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Photovoltaic Properties of Ultramicrostructure-Controlled Organic Co-Deposited FilmsMasahiro Hiramoto,¹ Kouji Suemori¹ and Masaaki Yokoyama¹Material and Life Science, Graduate School of Engineering, Osaka University
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Yamadaoka, Suita, Osaka 565-0871, Japan**1. Introduction**

Ability of the photocarrier generation of organic semiconductors can be enhanced by mixing two kinds of organic molecules. For example, we observed high photo-electric conversion efficiency in the mixed organic films of metal-free phthalocyanine (H₂Pc) and perylene (Me-PTC) pigments (Fig. 1), which were fabricated by the co-deposition technique [1, 2]. However, carrier generation in such co-deposited films is expected to be crucially affected by the nano-level structure of mixed two pigments. From this standpoint of view, we have tried to control the ultramicrostructure of co-deposited films by the substrate temperature. Remarkable improvements of photovoltaic properties was achieved for the films co-deposited on the cooled substrate.

2. Experiment

Mixed layers were prepared by the co-deposition of H₂Pc and Me-PTC from two separate controlled sources onto the substrate at temperatures of 50, 25, -80, and -170°C. Mixing ratio of two pigments was always 1:1. Photovoltaic properties were evaluated by the single-layered cells (Fig. 2) and by the three-layered cells (Fig. 3). Three-layered cells, in which the co-deposited layer was incorporated as an interlayer sandwiched between p-type H₂Pc and n-type Me-PTC layers, have quasi-pin energy structure [2]. Ultramicrostructure of co-deposited films were evaluated by atomic force microscopy (AFM) and by X-ray diffraction (XRD).

3. Results*3.1 Photovoltaic Properties of Co-deposited Films*

Figure 2 shows the substrate temperature dependence of the short-circuit photocurrent (J_{sc}) of single-layered H₂Pc:Me-PTC (1:1) cells. Interestingly, the co-deposited film evaporated on the substrate cooled at -170°C revealed to be able to generate 15 times larger photocurrent compared to that evaporated on the substrate at room-temperature. High performance of low-temperature fabricated film was also appeared in the three-layered cells, i.e., about 5 times larger J_{sc} of 2.42

mAcm⁻² (100 mWcm⁻², AM1.5) compared to the room-temperature fabricated film was observed (Fig. 3). Maximum quantum efficiency of J_{sc} reaching 50% was observed for the monochromatic light of 420 nm irradiated on the Au electrode.

3.2 Ultramicrostructure of Co-deposited Films

Enhancement of the photocurrent was revealed to be closely related to the ultramicrostructure of co-deposited films. Figure 4 shows the AFM images for the surface of co-deposited films fabricated at room temperature (a) and at -170°C (b). The former film was considerably flat and revealed to be amorphous. On the contrary, the latter film contains a lot of nanoparticle of the diameter of about 20 nm. XRD measurements showed that these particles are Me-PTC nanocrystals and H₂Pc is amorphous.

4. Discussion

We concluded that room-temperature fabricated films have structure of molecular-level mixture of two pigments, while low-temperature fabricated films have structure containing Me-PTC nanocrystals surrounded by amorphous H₂Pc (Fig. 5). Efficient photocurrent generation seems to be due to the large area contact between two pigments and the formation of individual routes for photogenerated electrons and holes. Advantage of low-temperature fabricated films is conspicuous for the thicker films (Fig. 6). Since considerable portion of the incident light can be absorbed by the 100 nm-thick co-deposited film, we think that the present co-deposited films with both high photocarrier generation efficiency and sufficient thickness for light absorption have a potential to make efficient photovoltaic cells.

References

- [1] M. Hiramoto, H. Fujiwara, and M. Yokoyama, *Appl. Phys. Lett.*, **58**, 1062 (1991).
- [2] M. Hiramoto, H. Fujiwara, and M. Yokoyama, *J. Appl. Phys.*, **72**, 3781 (1992).

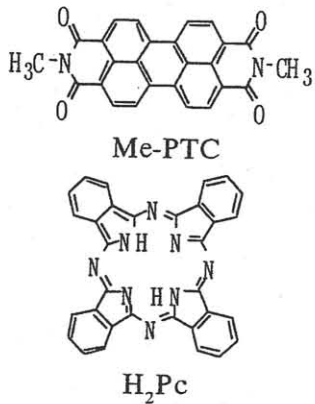


Fig. 1 Organic semiconductors used.

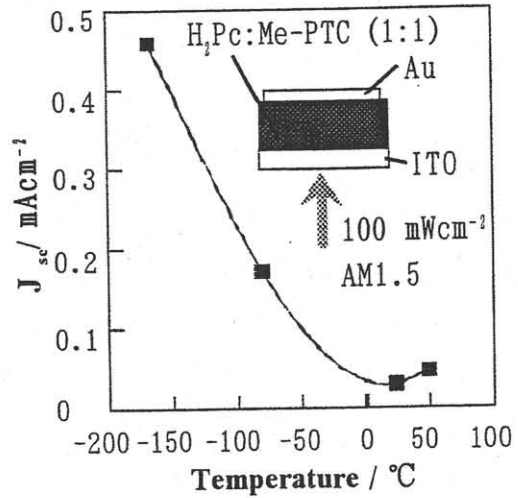


Fig. 2 Substrate temperature dependence of short-circuit photocurrent (J_{sc}) of single-layered H₂Pc:Me-PTC cell.

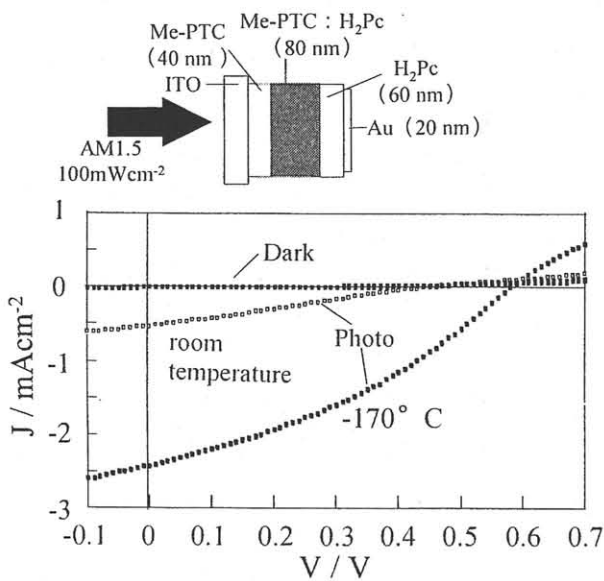


Fig. 3 Photocurrent-voltage (J-V) characteristics of three-layered cells.

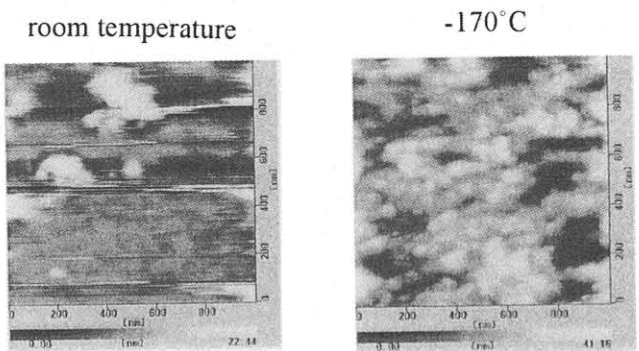


Fig. 4 AFM images (1000 x 1000 nm) of H₂Pc:Me-PTC films.

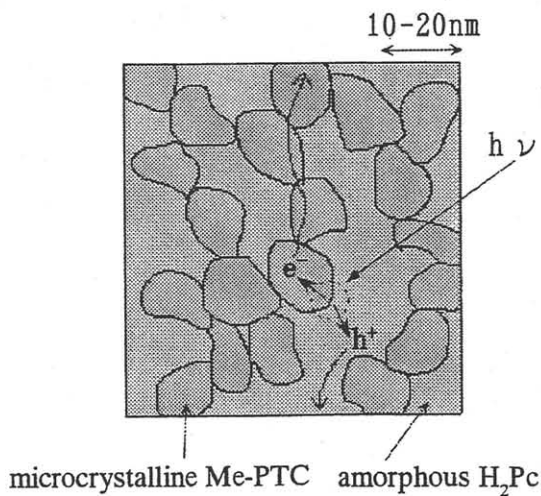


Fig. 5 Ultramicrostructure of H₂Pc:Me-PTC film co-deposited on cooled substrate.

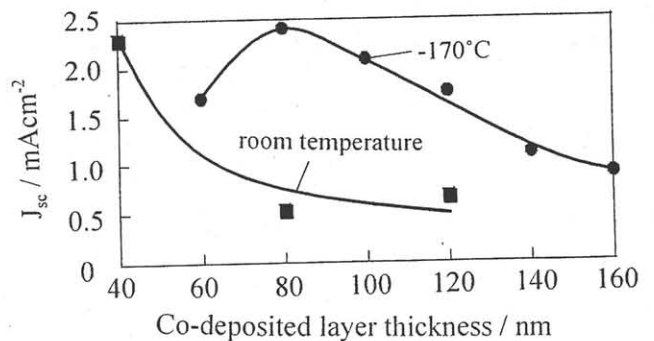


Fig. 6 Dependence of J_{sc} on the co-deposited layer thickness in three-layered cells.