Atomization characteristics of impinging jets of gel material containing nanoparticles

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ABSTRACT

We have investigated the spray and atomization behavior of 0.35% C934 Carbopol gels with or without 15 wt.% SUS304 nanoparticles of 100 nm using a doublet like-on-like impinging jet system (orifice diameter of 0.7 mm, impingement angle 2θ of 90°) and compared with the spray behavior of water. The spray patterns of gel materials were qualitatively different from those of water and could be divided into four types: open rim and no shedding drop; rimless sheet with ray shaped shedding; sheet with ligament separation; fully developed. The sheet sizes of water and the pure Carbopol gel increased with the increase in jet velocity, but the sheet size of the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio (lmax/Wmax) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and the spreading angle was smaller when compared with water. The breakup length of the pure Carbopol gel was longer than that of water. However the breakup length of the Carbopol gel containing SUS particles was remarkably shorter than that of the pure Carbopol gel due to the increase in impact energy by the heavy particles as well as the decrease in gel strength by the addition of nanoparticles. The drop Sauter mean diameter (SMD) decreased with the increase in jet velocity, and it converged to an asymptotic value. The asymptotically convergent drop SMD of water was about 100 µm while that of Carbopol gels was 75–77 µm. The elasticity of gels appears to reduce the asymptotic drop size.

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1. Introduction

There has been a growing interest on the development of rocket propellant using gelled fuels and oxidizers because the rocket propellant using a gelled fuel has been considered to complement some drawbacks of conventional fuels or oxidizers. The drawbacks include the increasing hazard of fire or explosion due to continuous leakage when the storage tank is broken by external shocks and the performance limit of the liquid fuel i.e., a lower specific impulse affected by the design of nozzle, operation of the flow system and material properties such as yield stress and viscosity increase and hence the protections of the liquid propellant can be ensured against storage risks. Also the restraint of particle sedimentation by the increased viscosity and yield stress makes it possible to add metal particles to the liquid propellant, and subsequently to enhance the performance further. However it has been recognized that the rheological properties of gel material make it difficult to atomize gel propellant into fine drops [1].

For efficient combustion, fine drops have to be produced in a stable manner. To understand the atomization phenomena many studies have been conducted experimentally or theoretically on the shape of the liquid sheet, breakup length and drop size formed by the impinging jet. Until now the doublet like-on-like impinging jet has been widely used for atomization in the field of combustion and many experimental results have been reported on the atomization of Newtonian fluids [2–9]. In this process two jets collide to form a liquid sheet and the sheet becomes unstable and finally disintegrated into drops. The spraying and atomization processes are affected by the design of nozzle, operation of the flow system and material properties of the liquid. In spraying and atomization by the doublet like-on-like impinging jet, orifice diameter (d₀), impingement angle (2θ) and jet velocity (U) are known to be important as well as material properties such as viscosity (η), surface tension (σ) and density (ρ). Heidmann et al. [2] performed comprehensive experiments regarding how orifice diameter, jet velocity, impingement angle, pre-impingement length and liquid properties affected the spray structure of impinging jets. They classified the spraying patterns into four different types of closed rim, periodic drop, open rim and fully developed patterns, and realized the importance of liquid jet velocity and impingement angle in characterizing the spray structure. Dombrowski and Hooper [3]
performed impingent jet atomization experiments while changing the impingement angle from 50° to 140°. They showed that the disintegration generally resulted from the formation of unstable waves of aerodynamic and hydrodynamic origins. Lai et al. [4] used the photographic image technology to examine the like-doublet impinging jet of water and showed that a liquid sheet was formed when the momentum of impinging jets was large enough to overcome the surface tension of liquid. They found that the maximum length (L\text{max}) and width (W\text{max}) of sheet increased in the form of the 2nd order polynomial as jet velocity increased. They also discovered that sheet length was independent of the impingement angle although sheet width increased with impingement angle. However their ratio (L\text{max}/W\text{max}) was constant under a given impingement angle independent of jet velocity. Lai et al. [5] investigated how viscosity and surface tension of the working fluid affected the atomization characteristics of the like-doublet impinging jet spray. They reported that ten different spray patterns were observed by varying the viscosity and surface tension of fluid. Especially a new pattern named as "the incredible periodic pattern" was found in addition to the four spray patterns suggested by Heidmann. Lai et al. [5] also revealed that the length-to-width ratio of the sheet formed at the closed-rim mode was constant within the velocity range to maintain the closed rim. The length-to-width ratio increased with the increase of viscosity while this ratio varied little with the change in surface tension. High viscosity and high surface tension fluids made large drops when the velocity was small, whereas the viscosity and surface tension did not influence the drop size if the jet velocity was over 20 m/s (this velocity may be called the critical velocity). Moreover, in case of fully developed spray with a high enough velocity over 30 m/s, drop size was not influenced by the viscosity and surface tension of fluid, and it approached to an asymptotic value (about 50 μm in their case) which was strongly dependent on orifice diameter.

To understand the atomization process theoretical studies on breakup length and drop size have also been performed for Newtonian fluids. It has been known that the spatial distribution of drops is important for the combustion efficiency and flame stability, and the drop size and its distribution are dependent on the size and thickness of sheet as well as the breakup length of sheet [7]. In this case the linear stability theory is most commonly used to understand the breakup of liquid sheets formed by the impingement of two liquid jets. Squire [10] performed a hydrodynamic stability analysis on the constant thickness and inviscid liquid sheet. He derived the most unstable wave number for the anti-symmetric mode and the corresponding maximum growth rate factor to become ligaments. But he did not consider the formation of droplets from the ligaments. Dombrowski and Johns [11] formulated the drop-size relation by the analysis of aerodynamic instability. They assumed that drops were generated by three steps: the growth of waves on liquid sheet, fragmentation and formation of ligaments and finally breakdown of ligaments into drops. They obtained the drop diameter as follows:

\[ d_{D} = \left[ \frac{3\pi}{\sqrt{2}} \right]^{1/3} d_{l} \left[ 1 + \frac{3\eta}{\rho \sigma d_{l}} \right]^{1/3} \]  

(1)

where the ligament diameter \( d_{l} \) is given by the following formula

\[ d_{l} = 0.9614 \left( \frac{K^{2/3} \sigma}{\rho \mu^{1/3} U^{1/3}} \right)^{1/3} \left[ 1 + 2.66 \rho \mu^{1/3} U^{1/3} \right]^{1/3} \]  

(2)

In the above equation \( K \) is a constant which determines the thickness \( h \) at any point \( x \) by \( h = K/\kappa \) for a radiating sheet of uniform velocity. Weihls [12] considered the radially spreading liquid sheet with viscosity. He found that, in addition to the conventional Kelvin–Helmholtz instability, a new type of instability called spatial instability also appeared. He also showed that waves on the sheet should grow to a certain distance and then damped further away from the origin. This was already observed experimentally by Crapper et al. [13]. He also predicted that for viscous liquids (kinematic viscosity of 1 St) the wave length at the maximum instability would be much longer than that of less viscous fluid. Anderson et al. [14] and Ryan et al. [15] found that the deviation in the wave number predictions between the viscous models and the inviscid models increased with increasing sheet thickness and jet velocity. However the maximum growth rates predicted by the inviscid and viscous analyses were the same for the conditions of their study. Finally, they derived an explicit expression for the breakup length (\( X_{b} \)) and the average drop size (\( d_{0} \)) as follows:

\[ X_{b} = 5.451 \frac{s}{d_{0}} \left( \frac{\text{We}}{\text{f}(\theta)} \right)^{-1/3} \]  

(3)

\[ d_{0} = \frac{1.144 s}{\text{We}} \left( \frac{\text{f}(\theta)}{\text{f}(\theta)} \right)^{-1/3} \]  

(4)

where \( d_{0} \) is the orifice diameter, \( s \) is the ratio of gas density to liquid density, \( \text{We} \) is the Weber number based on liquid properties, jet velocity and orifice diameter, and \( \text{f}(\theta) \) is given by

\[ \text{f}(\theta) = \frac{(1 - \cos \theta)^2}{\sin^2 \theta} \]  

(5)

where \( \theta \) is the impingement half-angle. Therefore a very simple correlation was obtained without including the effect of the viscosity. It was suggested that viscosity had a damping effect on wave growth. Ibrahim and Przekwas [16] analytically treated the discretization of the liquid sheet formed by two impinging jets. They suggested that two breakup regimes should be present based on Huang’s [17] experiments on opposed jets. In addition they also suggested that, at a low Weber number regime (\( \text{We} < 2000 \)) the stationary anti-symmetric wave theory should determine the shape of the sheet, while at a high Weber number regime (\( \text{We} > 2000 \)) the linear stability theory should be used. Thus a general expression for the sheet thickness formed by two impinging jets at low Weber numbers is as follows:

\[ r_{s} = \frac{d_{h_{0}}}{2h_{c} \sin \theta} \]  

(6)

where \( r_{s} \) is the distance from the impingement point to the sheet edge, \( h_{0} \) is the initial sheet thickness, and \( h_{c} \) is the thickness at the edge of the sheet. Both \( h_{0} \) and \( h_{c} \) are functions of angular position.

The theoretical approach on the atomization and spray of gel material involves a large amount of computational works, and it will be even more complicated for gel materials containing particles. Therefore many studies regarding spray and atomization of gel materials have relied on experimental works. For example, Chojnacki and Feikema [18,19] used a Carbolpol gel as a simulant of gel propellants and subsequently controlled the impinging angle and jet velocity under the turbulent jet condition. They reported that the critical Weber number (\( \text{We} = \rho U^{2} d_{0} \sin^2 \theta/\sigma \) at which the hydrodynamic wave began to appear was in the range of 400–500 when using 0.5 wt.% Carbolpol 941 gel. In fact, Dombrowski and Hooper [3] used the Weber number first and showed that the critical Weber number was in the range of 66–165 for water. Chojnacki and Feikema [18,19] attributed the difference to the higher viscosity of the gel and the change in the velocity profile. They also found that a cardioid shaped wave structure appeared below the Weber number, but a circular wave emanated from the point of impingement and propagated through the sheet with short wavelengths above this Weber number. However their experimental results on the occurrence of instability were re-
rectified to low velocity regimes and therefore they did not consider the generation of droplets with the gel. Recently Lee and Koo [20] reported an experimental study on the break-up characteristics of Carbopol gels. They noticed the importance of the rheological properties of gel materials, but their studies were generally qualitative rather than quantitative. Jayaprakash and Chakravarthy [21] tested a gelled fuel composed of kerosene and aluminum particles. They studied atomization characteristics of gel propellants using two orifices (1 and 1.5 mm in diameter) and varying impinging angles (50°, 60° and 70°). The injection pressure was in the range of 3–7 bar, which corresponds to the jet velocities of 10–20 m/s. They found that the spray angle and the breakup length decreased with the increase in injection pressure and this tendency was prominent at low impinging angles of 50° and 60° rather than at 70°. When calculating drop size using images taken at 75 mm downstream from the impinging point, the drop size distribution was broader and the mean drop size was larger at the low impinging angle compared to the high impinging angle. They argued that all of these results could relate to the spray angle and the breakup length. It is noted that, although drop size decreases with the increase in injection pressure within the velocity range they have tested, the velocity range is relatively narrow and the measurement of the drop size may not be accurate enough because one pixel size of the image is almost 25 μm, just one half of the drop size. von Kampen et al. [22] and Madlener et al. [23] studied the effect of aluminum particle content on spray and combustion characteristics of gelled jet A-1 fuel. The particle size was 10 or 80 μm and the aluminum content was 0–40 wt.%. From the atomization experiment, they identified three types of spray patterns, i.e. ray-shaped pattern, ligament pattern and fully developed pattern. This result is different from Heidmman’s report in that the closed rim and periodic drop pattern were absent while the ray-shaped pattern newly appeared. They observed that the generalized Reynolds number (based on the power-law) at the point of transition from the ray-shaped pattern to the ligament pattern was similar to the critical Reynolds number (Re) discriminating between laminar and turbulent flows. They also found that the breakup length and width of the gel sheet reduced when Re was increased and both break-up length and sheet width further increased for the gel with higher aluminum content.

Until now there has been nearly no study on the atomization of gel material containing nano-sized particle except two reports on the combustion characteristics of real gel propellant containing nanoparticles. Tepper and Kaledin [24] revealed that the addition of nano-sized aluminum particles in a kerosene gel propellant for an air breathing system reduced the ignition delay time of the gel propellant. Mordosky et al. [25] observed that the use of nano-sized aluminum powder in gel propellants increased the efficiency of combustion as well as the performance through complete combustion. Although the addition of nanoparticle has some advantages in combustion, we may still encounter difficulties in the understanding the spray and atomization of gel materials containing nanoparticles. Therefore it should be very important to investigate these processes as they are deeply related to the combustion characteristics. In the present study we have characterized the doublet like-on-like impinging jet atomization of gel material containing nanoparticle. In addition we have studied the change of spray and atomization characteristics with rheological properties by performing a series of experiments with water (Newtonian fluid) and Carbopol gels with or without nanoparticles (non-Newtonian fluids). The result shows that the instability characteristics are quite different depending on fluids especially at low velocities and the average drop sizes of gel materials at the asymptotic condition are smaller than that of water. The atomization is an old but very important topic in the case of Newtonian fluids. But there have been no systematic studies on gels especially with nanoparticles. The present study can shed light on this field and contribute to the development of better propellants.

2. Experiments

2.1. Materials

Three kinds of fluids were tested: water representing Newtonian fluid; pure 0.35 wt.% Carbopol gel; and Carbopol gel containing 15 wt.% stainless steel (SUS304) nanoparticles. The master solution was prepared by dissolving 1 wt.% of Carbopol C934 (provided by Noveon, USA) powders in de-ionized water and mixing with a magnetic stirrer over 2 days. Gel samples used for the present study were prepared by diluting the master solution and agitating with a 3-impeller mechanical stirrer of 100 mm at approximately 750–800 rpm. Triethanolamine (TEA: purity 98%, Aldrich Chem. Co.) was added as a neutralizer and the pH was 8.0 ± 0.3 at the final gel state. As the nanoparticles SUS304 particles (provided by Nano Technology Inc., Korea) were used. The particles have the average diameter of 100 nm and the bulk density of 7930 kg/m³. Bubbles in the gel materials which were produced during the gel preparation stage were removed by keeping the filled reservoir in a vacuum at 740 mm Hg for 15 min.

The detailed rheological properties have been studied recently in our laboratory and hence the paper could be referred for detailed information [26]. But to be more readable we have briefly reviewed our previous paper focusing on the issues relevant to the present research as follows: We explored the rheological characteristics of Carbopol C934 gels with or without SUS 304 spherical nanoparticles of 100 nm. In comparison with the pure Carbopel gel, the SUS nanoparticle filled Carbopol gel exhibited stronger shear thinning and higher yield stress. As the concentration of nanoparticles increased yield stress increased, but viscosity and storage modulus increased first and then decreased abruptly beyond the critical limit. Also as the concentration of nanoparticles increased there was a transition in material characteristics from the ductile type to the brittle type, which means that highly filled Carbopol gels lost the structure almost instantaneously as the imposed stress was larger than the yield stress. This also means that the nanoparticle-filled gel is very fragile. The cryo-SEM analysis revealed that particles are evenly attached to gel surface without clumping and the network structure changed abruptly when the rheological properties changed abruptly. The change in gel structure is attributed to the nanoparticles that compete with Carbopol chains in forming networks.

2.2. Atomization equipment

The experimental setup used for our study is shown in Fig. 1 and the detailed design of the jet injector is shown in Fig. 2. The liquid for impinging jet atomization was filled in an approximately 3-liter cylindrical reservoir and pressurized by a piston. The pressure was controlled by the regulators of two parallel connected nitrogen tanks, and the flow of the material was controlled by a solenoid valve at the bottom of the reservoir. The flow was divided into two tubes which a jet injector was finally coupled to. For atomization a doublet like-on-like impinging jet injector device was used. The orifice diameter of the injector was 0.7 mm and the length to diameter ratio of the orifice was 10. The entrance wall was inclined by 25° to reduce the formation of vortex by the abrupt contraction. The distance from the orifice exit to the impinging point was 10 mm and impingement angle (20°) was fixed at 90°. Fig. 3 shows the shape of the liquid sheet formed by two impinging jets. Shadowgraph images were taken from two perpendicular directions to show the spray behavior. A CMOS high speed camera
3. Results and analysis

3.1. Material properties and jet velocity

A propellant can experience a wide range of shear rate. At the storage state the shear rate almost vanishes while it can be very large when subjected to an atomization process. Especially at the impinging point of the two colliding jets the shear rate can be as large as $10^3$ s$^{-1}$. In the case of gel materials, the solid-like properties such as yield stress and high viscosity at the low shear regime enhance the storage stability and helpfully restrict the sedimentation of particles. At the high shear rate regime, the gel material should have a viscosity low enough to flow and atomize like a common liquid propellant. It is known, however, complex rheological properties can cause adverse effects on feeding under the required high pressure. They can also complicate the system in controlling mass flow rate [1].

In the present study, we will focus on the effect of viscosity, particularly at the high shear rate regions. Then the relevant dimensionless groups should be Reynolds number and Weber number defined as follows:

$$Re_{gen} = \frac{\rho U^2 d_0}{K \left( \frac{n+1}{n} \right) \frac{3}{8} n^{\frac{3}{4}}}$$

$$We = \frac{\rho U^2 d_0}{\sigma}$$

Here the generalized Reynolds number ($Re_{gen}$) is defined for the power-law fluid with consistency $K$ and power-law index $n$. The average shear rate can be estimated by the equation $\dot{\gamma} = \frac{8}{d_0} \frac{U}{U}$, where $d_0$ is the injector diameter and $U$ is the jet velocity. In the present study $\dot{\gamma}$ is in the range of $10^4$–$10^5$ s$^{-1}$. Fig. 4 shows the viscosities of the Carbopol gels measured by a modified capillary viscometer (RH7, Malvern Instrument). The Carbopol gels show highly shear-thinning characteristics and obey the power-law within the measured range. The power law indices ($n$) of the Carbopol gels with and without particles are 0.36 and 0.30, respectively. The viscosity of the Carbopol gel containing 15 wt.% SUS nanoparticles is more than ten times the viscosity of pure Carbopol gel. At the shear rate of $10^4$ s$^{-1}$, the particle containing gel is 1000 times more viscous than water.

Surface tension is another important material property in the atomization process besides viscosity. Some of the most frequently used methods to measure the surface tension of liquid are the ring method and the Wilhelmy slide method. But these methods were
found to be not appropriate for the gel materials with strong elasticity since the liquid column did not break at the critical point when the surface tension force and the gravity force became the same. Rather the liquid column extended almost indefinitely. Therefore to measure the surface tension of the gel we utilized the Laplace–Young equation directly:

$$2\kappa_m = \frac{\Delta \rho}{\sigma} g \cdot x + B$$  \hspace{1cm} (9)

In the above equation $\kappa_m$ is the mean curvature, $g$ is the gravitational acceleration, $x$ is position vector and $B$ is the total curvature at $x = 0$. To measure the surface tension by applying this equation a small pendant drop was generated by using a syringe and the shape of the drop was analyzed by taking an image as shown in Fig. 5. We took two points to determine the curvature: one at the bottom of the drop ($x = 0$) and the other at the side of the drop as shown in Fig. 5a. At point A, one of the curvatures is the inverse of the maximum radius and the other curvature can be obtained by fitting the shape of the drop. Then the two curves should be perpendicular to each other at the point A. The curvatures at the bottom and the side were determined by fitting the outline of the drop to second order polynomials. Because the curvature was very sensitive to the shape of the outline, the position of the drop outline was determined to the sub-pixel value by using the linear interpolation method. The sub-pixel value was chosen so that the boundary was located at the gray value of 150. The maximum radius and the length between the bottom and the position where the maximum occurs were determined from the difference in sub-pixel values. When this procedure was used the surface tension of water was 69 mN/m which was quite close to the literature value of 71. Using the same procedure the surface tensions of Carbopol gels were measured from the images shown in Fig. 5b–g. The reason why less concentrated Carbopol gels were measured is to confirm whether our method can be applied to highly concentrated gels with yield stresses. In the case of materials with yield stresses the equilibrium shape of the pendant drop may not be obtained since the deformation will stop when the stress due to the flow reaches the yield stress before reaching the equilibrium. With the increase in Carbopol concentration, surface tension decreases slightly first then decreases abruptly when Carbopol concentration becomes larger than 0.25%. Actually this value is much the same as the appearance of yield stress [26]. We believe that the surface tension values over 0.25% contain some artifacts due to yield stress. Rather the dotted line in Fig. 6 is believed to be closer to the true value. To measure the surface tension of the gel the Zisman method [27] can be used for determining the solid surface tension noting that the Carbopol gel behaves as a solid under the rest state. In this method a liquid drop is placed over the gel surface and the wetting behavior is observed. When it is fully wetted (the contact angle is zero) the surface energy of the solid surface is larger than the surface tension of the liquid; when it is not fully wetted, the surface energy of the solid surface is smaller than the surface tension of the liquid. Therefore if a series of experiments are performed while changing the liquid with differing surface tension, the surface tension of the gel can be determined at least theoretically. However, since the gel is not a real solid the liquid can be imbibed into the gel. Also most of the organic liquids that do not mix with water (the base liquid of the gel) have much lower surface tension than the gel. So only the lower bound can be determined if these organic liquids are used. When a drop of silicone oil (surface tension: 23 mN/m) was placed over the gel surface it was fully wetted. This means that the surface tension of the gel is at least 23 mN/m which is much larger than the value obtained by the pendant drop method of 6 mN/m. This, in turn, means that the value obtained by the pendant drop method contains a large error. When a drop of water was placed the contact angle was finite. Since the temporal equilibrium was attained

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**Fig. 5.** Pendant drop images of (a) water, (b–g) pure Carbopol gels (0.1 wt.%, 0.15 wt.%, 0.2 wt.%, 0.25 wt.%, 0.3 wt.%, 0.35 wt.%) and (h) SUS containing gel.

**Fig. 6.** Surface tension variation of Carbopol gel with concentration. The dotted line is the estimated surface tension for more concentrated Carbopol gel.
within a second, we supposed that water was not imbibed during this short time. This means that the surface tension of the gel should be smaller than the surface tension of water. Hence we regard that the extrapolated value could be sufficiently close to the true value. Also the surface tension value was used only in calculating Weber number and since the material is fixed, there appears to be no confusion in the order of sheet transition forms. Hence we will use the extrapolated value of 54 mN/m as the surface tension of the 0.35% Carbopol gel. In the case of Carbopol gels containing nanoparticles no rounded drop shape could be obtained. The liquid behaved like tooth paste as shown in Fig. 5h and therefore there was no way to measure the surface tension of this material. Therefore in calculating the Weber number of the flow of gels with nanoparticles we will use the same surface tension value of 54 mN/m of pure Carbopol gel without nanoparticles.

Since the atomization process is basically an unsteady process the more relevant surface tension should be the dynamic surface tension. In the atomization process the typical velocity and length scales are 50 m/s and 100 mm (The travelling length of liquid along the sheet formed by two jets before forming ligaments), respectively, as described below. This means that the typical timescale (surface age) is 2 ms. As far as the authors are aware of, this time scale is too small to measure the dynamic surface tension with the currently available methods. However, it is noted that there may be no large difference in Weber number even if we use equilibrium value rather than the dynamic surface tension since the equilibrium surface tension is not much reduced from the water value (72–54 mN/m in the present case) as in the case of surfactant solutions that have been studied most on dynamic surface tension (usually 72–20 mN/m). Chemically we may not assume that the dynamic surface tension will be close to the water value since the Carbopol gel has a network structure and when a fresh surface is formed it is not certain that the surface is free of Carbopol polymers. Also all the previous reports used the equilibrium surface tension. So to be more consistent with literatures it appears to be reasonable to use the equilibrium surface tension in calculating Weber number.

In the analysis of an atomization process, the jet velocity is a very important parameter. To know the average jet velocity, we have to know the flow rate of the fluid. Since it is difficult to measure the flow rate using a conventional flowmeter in the case of non-Newtonian fluids such as a gel material having a yield stress and a shear rate dependent viscosity, we used a simple and robust catch-and-weigh method. By weighing the ejected fluid during a fixed time interval (10 s) under a certain pressure, we can calculate the jet velocity from the following equation:

\[ U = \frac{Q}{4\pi d_0^2}, \quad Q = \frac{\dot{m}}{\rho_l} \]  

(10)

Figs. 7 and 8 illustrate the jet velocity variations with the inlet pressure of the injector inlet for water and Carbopol gels, respectively, together with the correlation equation for each curve. The jet velocity was calculated from the correlation equation. The pressure difference between two injectors was found to be negligibly small, and hence the present system should be considered to be a like-on-like impinging jet. The gel materials tested in the present study have yield stresses which have to be overcome to flow. The presence of yield stress can be confirmed by the negative value of \( y \)-intercept in the correlation equation between inlet pressure and jet velocity. In other words the inlet pressure has to reach the critical inlet pressure in order to make the gel flow. This can be seen from the positive \( x \)-intercept in Fig. 8. The jet velocity of the Carbopol gel containing 15 wt.% SUS particles was lower than that of the pure Carbopol gel at the same inlet pressure as a natural consequence by viscosity difference. However the relation between jet velocity and inlet pressure was not affected qualitatively by particle addition.

3.2. Spray patterns and breakup length

3.2.1. Newtonian fluid

Before investigating the atomization of gel materials, the spray and atomization of water were considered first to validate our jet impinging device by comparing with the previous results obtained by Lai et al. [4] and Ciezi et al. [8]. Fig. 9 shows the images taken from both front and side directions at six different ejection velocities. First of all, from the side view images, we can see the transition to instability. Fig. 9a and b are the representative images of the "closed rim with shedding drops" pattern suggested by Dombrowski and Hooper [3]. This pattern is the combined pattern of "closed rim" and "periodic drop" suggested by Heidmann et al. [2]. In this pattern the liquid sheet surrounded by a thick rim and drops are detached tangentially off the sheet periphery. The ratio of the maximum length (\( L_{\text{max}} \)) to the maximum width (\( W_{\text{max}} \)) of sheet is approximately 2.1, which is similar to 2.2 from Lai et al.’s [4] result. In Fig. 9b, some waves appear from the impinging point first, decay along the downstream direction and then grow again. This is the same pattern already observed by Crapper et al. [13] and predicted by Weihs [12]. Dombrowski and Hooper [3] reported that the wave could appear at the lower jet velocity in the case of turbulent flows.
In our experiment the wave begins to appear when the velocity becomes 5 m/s which corresponds to We = 126. This value lies inside the critical Weber number range of 66 and 165 of Dombrowski and Hooper [3].

Fig. 9c–e shows the “open rim with ligament separation” pattern, where the distinct rim disappears and the length of sheet becomes shorter. As the velocity increases a bow-shaped ligament appears at the bottom. In addition, when viewed from the side, the liquid sheet flaps like a sine wave and this phenomenon is enlarged at the downstream. This means that the sinuous mode is more unstable than the varicose mode. This sine wave is amplified with the increase in jet velocity. Spreading angle also increases with the increase in jet velocity. Fig. 9f shows the spray pattern at the jet velocity of 33.3 m/s. In this pattern, it is difficult to see the shape of the sheet because it splits into ligaments and then droplets immediately after impingement. This breakup pattern is defined as “fully developed”. Though the universal jet velocity at which the fully developed pattern begins to appear cannot be defined due to the difference in the experimental conditions among literatures, the fully developed pattern shows when the jet velocity reaches 30 m/s or higher in our experiment. In this case the spreading angle is 70° as can be seen in Fig. 9f. The present set of experiments on the spray and atomization of water is virtually the same as reported in the literatures [4,5,8]. Hence the experimental setup is considered to work properly. In what follows, this result will be compared with the spray and atomization of gel materials.

3.2.2. Carbopol gel without nanoparticles

Fig. 10 shows the spray and atomization patterns of the Carbopol gel without nanoparticles at different jet velocities. When the jet velocity is less than 10 m/s (Fig. 10a), despite the sheet formation, the closed rim as shown in Fig. 9a for water does not exist and the lower part of the sheet is opened. This is the “open rim and no shedding drop” pattern. Huang [17] argued that the closed rim is formed by the equilibration between the inertia of impinging jet and the surface tension of material. So the following two reasons can be considered why the closed rim is not formed at the small jet velocity regime (i.e. low Weber number) in the case of Carbopol gel. First of all, the highly viscous material like Carbopol gel restrains the sheet expansion by damping the inertia force of impinging jet, and consequently the sheet is thickened and the maximum length-to-width ratio becomes larger as Lai et al. [5] argued. Secondly the lower surface tension of Carbopol gel makes it difficult to close the periphery of the lower part of the sheet. So we may argue that the high viscosity and relatively low surface tension of Carbopol gel are the cause of rim opening at the low jet velocity regime.

As the jet velocity reaches 10 m/s, a closed sheet (see Fig. 10b and c) is formed. It is defined as “rimless sheet with ray shaped shedding.” The rim does not appear to be as thick as in the case of water. Web-like ligaments are generated at the periphery of the rim and droplets are spurted in the radial direction from the end of the ligaments. This pattern is similar to the spray result [22,23] of gel propellant containing aluminum particles. In this pattern the sheet may not be formed by the equilibration between surface tension and inertia considering that inertia is much larger than the case of water while surface tension is smaller or at most the same if we consider the maximum dynamic surface tension the gel can have. It appears that the inertia force of impinging jet is balanced by the high extensional stress. Even though we have not been able to measure the extensional viscosity of the Carbopol gel, it should be reasonable to assume that the extensional viscosity of elastic gel is large. The extensional viscosity issue is considered later in more detail. Under this condition, the maximum length-to-width ratio of the sheet is about 1.7, i.e. width is enlarged relatively compared with the case of water. On the other
hand, waves that appeared in Fig. 9b for water on the sheet surface from the impinging point are not observed in Fig. 10c even though jet velocity is high (15.6 m/s). It appears that the high viscosity of the gel mitigates the impinging shock.

Fig. 10d–g shows the third pattern. In this case waves of concentric circles appear on the sheet surface as observed by Chojnacki and Feikema [18, 19] and ligaments are detached from the lower part of the sheet. This pattern can be called as “sheet with ligament separation.” The wave frequency increases and the breakup length decreases as jet velocity increases, which matches the “open rim with ligament separation” pattern of water. As seen in Fig. 10f, spreading is insignificant for jet velocity up to 44.5 m/s. It appears that the high viscosity of gel material obstructs the increase of wave amplitude. This result is unfavorable for the atomization efficiency because drops are distributed only to a certain limited space. Fig. 10g shows the spreading characteristics at 54.7 m/s of jet velocity. This is approximately seven times larger than jet velocity of water when the similar spreading occurs. This result is consistent with Ryan et al.’s [15] anticipation of the damping effect by the high viscosity of gel.

Fig. 10h shows the pattern at 62.3 m/s. From the front view image we cannot distinguish the breakup length and we can only observe well distributed drops. Hence it can be called as the “fully developed” pattern. The spreading angle is increased as well even though it is relatively small when compared with the case of water (Fig. 9f).

3.2.3. Carbopol gel containing nanoparticles

The spray patterns of Carbopol gel containing SUS nanoparticles are shown in Fig. 11. Due to the attachment of flying black gel drops onto the camera lens, it was impossible to take photographs from the side and hence Fig. 11 shows only front view images. First of all, the sheet begins to form at a higher jet velocity since the viscosity was increased by adding particles. The sheet width is smaller compared with the pure Carbopol gel (Fig. 10a) case at the “open rim and no shedding drop” pattern. Fig. 11b shows a unique pattern which does not appear in pure Carbopol gel. In this case drop shedding occurs only at the lower part of the sheet. Another unique feature is that the sheet appears to become unstable in two-dimensional fashion (two dimensional waves may exist). This is not observed in the case of water or pure Carbopol gel. In the case of the pure Carbopol gel, concentric waves begin to form from the impingement point as the sheet becomes unstable. Therefore the pattern should be a new one and
but the overall characteristics are the same as the “rimless sheet with ray shaped shedding” pattern of pure Carbopol gel. As can be seen in Fig. 11b and c, the sheet size is reduced from the pure Carbopol case. Moreover the sheet size does not change with jet velocity whereas the sheet of pure Carbopol gel becomes larger with jet velocity (Fig. 10b and c). However the maximum length-to-width ratio remains the same at 1.7 which is the same as the pure Carbopol gel. If the inertia and the viscosity are balanced, the sheet of the Carbopol gel with nanoparticles is supposed to be more expanded because the jet velocity is higher and the viscosity is higher than that of the pure Carbopol gel. However the sheet becomes disintegrated at a shorter distance from the impingement point by particle addition. It appears that the cohesive force of the gel becomes weaker by the addition of nanoparticles. The fragile nature of the nanoparticle filled Carbopol gels is reported in Baek and Kim [26]. Many of the disintegrated part of fluid do not have a spherical shape in this case reflecting the large viscosity and small surface tension.

Fig. 11. Typical spray images of Carbopol gel containing 15 wt.% SUS particles: (a) 7.3 m/s, (b) 9.0 m/s, (c) 13.3 m/s, (d) 15.9 m/s, (e) 19.8 m/s, (f) 33.4 m/s, (g) 50.0 m/s, (h) 58.8 m/s. The arrows on front views indicate the breakup length.
The concentric circular waves around impinging point begin to appear at the jet velocity of 15.9 m/s as shown in Fig. 11d. This value is lower than the jet velocity from which these waves appear when the pure Carbopol gel is used. In the lower part of the image some connecting ligaments between concentric ligaments can be seen. This suggests that even though the concentric circular waves grow to form ligaments, the two-dimensional characteristics of surface instability should still remain at the flow condition. As the jet velocity increases (Fig. 11e–g) distinct circular waves appear and breakup length decreases slightly. The atomization pattern in this regime is the same as pure Carbopol case of the “sheet with ligament separation.” When jet velocity goes beyond 58.8 m/s, the “fully developed” spray pattern appears. This value can be considered to be much the same as 62.3 m/s of the pure Carbopol case. At this condition the viscosity does not seem to play a significant role in the atomization process due to large inertia.

### 3.2.4. Breakup length

The breakup length is defined as the length from the impinging point to the edge of the intact sheet along the spray centerline. In Table 1, the breakup length and two dimensionless groups of Re and We at each jet velocity are summarized. Here Weber number was calculated based on the equilibrium surface tension even though the dynamic surface tension should be more relevant. This is because, as described above, the dynamic surface tension of the gel material cannot be measured and the equilibrium surface tensions have been used in the literatures. It can be theoretically approached by a linear stability model and a stationary anti-symmetric wave model as discussed in Introduction. At low Weber number under 2000 the breakup length increases with the increase in jet velocity as predicted by the stationary anti-symmetric wave-based model (Eq. (6)) for water. Two theoretical results by both linear stability-based model (Eq. (3)) and stationary anti-symmetric wave-based model (Eq. (6)) for water are displayed to gather. In the case of water, the theoretical results agree with the experimental results in trend but the theoretical breakup length is overestimated. In contrast, the breakup length of the gel materials cannot be identified due to the absence of the closed rim at the low jet velocity regime. So this explains why stationary anti-symmetric wave-based model was not applicable to the breakup length within low Weber number region. The breakup length was decreased with the increase in Weber number following the linear stability-based model. When compared with water, however, the breakup length of pure Carbopol gel increases probably due to the damping effect of high viscosity. On the other hand, the breakup length of the gel containing nanoparticle is considerably shorter than that of the pure Carbopol gel for Weber numbers considered here. It is considered that the impact energy generated by the impingement of high density particles accelerates the disintegration of the Carbopol gel. The viscosity increased by the addition of nanoparticles requires higher pressure for the atomization of the gel material, but the Carbopol gel with nanoparticles is more easily disrupted and consequently reaches fully developed pattern at the lower jet velocity.

### Table 1

<table>
<thead>
<tr>
<th>Jet velocity (m/s)</th>
<th>Re</th>
<th>We</th>
<th>Breakup length (mm)</th>
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<tr>
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<td></td>
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<tr>
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<td>7.8</td>
<td>5460</td>
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<td>12.3</td>
<td>8610</td>
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<td>16.7</td>
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<td>21.3</td>
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<td>10781</td>
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<tr>
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<td>565</td>
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<tr>
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### 3.3. Drop size and distribution

To measure the drop size we used an image processing method. Although this method is relatively time–costing, it is able to check the images directly and handle non-spherical or duplicated drops. We analyzed 2327 images to measure the drop size and its distribution at each jet velocity. We used the commercial software Image-Pro® for image processing and all images were treated with the same constraint. It was found that some of the droplets had non-spherical shapes and, in the case of gel materials, more droplets had non-spherical shapes due to high viscosity. In calculating the drop size by recognizing the drop perimeter and area from a 2-D image, the drop size can be underestimated and the degree of underestimation increases as non-sphericity increases. Therefore to obtain a better estimation, non-spherical particles were sorted out even though it could reduce the number of samples. To identify well focused spherical droplets, the gray range was fixed at 0–100, and the aspect ratio and the roundness (defined by 4π(area)/perimeter²) were set to 1.03 and 1.07, respectively, when using the software. The Sauter mean diameter (SMD, D_{32}) is defined as the diameter of a drop having the same [volume/surface area] ratio. We calculated the SMD for the average drop diameter by the following relation:
The Rosin–Rammler distribution is frequently used for the atomized drop size and its distribution [28]:

\[ 1 - Q = \exp \left( -\left( \frac{d}{X} \right)^q \right) \] (12)

where \( Q \) is the volume fraction of the drops the diameter of which are less than \( d \), \( X \) is the Rosin–Rammler diameter which is defined as the diameter in such a way that drops with sizes less than \( X \) occupy 63.2% of the total volume. \( q \) is a measure of the drop size distribution and is called the Rosin–Rammler exponent. We regard the Rosin–Rammler diameter as the SMD at each jet velocity condition. Therefore drop size distribution can be represented by \( X \) and \( q \). To determine \( X \) and \( q \), we first determined \( X \) so that the cumulative volume fraction was 63.2% and then \( q \) was determined by finding the value that fits the distribution curve in the following form:

\[ q = \frac{\ln(1 - (1 - Q))}{\ln(1/X)} \] (13)

In determining \( Q \) and \( d \) from the images the drop diameters for each set of images were calculated first and then the diameter values were classified into discrete intervals of 10 \( \mu \text{m} \) and then the volume fraction was calculated for each interval.

Fig. 13 shows the distribution of water drop size at various jet velocities. The solid lines are the best fitted Rosin–Rammler distributions. For each jet velocity, the drop size is well expressed by the Rosin–Rammler distribution. Table 2 summarizes the number of drops obtained from image processing, Rosin–Rammler diameter (\( X \)) and exponent (\( q \)) for each jet velocity of atomization. As the jet velocity increases, the Rosin–Rammler diameter decreases. On the other hand, the Rosin–Rammler exponent does not show a monotonic trend for the whole range of jet velocity but it clearly has a tendency to increase above 8.3 m/s, meaning that the drop size distribution becomes narrower over this velocity. In the fully developed pattern above the jet velocity of 30 m/s, the SMD of water atomization converges to approximately 100 \( \mu \text{m} \). The asymptotic convergence in drop diameter at high jet velocity result is consistent with many other experimental results. Lai et al. [5] also observed the converging drop diameter as jet velocity became large. The converging drop diameter was about 50 \( \mu \text{m} \) when the orifice diameter was 0.5 mm. Ryan et al. [15] reported that the non-dimensional mean drop size (SMD/\( d_0 \)) was convergent to 0.2 for atomization with 0.64 mm diameter of injector, i.e. drop size was approximately 128 \( \mu \text{m} \). In the present case, this ratio is 0.14 while in Lai’s case it is 0.1. Considering that there should be many factors affecting the mean diameter of droplets, our result appears to be much the same as reported in the literatures. Since the present investigation aims at comparing the atomization characteristics of water and Carbopol gels with and without nanoparticles, we will focus on the change of the drop SMD with the change in fluids properties under the same doublet like-on-like impinging jet atomization setup.

### Table 2

<table>
<thead>
<tr>
<th>Jet velocity (m/s)</th>
<th>Number of drops</th>
<th>Rosin–Rammler diameter, ( X ) (( \mu \text{m} ))</th>
<th>Rosin–Rammler exponent, ( q )</th>
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<tr>
<td>55.1</td>
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<td>75</td>
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</table>
In the case of Carbopol gels the drop size analysis was done only for large jet velocities of 10–60 m/s as shown in Table 2. This is primarily due to the higher incipient velocity for droplet formation for highly viscous gels. Also it was not easy to take photographs because the liquid sheet sustained a flat sheet without shedding drops until reaching a very high jet velocity compared with water and sometimes the sheet undulated by the flow of the surrounding air which appeared to be generated by the fast movement of the gel itself. Therefore the number of drops was smaller in the determination of the average drop size.

Fig. 14 shows the typical drop images at some jet velocities for Carbopol gel atomization. Fig. 14a and b are the representative images from the “rimless sheet with ray shaped shedding” region. Most drops are large (the diameters drops are in the range of several hundred or thousand microns) and often do not have the perfect spherical shape. At high jet velocities drops are in a near-spherical shape as shown in Fig. 14c–f.

Fig. 15 shows the Rosin–Rammiller distribution plot by varying jet velocity for the pure Carbopol gel. For each jet velocity, the relation between cumulative volume fraction and drop size is well fitted to the Rosin–Rammiller distribution. The Rosin–Rammiller exponent is relatively large for the pure Carbopol gel when compared with that of water, i.e. the drop size has a narrow distribution, but it is difficult to find any distinct correlation with jet velocity. The drop SMD decreases with the increase in jet velocity and the final convergent drop SMD of the Carbopol gel is 77 μm, less than that of water. The drop size should be determined by many factors. Compared to water the Carbopol gel has much higher viscosity while the surface tension remains much the same even when the dynamic surface tension remains much the same even when the dynamic surface tension is taken into account. In this case the drop diameter will become larger if Eq. (1) is applied, which is contrary to the present observation. The drop size appears to be affected much by the elasticity of the Carbopol gel. There have been many reports on the instability characteristics of viscoelastic materials. They reported that viscoelastic liquid jets are more unstable than Newtonian jets with the same viscosity and the anti-symmetric disturbances (sinuous mode) are more unstable than the symmetric disturbances (varicose mode). [29–32]

Especially the most unstable wave number for viscoelastic fluid is larger than that for Newtonian fluid. Brunn and Ryssel [32] have reported that the viscoelastic jet is more unstable than viscous jet. All of these reports imply that the drop size of viscoelastic fluids should be smaller than that of Newtonian fluid, considering that liquid sheet becomes ligaments which further become unstable to generate droplets by instability. Since the Carbopol gel has viscoelasticity, shear thinning viscosity and even a yield stress, it may not be possible to apply their theories directly to the present problem. However it is certain that the reduction of drop size should be affected by the viscoelastic characteristics of the gel.

The Rosin–Rammiller distribution plot at various jet velocities for the Carbopol gel containing SUS nanoparticles is shown in Fig. 16. The general trend is similar to the pure Carbopol gel case and the finally converging drop SMD in the fully developed region is around 75 μm, which is virtually the same as the drop size of the pure Carbopol gel. The change in drop size by added particles is a very important issue especially from the combustion point of view. Since the drop size is related directly to the combustion efficiency, smaller drops are more favored. Bartels et al. [33] reported that the drop size of gel containing 15 wt.% aluminum particles is approximately 10–15 μm larger than the pure gel drop when performing atomization using jet A-1/Thixatrol ST gel with 5–15 μm (~315 mesh) aluminum particles. Until now there has been no report on the drop size of gel material containing nanoparticles. However considering that viscoelastic gels are more unstable to larger wave number disturbances and gels with nanoparticles are very fragile, it is conceivable that the asymptotic drop size of gels with nanoparticles can be smaller than that of water. The detailed theoretical analysis will be a formidable task and beyond the scope of the present experimental study.

Fig. 17 shows variations of the dimensionless drop diameter (SMD/μm) with non-dimensional scaling parameter (We/f(θ)) for the present experimental results as well as the theoretical prediction of water based on the linear stability model (Eq. (2)). For all materials tested in this experiment, the drop SMD decreases with the modified Weber number, and it approaches an asymptotic value. The variation in drop SMD of Carbopol gel containing SUS...
nanoparticles is very similar to that of the pure Carbopol gel, but there is a slight difference in jet velocity at which the finally converging drop SMD is obtained. It is considered that the finally converging drop SMD is obtained in the lower jet velocity region due to the fragile nature of gels with nanoparticles.

Until now we have considered mostly the shear properties of gel materials even though elongational and biaxial extensional properties should affect the process somehow. However it is usually more difficult to measure elongational and biaxial viscosities of materials than shear properties. Regarding the present study, it was found that the capillary break-up method did not work due to the yield stress. From the general extensional properties of polymeric liquids we may argue that the extensional property may affect the process as follows: The larger biaxial extensional viscosity of polymer solution will retard the biaxial extension of sheet, and therefore the size of ligaments can be larger for polymer solutions. This may enlarge the drop size ultimately, but it has been found that the drop size is smaller for gels than water even though the surface tension is smaller. This is due to large inertial effect. This means that the extensional viscosity may not have strongly affected the ligament size. The larger elongational viscosity and extensional thickening property of polymer solution should not strongly affect the size of drop since the interfacial instability is caused by the shear flow and drop size should be determined primarily by the wave length of the instability. However the pinch off time will be strongly dependent on the elongational properties even though we did not measure such properties.

4. Summary and conclusion

We have investigated the spray and atomization behavior of C934 Carbopol gels with or without SUS304 nanoparticles using doublet like-on-like impinging jets and compared with the spray behavior of water. The spray patterns of gel materials were qualitatively different from those of water and could be divided into four types: open rim and no shedding drop; rimless sheet with ray shaped shedding; sheet with ligament separation; fully developed. The sheet sizes of water and the pure Carbopol gel increased with the increase in jet velocity, but the sheet size of the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio ($L_{\text{max}}/W_{\text{max}}$) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and consequently the spreading angle was smaller when compared with water. The breakup length of the pure Carbopol gel was longer than that of water reflecting that the breakup length is a strong function of viscosity. However the addition of nanoparticles to the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio ($L_{\text{max}}/W_{\text{max}}$) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and consequently the spreading angle was smaller when compared with water. The breakup length of the pure Carbopol gel was longer than that of water reflecting that the breakup length is a strong function of viscosity. However the addition of nanoparticles to the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio ($L_{\text{max}}/W_{\text{max}}$) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and consequently the spreading angle was smaller when compared with water. The breakup length of the pure Carbopol gel was longer than that of water reflecting that the breakup length is a strong function of viscosity. However the addition of nanoparticles to the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio ($L_{\text{max}}/W_{\text{max}}$) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and consequently the spreading angle was smaller when compared with water. The breakup length of the pure Carbopol gel was longer than that of water reflecting that the breakup length is a strong function of viscosity. However the addition of nanoparticles to the Carbopol gel containing SUS particles did not change with the increase in velocity. The aspect ratio ($L_{\text{max}}/W_{\text{max}}$) of the sheet was constant at 2.1 for water and 1.7 for two gel materials. Due to the high viscosity of gel materials, the amplification of sinusoidal waves was mitigated and consequently the spreading angle was smaller when compared with water.

We measured the drop SMD using an image processing method. The drop SMD was obtained from the Rosin–Rammler distribution at each jet velocity. The drop SMD decreased with the increase in jet velocity, and it converged to an asymptotic value. Under the same condition, the final convergent drop SMD of water was about 100 $\mu$m which was larger than that of 75–77 $\mu$m of Carbopol gels. The elasticity of gels appears to reduce the asymptotic drop size.

Until now there have been no theoretical reports on the atomization of various gel materials. Through the theoretical stability analysis for the Carbopol gel with nanoparticles while incorporating the rheological properties including shear thinning and yield stress properly, one may gain the insight into the physics of the
atomization and spray of gel materials. This will help to develop a better rocket propellant in a safer way.

Acknowledgment

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.jnnfm.2011.08.005.

References