and Thixcin® R were valid solutions to gel JET A-1. The optimal percentage of Thixcin® R was found to be 7% by weight, with no advantage of adding a co-solvent. On the contrary, a co-solvent was fundamental with THIXATROL® ST, as it prevented the formation of a gel that was too soft. The optimal percentage for THIXATROL® ST was also 7% by weight, and the same quantity was used for the polar co-solvent. Almost no differences were observed between other liquid co-solvents, except for octadecanol, which contributed to the increased hardness of the gels. In addition, the formulation with ethanol as a co-solvent was identified as one of the most interesting for future studies, because of the increasing attention to biomass-derived fuels within the aerospace sector. For this reason, the compositions with octadecanol and ethanol as co-solvents were subject to a thorough test campaign.

Several weight fractions of THIXATROL® ST and ethanol were investigated in the mixture. The results showed increased gel stability with an increasing amount of gelling agent, reaching a peak when ethanol and gelling agent concentrations were equal. The optimal composition was identified as 86%/7%/7% by weight of JET A-1, THIXATROL® ST, and ethanol, respectively. This is consistent with the results of the literature and in the gellant range indicated by Padwal and Mishra [13]. In general, THIXATROL® ST gels appeared to be more stable, with greater resistance to stresses. In terms of the manufacturing process, such gels needed to be heated to a certain temperature to maximize the bonding efficiency between the liquid propellant and gelling substance, ensuring a continuous mixing for better dispersion. The optimal temperature was found to be 58 °C for Thixcin® R gels, and 60 °C for THIXATROL® ST ones, perfectly aligning with previous studies [13]. As explained in Section 2.1, this temperature was maintained for about 15 min while mixing. In both cases, the preparation was performed about ten degrees below the melting point of the gellant because the material softens and bonds more easily, entrapping kerosene. In the current work, both positive and negative deviations from the optimal temperature resulted in softer and less stable gels. The mixing was necessary until the mixture reached ambient temperature, while the impeller angular velocity was not a crucial parameter of the preparation procedure.

Aluminum stearate behaved differently when mixed with kerosene. Upon reaching the critical temperature of 70 °C, the mixture transitioned from liquid to solid phase, clumping around the blade and exhibiting too sticky and gummy consistency. Furthermore, the mixtures prepared with 10 % to 25 % by weight of Al stearate separated after a few hours. Even after the separation of part of the liquid JET A-1, a gel of this type would be unsuitable for practical aerospace applications as excessive gel tension would prevent breakup, rendering atomization unfeasible. After the separation of JET A-1 was complete, re-mixing produced a stable gel. The different appearances of these compounds are depicted in Figure 4.

(**a**) After the first preparation. (**b**) After a few hours. (**c**) After final mixing.

Figure 4. Kerosene–aluminum stearate gels, highlighting the appearance in different preparation phases. Immediately after the original preparation (**a**); after a few hours (**b**); after the second mixing (**c**).

When zinc stearate was used with kerosene as a gelling agent at the weight fraction of 10 % to 25 %, without mixing, and after reaching the activation temperature of 93 °C, it formed a solid-like compound during the cooling phase. The temperature was some tens of degrees below the melting temperature of Zn stearate. Such solid-like compounds were characterized by extreme shear-thinning and thixotropic properties. As soon as the mixture was perturbed, it tended to liquefy. If the stress was applied for a non-negligible time, it did not fully recover the original appearance, resulting in a much more liquid substance. A similar result was obtained with gels based on copper stearate, following the same procedure adopted for Zn stearate mixtures. However, a superficial layer of solid-like material with an inner liquid part was observed. Hence, Cu stearate was discarded, and Al and Zn stearates were investigated in detail.

Octadecanol was added to the zinc stearate-based mixtures to attempt to yield more stability and rigidity to the compound and make it more similar to compositions prepared with JET A-1 and THIXATROL® ST. The result confirmed this effect and an optimal composition was obtained with 77.5%/15 %/7.5% by weight of JET A-1, zinc stearate, and octadecanol, respectively. Additional tests were performed by incorporating paraffin wax inside the gel. The mixture was heated to approximately 65 °C, which caused the melting of paraffin. Afterward, the blend was stirred with the impeller and heated to 93 °C. Finally, the mixture was allowed to cool undisturbed until ambient temperature. At least one hour was necessary to obtain a solid-like gel, and a longer time interval passed for full stabilization. The composition was optimized to maximize the kerosene and paraffin contributions, and the final weight fractions were set at 70%/20%/5%/5% by weight of JET A-1, paraffin wax, zinc stearate, and octadecanol, respectively. Finally, the simultaneous use of aluminum and zinc stearates was attempted to combine the bonding capabilities of Al stearate and the pronounced shear-thinning property derived from Zn stearate. Adding kerosene–Zn stearate mixture to kerosene–aluminum stearate gel led to an extremely sticky solution, which separated after cooling. However, after several hours of mixing, a very stable gel was obtained. The final composition was fixed at 85%/10%/5% by weight of JET A-1, aluminum stearate, and zinc stearate, respectively.

The progress on gelling agents for ethanol is more limited compared to that of kerosene-based mixtures. Thus, the present work focuses on advancing the current state of the art. A small fraction of HPC (i.e., below 5 % by weight) was sufficient to obtain a gel. The tube inversion test was passed only with gelling agent content from 5 % by weight and above. This composition was considered optimal. The manufacturing process did not require heating, thereby preventing excessive ethanol evaporation. The resulting gel exhibited excessive stickiness and a consistency similar to that of pre-aged kerosene–aluminum stearate. Similar observations apply to ethanol and Carbopol® 940. Excessive stickiness made the proposed solution impracticable for atomization purposes. Ethanol-based gels were not obtained with agar, xanthan gum, pectin powder, and sodium alginate. A gel based on ethanol was obtained from the combination of an ethanol–HPC gel with a kerosene–Zn stearate gel, in a proportion of 55 % and 45 % by weight, respectively. The compound was very stable, slightly softer than the materials made of THIXATROL® ST. The authors hypothesize that stickiness and compactness were granted by HPC while Zn stearate seems to confer the shear-thinning quality, needed to efficiently atomize the mixture. The optimal gel compositions for both kerosene-based and ethanol-based mixtures are outlined in Table 5.

Table 5. Optimal gel formulations and related gel code: nominal weight fractions.

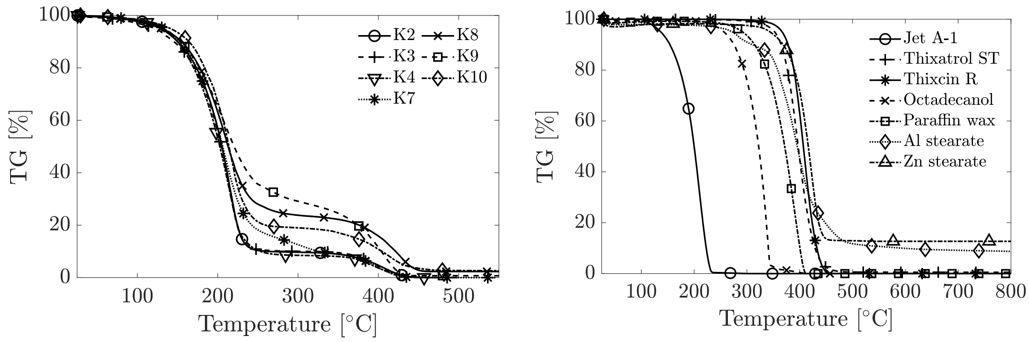
Component	**Percentage**												
	K1	**K2**	**K3**	**K4**	**K5**	**K6**	**K7**	**K8**	**K9**	**K10**	**E1**	**E2**	**E3**
JET A-1	93%	93%	86%	86%	86%	86%	86%	77.5%	70%	85%	-	-	38%
Ethanol	-	-	7%	-	-	-	-	-	-	-	95%	95%	52%
Fumed silica	7%	-	-	-	-	-	-	-	-	-	-	-	-
Thixcin® R	-	7%	-	-	-	-	-	-	-	-	-	-	-
THIXATROL® ST	-	-	7%	7%	7%	7%	7%	-	-	-	-	-	-
MIAK	-	-	-	7%	-	-	-	-	-	-	-	-	-
Acetone	-	-	-	-	7%	-	-	-	-	-	-	-	-
MEK	-	-	-	-	-	7%	-	-	-	-	-	-	-
Octadecanol	-	-	-	-	-	-	7%	7.5%	5%	-	-	-	-
Al stearate	-	-	-	-	-	-	-	-	-	10%	-	-	-
Zn stearate	-	-	-	-	-	-	-	15%	5%	5%	-	-	7%
Paraffin wax	-	-	-	-	-	-	-	-	20%	-	-	-	-
HPC	-	-	-	-	-	-	-	-	-	-	5%	-	3%
Carbopol® 940	-	-	-	-	-	-	-	-	-	-	-	5%	-

Table 6 highlights the test matrix. Some of the gels were not satisfactory for storage or featured excessive stickiness, thus being unsuitable for practical use. For this reason, K1, E1, and E2 were not subject to any experimental test. Among gels containing THIXATROL® ST, only K3 and K7 (exploiting ethanol and octadecanol as co-solvents, respectively) were thoroughly investigated. All the novel compositions were subject to complete experimental characterization. Steady rheological analysis was not performed for state-of-the-art formulations, as several results are already available in the literature [11,22,42].

Table 6. Test matrix for the investigation of the optimal gelled fuels listed in Table 5.

Test	K1	K2	K3	K4	K5	K6	K7	K8	K9	K10	E1	E2	E3
TG-DTA		×	×	×			×	×	×	×			
Frequency-sweep rheometry		×	×				×	×	×	×			×
Amplitude-sweep rheometry		×	×				×	×	×	×			×
Steady rheometry							×	×	×	×			×
Vibration		×	×				×	×	×	×			×
Ideal performance	×	×	×	×			×	×	×	×			×

Because mixing and heating procedures can affect the final composition, thermogravimetry was used to quantify the volatile fraction of gel formulations. The results are shown in Figure 5.



(**a**) Gels. (**b**) Single components.

Figure 5. Thermogravimetry analysis results for the most promising gels (**a**) and for gel components (**b**).

All gels showed a first substantial mass decrease between 110 °C and 230 °C, which is the temperature range of JET A-1 evaporation. Above such an interval, a plateau was observed for most cases, especially for the gel made of two components (i.e., K2) and for the mixtures with THIXATROL® ST and a liquid co-solvent (K3, K4). The plateau is representative of the gelling agent mass fraction. Typically, this value exceeds the nominal one by a few percentage points, confirming the slight increase of solid phase inside the gels due to evaporation during production. Since the evaporation temperature ranges overlap in some multi-component gels, similar trace interpretation is still valid, although with a lower accuracy. This is the case of stearates (i.e., K10). In general, it was possible to appreciate a good consistency of the overall solid phase fraction with respect to the nominal one. All gels completely evaporated before reaching 500 °C except for those containing Al or Zn stearate. As discussed in Section 3.1, these two components do not vaporize completely, leading to 8.5% and 12.7% by weight of residual for Al stearate and Zn stearate, respectively.

3.2. Rheological Characterization

The $|\eta^{\star}|$ profile for the selected formulations is depicted in Figure 6 as a function of frequency, for two strain percentages, 2 % and 20 %. The reader should be aware that several gels were tested only once. This analysis aimed at preliminary screening and comparison with the open literature, enabling the identification of those gelled compositions characterized by promising features. Gels made of castor oil derivatives and of the stearates featured a smooth profile, showing a remarkable shear-thinning property. A lower viscosity—up to two orders of magnitude—was obtained for K8, K9, and E3. Substantial data scattering was originated by limits of instrument sensitivity. For this reason, these data are not reported.

Measurements for the other gels were reliable and confirmed trends and viscosity values identified by other researchers for similar applications and compounds [22,26,42]. For instance, the viscosity of K3 was found between 10^5 and 10^2 Pa s, according to Figure 6. Similar values were identified at medium shear rates for the same compound (10^3 Pa s at about $100\,s^{-1}$ [42]) or for a comparable mixture (10^4 Pa s at about $1\,s^{-1}$ [26]). Such a composition reportedly provided satisfying atomization performance with an internal impinging injector.

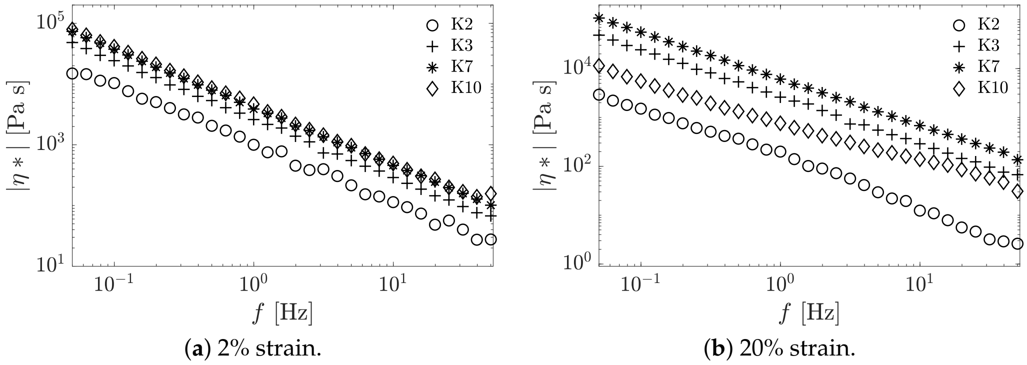


Figure 6. Frequency-sweep dynamic measurements for different strain percentages.

Figure 7 summarizes shear stress (τ), storage modulus (G'), and loss modulus (G'') measurements through dynamic strain–sweep rheological analyses. The yield stress (τ_{yield}) can be obtained as the stress at which the slope change of the G' curve occurs. It should be noted that some of the gels mentioned in Table 6 are not shown in the figure either for readability or for data dispersion.

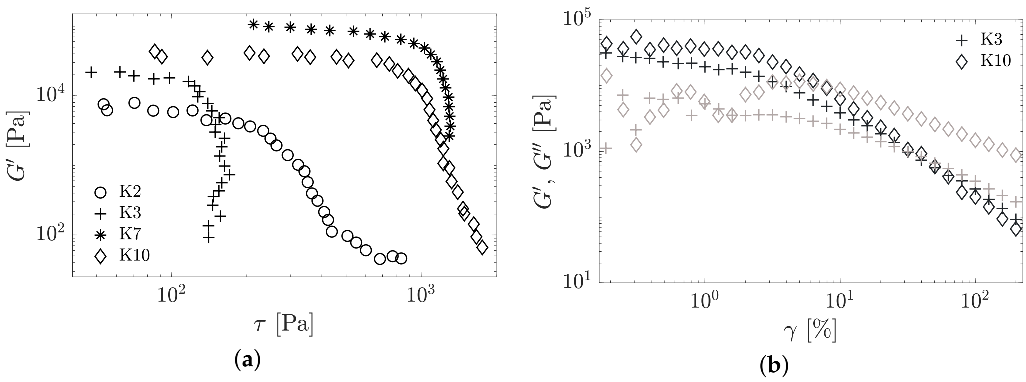


Figure 7. Dynamic strain–sweep analysis outcomes. (**a**) Storage modulus–shear stress curves, from which the yield stress τ_{yield} can be derived; (**b**) storage modulus (black) and loss modulus (gray) profiles, identifying the flow point γ_{flow}.

Gels based on Thixcin® R, THIXATROL® ST, and stearates were characterized by high-yield stresses. The K7 composition was characterized by the highest value (about 650 Pa). A slightly smaller value was obtained for K10. For this reason, these two formulations might be considered the most suitable for storage. Yield stress values of about 160 Pa and 100 Pa were obtained for K2 and K3, respectively. Other gels (i.e., K8, K9, and E3) did not provide reliable measurements because of data scattering.

The flow points of gels K3 and K10 were 31.7% and 7.9% strain percentages, respectively. Data scattering did not allow for reliable measurements for K8, K9, and E3 compositions. Concerning K3, it should be noted that the G' plateau between 10^4 and 10^5 Pa at a low strain rate is consistent with the measurements collected by Padwal and Mishra [26] for a similar compound, including MIAK, in place of ethanol as the co-solvent.

Steady tests were performed to obtain the shear rate–shear viscosity profiles and compare them through the PL model with the formulations that were already characterized in the literature. Relevant results are shown in Figure 8, while the reader is encouraged to refer to Refs. [11,22,26,42] for the $\dot{\gamma}$-η profile and the K, n parameters of the remaining gels. As highlighted by Figure 8, K7 and K10 exhibited an exponentially decreasing viscosity profile with increasing shear rate. A similar trend was observed for K8, K9, and E3; however, only qualitative conclusions could be drawn due to significant data scattering.

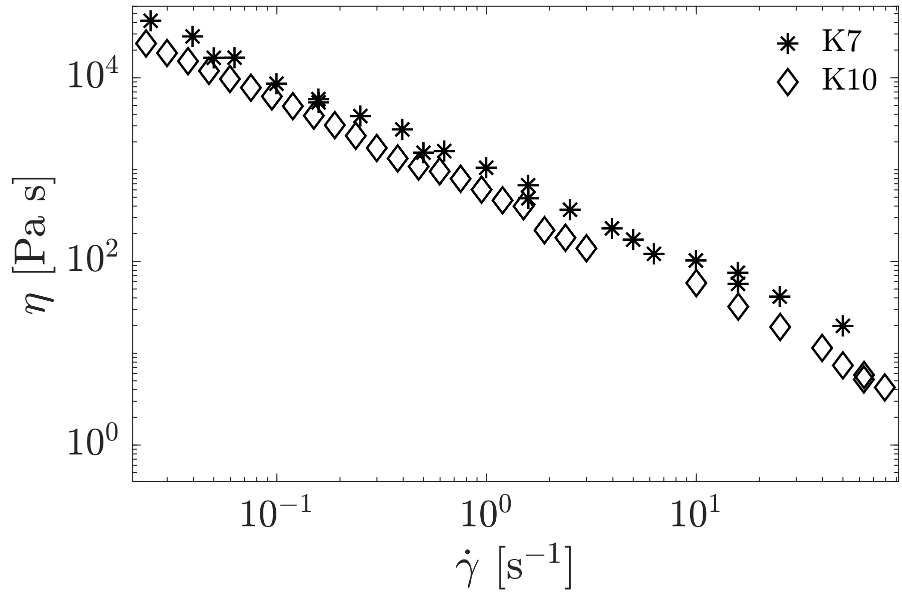


Figure 8. Rheological measurements from steady tests represented in logarithmic axes.

The numerical values for the coefficients of the PL rheological model are listed in Table 7. The table also includes τ_{yield} and γ_{flow}. The consistency indices K represent gel viscosity, with the analysis confirming K7 as the most viscous. Gel K10, based on zinc stearate, has a lower viscosity and demonstrates pronounced shear-thinning behavior, consistent with observations made during its preparation; refer to Section 3.1 for details. The consistency index of K7 is about one order of magnitude greater than those of K3 and K4. This observation aligns with the increased hardness of the mixture caused by the addition of octadecanol as a polar co-solvent in place of ethanol or MIAK. Some of the pseudoplasticity indices n are negative. This can be caused either by a severely negative slope of the viscosity profile at higher shear rates—a negative profile for shear stress as well—or by experimental uncertainties. However, all gels had a value of n quite close to 0, similar to compositions developed with THIXATROL® ST and Thixcin® R. Thus, it can be concluded that the dynamic rheological campaign is consistent with the results of the steady tests shown in Figure 6.

Table 7. Rheological campaign results - the values for the consistency index K and the pseudoplasticity index n of gels K1, K3, and K4 were retrieved from Santos et al. [11], Deng et al. [42], and Madlener et al. [22], respectively. Nomenclature: N.A - Not Available.

Gel	Steady Tests		Dynamic Tests	
	K [Pa s^n]	n [-]	τ_{yield} [Pa]	γ_{flow} [%]
K1	850	0.02	-	-
K2	N. A.	N. A.	160	19.9

Table 7. *Cont.*

Gel	Steady Tests		Dynamic Tests	
	K [Pa s^n]	n [-]	τ_{yield} [Pa]	γ_{flow} [%]
K3	80.49	0.065	100	31.7
K4	37.78	0.12	-	-
K7	929.6	0.018	650	5.0
K10	470.7	−0.093	550	7.9

3.3. Stability

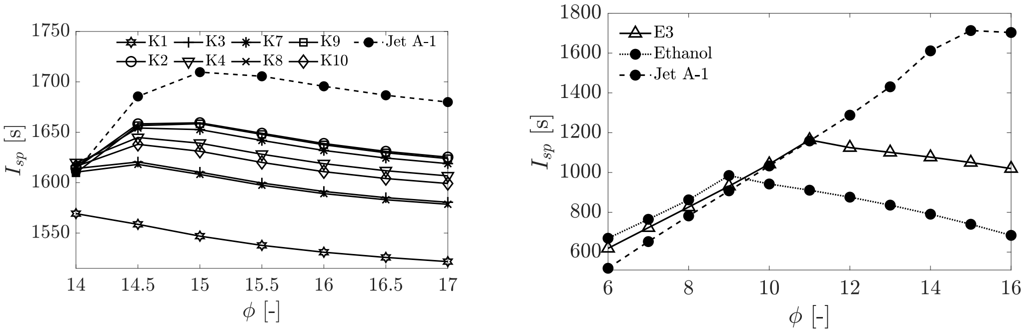
Vibration test ranking is available in Table 8, according to the requirements discussed in Section 2.3. At 2.28 Hz, none of the gels exhibited any changes in the structure, while different behavior was observed at 70 Hz. Indeed, K2, K3, K7, and K10 settled on the bottom of the test tube but did not show any kind of separation or liquid-like behavior. On the contrary, gels containing zinc stearate became liquid after a few minutes. In addition, their volume increased as the liquefied zinc stearate gels trapped air, which compromised their mechanical properties. After the vibration tests, the gels were allowed to rest for a few hours to check if the recovery of solid-like properties occurred. All the gels passed the test tube inversion method after a few minutes, except for K8, which remained liquid for several hours and never returned to the original state. Propellants containing THIXATROL® ST or Thixcin® R appeared slightly softer than their original thickness, while the one containing octadecanol as co-solvent (K7) presented a slight loss in structural rigidity, confirming the thixotropic property. Similar considerations applied to K10. Finally, it is worth mentioning that E3 did not show any alterations.

Table 8. Partial and total scores for gelled propellants under a vibration test.

Gel	Grade A	Grade B	Total
K2	2	2	4
K3	2	2	4
K7	2	3	5
K8	1	1	2
K9	1	2	3
K10	2	3	5
E3	3	3	6

3.4. Ideal Performance

The ideal gravimetric specific impulses I_{sp} of kerosene-based and ethanol-based gelled fuels are shown in Figure 9, along with pure liquid fuels; refer to Section 2.4 for the reference flight conditions. As expected, the addition of gelling agents to pure kerosene causes a reduction in the specific impulse, especially when fumed silica is used. Except for K1, the maximum I_{sp} is in the range of 1618 s to 1659 s for the other kerosene-based gelled fuels, and 1710 s for pure JET A-1. Despite the significant difference in kerosene fractions in K2 and K9 gels (93 % and 70 % by weight, respectively), they feature almost the same performance. Paraffin wax in K9 positively contributes to the global combustion enthalpy. According to thermochemistry, the aluminum contained in Al stearate partakes in the reaction and contributes to I_{sp}. The E3 composition, characterized by 52 % ethanol and 38 % kerosene, features intermediate properties between the two substances. The peak specific impulse is granted by a mass ratio of about 11, which is interposed between the two values for pure ethanol and for pure JET A-1 (around 9 and 15, respectively). The maximum theoretical value of the gravimetric specific impulse is 1164 s. In general, ethanol provides lower thermochemical performance when compared to JET A-1. This grading is inherited by gels.



(**a**) Kerosene-based gelled fuels. (**b**) Ethanol-based gelled fuels.

Figure 9. Gravimetric specific impulse for gelled propellants and pure liquid fuels as a function of the air-to-fuel mass ratio.

Finally, it is worth mentioning that gelled fuels may not be directly used in common airbreathing or rocket engines. As highlighted in Section 1, an advantage lies in the possibility of suspending metal particles (e.g., aluminum or boron), increasing energetic content, density, and volumetric specific impulse. Further research should be carried out to assess such performance variation, in order to find the proper combustion conditions that allow to maximize the efficiency.

4. Conclusions

This current work focuses on the investigation of gelled fuels for airbreathing applications (e.g., ramjet), particularly targeting the formulation analysis and the characterization of gels. The main constituent used was either kerosene (i.e., JET A-1) or ethanol. The preparation procedure and gelling agent quantity were optimized for each composition. Several stable formulations were obtained, with both organic and inorganic gellants. Thixcin® R and THIXATROL® ST were used to gel JET A-1, successfully verifying state-of-the-art gels. Different polar substances were employed as co-solvents for THIXATROL® ST-based gels, such as ethanol and octadecanol. Finally, several metallic salts were tested to improve the energetic content of the produced fuels, possibly in combination with other substances (e.g., paraffin wax and octadecanol). An optimal formulation was also obtained by mixing a JET A-1–Zn stearate gel with an ethanol–HPC gel, demonstrating the possibility of blending different solutions and achieving intermediate properties in the compound. Gelled fuels were subject to a preliminary rheological test campaign, investigating shear-thinning behavior, yield stress, flow point, and power-law parameters. Parts of the results were discarded since instrument sensitivity did not capture too low viscosity data. This paper addresses the stability of gels under vibration stimulus. This novel approach is aimed at reproducing the most significant shaking profiles for different phases of a ramjet mission, and qualitatively establishing the tendency of separation under relevant use case conditions. The attitude of recovering the original properties at the end of the disturbance could also be observed. Finally, the ideal combustion performance of the gels was compared to that of pure liquid fuels.

The outcomes of such analyses were critically analyzed and combined. Composition E3 (52 % ethanol, 38 % JET A-1, 7 % Zn stearate, 3 % HPC) provided the best performance from the stability viewpoint. Alternatively, K10 (85 % JET A-1, 10 % Al stearate, 5 % Zn stearate) featured similar stability and a more pronounced shear-thinning property, as well as better thermochemical performance. Solutions made of Thixcin® R or THIXATROL® ST confirmed their good performance, thoroughly discussed in the literature [23], whereas stability and rheology of gels with Zn stearate (K8, K9) did not provide remarkable advantages.

As a natural extension to this work, the suspension of metal particles in the proposed gels should be studied to identify and characterize potential changes in rheological and stability behaviors, as well as to assess the optimal quantity of metal additives. According to the open literature, aluminum and boron are the two most promising solutions, as they have already proven significant energetic contributions in ramjet applications [4,43,44]. Injection performance should be thoroughly assessed. Indeed, the pressure drop used to atomize gels is higher than the one needed for liquids due to their enhanced viscosity. This could also cause the formation of agglomerates, ligaments, and net-like structures [45]. The long-term stability of the gels should be assessed under both static and dynamic conditions, replicating ground and in-flight storage, respectively. Finally, the potential of "hybrid" gels, obtained by blending two or more gelled fuels (e.g., K10, E3), can be of interest as mixed products may inherit the features of the original components.

Author Contributions: Conceptualization, F.M. (Filippo Maggi) and S.C.; methodology, S.D., F.M. (Francesco Morando) and S.C.; formal analysis, F.M. (Filippo Maggi) and S.C.; investigation, S.D. and F.M. (Francesco Morando); resources, F.M. (Filippo Maggi); data curation, S.D. and F.M. (Francesco Morando); writing—original draft preparation, S.D. and F.M. (Francesco Morando); writing—review and editing, S.D., F.M. (Francesco Morando), S.C. and F.M. (Filippo Maggi); supervision, F.M. (Filippo Maggi) and S.C.; funding acquisition, F.M. (Filippo Maggi). All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

Acknowledgments: The support provided to the experimental campaign by senior technicians Alberto Verga and Maria Rosaria Pagano is gratefully acknowledged.

Conflicts of Interest: The authors declare no conflicts of interest.

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