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Preparation of Polymer Light-Emitting Devices by Electrophoretic Deposition

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It has been shown that the conjugated polymer films for electronic devices can be prepared by the electrophoretic deposition with polymer suspensions derived from dilute polymer solutions which are so dilute that the conventional spin-coating technique is not applicable. For example, a few hundreds nm-thick film can be prepared on an indium-tin-oxide electrode from a suspension derived from a solution containing 0.1 g/L of the polymer. The polymer light-emitting devices with various thickness of the emission layer prepared by the electrophoretic deposition have been demonstrated.

Keywords: conjugated polymer; dilute solution; electrophoretic deposition; polymer light-emitting device

1. INTRODUCTION

Major advantages of conjugated polymers as semiconductors against conventional inorganic semiconductors may be in their unique properties such as mechanical flexibility and solubility. Especially, the latter feature enables the preparation of semiconductor films under the atmospheric pressure by wet-processes, motivating a number of researches on the application of conjugated polymers for light-emitting devices [1–3], photocells [4,5], field-effect transistors [6,7] and other electronic devices, or "printed electronics."

For the application of the conjugated polymers to large-area electronic devices, it is important to develop high-throughput and efficient

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technologies of coating. Although the electrophoretic deposition technology is widely used in the industrial coating process, it has not caught the adequate attentions of the researchers of organic electronics for many years. The principle of electrophoretic deposition is quite simple; the electric field accelerates the colloidal particles in the suspension of material, and the particles reached to the electrode form deposit. It is obvious that the resultant films by electrophoretic deposition are particulate, since they are just accumulated colloidal particles. We have reported the electrophoretic deposition of conjugated polymers, from suspensions which are prepared by simple re-precipitation technique, as a method to obtain nanostructured conjugated polymer films [8,9].

Recently, we have found that the morphology of the films of a polyfluorene-type conjugated polymer poly[(9,9-dioctyl-2,7-divinylene fluorenylene)-alt-(2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylene)] (PDOF-MEHPV) by electrophoretic deposition strongly depends on the content of good solvent of suspension used [10]. The polymer films from suspensions containing less than 20% of toluene has apparently rough surface and are frosted. On the other hand, the films from the suspensions with more than 30% of toluene are transparent. The atomic force microscopy study has indicated that the latter films with approximately 100 nm in thickness have the rms-roughness below 10 nm. It has been also confirmed that the light-emitting devices with ITO/PEDOT:PSS/PDOF-MEHPV/MgAg structure, where PEDOT:PSS denotes poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) salt, using the latter type of films show uniform emission.

From the viewpoint of the required concentration of the polymer solution in good solvent to make suspensions, this result is important. For example, while the suspension consisting of 90% of poor solvent, 10% of good solvent and 0.1 g/L of the polymer is made by mixing a unit of the polymer solution containing 1.0 g/L of the polymer with 9 units of the poor solvent, the suspension consisting of equivalent volumes of the poor and good solvents with the same polymer content requires the polymer solution containing just 0.2 g/L, 5 times thinner than the aforementioned one, of the polymer.

In this study, the preparation of the polymer light-emitting devices with various thickness of the emission layer from dilute polymer solutions by using the electrophoretic deposition technique has been performed.

2. EXPERIMENTAL

The polymer suspensions were prepared by mixing equivalent volumes of the toluene solution of the polymer and the acetonitrile.

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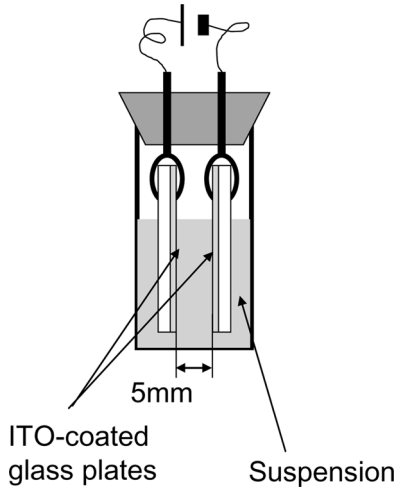

FIGURE 1 Setup for the electrophoretic deposition.

The electrophoretic deposition is carried out by applying DC voltage between a couple of indium-tin-oxide (ITO) electrodes coated on glass plates which are soaked in the suspension charged in a glass cuvette as shown in Figure 1. The deposition of PDOF-MEHPV occurs on the positively biased electrode or the anode, indicating that the colloidal particles in the suspension are negatively charged. Since the suspension used contains equivalent volumes of good and poor solvents, dense and homogeneous thin films are obtained by the natural drying in air, as mentioned in the previous paper [10]. To improve the device performance, The ITO electrodes served as anode are coated with approximately 50 nm-thick PEDOT:PSS layer by spin-coating the aqueous suspension.

The thickness of the PDOF-MEHPV films is estimated by using the relationship that unit peak absorbance approximately corresponds to 120 nm in thickness.

The light-emitting devices with a ITO/PEDOT:PSS/PDOF-MEHPV/MgAg structure were fabricated with various thickness of PDOF-MEHPV films by the electrophoretic deposition. PDOF-MEHPV films have been deposited on the PEDOT:PSS-coated ITO films by the application of DC 300 V between 5 mm in a suspension containing 0.05 g/L of the polymer for 4–10 s, resulting in the PDOF-MEHPV films with thicknesses ranging from 80 nm to 250 nm. The emission area of the devices is approximately 3 mm $\times$ 3 mm square, and the emission intensity was measured with a Si-photodiode attached on the emission face of the

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devices. The device structure is similar to those reported in our previous paper [11]. The vacuum deposition of cathode composed of Mg and Ag, as well as the characterization of the devices was performed in a glove-box filled with nitrogen. The devices with the same structure using spin-coated PDOF-MEHPV films were also prepared for comparison.

3. RESULTS AND DISCUSSION

Typical emission intensity-voltage and the emission intensity-current characteristics of the devices are shown in Figures 2(a) and (b), respectively. Since the emission onset voltage as well as the quantum efficiency of the devices does not seem to seriously depend on the preparation method. It can be concluded that the characteristics of the devices fabricated by the electrophoretic deposition are comparable to those by the spin-coating.

The dependence of the emission onset voltage on the polymer thickness is plotted in Figure 3. The onset voltage seems to be determined by the film thickness, and there is no significant difference due to the difference in the preparation method. The onset voltage is monotonically increased with the increasing thickness of the PDOF-MEHPV

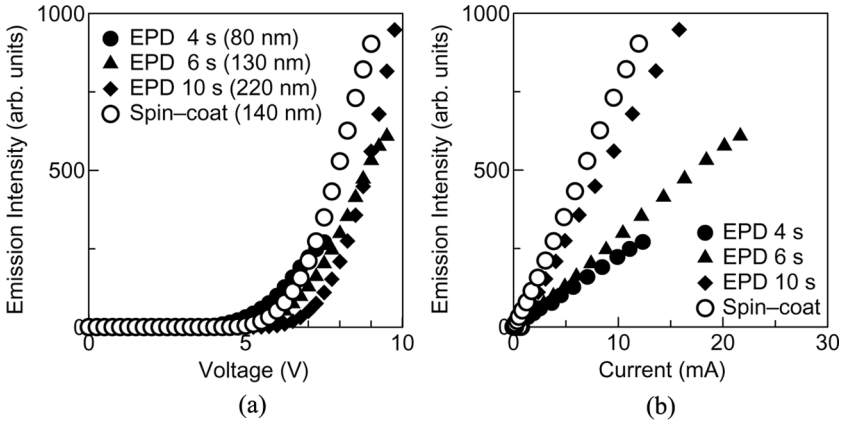

FIGURE 2 (a) Emission intensity-voltage characteristics of the light-emitting devices using PDOF-MEHPV films prepared by the electrophoretic deposition with various deposition periods. The case of spin-coated PDOF-MEHPV film is also indicated. The thicknesses of PDOF-MEHPV are indicated in the parentheses. Accompanying emission intensity–current characteristics are shown in (b). The emission area of the devices is approximately 3 mm × 3 mm.

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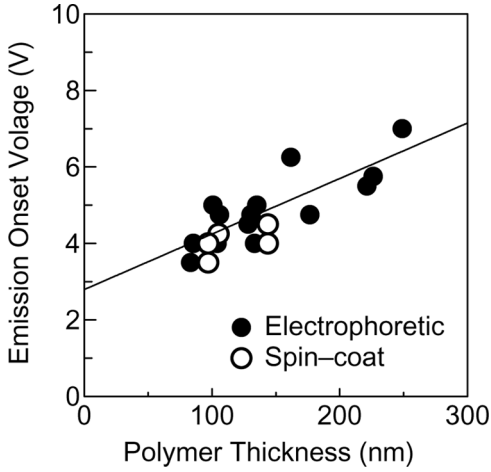

FIGURE 3 Dependence of the emission onset voltage on the PDOF-MEHPV thickness of the light-emitting devices using PDOF-MEHPV films prepared by the electrophoretic deposition. The cases of spin-coated PDOF-MEHPV films are also indicated. The line shows the linear fitting.

layer, as commonly observed in the study of polymer light-emitting devices [12].

For the preparation of the devices by the spin-coating technique, a chloroform solution containing 10 g/L of the polymer and the spin-speeds ranging from 1000–5000 rpm have been employed. Generally, the spin-coating from the chloroform solution of a polymer gives thicker film easily than that from the toluene solution at a constant polymer concentration because of the difference in the evaporation rate, and a spin-speed below 1000 rpm results in inhomogeneous films. Despite of the concentrated polymer solution used, the maximum thickness of the spin-coated films obtained was below 150 nm.

In conclusion, it has been mentioned that the electrophoretic deposition using a suspension derived from a toluene solution containing only 0.1 g/L of the polymer can yield films with thicknesses >200 nm, which were not obtained by a single spin-coating shot with a 10 g/L chloroform solution. The light-emitting devices using the electrophoretically deposited polymer films show similar performance to those using spin-coated films. These features indicate that the electrophoretic deposition can be an important candidate for the film deposition technique for polymer-based electronic devices.

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