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# PEDOT:PSS Films Uniformly Coated on Polyimide Substrate without Repellence

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## Abstract

PEDOT:PSS films were grown on polyimide substrates by means of a bar-coating method. The biggest problem is repellency resulting in an obstacle to forming a uniform thin film. Starting PEDOT solution mixed with ethanol, $CuSO_4$ water solution and ethylene glycol at an appropriate ratio was found to be able to reduce repellency prominently to bear a uniform film on plastic substrates. The resistivity of the film becomes lower by 22% when it is heated on a hot plate during drying.

## Keywords

Bar Coat, Polyimide Substrate, Printing Electronics, PEDOT:PSS

## 1. Introduction

Organic electronics have been developed so far; organic electroluminescence displays are the most common examples in our everyday life. Organic material usually does not include toxic elements such as Hg, Cd and so on. It does not include rare earth elements at the same time. Organic materials are actually suitable for our sustainable society nowadays. It is a great advantage to be able to fabricate the material through chemical reactions. When we need to dispose of organic electronic devices, they can be easily burned to ashes.

In order to realize electronic devices, it is inevitable to form electrodes or electronic conductors. There are a few organic materials for electronic conductors actually. PEDOT:PSS is a good candidate material as an organic electronic conductor. PEDOT:PSS consists of two kinds of polymers of poly(3,4-ethylene dioxythiophene(PEDOT) and poly styrene sulfonate (PSS), which are usually used as a starting material in the form of dispersion in water. And then it is easy to be diluted or treated because we do not need any special solvent.

One application of PEDOT:PSS is an electrode for organic light emitting devices. In this case, PEDOT:PSS is coated on indium tin oxide film on glass substrates [1] [2]. Micropatterns were also challenged in this field [3]. It is also used in a white LED or an electroluminescence device with a polymer activating layer [4]-[6]. PEDOT:PSS is also utilized in organic field effect transistors [7] [8] and organic thin film transistors [9]-[11]. The advantage of using polymer as a starting material for organic electronic devices is that offset [7] or inkjet printing techniques [9] [11] [12] can be applied. These techniques need no vacuum environment, which requires a high-cost apparatus and long time for forming films. In addition, the spray coating method was also applied to PEDOT:PSS [13].

PEDOT:PSS has been widely applied to other devices such as supercapacitors [14], thermoelectric nanocomposites [15], and actuators [16]. The characteristics of PEDOT:PSS have also been studied so far. For example, PEDOT:PSS on a silicon substrate was analyzed by means of ellipsometry [17] [18], or the work function was studied [19]. It was also reported that the hole injection barrier between pentacene and PEDOT:PSS is lower than that of a pentacene-Au interface [20] and that the ionization potential of PEDOT:PSS varies after acid or base treatment [21]. Anyway, PEDOT:PSS is now known as a good candidate for an organic conductor.

When we realize organic electronic devices, it is a great advantage to use plastic or organic polymer substrates instead of glass plates or inorganic ones as silicon crystalline wafers. The most prominent characteristic is to be curved easily so that the electronic devices can be installed in a curved area.

The starting liquid material of PEDOT:PSS is widely used in the form of dispersion in water. As a first step, this is put on the substrates, spread all over by means of a bar-coating method, and then dried. The problem is that the liquid is often repelled by plastic substrates, which makes it so difficult to form a thin uniform liquid film.

Seo *et al.* have studied the formation of an ionic complex of $CuSO_4$ and PEDOT:PSS to become a better conductor [2]. So in this study, it was used the mixture of PEDOT:PSS, $CuSO_4$ water solution, ethanol and ethylene glycol. Both ethanol and ethylene glycol were used for the purpose of the reduction of surface tension. The ratio of each component was varied to find the best ratio. The biggest purpose of this article is to obtain a uniform liquid layer without repelling. Forming a thin film without repelling has a great advantage regardless of the coating method because organic films often repel water solutions.

## 2. Experimental

Polyimide films (Kapton by DuPont) were used as a substrate because polyimide is highly resistant to temperature and endures heat treatment in a process. The size of the substrate was rectangular in shape by 7 and 3.5 mm where the thickness was 0.125 mm. It was rinsed with water and then treated with ethanol vapor. $CuSO_4$ solution was prepared by putting copper sulfate pentahydrate (25 g) into water (150 g). The concentration is then 9.14 wt. % owing to water of hydration. As PEDOT:PSS was used Baytron PH 500 by H. C. Starck. To 1 $mm^3$ of PEDOT:PSS, x $mm^3$ of ethanol, y $mm^3$ of $CuSO_4$ solution and z $mm^3$ of ethylene glycol were added. This mixture is indicated by S(x,y,z).

The mixture liquid of 30 $mm^3$ was put on a substrate with a pipet and then a bar was swept from one edge to the other while keeping contact carefully for several seconds. The bar consists of a stainless-steel rod wrapped with a stainless-steel wire without any gap. With this bar, a film thickness of 0.027 mm (a nominal value) is obtained.

In addition, two samples with uniform films were dried and then resistivity was measured by Van der Pauw method. One sample was dried naturally and the other with a hot plate at 140 degree Celsius for 5 minutes.

## 3. Results and Discussion

Two photos of samples S(0, 0, 0) and S(2, 0, 0) are shown in **Figures 1**. The polyimide substrate is a brown sheet and PEDOT:PSS liquid mixture shows a black color. Round islands of black color were clearly observed in S(0, 0, 0). On the other hand, S(2, 0, 0) also shows black islands but the shape is not so round as S(0, 0, 0) but a sawtooth one.

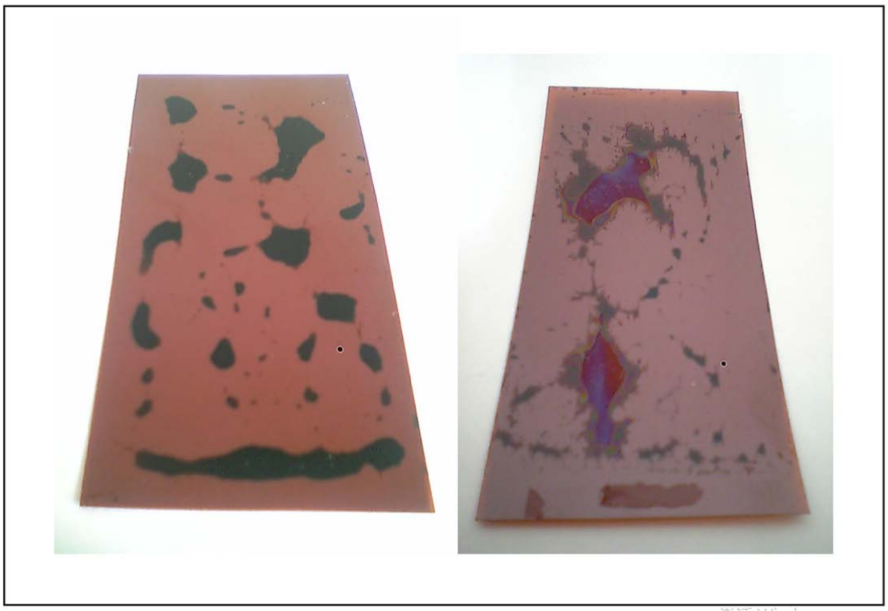

**Figure 1.** Photos of S(0, 0, 0) and S(2, 0, 0) from the left to the right. Ethanol controls repellency to a slight extent.

The two samples to which 0.025 $cm^3$ of $CuSO_4$ solution was added are shown in **Figure 2**. Ethylene glycol (0.5 $cm^3$) was also added in S(2, 0.025, 0.5); the right one in **Figure 2**. In the two samples, repellency was controlled to a great extent compared with the samples in **Figure 1**. Actually Seo *et al.* reported the addition of $CuSO_4$ to PEDOT:PSS solution lowers resistivity, but they did not describe about repellency or the liquid state during a coating process. The addition of $CuSO_4$ surely contributes to the control of repellency. The appearance of the two samples is almost the same regardless of the addition of ethylene glycol. The resistance was quite different between the two by checking electric resistance.

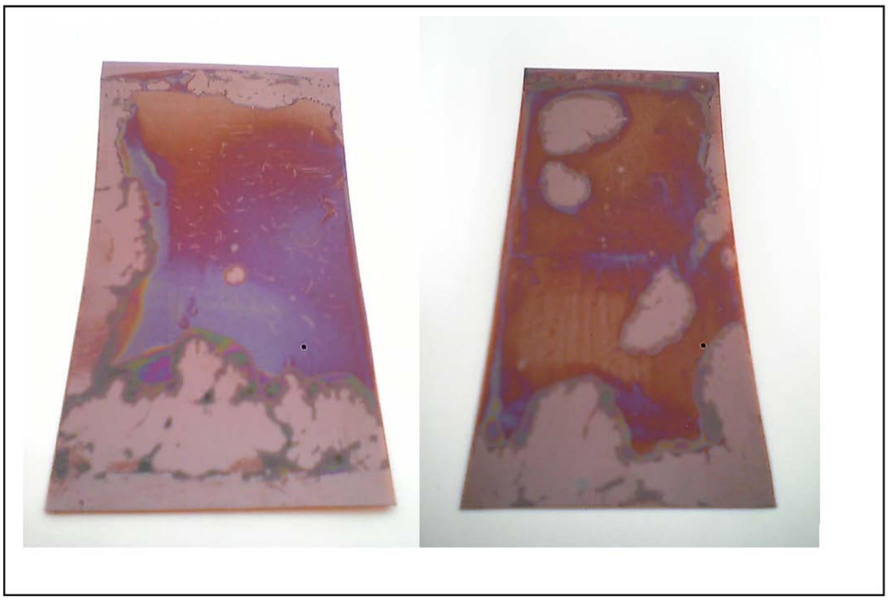

**Figure 2.** Photos of S(2, 0.025, 0) and S(2, 0.025, 0.5) from the left to the right. Ethanol controls repellency to a slight extent.

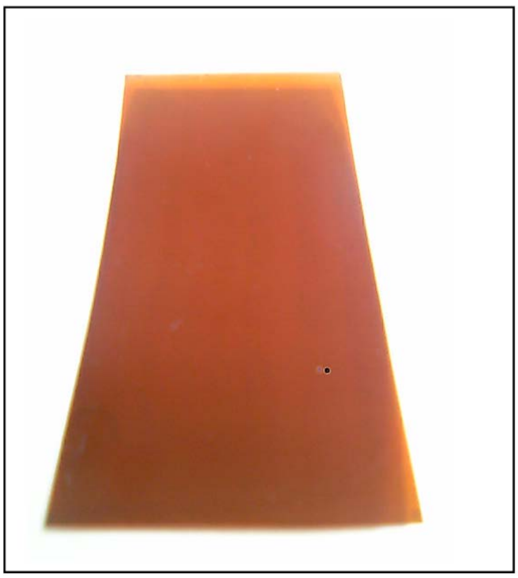

**Figure 3.** Photo of S(1, 0.05, 0.3) that shows a perfectly uniform surface of the coating layer without any repellency.

Simply for the longest span. The addition of ethylene glycol improved it to 5 k ohm in S(2, 0.025, 0.5); it is one thousandth of that of S(2, 0.025, 0).

The combination of x, y and z was varied in the range of 0 - 2, 0 - 0.2, 0 - 1, respectively. It was found through trial and error that the sample of S(1, 0.05, 0.3) has the most uniform layer by an eye as shown in **Figure 3**. In the mixture with much $CuSO_4$ solution, many solid particles appear. The particles become an obstacle to sweeping the bar during a coating process. The particles were ground to a smaller size before the coating process. **Figure 3** shows a perfectly uniform and transparent layer without any repellency.

Two samples of S(1, 0.05, 0.3) were dried and then resistivity was measured by means of Van der Pauw method. One was dried naturally for a long enough time (SN) and the other was dried on a hot plate at 140 degree Celsius for 5 minutes (SH). The SN and SH samples were found to have the resistivity of 1.76 and 1.33 mohm cm, respectively. From this result, resistivity was lowered 22% by heating during drying.

Ahn *et al.* reported that thermal annealing enhances efficiency in organic light-emitting diodes of a blend polymer with a PEDOT:PSS electrode [22]. The annealing was performed between 20 and 200 degree Celsius. They did not describe the annealing effect on PEDOT:PSS only. So our results cannot be compared with theirs. The next interesting point is whether thermal annealing after naturally dried samples decreases the resistivity.

## 4. Conclusion

PEDOT:PSS layers were fabricated on polyimide substrates by means of a bar coating method. Since plastic plates such as polyimide usually repel liquids, repellency is a serious problem for forming a uniform liquid layer. It has been achieved to make it by adding ethanol, ethylene glycol and $CuSO_4$ solution to PEDOT:PSS dispersion liquid at a proper rate. In addition, the resistivity of PEDOT:PSS layer was lowered by 22% by heating during drying compared with no heating drying. But it is not clear yet, whether heating after drying the solvent makes sense.

## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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