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Direct observation and characterization of the generation of organic solvent droplets with and without triglyceride oil by electrospraying

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ABSTRACT

The primary aim of this study was to investigate the generation characteristics of organic solvent droplets with and without triglyceride oil by electrospraying using single-nozzle and nozzle-array devices. The liquids used were ethanol (10 to 99.5%), ethanol (99.5%) solutions containing a triacylglyceride oil, ethyl acetate, hexane, and heptane. The droplet generation behaviors from an electrode nozzle were observed using high-speed video cameras. Unstable micro-dripping mode was observed when using ethanol (<50%), hexane, and heptane. In contrast, the stable electrospraying by cone-jet mode was achieved when using ethanol (>66.7%) and ethyl acetate. The jet diameter values mostly ranged between 10 and 20 μm, which is similar to the estimated ones. High-speed photographic observation at a frame rate of 10^6 s^{-1} demonstrated the generation of ethanol droplets at very high frequencies of 3.0x10^5 to 5.0x10^5 s^{-1} by breaking up the jet. The ethanol jet diameter increased with increasing the flow rate (1 to 10 mL/h). The nozzle number per device did not affect the jet diameter when the flow rate per nozzle is the same. The stable cone-jet mode was also achieved when using ethanol solution containing 0.1% triacylglyceride oil. Triacylglyceride oil droplets with diameters of about 2 μm were collected after evaporating ethanol from the droplets generated by electrospraying.

Keywords: Electrospraying; Organic solvent; Triglyceride oil; Droplet generation; Direct observation; Nozzle array
1. **Introduction**

Electrospraying (electrohydrodynamic spraying) is a method of liquid atomization induced by electrical force [1]. With electrospraying, liquid surface tension is balanced by electrical force at each point on the liquid surface. When electrical force and gravity force that act on a drop overcome the surface tension, the meniscus of the liquid caused the jet formation. The cone-jet leads to fission and subsequently disrupts into fine droplets due to the instability of the jet. From the energy viewpoint, liquid forms a jet when the kinetic energy of the liquid is greater than the surface energy required for creating the surface of the jet [2]. Thus, variations in normal electric stress at the apex of the meniscus, the energy gain per unit area of the liquid, and the tangential electric stress at the meniscus lateral are evaluated in the forms of electrospraying modes. When the tangential electric stress is intense enough, a cone-jet is formed [3].

Electrospray systems have the following advantages over mechanical atomizers. The droplet size can be smaller than 1 μm. The size distribution of the droplets can be nearly monodisperse [4]. The charge and size of the droplet can be easily controlled (including deflection or focusing) to some extent by adjusting the flow rate and voltage applied to the nozzle [5].

Electrospraying can be widely applied to both industrial processes and scientific instrumentations. Interest in industrial or laboratory applications has recently prompted the search for effective techniques that enable control of the processes in which droplets are involved. Electrospraying techniques are aimed at developing new drug-delivery systems, medicine production, and ingredient dosage in the cosmetic and food industries. In particular, electrospraying has recently been adapted for many applications (e.g., medical powder production [6], fine metal powder production [7], film coating [8]).
Electrospraying is often performed in typical modes. Among these modes, many studies have focused on the cone-jet mode, in which the liquid meniscus appears to have a Taylor cone [11-15]. The size distribution of the droplets produced in cone-jet mode depends on the jet diameter and the type of jet instabilities (e.g., varicose instability and kink instability) [16]. The jet diameter is a strong function of the axial distance, which may be due to solvent evaporation. Therefore, it is important to make comparisons at a fixed point below which there is negligible change in jet diameter [17]. Xie et al. [18] also developed a hypothesis explaining the characterization of particles obtained by electrospraying under various operating conditions (e.g., polymer solution flow rate, polymer concentration, and type of chemical solvent). Electrospraying are also generally performed using polar liquids (e.g., water, ethanol, glycerol, and polyethylene glycols). Ethanol has been used for understanding fundamental properties of electrospraying and as a carrier fluid for producing micro/nanoparticles [19-21]. Martin et al. [19] reported how liquid flow rate and nozzle and flat plate voltages influence the cone-jet domain for electrospraying using pure ethanol.

Droplets generated by electrospraying have potential food applications as forms of micro/nano-particles and micro/nano-dispersions. However, few electrospraying researches using food-grade materials have been conducted [22,23]. Therefore, the present work attempts to employ the electrospraying technology to generate the fine droplet consisting of organic substances. Here, we investigate the production process focusing on the type of organic solvents, the effects of device and operating conditions, high-speed camera observation of droplet generation. We also investigated the generation of droplets consisting of an organic solvent and a triacylglyceride oil as well
as the formation of triacylglyceride oil droplets by solvent evaporation from the droplets.

2. Materials and methods

2.1. Experimental setup

2.1.1. Electrode nozzle devices

Stainless-steel 24×24-mm devices with different numbers (1, 4, or 12) of electrode nozzles were designed and fabricated for this study (Fig. 1). Initially, each circular through-hole with a diameter of 600 μm on a polished metal plate was fabricated by microdrilling. Subsequently, each electrode nozzle with an inner diameter of 230 μm and a length of 13 mm was connected to the through-hole by solder bonding.

2.1.2. Electrospraying module and peripherals

Fig. 2 is a simplified schematic diagram of the experimental setup for electrospraying using electrode nozzle devices. It consists of a holder equipped with the device, a metal ring with a diameter of 100 mm as the extractor electrode, a syringe pump (Model-11, Harvard Apparatus, USA) for feeding an organic liquid, a high voltage DC power supply (KHV-U5003, Kyoshin Electric Co., Ltd., Japan) for applying an electric field to the liquid, a high-speed video camera with a maximum frame rate of 2.5×10^5 s⁻¹ (Fastcam SA1.1, Photron Ltd., Japan) or 10^6 s⁻¹ (HyperVision HPV-1, Shimadzu Co., Japan), and a metal halide light source (LS-M210, Sumita Optical Glass, Inc., Japan).

2.2. Chemicals and solution preparation
The organic solvents used in this study are ethanol (purity 99.5%, Nacalai Tesque, Inc., Japan), n-hexane (Wako Pure Chemical Industries Ltd.), n-heptane (Wako Pure Chemical Industries Ltd.), and ethyl acetate (Wako Pure Chemical Industries Ltd.). Ethanol solutions with a concentration of 10 to 99.5% were prepared by mixing 99.5% ethanol with deionized and filtered water (Milli-Q water). Refined soybean oil (Wako Pure Chemical Industries Ltd.) as a model long-chain triglyceride, and medium-chain triglyceride (MCT) oil (MCT-7, Taiyo Kagaku Co. Ltd., Japan) were also dissolved in 99.5% ethanol at a concentration of 0.1% and a room temperature of 25 °C.

2.3. Experimental procedure

Prior to the electrospraying experiments, an electric nozzle device was fixed in the holder, and the tip of each electrode nozzle was positioned at 10 mm over the ring grounding electrode. Each liquid was contained a 10-mL glass syringe and was introduced into the module using a syringe pump at a liquid flow rate ($Q_L$) of 1 to 10 mL/h for the single nozzle device, 6 mL/h for the 4-nozzle device, and 18 mL/h for the 12-nozzle device. Next, the liquid was injected from the nozzle tip under positive DC voltage ($V_{DC}$) (3.7 to 14 kV) in order to generate organic solvent droplets. At that moment, the nozzle tip was positively electrified, while the ring was grounded. All experiments were conducted at 25 °C. Electrospraying characteristics were microscopically observed using a high-speed video camera at 9000 to 15000 fps for Fastcam SA1.1 or 10^6 fps for HyperVision HPV-1.

Image-analysis software (WinRoof version 5.6, Mitani Co., Japan) was used to measure the diameters of the droplets on the images captured during experiments. The mean diameter of the droplets generated under each condition was determined using the
measured data of 30 droplets.

2.4. Determination of viscosity, electrical conductivity, and surface tension

A Sine-wave vibroviscometer (SV-10, A&D Co., Ltd., Japan) was used to determine the absolute viscosities of the liquids (defined as the ratio of liquid viscosity to liquid density). The electrical conductivities of the liquids were measured using a conductivity electrode (Model 9382-10D, Horiba Ltd., Japan). The surface tensions of the liquids were determined using a fully automatic surface tensiometer (CBUP-Z, Kyowa Interface Science Co., Ltd, Japan) that adopts the Wilhelmy Plate method. The contact angles of the liquids to a stainless-steel plate were determined using a fully automatic interfacial tensiometer (PD-W, Kyowa Interface Science Co., Ltd, Japan). Measurements of liquid viscosity, electrical conductivity, surface tension, and contact angle were repeated at least three times at 25 °C; their mean values are presented in Table 1.

3. Results and discussion

3.1. Effect of organic solvent type

3.1.1. Electrospraying characteristics

Electrospraying experiments were first conducted using four different organic solvents. Fig. 3 depicts microscopic snapshots of electrospraying from a single nozzle. The $Q_L$ applied here was 1 mL/h for ethanol, hexane, and heptane, and 6 mL/h for ethyl acetate. Ethanol (99.5%) ejected from the nozzle tip formed a stable cone and jet (cone-jet mode) at a $V_{DC}$ of 3.7 kV (Fig. 3a). Due to electrostatic force, this jet broke up into droplets (see Sect. 3.2). The use of ethyl acetate also resulted in the formation of
the cone-jet mode at a $V_{DC}$ of 3.7 kV (Fig. 3b). It should be noted that ethyl acetate rapidly evaporated from the nozzle tip at a $Q_L$ of 1.5 mL/h, as the viscosity of ethyl acetate is about one-third of that of ethanol. The boiling point of ethyl acetate (77.2 °C) is similar to that of ethanol (78.4 °C). By contrast, heptane and hexane ejected from the nozzle irregularly formed large droplets with diameters of >100 μm nearby the cone tip (microdripping mode) at a $Q_L$ of 6.5 kV for hexane and 5.8 kV for heptane (Fig. 3c and d). The difference in these electrospraying characteristics could be explained using the properties of the organic solvents (Table 1). There was unpronounced difference in the surface tension of the organic solvents. The viscosity values of hexane and heptane were similar to that of ethyl acetate, while remarkable solvent evaporation was not observed for hexane and heptane ejected from the nozzle tip. These results indicate that their surface tension and viscosity are not dominant factors affecting the electrospraying modes in this case. The solvent conductivity was appreciably influenced by solvent type. The conductivities of ethanol and ethyl acetate were at least two orders of magnitude greater than those of hexane and heptane. Earlier studies reported that liquid conductivity plays a critical role in electrospraying characteristics, and that a stable cone-jet mode can be obtained using liquids with a conductivity of $10^{-7}$ to $10^{-2}$ S/m [12,16,21]. It is currently assumed that liquids can be electrosprayed in cone-jet mode at conductivities of $10^{-4}$ to $10^{-8}$ S/m [5]. The conductivities of ethanol and ethyl acetate are thus considered to be appropriate for stable electrospraying in cone-jet mode.

3.1.2. Estimating jet diameter

To achieve stable electrostatic atomization, the electrical relaxation time ($t_e = \beta \varepsilon_0 / K_L$) must be much smaller than the hydrodynamic time ($t_h = l_d j^2 / Q_L$) [24]:

\begin{equation}
\frac{t_e}{t_h} = \frac{\beta \varepsilon_0}{l_d j^2 Q_L}
\end{equation}
where $\beta$ is the relative permittivity, $\varepsilon_0$ is the permittivity of free space ($8.854 \times 10^{-12}$ F/m), $K_L$ is the electrical conductivity, and $l_j$ and $d_j$ are the jet length and diameter. Hence, the dimensionless viscous parameter ($\delta_m$) is defined by Ganan-Calvo et al. [25] as

$$\delta_m = \frac{\rho_L\varepsilon_0\gamma^2}{K_L\eta_L^3}.$$  

(2)

where $\rho_L$ is liquid density, $\gamma$ is surface tension, and $\eta_L$ is viscosity of liquid. For $\delta_m \gg 1$, the jet diameter ($d_j$) in stable cone-jet mode can be estimated by [25]:

$$d_j \approx \sqrt[3]{\frac{\rho_L\varepsilon_0\gamma^2}{\gamma K_L}},$$  

(3)

Substituting the physical values of ethyl acetate in Eq. 2 indicates that $\delta_m$ for the ethyl acetate is $\gg 1$ and therefore, $d_j$ is 15 $\mu$m at a $Q_L$ of 6.0 mL/h. Conversely, for $\delta_m \ll 1$, $d_j$ is given by [25]:

$$d_j \approx \sqrt[3]{\frac{\beta - 1}{2} Q_L \varepsilon_0},$$  

(4)

Substituting the values of 99.5% ethanol in Eq. 4 yields a $d_j$ of 8 $\mu$m at a $Q_L$ of 1.5 mL/h. Based on these results, the measured jet diameters of ethyl acetate and ethanol are similar to the estimated values.

3.1.3. High-speed camera analysis of droplet generation by cone-jet mode

A high-speed camera with a maximum frame rate of $10^6$ s$^{-1}$ was used to investigate
droplet generation of 99.5% ethanol using the single-nozzle device. During electrospraying, the liquid formed a conical meniscus at the nozzle tip, and the jet ejected from the nozzle broke up into uniformly sized droplets. High-speed photographic observation (Fig. 4) indicated that fine droplets were generated at remarkably high frequencies of $3.0 \times 10^5$ to $5.0 \times 10^5$ s$^{-1}$ under the $Q_L$ of 1.0 mL/h and the $V_{DC}$ of 3.7 kV. In stable electrospraying, the Rayleigh law is defined as [26]

$$\frac{d_d}{2d_j} = 1.89$$

(5)

where $d_d$ is the droplet diameter. The $d_d$ calculated using Eq. 5 was 14 µm, which is similar to the $d_d$ measured from the experiment.

3.2. Effect of ethanol concentration and flow rate

Ethanol concentration affects many physical properties of a liquid (e.g., viscosity, density, surface tension, and conductivity); therefore, it is expected to have a great influence on electrospraying characteristics. Single-phase ethanol with a concentration of 10 to 99.5 wt% was made to flow through a single electrode nozzle, 4 electrode nozzles, and 12 electrode nozzles (Fig. 1). The flow rates of all nozzles were the same; 1.5 mL/h for the single nozzle, 6 mL/h for the 4-nozzle array, 18 mL/h for the 12-nozzle array. The $V_{DC}$ values were 4.2 kV for the single nozzle, 9.0 to 11.0 kV for the 4-nozzles, and 12.5 to 14.0 kV for the 12-nozzles. Figure 5a shows the influence of ethanol concentration on the droplet/jet diameter obtained by electrospraying. The influence of ethanol concentration on the surface tension, conductivity, and viscosity of ethanol are also presented in Fig. 5b, c. The processed liquids exhibited microdripping at concentrations of 50% or less (Fig. 5a), which be mainly due to higher surface tension.
This may have prevented electrospraying in the stable cone-jet mode at the flow rates and voltages applied here. Electrostatic atomization in cone-jet mode is difficult with liquids containing a high proportion of water [14,27,28] unless specified conditions (e.g., flow rate, surrounding atmosphere, and conductivity) are met [28,29]. The use of ethanol solutions with a concentration of 67% or higher exhibited electrospraying in the cone-jet mode with a narrower jet diameter ranging between 10 and 20 µm (Fig. 5a), indicating that the diameter of ethanol droplets generated in this cone-jet mode decreased slowly with increased the ethanol concentration. The stable cone-jet mode was achieved at surface tensions of 22 to 27 mN/m, conductivities of 1.6x10$^{-7}$ to 8.3x10$^{-7}$ S/m, and viscosities of 0.8 to 1.8 mPa s. It should be noted that the viscosity values of ethanol for the microdripping mode were similar to those for the cone-jet mode (Fig. 5b). The results in Fig. 5a also demonstrated that the resultant jet (and droplet) diameter was hardly affected by the nozzle number at an ethanol concentration of 67% or higher.

Jayasinghe et al. [30] suggested that the flow rate has to be minimized to obtain the finest droplets. However, no stable cone can be formed at flow rates being too low. The values of $\beta$, $\varepsilon_0$, and $K_L$ in Eq. 4 do not change for ethanol, and the jet diameter of ethanol increases monotonically with increased flow rates. Among the various parameters, flow rate is the simplest and most effective variable for controlling jet diameter. Therefore, to analyze the influence of 99.5% ethanol flow rate, the applied voltage was kept constant throughout the investigation at 4.4 kV by using the single-nozzle device. Ethanol that ejected from the nozzle tip formed the cone-jet mode at all the flow rates (1 to 10 mL/h). The cone depth, defined as distance from the nozzle tip to the apex of the cone, increased gradually from 150 to 670 µm as the flow rate
increased (Fig. 6a). The ethanol flow rate also affected the jet diameter which ranged between 9.0 and 16.1 μm (Fig. 6b). The jet diameter increased slowly with increasing the flow rate in its range of 2 to 10 mL/h, whereas the slope of the jet diameter became higher at the low flow rates of < 2 mL/h.

3.3. Electrospraying characteristics of ethanol solution containing triacylglyceride oil

3.3.1. Effects of oil type and flow rate

Electrospraying an ethanol solution containing MCT (0.1%) or soybean oil (0.1%) formed a stable cone-jet using the single nozzle at a $Q_L$ of 1 mL/h and a $V_{DC}$ of 4.0 kV (Fig. 6). The effect of the flow rate of the ethanol solutions on the cone depth and jet diameter is also shown in Fig. 6. The values of the liquid properties are listed in Table 1. The shape and the wettability of the nozzle material can pronouncedly influence the shape of the cone formed, the stability of the jet, and the electrospraying mode [30,31]. The contact angle is an indicator of the wettability of a liquid to the nozzle surface. The wettability of the liquids became poor with increased contact angle on the stainless-steel plate. The static contact angle of 99.5 wt% ethanol to the stainless-steel plate was <10°, indicating that the nozzle surface is easily wetted by ethanol during electrospraying. The static contact angle of MCT on the stainless-steel plate was 20°, and that of soybean oil was 15°. The formed cone and jet of the ethanol solution containing soybean oil were somewhat larger (Fig. 6), which may be attributed to its higher wettability to the nozzle surface.

At the $Q_L$ of 1 mL/h, the jet diameter was 13 μm for MCT and 18 μm for soybean oil. The jet diameters of the ethanol solutions increased with increasing the $Q_L$ in ranges of >2 mL/h or more for MCT and >1 mL/h for soybean oil. The slope of the lines in Fig...
6b became greater between 1 and 2 mL/h for MCT and 0.5 and 1 mL/h for soybean oil.
The jet diameters estimated by Eq. 4 that increased with increasing $Q_L$ agreed with the
measured results.

3.3.2. Optical microscopy of triglyceride oil droplets

Under adequate electrospray condition, droplet relics were collected by swiftly
moving a glass slide below the ring ground electrode or over the ring ground electrode.
This collection procedure was repeated at least twice. Soon after deposition, the
collected droplet relics were analyzed using optical electron microscopy (Leica DMIRM,
Leica Microsystems Wetzlar GmbH, Germany). The jet was confirmed to break up into
droplets above the ground electrode (Fig. 7a and c). With increasing the distance
between the nozzle tip and the glass slide, ethanol evaporated more from droplets, and
the size of the collected droplets decreased. The average diameters of the resultant
triglyceride oil droplets were about 2 μm, which is reasonable considering the oil
concentration in ethanol solution.

4. Conclusions

The results have demonstrated that successful electrospraying in cone-jet mode can
be achieved when using organic solvents (especially ethanol) with liquid conductivities
of $>10^{-8}$ S/m and applying appropriate $V_{DC}$. The electrospraying behaviors were affected
by ethanol concentration as well. High-speed photographic observation demonstrated
that the jet formed by electrospraying broke down into fine 99.5% ethanol droplets at a
maximum generation frequencies of $5 \times 10^5$ s$^{-1}$. The jet diameter was affected by the type
and composition of organic solvents and increased with increasing $Q_L$. The jet diameter
obtained from experimentation was similar to the estimated jet diameter. The successful cone-jet mode was achieved using the 4-nozzle and 12-nozzle devices when the $Q_L$ per nozzle was the same. This finding is useful for improving low droplet productivity in electrospraying. Fine triglyceride oils with diameters of about 2 µm were also obtained by electrospraying of ethanol solution containing triglyceride oils and subsequent solvent evaporation from the generated droplets. This combined process could be applicable for forming a thin edible oil layer in foods and pharmaceuticals.

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Table 1 Surface tension, conductivity, and viscosity of organic solvent solutions with and without triacylglyceride oil used for electrospraying.

<table>
<thead>
<tr>
<th>Solution type</th>
<th>Surface tension (mN/m)</th>
<th>Conductivity (S/m)</th>
<th>Viscosity (mPa s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethanol</td>
<td>22.2</td>
<td>1.6\times10^{-5}</td>
<td>0.83</td>
</tr>
<tr>
<td>Ethanol (0.1% MCT)</td>
<td>22.0</td>
<td>1.7\times10^{-5}</td>
<td>0.78</td>
</tr>
<tr>
<td>Ethanol (0.1% soybean oil)</td>
<td>22.2</td>
<td>9.0\times10^{-6}</td>
<td>0.82</td>
</tr>
<tr>
<td>Ethyl acetate</td>
<td>23.4</td>
<td>&lt;1\times10^{-7}</td>
<td>0.45</td>
</tr>
<tr>
<td>Hexane</td>
<td>18.3</td>
<td>&lt;1\times10^{-13}</td>
<td>0.32</td>
</tr>
<tr>
<td>Heptane</td>
<td>19.7</td>
<td>&lt;1\times10^{-9}</td>
<td>0.41</td>
</tr>
</tbody>
</table>
Figure captions

Fig. 1 Schematic illustrations of stainless-steel devices with different numbers (1, 4, and 12) of electrode nozzle. (a) Single-nozzle device. (b) 4-nozzle device. (c) 12-nozzle device.

Fig. 2 Schematic representation of the setup for electrospaying.

Fig. 3 Typical electrospaying behaviors of different organic solvents: (a) ethanol, (b) ethyl acetate, (c) hexane, and (d) heptane. Each liquid was ejected using a single nozzle. Scale bars are 100 μm.

Fig. 4 Typical image of stable cone-jet mode and generation of 99.5% ethanol droplets. This image was taken at a frame rate of 10^6 s⁻¹.

Fig. 5 Variations in jet and droplet diameters (a), surface tension and viscosity (b), and conductivity (c) as a function of ethanol concentration.

Fig. 6 Variations in cone depth (a) and jet diameter (b) as a function of flow rate. The images in (c and d) are examples of electrospaying of ethanol solutions each containing a triglyceride oil. The cone depth and jet diameter are defined in (c).

Fig. 7 Optical micrographs of ethanol droplets containing 0.1% MCT (a and b) or 0.1% soybean oil (c and d) collected on a glass slide. The distance in (a and c) was 0.5 cm.
below the ring electrode, and that in (b and d) was 2 cm over the ring electrode. Scale bars are 10 μm.
Fig. 1

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Fig. 2

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Fig. 3

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Fig. 4

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Fig. 5

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Fig. 5 (Cont.)

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Fig. 6

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Fig. 7

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