Supporting Information for
Rotating Motion of an Oil Droplet in a Circular Channel Subjected to a Transverse Alternating Electric Field

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Experimental Setup

Figure S1 shows a schematic diagram of the experimental setup used to investigate the behavior of a silicone oil droplet in castor oil. The setup, which without some parts is almost the same as that used in a previous study, is described here. The main part of the experimental setup is a transparent Pyrex glass tube (1.0 mm ID, 1.6 mm OD, 210 mm long), which is referred to as the main tube. The main tube passes vertically through a hollow rectangular container fabricated with transparent poly(methyl methacrylate) (PMMA) plates and is set so that the longitudinal axis of the container coincides with the gravitational direction and with the longitudinal axis of the main tube. Another transparent glass tube (0.54 mm ID, 0.9 mm OD, 50 mm long), which is referred to as the contraction tube, is inserted from the upper end of the main tube to contract the cross-sectional area of the main tube to ensure that the diameter of droplets formed into the main tube from the nozzle noted below is less than 0.54 mm. The height of the upper end of the contraction tube is 5 mm higher than that of the main tube. A 30-gauge stainless steel tube (0.31 mm OD, 0.13 mm ID), of which the tip surface is cut to be normal to the axis of the tube, is inserted from the upper end of the contraction tube as a nozzle to form silicone oil droplets in the castor oil medium. The tip of the nozzle facing downward is 25 mm below the upper end of the contraction tube. The nozzle is connected to a syringe infusion device (YSP-301; YMC Co. Ltd., Kyoto, Japan) equipped with a syringe (1710TLL; Hamilton Co., Nevada, USA), and silicone oil (KF96-300cS; Shin-Etu Chemical Co. Ltd., Tokyo, Japan) is supplied to the nozzle from the syringe. The gap between the outer wall of the nozzle and the inner wall of the contraction tube is connected with another syringe infusion device (YSP-301; YMC Co.
Ltd., Kyoto, Japan) equipped with a syringe (1002TLL; Hamilton Co., Nevada, USA), and castor oil (extra pure grade; Kanto Chemical Co., Tokyo, Japan) is supplied to the main tube from this syringe and flows down into the main tube. The tip of the nozzle is shielded with a grounded stainless steel tube (2.40 mm ID, 3.40 mm OD, 50 mm long), which is set around the main tube, to prevent electrostatic forces acting on the silicone-oil/castor-oil interface of a droplet growing on the tip of the nozzle. The volumetric flow rates of castor oil and silicone oil are 5 µL/min and 0.01 µL/min, respectively (the superficial velocity of castor oil in the main tube was ca. 0.11 mm/s). Note that the interval between the formation of silicone oil droplets at the nozzle is not uniform, i.e., droplets were formed irregularly. Droplets thus formed fall into the main tube with castor oil flowing down in the main tube and are then drained off with the castor oil after they pass through the main tube. Silicone oil and castor oil once passed through the main tube are not used again. Table S1 lists the physical properties of the castor oil and silicone oil used in this work.

A pair of parallel electrodes made of metallized glass plates (26.8 mm wide, 2 mm thick, 94 mm long) are contained in the hollow rectangular container, the longitudinal axes of which are parallel with the main tube, and are placed ca. 20 mm below the tip of the nozzle so that the main tube passes through the spacing between the pair of parallel plate electrodes; the spacing between the electrodes is filled with the same liquid that passes through the main tube. The distance between the electrodes was set at 2.1 mm. One of the electrodes is grounded, while the other electrode is connected with a high-voltage bipolar amplifier (HOPP-3B1; Matsusada Precision Inc., Shiga, Japan) and a waveform generator (WF1973; NF Co., Yokohama, Japan) to supply high voltage to the electrode. The electrical potential difference between the electrodes is
measured at the monitor terminal of the amplifier with an oscilloscope (TDS2001C; Tektronix Inc., Oregon, USA).

Observations were performed at the mid-height of the spacing using a high-speed video camera (HAS-L1; Ditect Co., Tokyo, Japan) equipped with a 55 mm macro zoom lens assembly (MTE-55; Moritex Co., Saitama, Japan), a 2× magnification lens assembly (MTE2, Moritex Co., Saitama, Japan), and a 25 mm extension tube. The optical axis was set to be normal to the electric field direction and the tube axis. An LED light assembly (VLP-9000X; LPL Co. Ltd., Saitama, Japan) was set at the opposite side of the main tube to configure a backlighting condition to obtain images. Images thus obtained were later analyzed to compute the aspect ratio and orientation of the droplets, as defined in the main text. Observations were conducted at room temperature.

Figure S1. Schematic diagram of the experimental apparatus.
Physical Properties of Liquids Used

|                    | Density (kg/m³) | Kinematic viscosity (mm²/s) | Relative permittivity (at 50 Hz) | Resistivity (Ω·m) | Interfacial tension against castor oil (mN/m) |
|--------------------|----------------|-----------------------------|---------------------------------|-------------------|-----------------------------------------------|
| Castor oil         | 956ᵇ           | 732ᶜ                        | 4.56                            | 4.9×10¹⁰          |                                               |
|                    |                |                              |                                 |                   |                                               |
|                      |                |                              |                                 |                   |                                               |
| Silicone oil (KF96-300cS) | 970ᵉ           | 300ᵉ                        | 2.75ᵉ                           | 4.5×10¹⁴ᵈ         | 6.09ᶠ                                         |

ᵃ Measured properties are those for non-used liquids with lot numbers that are different from those used in the experiments.  
b Measured with a pycnometer.  
c Measured with an Ubbelohde viscometer.  
d Measured according to Japanese Industrial Standard C2101:1999⁷ except for the temperature of the liquids.  
e Manufacturer data.⁸  
f Measured by the pendant drop method.
Effect of Inner Channel Wall on Behavior of Droplet Passing Through the Channel

It is possible that the inner wall of a channel (a glass tube with an inner diameter of 1.0 mm) can affect the behavior of a droplet because the silicone droplet passes through the channel containing castor oil, and the equivalent spherical diameter of the droplet is approximately one-third of the inner diameter of the channel. For example, when a droplet elongates along the radial direction of the main tube, castor oil present between the tip of the elongating droplet, i.e., the area near the pole of an ellipsoidal droplet, and the inner wall surface facing the tip of the elongating droplet must be drained off. The drag force against this drainage may prevent elongation of the droplet. In addition, shear flow affects the electrical deformation of a droplet when a hydraulic shear field and electric field are both imposed on a droplet.4

In Table S2, the mean translational velocity of a droplet along the axis of the main tube during a single period $t_c$ is summarized for different $t_c$ values. The velocity has a value of 0.20–0.23 mm/s, which is independent of either $E_a$ or $t_c$. This indicates that flow induced by the electric field around a droplet does not affect the mean translational velocity along the axis of the main tube. The velocity profile for castor oil flow in the main tube is presumed to follow Hagen-Poiseuille flow, for which the superficial velocity is ca. 0.11 mm/s; therefore, the maximum velocity (along the central axis of the main tube) is ca. 0.22 mm/s, i.e., the mean translational velocity of a droplet along the tube axis is almost the same as the maximum velocity for Hagen-Poiseuille flow.
Figure S2(i) shows $L^*$ vs $t$ under an intermittent alternating electric field at $E_a = 1.4$ MV/m and $t_c = 4$ s. $L^*$ indicates the position of the center of mass of a droplet measured from the origin on the inner wall surface of the tube, which is in the nearest position to the surface of the electrode connected to the high-voltage bipolar amplifier, i.e., the left inner wall surface shown in Figure S3. $\theta$ vs $t$ and $a/b$ vs $t$ are shown in Figures S2(ii) and S2(iii), respectively. The maximum $a/b$ is ca. 1.17 ($|\theta| \approx 72.3^\circ$) when the droplet approaches $L^* \approx 0.33$ mm (Label a), while the maximum $a/b$ is ca. 1.18 ($|\theta| \approx 64.8^\circ$) when the droplet approaches $L^* \approx 0.48$ mm (Label b). At $E_a = 1.4$ MV/m and $t_c = 2.86$ s, the maximum $a/b$ is ca. 1.18 ($|\theta| \approx 64.1^\circ$) when the center of mass of the droplet approaches $L^* \approx 0.29$ mm, while the maximum $a/b$ is ca. 1.21 ($|\theta| \approx 66.1^\circ$) when the center of mass of the droplet approaches $L^* \approx 0.43$ mm (results not shown). Although the effect of the inner wall on the behavior of a droplet cannot be completely denied, it is considered to be negligibly small in this study when compared with the uncertainty of the measurements. On the other hand, the effect of the wall will not prevent a droplet from returning to a spheroidal shape between the first and second deformations. That is, with the effect of the wall, the reason why a droplet does not return to a spheroidal shape between the first and second deformations in Figures 2(i) and 6(i) cannot be interpreted.

Recently, Mandal and Chakraborty\textsuperscript{4} studied the deformation of a droplet in a shear flow with a linear velocity profile and under a dc field. The direction of the dc field was normal to that of the flow. The center of mass of the droplet shown in Figure 9(i) is located at $L^* = 0.429$ mm, i.e., the smallest $L$ for the droplet ($D_0 = 0.33$ mm)/surrounding castor oil interface $L_1$, is considered to be 0.264 mm, while the largest $L$, i.e., $L_2$, is 0.594 mm (see Figure S3). $L$ indicates the position measured from the inner
wall of the channel; the origin of $L$ is the same as that of $L^*$. The velocity profile for the
castor oil flow in the main tube is supposed to follow Hagen-Poiseuille flow; therefore,
the velocity of castor oil flow at $L = 0.264$ mm is estimated to be $v_1 \approx 0.171$ mm/s,
while that at $L = 0.594$ mm is estimated to be $v_2 \approx 0.212$ mm/s. The velocity profile for
Hagen-Poiseuille flow is not linear; however, in this study, it is assumed that the droplet
exists in a shear flow with a constant velocity gradient $G$, defined as $(v_2 - v_1)/(L_2 - L_1)$
(see Figure S3). It is also assumed that a dc field with $E_a = 0.44$ MV/m is applied to the
droplet, although in the experiment shown in Figure 9(i), an ac field with $E_a = 0.44$
MV/m is applied to the droplet. $G \approx 0.125$ s$^{-1}$, $E_a$, the physical properties of castor and
silicone oils, and the experimentally measured equivalent spherical diameter of a droplet
are used with the theory presented by Mandal and Chakraborty, by which it is
estimated that the inclination angle of the symmetric axis for an oblate deformed
droplet in a shear flow from that of a deformed droplet under no flow condition (i.e.,
condition under which deformation of a droplet is induced only by a dc field) is ca. $2.3^\circ$.
This inclination angle estimated here is small; therefore, the effect of shear stress on the
behavior of a droplet at $E_a = 0.44$ MV/m is considered to be negligibly small. At $E_a =
0.44$ MV/m, the Mason number, which shows the electric stress/hydrodynamic stress
ratio and is defined as $\varepsilon E_a^2/(\mu G)$ in ref 4, is ca. 89 (where $\mu$ is the dynamic viscosity
of the surrounding liquid). For $E_a = 1.4$ and 1.5 MV/m, the theory presented by Mandal
and Chakraborty cannot be applied, because $E_a$ is out of range for application of the
theory. Therefore, a dimensionless parameter, i.e., the Mason number, is used for $E_a =
1.4$ and 1.5 MV/m. The minimum Mason number at $E_a = 1.4$ MV/m is ca. 378 (with the
assumption of $G \approx 0.30$ s$^{-1}$, which is estimated from the experimentally measured $L^*$),
while that at $E_a = 1.5$ MV/m is ca. 254 (with the assumption of $G \approx 0.51$ s$^{-1}$, which is
estimated from the experimentally measured $L^\ast$). These results for the Mason number indicate that the electrical stress is much stronger than the hydrodynamic stress at $E_a = 1.4$ MV/m and 1.5 MV/m. Therefore, the effect of shear stress induced by the flow of castor oil at $E_a = 1.4$ and 1.5 MV/m can be ignored in this study because the effect of shear stress when the Mason number is ca. 89, i.e., when $E_a = 0.44$ MV/m, is considered to be negligibly small (previously estimated inclination angle was ca. 2.3°).

Table S2. Mean translational velocity for a droplet along the axis of the main tube

| No electric field | $E_a = 1.4$ MV/m (intermittent electric field)$^a$ |
|-------------------|-----------------------------------------------|
|                   | $t_c = 4$ s | $t_c = 2$ s | $t_c = 0.4$ s |
| 0.23±0.01 mm/s$^b$ | 0.21±0.02 mm/s$^c$ | 0.20±0.03 mm/s$^d$ | 0.21±0.01 mm/s$^e$ |

$^a$ Mean translational velocity in cycle of applied intermittent alternating electric field, i.e., during $t_c$.

$^b$ Mean value ± 2 standard deviations computed with 7 different droplets.

$^c$ Mean value ± 2 standard deviations computed with 6 different droplets.

$^d$ Mean value ± 2 standard deviations computed with 5 different droplets.

$^e$ Mean value ± 2 standard deviations computed with 3 different droplets.
Figure S2. Transient behavior of silicone oil droplet in castor oil medium under intermittent electric field at $E_a = 1.4$ MV/m and $t_c = 4$ s; (i) $L^\ast$ vs $t$, (ii) $\theta$ vs $t$, and (iii) $a/b$ vs $t$. The time origin was arbitrarily chosen as 10 s or more after application of the electric field.
Figure S3. Geometrical arrangement of $L$ and $L^*$. 

$v_1 = 0.171$ mm/s  
$v_2 = 0.212$ mm/s  

Velocity profile  

Glass tube  

Plate electrode  

High-voltage bipolar amplifier  

Droplet  

Center of mass  

L = $L_1 = 0.264$ mm  

$L^* = 0.429$ mm  

$L = L_2 = 0.594$ mm  

Inner wall of the main tube  

Castor oil flow  

GND
Time Evolution of Silicone Oil Droplet Aspect Ratio $a/b$ and Orientation $\theta$ under Sinusoidal Alternating Electric Field at $E_a = 1.5$ MV/m and $0.05 \text{s} \leq t_c \leq 0.2 \text{s}$

Figure S4. Transient behavior of silicone oil droplet in castor oil medium under ac field at $E_a = 1.5$ MV/m; $t_c =$ (i) 0.2, (ii) 0.13, and (iii) 0.05 s. Note that the variations in $\theta$ and $a/b$ with time are drawn for two cycles of an ac field. The time origin was arbitrarily chosen as 10 s or more after application of the electric field.
Time Evolution of Silicone Oil Droplet Aspect Ratio $a/b$ and Orientation $\theta$ under a Sinusoidal Alternating Electric Field at $E_a = 0.44$ MV/m and $0.05 \text{ s} \leq t_c \leq 0.2 \text{ s}$

Figure S5. Transient behavior of silicone oil droplet in castor oil medium under ac field at $E_a = 0.44$ MV/m; $t_c =$ (i) 0.2, (ii) 0.13, and (iii) 0.05 s. Note that the variations in $\theta$ and $a/b$ with time are drawn for two cycles of an ac field. The time origin was arbitrarily chosen as 10 s or more after application of the electric field.
Prolate Deformation of Castor Oil Droplet in Silicone Oil Medium under Sinusoidal Alternating Electric Field

For comparison of oblate–spherical deformation under an ac field, deformation of a castor oil droplet in a silicone oil medium under an ac field is studied. In a weak dc field, a silicone oil droplet in castor oil undergoes oblate deformation, while a castor oil droplet in silicone oil undergoes prolate deformation.

The experimental setup used here is similar to that described in the Experimental Setup section. The differences between the experimental setup for investigation of the behavior of a silicone oil droplet in castor oil and that for a castor oil droplet in silicone oil are the main tube material and the direction of the surrounding liquid flow in the main tube. The major differences in the setup for observations of a castor oil droplet and for a silicone oil droplet are noted below. When the behavior of a castor oil droplet in silicone oil is investigated, the direction of the experimental setup is installed upside down; the nozzle and the contraction tube are inserted from the lower end of the main tube, i.e., the tip of the nozzle faces upward. A polytetrafluoroethylene (PTFE) tube (0.96 mm ID, 1.56 mm OD, $\varepsilon = 2.1$ at 60 Hz for PTFE) was used as the main tube, and a PTFE tube (0.46 mm ID, 0.92 mm OD) was used as the contraction tube. The same castor oil as that noted in the main text is supplied to the nozzle to form droplets, while the same silicone oil as that noted in the main text is supplied to the main tube as the surrounding liquid. The surrounding liquid, i.e., silicone oil, flows upward in the main tube and castor oil droplets rise in the main tube. The spacing between the parallel plate electrodes was filled with the same silicone oil as that passing through the main tube. The volumetric flow rates of silicone oil and castor oil are 5 $\mu$L/min and 0.02 $\mu$L/min,
respectively. The apparatus used to supply the silicone oil and castor oil, to capture images of droplet behavior, and for application of the ac electric fields to droplets are the same.

Figure S6 shows \((a/b)_{\text{max}}\) and \((a/b)_{\text{min}}\) vs \(t_c\) at \(E_a = 0.44\) MV/m. A silicone oil droplet in castor oil deforms to/from a prolate ellipsoid from/to a quasi-spherical shape. \((a/b)_{\text{max}}\) indicates the maximum magnitude of \(a/b\), while \((a/b)_{\text{min}}\) indicates the minimum magnitude of \(a/b\), where \(a\) is the length of the axis parallel to the electric field direction, and \(b\) is the length of the axis normal to the electric field direction. Figure S6 shows mean values and two standard deviations computed from 6 to 12 different droplets. \((a/b)_{\text{max}}\) and \((a/b)_{\text{min}}\) predicted according to the theory presented by Torza et al.\(^1\) are also plotted. At \(t_c \geq 4\) s, a droplet shows two alternative values of \((a/b)_{\text{max}}\) in the cycle of an electric field: one is large (shown as open triangles in Figure S6) and the other is small (shown as open inverted triangles in Figure S6). It is considered that the two peaks do not represent the nature of deformation and are instead caused by the experimental setup. In experiments outlined here, the main tube does not pass through the center portion of the space between the pair of parallel plate electrodes. The distance between the surface of one electrode that was connected to a high-voltage bipolar amplifier and the neighboring outer surface of the main tube was ca. 0.39 mm, while the distance between the surface of the other grounded electrode and the neighboring outer surface of the main tube was ca. 0.17 mm. Forster\(^6\) showed the electrical potential profile in a spacing between a pair of electrodes that was filled with benzene; the local gradient of the potential against the position, which is measured from a positive electrode and is along the direction normal to the electrode surface, is not constant between the electrodes. The gradient of the potential in the portion near the positive electrode is much larger than
that in the other portion. The main tube is placed near the grounded electrode rather than the electrode connected to the bipolar amplifier; therefore, when the grounded electrode acts as a positive electrode during the application of an ac field, i.e., negative potential is supplied to the electrode connected to the bipolar amplifier, the electric field strength applied to a droplet becomes larger than that when the grounded electrode acts as a negative electrode, i.e., a positive potential is supplied to the electrode connected to the bipolar amplifier. Thus, a droplet that is alternatively subjected to two different electric field strengths in a cycle of an ac field would alternatively show two different values of \((a/b)_{\text{max}}\) in a cycle. Even though the experimental setup has such a defect, it was determined that prolate deformation of a castor oil droplet in silicone oil at \(E_a = 0.44\) MV/m does not fully develop at \(t_c \leq 10\) s, as shown in Figure S6. In addition, the predicted \((a/b)_{\text{max}}\) underestimates the experimentally obtained \((a/b)_{\text{max}}\), and the underestimation is the same as that shown in Figure S7.
Figure S6. Transient behavior of a castor oil droplet in a silicone oil medium under an alternating sinusoidal electric field at $E_a = 0.44$ MV/m and $D_0 = 0.33$ mm. A castor oil droplet in silicone oil undergoes prolate deformation in a weak dc electric field. Open circles, open triangles, and open inverted triangles indicate $(a/b)_{\text{max}}$, while closed circles indicate $(a/b)_{\text{min}}$. Error bars for each data point indicate two standard deviations. If the range of the error bar is smaller than the size of the plots, then the error bar is not shown. At $t_c \geq 4$ s, droplets show two magnitudes of $(a/b)_{\text{max}}$, indicated by open triangles and open inverted triangles, in a cycle of an electric field due to the arrangement of the main tube and the pair of parallel plate electrodes. The solid line indicates $a/b$ under no electric field and the shaded band shows the range of two standard deviations. The dashed-dotted line indicates $(a/b)_{\text{max}}$ predicted according to the theory presented by Torza et al.\(^1\) and the dashed line shows $(a/b)_{\text{min}}$ predicted with the theory presented by Torza et al.\(^1\)
Comparison of Experimentally Obtained Time-Dependent Variation of Oblate Deformation of Silicone Oil Droplet in Castor Oil under AC Field and Prediction by Torza et al.¹

Figure S7. Comparison between experimentally obtained transient behavior of a droplet under an alternating sinusoidal electric field and that predicted using the theory presented by Torza et al.¹ $E_a = 0.44$ MV/m, $t_c = 10$ s, and $D_0 = 0.33$ mm. Open circles indicate experimentally obtained $a/b$ and the dashed line shows $a/b$ predicted according to the theory presented by Torza et al.¹ Experimentally obtained data shown are the same as those shown in Figure 9(i). According to a theory by Torza et al.,¹ there is a phase difference between the variation of droplet deformation with time, i.e., $a/b(t)$, and the variation of the electric field with time, i.e., $E(t)$. The phase difference is not known in this study; therefore, it is assumed that the predicted $a/b$ peaks coincide with the experimentally obtained $a/b$ peaks. The time origin was arbitrarily chosen as 10 s or more after application of the electric field.
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