Dynamic Rheological Characteristics of Ethanol Based Gel Propellant

Dong Gi Lee, B.V.S. Jyoti and Seung Wook Baek
*Division of aerospace Engineering, School of Mechanical, Aerospace and Systems Engineering, Korea Advanced Institute of Science and Technology (KAIST), 291 Daehak-ro, Yuseong-Gu, Daejeon 305-701, Republic of Korea
*Corresponding Author: swbaek@kaist.ac.kr

ABSTRACT
Because of the high toxicity of conventional propellant, the interest about low toxic and eco-friendly propellant is getting higher than before. Gelled propellant is the most promising solution at the time, and we chose ethanol as parent fuel which is liquid propellant of low toxic. In present study, the experiments had been performed to analyze dynamic rheological characteristics of ethanol gel propellant. The gelling agent used for gelation process was methylcellulose, and its concentration was three different wt% of 9, 10 and 11. To check the linear viscoelastic range of ethanol gels, strain sweep test had been conducted. The intra 3D network structure of ethanol gels were starting to collapse around 20 Pa. Phase angle of ethanol gels were checked to determine and verify the yield point. Frequency sweep test had been conducted to analyze elasticity and viscosity of ethanol gels. From the result of frequency sweep test, we can identify that elasticity is dominant in this viscoelastic gel.

Keyword: dynamic rheological characteristics, ethanol, methylcellulose, gel

1. Introduction
It is imperative that we build and develop a safer, more convenient and cost-effective environment for exploring new frontiers in space research. Also, because of the high toxicity of conventional propellant, interest in developing low toxic and eco-friendly propellant is getting higher than ever. Gelled propellant is currently the most promising solution, for it has high energy density and high specific impulse. Gel propellant has complex thixotropic nature of which chemical properties had been altered from parent liquid fuel by addition of suitable gelling agent. It is compatible with the conventional liquid propellant systems and easy to synthesize. Gel propulsion systems offer various advantages compared with both conventional liquid and solid propulsion systems. They offer not only high performance and energy management of liquid propulsion system, but also stable storability and density impulse of solid propulsion system. A gelled bipropellant propulsion system resembles a liquid propulsion system except for that the separately stored fuel and oxidizer are in gelled state. If we choose non-toxic fuel with suitable gellant, the low toxic eco-friendly gel propellant can be made.

Gel propellants are being actively studied in recent years, but research on low toxic gel propellant has not been done yet. To achieve eco-friendly nature, fuel has been chosen as an ethanol, which is used as low toxic liquid propellant. The gelling agent used here for gelation process was methylcellulose for three different wt % of 9, 10 and 11. The experiments had been performed to analyze dynamic rheological
characteristics of ethanol-methylcellulose gel propellant with a rotational rheometer.

2. Methods

In order to design and develop a gel propulsion system, a study about the rheological properties should be conducted. From the rheological properties, understanding and estimation of gel propellant flow behavior in pipes and combustion chamber is possible while the gel propulsion system is in operation. Also in the case of gel propellant, which is non-Newtonian fluids, the importance of rheological property test is much higher than conventional propellant.

Dynamic rheological characterization of ethanol-methylcellulose gel was performed using a rotational rheometer (HAAKE RS600). Rotational rheometer is basically used for the rheological characterization of non-Newtonian, thixotropic fluids. A set of parallel plate geometry was used for the present investigation since it is good for linear viscoelastic fluids. A temperature control system inside the equipment has ensured a constant temperature condition during all rheological measurements. The temperature was kept with 298±1 K during all measurements.

![Rotational Rheometer (HAAKE RS600)](image)

In order to investigate the viscoelastic characteristics of ethanol-methylcellulose gel, the oscillation (dynamic) tests were performed to determine the storage modulus ($G'$) and the loss modulus ($G''$) at different frequency and amplitude. The gel samples were exposed to harmonic oscillation, while the storage and loss moduli would be observed by the change of frequency or amplitude of harmonic oscillation in dynamic sweep study. In the case of viscoelastic materials, both viscosity and elasticity affects the flow behavior of sample at the same time, and it will cause a gap between force applied timing and reaction timing. From that difference, phase angle, which is bigger than 0° and smaller than 90° in the graph of storage and loss moduli versus time, will give the information about the dominance property between viscosity and elasticity. Also we can identify the range which shows stable behavior of rheological properties.

Strain sweep test and frequency sweep test were also conducted in the present study. In case of the strain sweep test, the amplitude of harmonic oscillation applied to the sample was increased by logarithmic scale while the frequency was fixed. From the
graph of storage modulus versus shear stress, linear viscoelastic range could be identified. Linear viscoelastic range gives the information about strength of the sample, and the sample shows predictable behavior in this range. Frequency sweep test is one of dynamic sweep tests; it enables the viscoelastic properties of a sample to be determined as a function of timescale. In case of the frequency sweep test, the frequency of harmonic oscillation applied to sample was increased by logarithmic scale, while the amplitude was fixed. By comparison of storage modulus with loss modulus, dominant property between viscosity and elasticity can be determined. Also by checking the shape of storage and loss moduli versus frequency graph, intra structure of the gel can be estimated whether the molecules involved form a strong 3D network or a well dispersed collide.

3. Results and Discussion

3.1 Strain sweep test

The study for strength property of gel propellant is essential, because relocation or breakage of internal 3D network structure of gel propellant is relatively frequent than liquid and solid propellants. Strain sweep test had been performed to determine linear viscoelastic range (LVR). LVR means the range that the storage modulus \( G' \) is independent of shear stress. Within LVR, rheological properties of ethanol gel are stable, which means that its flow behavior is predictable.

Figure 2 shows the result of strain sweep test of three ethanol gels. The frequency had been fixed at 5 Hz, while the range of shear stress is 1-1000 Pa.

![Figure 2 Strain sweep test of ethanol gels](image)
The value of \( G' \) is an order of concentration of gellant. Higher concentration of gellant means larger branch of 3D network and it also means stronger bond established between ethanol and methylcellulose molecules.

The deviation in the range which shows linear behavior of \( G' \) is smaller than the average of first five points by 10%. The criterion of decline is that \( G' \) is smaller than 85% of its average. All three gels show linear behavior at first, but \( G' \) for 9 wt % gel decreased dramatically when the shear stress passes 20 Pa. When the shear stress reaches 50 Pa, \( G' \) for 10 and 11 wt % gels was also starting to drop significantly.

Increased shear stress makes amplitude change, which is too big to sustain 3D network structure of ethanol gel. That is why the structural break down goes on in the gels from this moment. When it begins, \( G' \) and concentration of gellant are meaningless because those are unpredictable.

The 9 wt % gel has the shortest LVR because its concentration of gallant is the smallest, but 10 and 11 wt % gels have almost the same LVR. Based on this, it can be assumed that there is a physical limit to a reinforcement of the strength of chemical bonding using more concentration of gellant. Therefore it is not necessary to consider higher concentration than 10 wt %.

Figures 3, 4 and 5 depict \( G' \) versus phase angle (\( \phi \)) for three ethanol gels. Phase angle means a difference between storage modulus \( G' \) and loss modulus \( G'' \), which represents an information that can determine a yield point. If two trend lines are drawn along the linear range from the beginning point and the other linear range from the endpoint except for yield point area, the intersection point is the yield point. The yield point can be a reference to estimate precise LVR. It had been calculated from the function in HAAKE RheoWin Data Manger which is operating software of rheometer.

![Figure 3](image-url)  
**Figure 3** Storage modulus and phase angle of 9 wt% ethanol gel
In the case of 9 wt% gel in figure 3, the phase angle is linear but it increased from 10 Pa to the end. The calculated yield point is 13.86 Pa. The yield point for 10 wt% gel is 30.08 Pa, and for 11 wt% gel it is 26.57 Pa.
3.2 Frequency sweep test

Frequency sweep test was conducted to study viscoelastic property of gel network structure by comparing $G'$ and $G''$ by frequency change. It was performed from 0.5 to 50 Hz while the shear stress was fixed at 10 Pa which is in the LVR.

Figure 6 Frequency sweep test of ethanol gels

Figure 6 depicts the result of frequency sweep test for three ethanol gels. $G'$ is much larger than $G''$. Storage modulus $G'$ means the energy which is kept in elasticity part, and loss modulus $G''$ means the energy which is kept in viscosity part. If $G'$ is larger than $G''$, it means that the strong interaction between molecules and hydrophobic bond has much more resistance to the breakage of structure. Therefore for all the three gels, elasticity is more dominant than viscosity. Moreover, the structure of ethanol gel, which is based on cohesion of ethanol and methylcellulose, forms 3D network from the fact that both moduli are linear versus frequency in a logarithmic scale.

4. Conclusion

In this present study, the dynamic rheological characteristics of ethanol based gel propellant, which used methylcellulose as a gelling agent, have been investigated experimentally. Dynamic testing with rotational rheometer was also conducted to better understand micro structure of gel propellants. It was observed that the gellant concentration is one of the major factors which affect the dynamic rheological properties of gelled propellants. Linear viscoelastic range was determined via strain sweep test. Yield point of each ethanol gel was estimated from the value of storage modulus and phase angle. From the frequency sweep test, it was found that the
elasticity was dominant in this ethanol gel. To achieve the reliability in the gel propellant system, combustion study and stability test are required.

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