Surface morphology of fluorene thin film fabricated by electrospray deposition technique using two organic solvents: application for organic light-emitting diodes

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Abstract

We achieved the reduced the surface roughness of poly[9,9-dioctylfluorenyl-2,7-diyl]-co-1,4-benzo-(2,1,3)-thiadiazole (F8BT) thin film fabricated by the electrospray deposition technique, using two organic solvents. The root mean square (RMS) roughness of the F8BT thin film was reduced by the addition of dimethyl sulfoxide (DMSO) in tetrahydrofuran (THF). A minimum RMS roughness of 23 nm was obtained by optimizing the ratio of DMSO in THF. We found that the low evaporation pressure of the additional organic solvent is an important factor in attaining a smooth surface for F8BT thin film. The current density-voltage-luminance characteristics of the organic light-emitting diode was improved by reducing the surface roughness of the F8BT layer. A maximum luminance of 36.7 cd/cm^2 was obtained, and it was more than 14 times higher than the reference device fabricated by the conventional single organic solvent technique.

Key words: Organic light-emitting diode, Surface morphology, Organic solvent, Electrospray deposition, Solution process, Vapor pressure

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1. Introduction

Since many organic devices can be fabricated by a roll-to-roll printing process, researchers have investigated several solution-processed organic devices, such as organic light-emitting diodes (OLEDs) [1], photovoltaic cells [2], image sensors [3], and thin-film transistors [4]. In addition, several printing techniques have also been investigated to achieve a lower fabrication cost and a larger device area compared to conventional Si-based semiconductor devices [5, 6, 7]. This indicates that in the near future, solution-processed organic devices will probably be used in practical applications.

In general, most organic materials can be prepared using solution processes such as gravure printing, dip-coating, screen printing, and conventional spin-coating. A serious drawback of these processes is the dissolution of the underlying organic layer while coating the upper organic layer. This implies that a multilayer structure is difficult to form with different polymers using a conventional solution process, and results in the low performance of the solution-processed organic device [8, 9, 10]. Recently, many solution processes have been developed to fabricate organic devices [11, 12, 13, 14, 15, 16, 17], improving their performance. An intermixing method of the solution process is important technique to improve the device performances; however, the combination of the multilayer structure and the intermixing of organic materials is the useful method for further improved device performances of OLEDs in the near future.

In the case of the spray deposition technique, the smaller particles in the organic solution are important for reducing the surface roughness of the organic thin film, thus improving the performance of the organic device. We investigated electrospray deposition (ESD), a spray method, to fabricate organic thin-film devices [18]. In this process, a high voltage of several kV is applied to the polymer solution, which is then divided into several particles with diameters of less than 1 μm [19]. As a result, the organic solvent evaporates before reaching the substrate, and the organic thin film can be deposited using a thermal evaporation method, without dissolving the underlying organic layer. Other advantages of this technique are that it has (1) a simple experimental setup without special conditions (high temperature and vacuum chamber) and (2) the potential for thick films with a high uniformity and morphology [20].

Recently, J. S. Kim reported an optimized organic bulk heterojunction photovoltaic cell fabricated with the ESD method [21]. The pancake-like

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structures with diameters of over 10 μm were obtained by ESD; however, the surface morphology was improved using the solvent vapor soaking technique. In addition, the surface morphology of the organic thin film was improved by using two organic solvents (*N,N*-dimethylformamide (DMF) and toluene with different evaporation temperatures, and the ESD-based OLED was demonstrated [22]. To further improve the performance of OLEDs, the surface roughness needs to be reduced by choosing an additional solvent and by optimizing the fabrication condition.

In this paper, we investigated the way in which to reduce the surface roughness of the poly[9,9-dioctylfluorenyl-2,7-diyl]-co-1,4-benzo-(2,1,3)-thiadiazole (F8BT) thin film through the addition of several organic solvents. F8BT has high photoluminescence quantum efficiency [23] and carrier mobility [24]; therefore, it is of interest for use in several organic devices. In the ESD method, the spray diameter is an important factor for the future mass production system; therefore, the influence of the relative dielectric constant of the organic solvent on the spray diameter was measured for single and double organic solvent techniques of the ESD method.

2. Experimental details

A schematic configuration of the experimental setup for the ESD method is shown in Fig. 1. A glass capillary was fabricated using a puller (PC-10, Narishige) and a microforge (MF-900, Narishige). The tip of the glass capillary was approximately 50 μm in size. A red light was irradiated around the tip of the glass capillary, and a spray area was captured using a charge-coupled device (CCD) camera (WAT-902B, Watec). The inset of Fig. 1 shows the CCD image while spraying an organic solution. We estimated the spray diameter at distances of 2 mm, 4 mm, and 6 mm from the tip of the glass capillary in order to investigate the influence of the relative dielectric constant on the spray diameter.

A positive high voltage was applied to a conductive wire in the organic solution, using a high voltage source (HJPQ-30P1, Matsusada Precision). The ground line was connected to an indium tin oxide (ITO) layer on top of the glass substrate. The organic solution was atomized into small charged droplets by applying the high voltage between the organic solution and the ITO layer [22]. We used 18 different organic solvents: ethanol, methanol, acetone, 2-ethoxyethanol, 1-penthanol, 1-propanol, tetrahydrofuran (THF), ethylene glycol, benzene, toluene, hexane, cyclohexane, monochlorobenzene,

o-dichlorobenzene, 1,2-dichloroethane, dichloromethane, methoxybenzene, and DMF. The applied high voltage ranged from 4.5 to 7.0 kV because the stable condition of the Taylor cone is affected by the viscosity, conductivity, and surface tension of the deposited material [25, 26, 27]. We selected the optimized applied voltage when the spray diameter was largest and stable. The distance from the glass capillary to the ITO layer was 5 cm.

We also investigated the spray diameter of THF with an additional organic solvent. The measurement position was 6 mm from the tip of the glass capillary. The ratio of the second organic solvent was 5 vol.%. The applied high voltage was fixed at 3.6 kV for all the experimental conditions, and this voltage represented the stable spray condition in the case of THF only. The spray diameter was also measured in the same way as above. We also estimated the surface morphology of the F8BT thin film by adding the second organic solvent. After dissolving F8BT (American Dye Source, Inc.) in THF at a content of 0.1 vol.%, we then added the second organic solvent to the resulting solution. The deposition time was 3 min to observe the surface morphology of the F8BT neat film using a scanning electron microscope (SEM) and an atomic force microscope (AFM). The F8BT solution was deposited on the ITO-coated glass substrate.

An organic LED was fabricated in a subsequent process. An ITO layer with a thickness of 150 nm was sputtered on the glass substrate. The prepared glass substrate was then cleaned in organic solvent/deionized water under ultrasonic waves and was treated with ultraviolet ozone cleaning for 20 min. PEDOT:PSS (P AI 4083, H.C. Stark) was spin-coated on the ITO layer as a hole transport layer. The rotation speed was 2000 rpm, and the thickness of the PEDOT:PSS layer was 50 nm (measured by the surface profiler system [Alpha Step IQ, KLA Tencor]). After annealing at 120 °C for 30 min in a nitrogen atmosphere, the F8BT layer was deposited using the ESD method. The applied voltage was 4.5 kV, and the distance from the glass capillary to the ITO layer was 5 cm. The deposition time was 3 min, and the thicknesses of the F8BT layers were 100 nm and 80 nm for devices A and B, respectively. The organic solutions were a mixture of dimethyl sulfoxide (DMSO) and THF for device A and THF only for device B. The concentration of DMSO in THF was 20 vol.%. The sample was annealed at 120 °C for 30 min to remove the organic solvent in a nitrogen atmosphere. Finally, LiF (1 nm) and Al (100 nm) were thermally evaporated, successively, to obtain an efficient carrier transport between a metal cathode and an adjacent organic layer (F8BT) [28]. The deposition rates of LiF and Al, measured with a

quartz crystal monitor, were 0.05 and 0.3 nm/s, respectively. The device areas were 9 mm^2. The current density-voltage-luminance characteristics were measured using a DC voltage current source/monitor (6241A, ADCMT) and a luminance meter (LS-100, Konicaminolta).

3. Results and discussion

Figure 2 shows the relationship between the relative dielectric constant of the organic solvent and the sprayed diameter. The spray diameters were measured at different distances (2 mm, 4 mm, and 6 mm) from the tip of the glass capillary. The spray diameter increased with an increase in the relative dielectric constant of the organic solvent at all distances. This is because the charged droplets spread due to Coulomb repulsion [29, 30, 31]. The highly charged droplets are pushed towards the outer side by the static electrical force [32], resulting in a large spray diameter. The spray condition is also affected by the properties of the organic solvent, including its viscosity, conductivity, and surface tension [25, 26, 27]. Since the relative dielectric constant cannot be changed by changing the organic solvent, there was fluctuation in the spray diameter against the relative dielectric constant, as shown in Fig. 2. However, the spray diameter was almost proportional to the relative dielectric constant. This result indicates that the relative dielectric constant is the dominant factor affecting the spray diameter in the ESD method.

The spray diameter increased with increasing distance from the tip of the glass capillary. This phenomena was caused by the charged droplets [32], which were spread due to the static electrical force. We achieved the largest spray diameter (21.0 mm) at 6 mm from the glass capillary. Therefore, the high relative dielectric constant of the organic solvent is required for the roll-to-roll printing process.

We deposited the F8BT thin film on the ITO-coated glass substrate. The applied voltage was 3.6 kV, and the distance from the glass capillary to the glass substrate was 5 cm. In addition, we selected the deposition time of 6 min to estimate the shape of the deposited F8BT particles on the glass substrate. Figure 3 shows the SEM image of the deposited F8BT particles using THF as the only organic solvent. This is because THF has the highest relative dielectric constant and solubility against F8BT, compared to all the organic solvents used in Fig. 2. The relative dielectric constant of THF is 7.9.

As clearly shown in Fig. 3, particle structures were obtained. The diameter of these dropped F8BT particles were several 100 nm. A smooth surface is an important factor for organic thin film devices such as OLEDs and organic photovoltaic cells [33]. A large surface roughness will cause a short circuit between the two electrodes (ITO/Al) and a fluctuation in the device performance. The evaporation speed and spray diameter will be dominant factors affecting the surface roughness of an organic thin film. In a previous paper, Ju et al. demonstrated that the surface roughness can be reduced by adding DMF as a second organic solvent into toluene [22]. Therefore, in this study, we investigated the reduction of the surface roughness by the addition of a second organic solvent in the THF solution.

We investigated the spray diameter of the THF solution by adding the second organic solvent. Figure 4 shows the relationship between the relative dielectric constant of the second organic solvent in THF and the spray diameter at 6 mm from the glass capillary with a tip size of 50 μm. The applied voltage, and the concentration of the second organic solvent were 3.6 kV, and 5 vol.%, respectively. As was the case in Fig. 2, the spray diameter increased with an increase in the relative dielectric constant of the second organic solvent. This result indicates that the high relative dielectric constant results in the spread of the organic solution (due to the high Coulomb repulsion force [29, 30, 31]). The concentration of the second organic solvent was 5 vol%; however, the maximum spray diameter was 2.3 times larger than the reference condition without the second organic solvent (i.e., THF only). A maximum diameter of 20.0 mm was obtained when the second organic solvent was DMSO with the relative dielectric constant of 47.

Figure 5 shows SEM images of F8BT thin films deposited with various second organic solvents such as DMSO, toluene, 1,2-dichloroethane, hexane, acetone, and dichloromethane. The applied voltage, the concentration of the second organic solvent, and the deposition time were 3.6 kV, 5 vol.%, and 6 min, respectively. As clearly shown in Fig. 5, aggregated structures with diameters of several hundred nm were observed when hexane, acetone, and dichloromethane were used as the second organic solvent. The vapor pressures of DMSO, toluene, 1,2-dichloroethane, hexane, acetone, and dichloromethane are approximately 2.7, 2.9, 8.7, 17.0, 24.0, and 47.4, respectively, at room temperature. One possible explanation for this result is that the high vapor pressure of the second organic solvent causes the aggregated F8BT. This is because the F8BT particles evaporate before reaching the ITO layer due to their smaller particle size (less than several μm [32]).

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Wet droplets were observed when DMSO, toluene, and 1,2-dichloroethane were used as the second organic solvent. The sprayed F8BT particles are thought to reach the ITO layer with less evaporation of the organic solvent. Thus, the relatively wet F8BT droplets are deposited on top of the ITO layer, as shown in Fig. 5. In this experimental result, we found that DMSO is most suitable as the additional organic solvent to form the smooth F8BT thin film.

In order to achieve the smooth surface of the organic thin film, we investigated the concentration dependence of DMSO on the surface morphology of the F8BT thin film. The applied voltage and the deposition time were 4.5 kV and 3 min, respectively. Figure 6 shows the SEM images of the F8BT thin films with different concentrations of DMSO in THF. The surface roughness was drastically reduced with an increasing concentration of DMSO, and the smooth surface was achieved by adding DMSO at a concentration of 10 vol.%. However, the F8BT solution was aggregated when the concentration of DMSO was greater than 10 vol.%.

The relationship between the concentration of DMSO and the RMS roughness of the F8BT thin film is shown in Fig. 7. The RMS roughness was calculated using the AFM images. It was found to rapidly decrease with an increase in the concentration of DMSO up to 10 vol.%. The evaporation pressure of DMSO was 2.7, which was much lower than that of THF (19.3). This indicates that the sprayed F8BT particles do not easily evaporate when the concentration of the DMSO increases. Therefore, the wet droplets spread in a perpendicular direction against the glass substrate after depositing the F8BT particle, resulting in the reduced surface roughness. In addition, the RMS roughness of the F8BT thin film fabricated by the spin-coating process was 9.1 nm, and the smoothest RMS roughness of the ESD-based F8BT thin film was 23.0 nm.

Figure 8 shows the influence of the applied voltage on the spray diameter when the concentration of DMSO was changed from 0 to 10 vol.%. The spray diameter increased with an increase in the concentration of DMSO and the applied voltage. In general, the electrical charge of the F8BT droplets increased with an increase in applied voltage [25]. Therefore, the highly charged F8BT droplets were pushed towards the outer side by the static electrical force due to the Coulomb repulsion [29, 30, 31]. Furthermore, the spray diameter increased with the increasing concentration of DMSO, as shown in Fig. 8. This is because the relative dielectric constant of spread solution increased with increasing concentration of DMSO in THF. This in-

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dicates that the electrical charge of the F8BT droplets increased with the increasing concentration of DMSO, resulting in a large particle size [30]. The wet F8BT particles were deposited on the ITO layer due to slow evaporation speed when the concentration of DMSO increased [34], resulting in a smooth surface of the F8BT thin film(as shown in Fig. 8).

Figure 9(a) shows the current density-voltage-luminance characteristics of devices A and B. We used the mixed solution of DMSO and THF for device A; THF alone was used for device B. The current density of device A was higher than that of device B. This is because the surface roughness of device A (DMSO at 10 vol.%) was smoother than that of device B (DMSO at 0 vol.%), as shown in Fig. 7. In addition, the luminance of device A was drastically improved by the addition of DMSO due to the smooth surface of the F8BT layer. The maximum luminance of devices A and B were 36.7 and 12.5 cd/m^2, respectively. Furthermore, the current density rapidly increased to approximately 5 V for device B. This was due to the surface roughness of the F8BT layer [35], and this is not good for the organic thin film device. Figure 9(b) shows the current density-voltage-luminance characteristics of reference device fabricated by a conventional spin-coating process. The device structure is ITO/PEDOT:PSS (50 nm)/F8BT (100 nm)/LiF (1 nm)/Al (100 nm). The performance of the device A fabricated by the ESD method was lower than the reference device. The most likely cause of this result is that the relatively higher RMS roughness of the F8BT thin film causes the higher current density and the lower luminance compared to the reference device, as shown in Figs. 9(a) and (b).

4. Conclusion

We achieved the reduced surface roughness of the F8BT thin film fabricated by the electrospray deposition technique. The important method to reduce the surface roughness is the additional second organic solvent. We found that the low vapor pressure of the second organic solvent is important parameter to reduce the aggregated structure. In addition, the relative dielectric constant of the organic solvent affects the spray diameter for both cases of the conventional ESD method and the mixing organic solvents. Finally, we demonstrated the OLED with the maximum luminance of 36.7 cd/m^2.

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Figure captions

Figure 1 Schematic configuration of the experimental setup of the ESD method. The inset shows the CCD image while spraying the organic solution, and the spray diameter was estimated by the CCD image.

Figure 2 Relationship between the relative dielectric constant of the organic solvent and the spray diameter measured at the difference positions from the tip of the glass capillary. The dashed line indicates the regression line of the measured spray diameter.

Figure 3 SEM image of the F8BT thin film deposited by the ESD method. The organic solution consisted of F8BT dissolving into THF with the concentration of 0.1 vol.%.

Figure 4 Relationship between the relative dielectric constant of the additional organic solvent and the spray diameter measured at 6 mm far from the tip of the glass capillary. Dominant organic solvent was THF and the concentration of additional organic solvent was fixed 5 vol.%.

Figure 5 SEM image of the F8BT thin film deposited by the ESD method by adding different organic solvents. The concentration of the additional organic solvent was 5 vol.%.

Figure 6 SEM image of the F8BT thin film fabricated by the ESD method with the two organic solvent technique. The additional organic solvent was DMSO, and the concentration was changed from 0 to 10 vol.%.

Figure 7 Relationship between the RMS roughness of the F8BT thin film and the concentration of DMSO in THF.

Figure 8 Influence of the applied voltage on the spray diameter measured at 6 mm far from the tip of the glass capillary. The concentration of DMSO was changed as 0, 0.1, 0.5, 1, 5, and 10 vol.%.

Figure 9 (a) Current density-voltage-luminance characteristics of devices A and B. (b) Current density-voltage-luminance characteristics of the reference sample fabricated by the conventional spin-coating process.
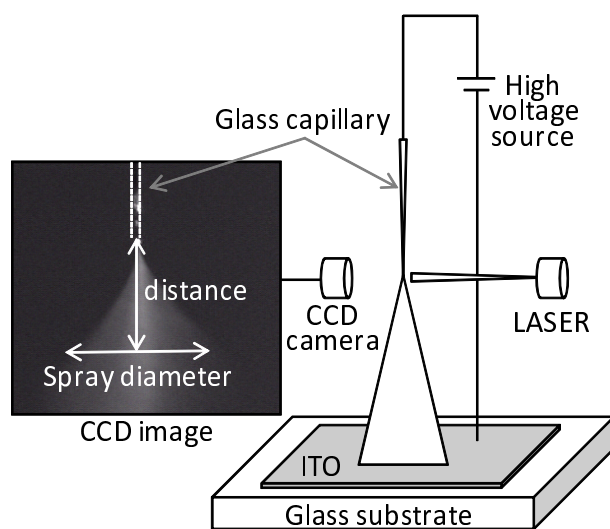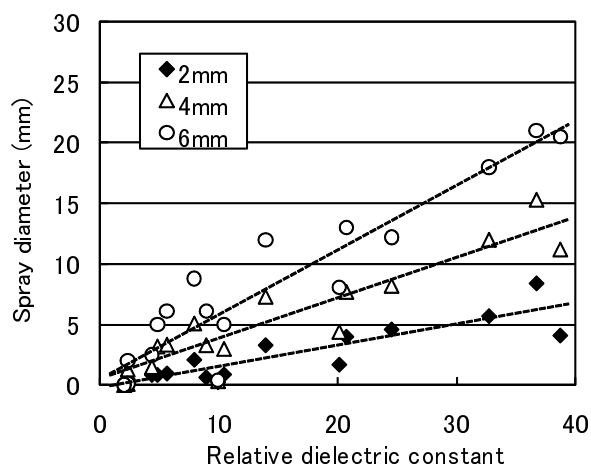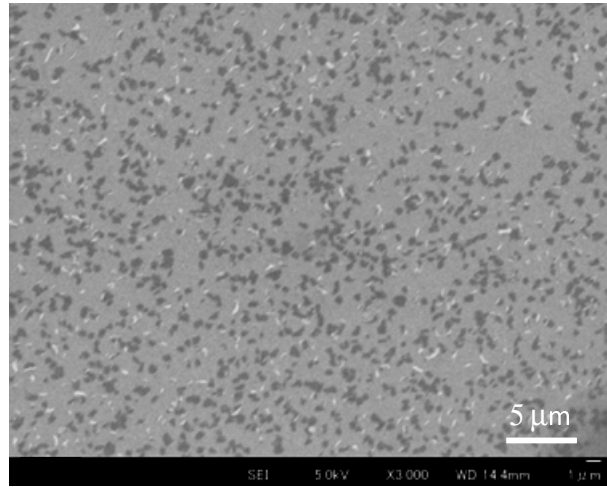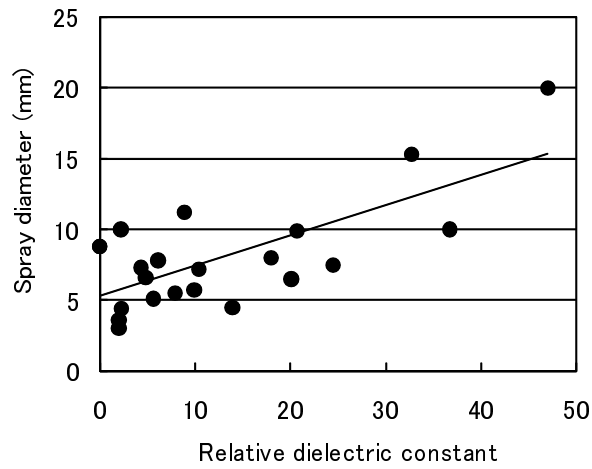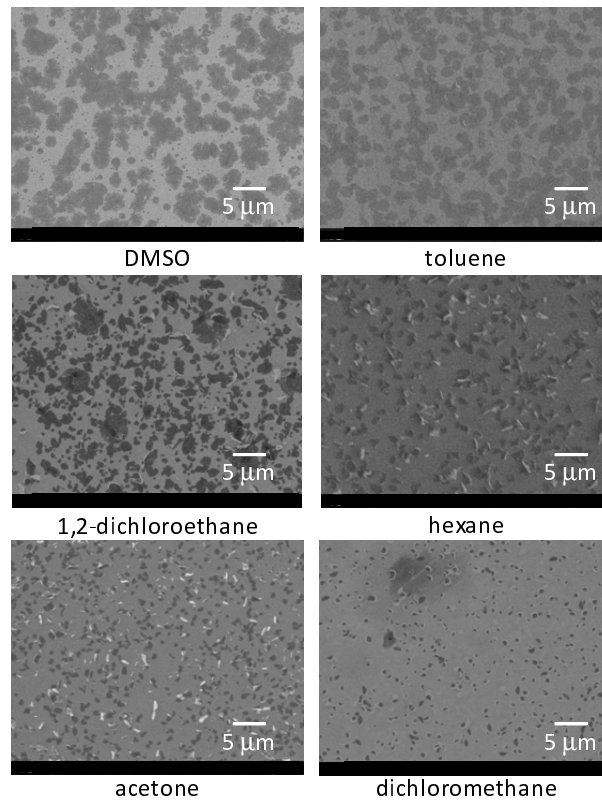
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Figure 5: SEM image of the F8BT thin film deposited by the ESD method by adding different organic solvents. The concentration of the additional organic solvent was 5 vol.%.

Figure 6: SEM image of the F8BT thin film fabricated by the ESD method with the two organic solvent technique. The additional organic solvent was DMSO, and the concentration was changed from 0 to 10 vol.%.
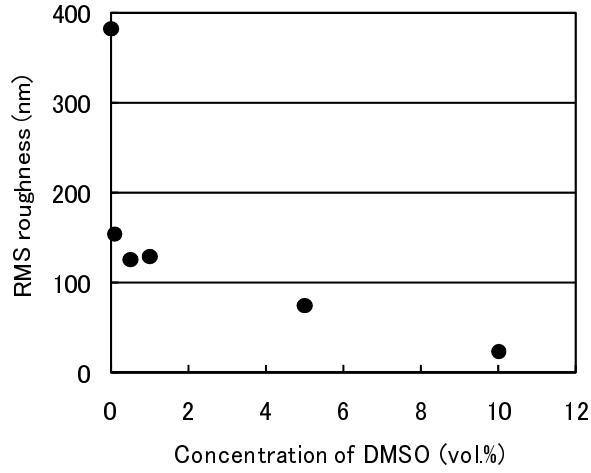
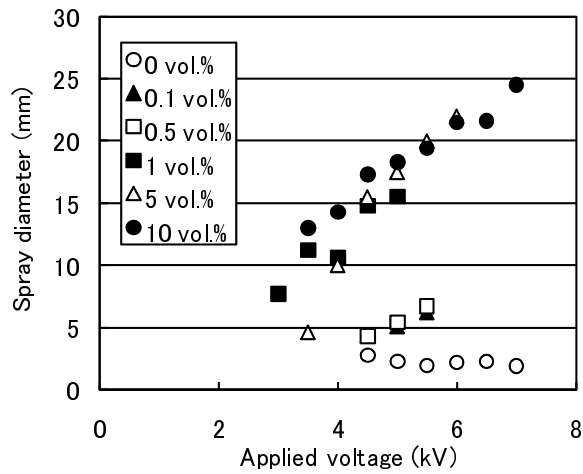Figure 7: Relationship between the RMS roughness of the F8BT thin film and the concentration of DMSO in THF.

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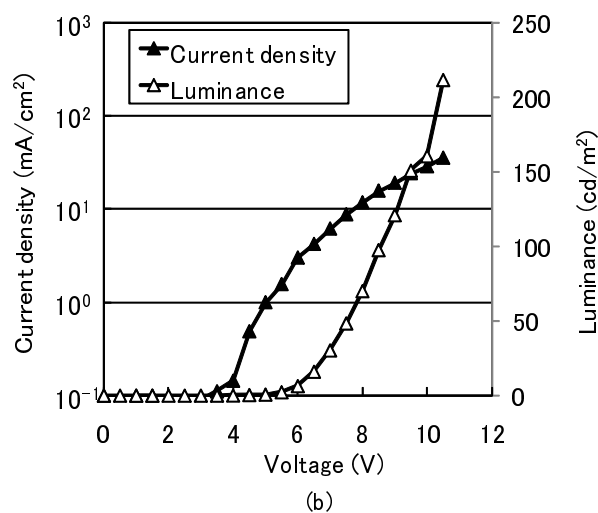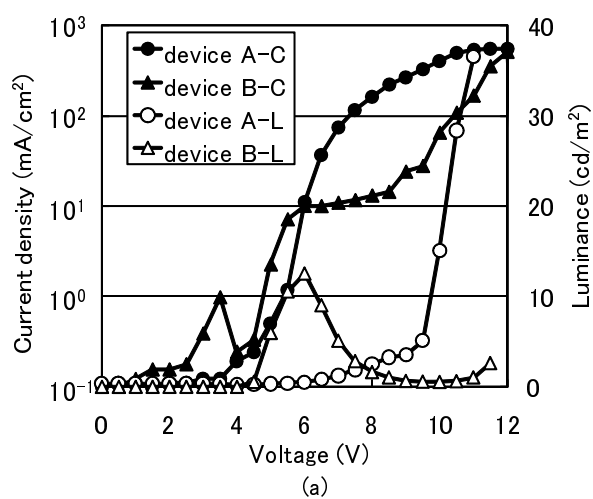
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