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Evaluation of Charge Transport in a Percolating Network of Disordered Organic Thin Films

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Abstract. Understanding effective carrier transport on percolating network is important for semiconductor device optimization. Structural as well as dynamical properties of percolation network such as hopping or tunneling length, conducting path, correlation length, system conductivity and resistivity are significant parameters that determine charge carriers mobility across the thin film module. In this work, we describe an alternative method to quantify the geometry of connectivity and the statistical variability of conductivity, and also show that the percolating transport exponent are highly correlated with fractal dimension of the active layer topology.

Keywords: Organic semiconductor; fractal geometry; percolation network
PACS: 73

INTRODUCTION

In the past ten years, the organic electronics have been significantly replacing inorganic semiconductors because of its lower fabrication cost for large area arrays, easy integration of heterostructures and composite materials, and also induced the possibility of using flexible substrate in organic semiconductor. The development of organic multi-layer structures in OLEDs has improved the efficiency of light-emission by achieving a better balance of the number of charge carriers of opposite sign and further lowered the operating voltage. The display and lighting applications prefer the organic light-emitting devices (OLEDs) due to their high efficiency and compatibility with high throughput manufacturing reason. There are more recent developments and research demands in OLED architecture to improve the quantum efficiency by using a graded hetero-junction. The organic molecules are responding sensitively to physical and chemical changes have encouraged the usage of the organic semiconductor in the bio-sensing field. For example, organic field effect transistors (OFETs) are widely used as a sensor in electrolytes and in vitro bio-sensing applications to avoid unwanted electrochemical reactions [1]. The use of disordered interfaces in organic based solar cell (OBSC) or organic photovoltaic solar cells (OPVC) architectures can significantly increase the occurrence of fast charge transfer rate (CT) at the interfaces and hence achieve higher quantum efficiency.

Morphologies of organic semiconductor can be studied with atomic force microscopy (AFM) and scanning electron microscopy (SEM). Morphology images of the disordered organic electronics can be characterized and modeled by using fractals and percolation theories. Over the years, fractal geometry provides a powerful tool to analyze signals in different applications, e.g. medical imaging applications, geographical image analysis, morphological study, physical properties of materials and other fields. Its efficiency has been demonstrated in classification and segmentation experiments where it was used as an additional texture parameter. Texture analysis and fractal methods provide 2D descriptors that are a good reflect of surface roughness and heterogeneity. The calculation of fractal dimensions and other related parameters are useful as quantitative morphological and developmental descriptors. The concept of fractal theories has been widely used in modeling nature objects with complexity in morphology, branching and irregular in shape [2]. The contribution of the "Minkowski cover" and "Kolmogorov box" methods to compute fractal characteristics for a texture roughness are convincingly presented in many research work. The greater the morphologic complexity of an object, the higher its fractal dimension.

In this study, percolation theory is also adopted because a surface topology tends to possess an increase in fractal dimension approaching percolation threshold which have been consistently agreed in many research work [3]. The most recent study has also showed that the percolation theory can be applied to the analysis of carrier mobility in an organic material, but the significant influence of a fractal-percolative network that affects the carrier transport in an organic thin film need to be further investigated.

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METHODS

Surface morphologies of the organic thin films are scanned using atomic force microscopy (AFM) for the solvent treated samples at different time duration which had been intensively studied in our previous work [4], and the images are currently computed by using HARFA software version 5.5 *(http://www.fch.vutbr.cz/lectures/imagesci/)*to generate a detailed computational analysis of fractal dimension at each intensity data point.

Thresholding technique is often used for image segmentation that transforms an image into black and white (BW) or binary image. The input image can be grayscale or color. The segmentation process will convert all data points below a certain threshold to zero and data points above that threshold to one. The binary image has grayscale intensity ranged from 0 to 255, where intensity of 0 has been categorized as black and 255 as white [5].

The grayscale image is then processed by using box counting method to generate fractal spectrum which indicates data points according to the grayscale intensity. Box counting methods are widely used to measure the fractal dimension of a structure. The method begins by superimposing regular grid of pixels of length L on a binary image and counts the number N of occupied pixels/boxes. This procedure is repeated by using different length value with defining power-law relationship: $N(L) = L^{-Df}$. The fractal dimension, D_f can be estimated from the slope of the logarithmic regression line:

$$D_f = -\frac{\log N}{\log L} \tag{1}$$

The clustering effect can be clearly visualized by using another image processing software, *ImageJ (http://rsbweb.riih.gov/ij/),* as shown in Figure 1. The color image is first converted to 8-bit grayscale and then processed under different stage of thresholding conversion to eliminate the shadowy region due to the minor unconnected clusters or other topographical irregularities.

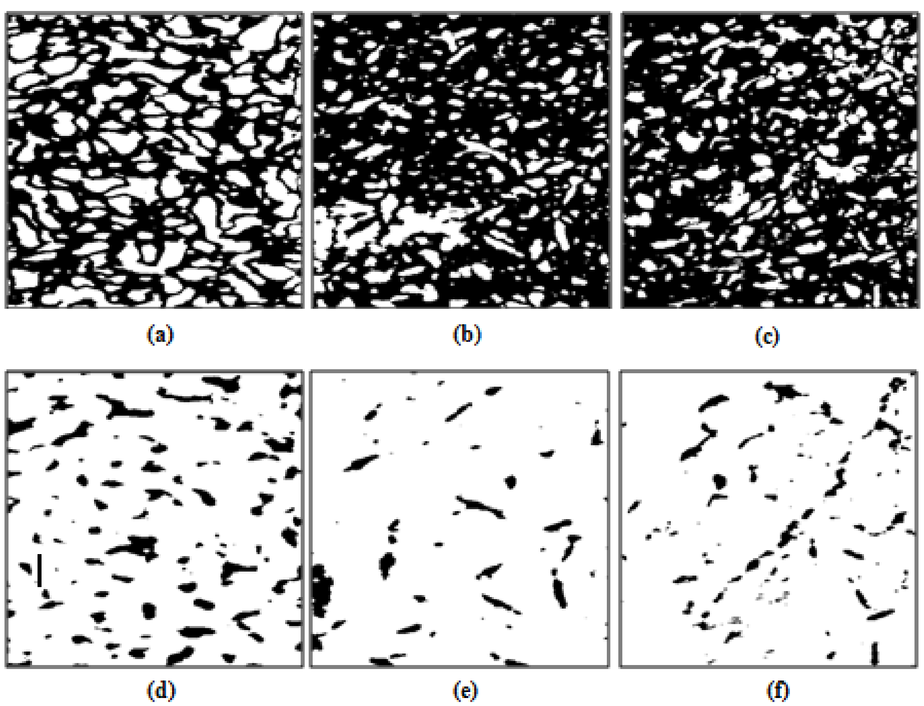

FIGURE 1: (a), (b), (c) are processed by thresholding technique from intensity 50 to 170, while (d), (e), (f) are under threshold 170 to 255, for 40 min, 80 min and 120 min thin films respectively.

RESULTS & DISCUSSION

Referring to the fractal spectrum as shown in Figure 2, no noticeable cluster or obvious fractals is formed between threshold intensity of 0 to 40. The charge transference has been restricted within the isolated cluster which is secluded from the others.The structure of connected clusters seem started to form only at intensity of 40 onwardsand the overall system begin exhibits fractal characteristic. The clusters at this stage are fractal and the carriers might still be trapped due to the existence of internal holes within the cluster, which are non-reachable from outside the cluster.

The black pixels region between 0 to 100 often represents the substrate structure that less contributes to the photocurrent density. However the finite clustering size effect is clearly seenwhen the fractal spectrum shows clear fluctuation towards the white pixels region from intensity of 150 to 200, which competently represents the connected network/clusters for charge transfer. The fractal spectrum at this region shows the consistency in the variation of the fractal dimension with the photocurrent density. It results the highest photocurrent of 0.825 $\mu A/cm^2$ in thin films treated for 40 min, following by 0.800 $\mu A/cm^2$ and 0.600 $\mu A/cm^2$ for 80 min and 120 min respectively. This also suggested that the connected clusters consist of percolative bonds which carry the electric current, often referred as 'backbone' of the cluster [6].

In the final region of white pixels (intensity of 200 to 255), the 40 min thin film continuously performs fractal structure while the other two thin films shows no fractal spectrum at all. This could due to the surface etching and smoothening after longer solvent treatment time for 80 min and 120 min thin. At this stage, the clusters are rather separated from each other.

The clustering size effect is much equally distributed as shown in Figure 1(d) for 40 min, as compared to the other two. The inter-distances between clusters is decreasing by the formation of similar sized cluster. The hopping distance for carriers is therefore shorten, possibly contributing to the highest charge transfer rate. The system conductivity is expected to increase when there are much charge hopping between neighboring conductive network. As seen in Figure 1 (e) and (f) which represents 80 min and 120 min thin film, the clusters are finite, self-similar and very randomly distributed from each other with larger chemical length, ℓ between two cluster sites [7].At the same time, the clusters seem overlapping with each other as shown in Figure 1 (b) and(c), causing disconnected percolation pathway that prevent the carriers to move to infinity across the module.

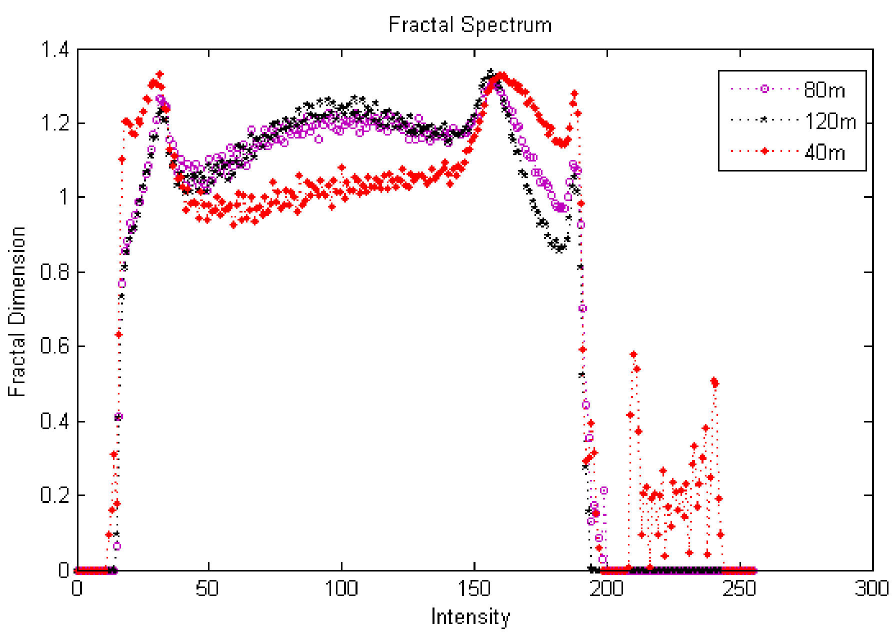

FIGURE 2: Fractal spectrum for all thin films processed by HARFA software.

CONCLUSION

Photocurrent density is highly dependence on the white pixels region which represents the most possible percolative network formed as compared to the other data points in the fractal spectrum. Thresholding technique is competent to provide labeling for almost-connected-component and also enable the visualization of the clustering effect at different stage. The local regularity of the thin films is characterized by the fractal dimension, which is shown to corroborate well the findings based on image analysis of the clustering effect from the AFM. According to our previous work, the fractal dimension is relatively higher when the surface is rougher. Higher conductivity is observed for thin films with higher fractal dimension but also enhanced when there exists spatially correlated morphologies in the form of percolative network enhances carriers transport at interfaces. In other words, the percolating transport exponent are highly correlated with fractal dimension of the active layer topology. However, further research and interpretation of the AFM image need to be carried out to relate efficiently the topology geometry of the active layer with the percolating systems to predict precisely the amount of photocurrent that enhances on the comprehensive conductivity of an active layer.

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