1. INTRODUCTION

Recently, an increasing attention has been paid to methods using electric field to control the physical properties and meso-scale structures of soft materials such as polymeric liquids (polymer blends, block copolymers), colloid dispersion systems, liquid crystals, surfactant solutions and vesicles. Among others, the changes of rheological properties, what is called, electro-rheological (ER) effect, have been intensively investigated for binary liquid blends and particle dispersion systems, and such electro-active fluids have been attempted to use as clutches of vehicle, the active dumper and brake, and so forth. It has been reported that, in an immiscible liquid blend, droplets dispersed in matrix are aligned along the direction of electric field and bridge between two electrodes, which is the origin of ER effect. Namely, the change of rheological properties in such fluids comes from a change of a meso-scale structure by electric field. On the other hand, there is another research trend to utilize or to control the structures themselves by applying an electric field in order to obtain a new functional material. Actually, the bridges observed in dielectric liquid blends can be applied to thermoplastic blend systems to develop new materials whose mesoscopic structures are controlled by the electric field. For instance, after obtaining solid columnar structures, such materials can be applied to create a porous medium and fibers with a controlled length by dissolving a matrix part and columnar part, respectively. We can also create elastically anisotropic materials by using a tough material for the columnar part. It has also been reported that the orientation of a mesophase such a lamellar or cylinder structure of block copolymer melt can be controlled by applying electric field, for instance, an orientation of cylinder structure can be aligned along the direction of applied electric field. For other applications, there are fabrications of nano-scale fiber using electric field, what we call, electro-spinning of polymer solutions and melts. Making of fibers and cylindrical objects has also been tried using patterned electrodes. Steiner group has reported that a pattern prepared on the one side of sheet-like electrode can be transferred to a polymeric material by applying electric field as shown in Fig. 1. In their system, one of two sheet-like electrodes has a periodically aligned uneven surface pattern, the other electrode is a flat sheet. A sample is between two sheet-like electrodes, and put on the flat sheet.

In the present paper we are addressing to develop a simpler method to control the surface pattern of liquid material without uneven surface patterns of electrodes. We used a...
periodically aligned electrode array as shown in Fig. 2(a), and expected that the liquid film will be transformed to that with periodically undulated surface depending on the pattern of electrode array by imposing a voltage between the electrodes as shown in Fig. 2(b). Actually, in the similar fashion to the mechanism on deformation of droplets dispersed in a host liquid under an electric field\(^2\)-\(^4\), the surface profile of a dielectric liquid film under an electric field will be determined by competition between the Maxwell stress acting on a dielectric liquid film surface and a surface tension between the liquid and air. Using the system shown in Fig. 2, we investigated changes of surface profile of a liquid film on an electrode array. The aim of this paper is to investigate (i) whether a change of surface profile is induced by an electric field generated above a periodically aligned electrode array, and (ii) how the change of surface profile depends on applied voltage, (in other words, strength of electric field), size of electrodes, initial height of the liquid film, viscosity of liquid, and so on.

2. EXPERIMENTAL

The samples we used here are silicone oil, Shin-Etsu Silicones, Silicone Fluid KF-96H-X series (X=5,000 cSt, 100,000 cSt, 500,000 cSt, and 1,000,000 cSt) with density \(\rho = 0.977 \sim 0.978 \times 10^3 \text{kg/m}^3\) at 25°C and dielectric constant \(\varepsilon = 2.76 \varepsilon_0\), \(\varepsilon_0\) being the permittivity of vacuum. The surface tensions \(\gamma\) of the samples are between 20-21 mN/m. An array pattern of electrodes is printed on a photosensitive print circuit board using a mask and then the pattern electrodes are obtained by etching a copper foil with 30 \(\mu\)m thickness on a glass epoxy resin board shown in Fig. 3(b), where the positive and negative electrodes are aligned alternatively. Films of the silicone oil are prepared by spreading a droplet of it with a suitable size using a coater. The thickness of the silicone oil film is controlled by number of stacked tape that works as spacer (see Fig. 4(a) and (b)). The observation system we used here to investigate changes of the surface shape of the silicone oil is shown in Fig. 4(c). The changes of surface shape of the silicone oil were observed by a microscope through a reflected image in a 45 degree inclined mirror made of Aluminum using physical vapor deposition rack method. To apply a high voltage between the electrodes, we used the high voltage amplifier, TOS 8700 WITH STANDING VOLTAGE TESTER (Kikusui-denshi Co. Ltd.).

3. RESULTS

We investigate firstly the effect of strength of electric field at the air-liquid interface on shape of dielectric fluid surface.
As shown in Fig. 5(a), we used a patterned circuit board on which electrodes with width \( L = 800 \) \( \mu \text{m} \) and distance to nearest neighbor electrodes \( d = 400 \) \( \mu \text{m} \) are set. On the circuit board, positive and negative electrodes are aligned alternatively along \( x \)-direction and have a translational symmetry along the \( z \)-direction as shown in Fig. 3. In the experiments to examine the effect of strength of electric field on the surface shape change, we used a silicone oil (KF-96H-100,000 cSt) with a kinetic viscosity \( \nu = 1.0 \times 10^{-4} \text{ m}^2/\text{s} \) and set the initial liquid height on the substrate to \( h_0 = 180 \) \( \mu \text{m} \). After applying a DC voltage \( \phi_a \) to the silicon oil between the adjacent electrodes, the shape of air-liquid interface is changed, and undulated surface is finally formed according to the electrode pattern as shown in Fig. 5(b). For various applied voltage, we observed a height difference \( \Delta h \) between the adjacent highest and lowest height of the surface undulated film (see Fig. 5(b)). Although we would like to relate the average of height difference \( \Delta h \) with the strength of electric field, as seen from the geometry of the system in Fig. 5, the electric field generated by the electrode array is not uniform. So, we refer to the position of initial interface at the middle point between two electrodes as “interface electric field”. Since we could not measure the field at two electrodes as \( \Delta h \). For various applied voltages by solving the equation for the electrostatic potential \( \phi(r) \):

\[
-\nabla \cdot (\varepsilon(r) \nabla \phi) = 0.
\]  

with the periodic boundary condition along the \( x \)-direction and \( \partial \phi / \partial y = 0 \) on \( CD \) as shown in Fig. 6. On the \( AB \) boundary, we gave a value of \( \phi \), i.e., \( \phi = -\phi_a/2 \) on \( G_1G_2 \) and \( \phi = \phi_a/2 \) on \( G_3G_4 \), and \( \phi \)'s on the lines \( G_1G_3 \) and \( G_2G_4 \) are given by the value which is estimated by the linear interpolation \( \phi(G_1) = \phi(G_3) = -\phi_a/2 \) and \( \phi(G_2) = \phi(G_4) = \phi_a/2 \). The permittivity \( \varepsilon(r) \) is assumed to be given by

\[
\varepsilon(r) = \frac{1}{2} \left( \varepsilon_l + \varepsilon_a \right) + \frac{1}{2} \left( \varepsilon_l - \varepsilon_a \right) \psi_a(r)
\]

where \( \varepsilon_l \) and \( \varepsilon_a \) are the permittivity of the silicone oil and that of air, respectively, and \( \psi_a(r) \) is a phase field which represents the liquid as \( \psi_a = 1 \) and the air as \( \psi_a = 0 \). When we estimate \( E_o \), \( \psi_a(r) = \tanh((h_c-y)/\xi) \) with a constant \( \xi \) is used. Using \( E_o \), estimated by solving numerically eq.(1), we plot in Fig. 7 the averaged height difference \( \Delta h \) as a function of the interface electric field \( E_o \). As seen from the Fig. 7, the filled circles representing the average of height difference \( \Delta h \) are

![Fig. 5. Schematic cross section of the system (a) before and (b) after applying a DC voltage.](image-url)

![Fig. 6. Schematic figure of the system and boundary conditions used in numerical analysis for the electric field.](image-url)

![Fig. 7. Height difference \( \Delta h \) of undulated surface as a function of the interface electric field \( E_o \).](image-url)
approximately proportional to square of the electric field, i.e., \( \Delta h \propto E_o^2 \), although the result has a large error bar for each data point. It is considered that the height difference is determined through the competition between the constant surface tension \( \gamma \) and the Maxwell stress \( \sigma_M \) (strength of \( \sigma_M \propto E_o^2 \)).

Next, we explored the effect of film thickness of silicone oil on the deformation of surface profile induced by an electric field. Owing to the geometry of the system, the electric field highly depends on the distance from the electrodes, i.e., the electric field becomes weaker as distance from the electrodes is larger. When we change the thickness of the liquid, the electric field at the surface also varies. Hence, depending on the initial film thickness \( h_o \) we used a numerically estimated voltage \( \phi_o \) which gives the almost same strength of electric field \( E_o \) at \( P \) for different \( h_o \)'s, actually, we adjusted the applied voltages so as to be \( E_o \cong 1.6 \text{ kV/mm DC} \) for various liquid thicknesses \( h_o \). Using \( L = 800 \text{ mm} \) and \( d = 400 \text{ mm} \), we found \( \phi_o = 800 \text{ V}, 1000 \text{ V}, 1200 \text{ V} \) and \( 1400 \text{ V} \) for (a) \( h_o = 100 \text{ \mu m} \), (b) \( h_o = 180 \text{ \mu m} \), (c) \( h_o = 240 \text{ \mu m} \) and (d) \( h_o = 280 \text{ \mu m} \), respectively, in order to realize \( E_o \cong 1.6 \text{ kV/mm DC} \). We show the microscope images of equilibrium surface shape of silicone oil with a kinetic viscosity \( \nu = 1.0\times10^{-1} \text{ m}^2/\text{s} \) for \( h_o = 100, 180, 240, \) and \( 280 \text{ \mu m} \) in Fig. 8 where the white thin solid lines stand for electrodes, and the white dotted lines denote the bare substrate. In Fig. 8, in order to easily see the shape deformation of liquid surface under electric fields, the scale of \( y \)-direction is magnified twice as large as the original one. It is found from Fig. 9 and visually from Fig. 8 that the average height difference \( \Delta h \) and the shapes of the liquid surface for the four different thickness cases are almost the same, when the applied voltages \( \phi_o \) are adjusted depending on \( h_o \) so that \( E_o \) gives almost the same value \( 1.6 \text{ kV/mm} \). We can attribute the reason why almost the same height differences are obtained to almost the same strength of Maxwell stresses (\( \propto E_o^2 \)) at the position \( P \) for the four cases of film thickness (a)-(d).

We also studied the effect of electrodes width \( L \) on the surface shape change of silicone oil induced by electric field. We used the following six types of periodically aligned electrode alloy with different width \( L \): (a) \( 200 \text{ \mu m} \), (b) \( 400 \text{ \mu m} \), (c) \( 800 \text{ \mu m} \), (d) \( 1600 \text{ \mu m} \), (e) \( 2400 \text{ \mu m} \) and (f) \( 3200 \text{ \mu m} \), while keeping the distance \( d \) between adjacent electrodes \( 200 \text{ \mu m} \). The kinetic viscosity of the silicone oil is \( 1.0\times10^{-1} \text{ m}^2/\text{s} \) and the film thickness is \( h_o = 180 \text{ \mu m} \). Using results of numerical analysis, we show the microscope images of undulated surface formed on the electrodes with width (a) \( 200 \text{ \mu m} \), (b) \( 400 \text{ \mu m} \), (c) \( 800 \text{ \mu m} \), (d) \( 1600 \text{ \mu m} \) (e) \( 2400 \text{ \mu m} \) and (f) \( 3200 \text{ \mu m} \) at the surface electric field \( E_o \cong 1.9 \text{ kV/mm} \). The white thin solid lines stand for electrodes, the white dotted lines are the bare substrate, and the white thick solid scale bars denote \( 500 \text{ \mu m} \).
calculations of eq.(1), we also adjusted the applied voltage \( \phi_o \) so that the interface electric field \( E_0 \) becomes 1.9 kV/mm for the six different widths (a)-(f) mentioned above. Figure 10 represents microscope images of undulated surface for the six cases (a)-(f) under \( E_o = 1.9 \) kV/mm. We also show the height difference \( \Delta h \) of undulated surface for the six cases (a)-(f) in Fig. 11. The data and error bars in Fig. 11 are evaluated using at least 90 data for each \( L \) for \( L \leq 800 \) \( \mu \)m, and using 40, 26 and 21 data for \( L = 1600, 2400, \) and 3200 \( \mu \)m, respectively. As seen from Fig. 11, the height difference \( \Delta h \) increases with the width of electrode \( L \) for \( L < 1600 \) \( \mu \)m, then decreases with \( L \) for \( L > 1600 \) \( \mu \)m. From the numerical analysis for electric field without change of the surface shape, the strength of electric field becomes weaker at the central region of electrode as the width \( L \) increases, which means the difference in the strength of electric field between that at the surface above the center of electrode and \( E_o \) at \( P \) increases with \( L \). The increase of the difference in strength of electric field between the two regions (i.e., the central region above an electrode and that above a bare substrate) brings the enhancement of the height difference \( \Delta h \). For \( L > 1600 \) \( \mu \)m, on the other hand, the increase in height of film at the middle position between two adjacent electrodes is approximately the same for the cases of different \( L \) since the difference in strength of electric field between the two regions reaches a constant, but the surface height at central region on an electrode does not much decrease as \( L \) becomes longer since the amount of liquid which moves to the region between two adjacent electrodes is almost the same for cases with different \( L \), which results in the decrease of the height difference \( \Delta h \).

In order to investigate time dependence of shape change of silicon film surface, we also used a periodically aligned electrode array where the width of each electrode is settled to 800 \( \mu \)m and the distance between the two adjacent electrodes is set to 400 \( \mu \)m. The kinetic viscosity of the samples are (a) \( 5.0 \times 10^{-3} \) m\(^2\)/s, (b) \( 1.0 \times 10^{-3} \) m\(^2\)/s, (c) \( 5.0 \times 10^{-1} \) m\(^2\)/s and (d) 1.0 m\(^2\)/s. Using results of numerical calculations of eq. (1), we also adjusted the applied voltage \( \phi_o \) so that the interface electric field \( E_o \) becomes 1.2 kV/mm DC for the four cases (a)-(d) mentioned above. We show the time evolution of the averaged height difference \( \Delta h(t) \) after applying the electric field \( E_o = 1.2 \) kV/mm in Fig. 12. For all the silicone oils with different viscosity, \( \Delta h(t) \) seems to reach equilibrium states. It took five seconds for the sample (a) to reach the equilibrium, and took ca.240 seconds for (d). As seen from Fig. 12, the finally reached \( \Delta h \)’s for (a)-(d) are different, even if the error bars are taken into consideration. When the samples are pure dielectric liquids, if the dielectric constant and the surface tension does not depend on the samples and they are the same among the samples, the viscosity influences only the time scale of the dynamics of the surface deformation, not the surface height difference. At the present stage, we cannot clearly explain the origin of the difference of \( \Delta h \) among the samples. In Fig. 13, we show the needed time \( t_{1/2} \) that the height difference \( \Delta h(t) \) reaches the half of the equilibrium height difference for (a)-(d) cases. As seen from Fig. 13, the time \( t_{1/2} \) is linearly proportional to the kinetic viscosity. The result is considered to be reasonable if the dynamics of liquid films is determined by the Stokes equation and the driving force of the surface change is given by the Maxwell stress and the surface tension of silicone oil to the air.

![Fig. 11. Height difference \( \Delta h \) of undulated surface for different width of electrodes \( L \) (a) 200 \( \mu \)m, (b) 400 \( \mu \)m, (c) 800 \( \mu \)m and (d) 1600 \( \mu \)m, (e) 2400 \( \mu \)m, (f) 3200 \( \mu \)m, while keeping the strength of electric field at \( P, E_o = 1.9 \) kV/mm DC.](image1.png)

![Fig. 12. Time evolution of the height difference \( \Delta h(t) \) of undulated surface for different kinetic viscosity (a) \( 5.0 \times 10^{-3} \) m\(^2\)/s (b) \( 1.0 \times 10^{-3} \) m\(^2\)/s, (c) \( 5.0 \times 10^{-1} \) m\(^2\)/s, and (d) 1.0 m\(^2\)/s under \( E_o = 1.2 \) kV/mm DC.](image2.png)
4. CONCLUSION

We investigated changes in surface profile of a liquid film put on a periodically aligned electrode array by imposing a voltage between adjacent electrodes. It is found that the surface of liquid film is undulated according to the periodicity of electrode array, and it rises on between the adjacent positive and negative electrodes, on the other hand, it goes down at the central region on each electrode. The averaged height difference of undulated surface is proportional to the square of the interface electric field $E_o$. When the width $L$ of electrode is changed while keeping the distance between two adjacent electrodes $d$ constant, the averaged height difference $D_h$ increases with the width $L$ of electrode for $L < 1600 \text{ m}$, decreases with $L$ for larger $L$, on the other hand, it goes down at the central region on each electrode. The averaged height difference $D_h$ did not change even for the cases with various thicknesses of liquid films, if the strengths of interface electric field $E_o$ in all the cases are adjusted to be the same. The time evolution of the height difference is also studied for samples with various kinetic viscosities. The time $t_{1/2}$ when the surface height difference reaches the half of $\Delta h$ is proportional to the viscosity of the samples.

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