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Stability Improvement of Photovoltaic Performance in Antimony Sulﬁde-Based Hybrid Solar Cells

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Sb2S3-based hybrid solar cells were prepared in the combination with electron transporting layer of TiO2 or ZnO nanoparticles in addition to poly(3-hexylthiophene)-2,5-diyl(3, 4-ethylenedioxythiophene): poly(styrene sulfonate), zinc phthalocyanine (ZnPc), or MoO3 for hole transporting layer. Photovoltaic performance and durability of the hybrid solar cells were compared each other with or without encapsulation by using glass and UV cutoff ﬁlm. Among these hybrid solar cells, it was found that a combination of glass-ITO/TiO2/Sb2S3/ZnPc/Au encapsulated with glass and UV cut ﬁlter has the highest durability with keeping the relative power conversion efﬁciency of 90% through the stability test under 1 sun at 63°C at a relative humidity of 50% for 1,500 h.

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Preparation of Sb2S3-based hybrid solar cells.—In the preparation of solar cell devices, indium-tin oxide (ITO)-coated glass substrates (Geomatec, 10 Ω cm−2) were spin coated with the above dispersed solution of TiO2 or ZnO at 4,000 rpm followed by annealing at 300°C for 10 min with a hot-plate. A ~100-nm-thick Sb2S3 was deposited by thermal evaporation of the Sb2S3 powder as the source under vacuum (5 × 10−2 Pa) onto the metal oxide layer. It was annealed at 260°C for 10 min ramping from 220°C under the reduced pressure (1 × 10−3 Pa). After cooling down to 220°C, HTL was subsequently coated onto the Sb2S3 layer as follows: 0.5 wt% P3HT in chlorobenzene was spin-coated at 4,000 rpm and 3 times diluted PEDOT:PSS solution (Clevios P) with methanol was then coated at 2,500 rpm followed by annealing at 110°C for 10 min. While ZnPc or MoO3 was deposited by thermal evaporation resulting in a 20-nm-thick layer. Thereafter Au electrode was deposited by thermal evaporation. The solar cells obtained as described above were encapsulated with glass and epoxy resin as sealant. A cut ﬁlter ( > 420 nm, Asahi Spectra Co., Ltd.) was utilized for cut off the UV region of the irradiation light.

Evaluations of photovoltaic performance.—Photovoltaic characteristics were measured under AM1.5 (100 mW cm−2) with solar simulator of HAL-320 (Asahi Spectra Co., Ltd.) in the combination with COSMO CIV-1000F applying a bias from −0.2 to 1.0 V in steps of 0.05 V and the current density was measured just after 0.5 s under biased conditions.

Results and Discussion

Photovoltaic performance of the Sb2S3-based hybrid solar cells for 1 d.—The photovoltaic performance of the Sb2S3-based hybrid solar cells with three different HTLs consisting of glass-ITO/TiO2/Sb2S3/HTL (HTL = P3HT/PEDOT: PSS, ZnPc, or MoO3)/Au were prepared and compared with each other. The current density-voltage (J-V) curves of the initial (viz., freshly prepared) photovoltaic performance (solid) and the curves after 1 d at 63°C and 50% RH in the dark (broken) are shown in Figure 1. PCEs of ca. 5% were attained with P3HT/PEDOT: PSS and ZnPc. While PCE of 0.25% was obtained when MoO3 was applied. Since relatively higher series resistance was observed in the case of MoO3, it might be estimated that the mobility in MoO3 is lower than other HTLs and resulted in low PCE. With regard to the stability, the PCE of P3HT/PEDOT: PSS decreased slightly after 1 d at 63°C and 50% RH even when there was...
no irradiation of light. This result is probably ascribed to the acidic property of the diluted PEDOT:PSS solution, which tends to lower the cell performance.25,26 On the contrary, there was no lowering the $PCE$s after 1 d at 63 °C and 50% RH in the dark both in the cases of ZnPc and MoO$_3$.

Effect of hole transporting layers (HTLs) on stability.—The durability test of the cells with UV cut filter was performed at 63 °C and 50% RH under 1 sun as shown in Figure 2. $PCE$ of P3HT/PEDOT:PSS decreased to the relative value of 10% after 100 h photo-irradiation. On the other hand, in the cases of ZnPc and MoO$_3$, $PCE$s kept constant until 300 h irradiation. However, that of MoO$_3$ started to decrease gradually after 300 h, while ZnPc indicated stable and 90% of $PCE$ was retained after 500 h. TEM-EDX observations of the cross-sectional views of glass-ITO/TiO$_2$/Sb$_2$S$_3$/MoO$_3$/Au before and after the durability test for 576 h revealed that Sb and Mo diffused opposite directions one another (Figure 3). These diffusions during the durability test might affect the lowering of $PCE$s.

Effect of electron transporting layers (ETLs) on stability.—Two ETLs were utilized and compared in terms of durability for the
Sb₂S₃-based hybrid solar cells with TiO₂ or ZnO nanoparticles as glass-ITO/TiO₂/Sb₂S₃/ZnPc/Au or glass-ITO/ZnO/Sb₂S₃/ZnPc/Au with UV cut filter at 63°C and 50% RH under 1 sun. The J-V curves of the initial photovoltaic performance and after 7 d and the time-course changes of relative PCEs through the durability test (Figure 4) indicate that almost constant stability of TiO₂-based cell and remarkable lowering of the PCE and lability of ZnO-based one. In particular, the fill factor (FF) and the series resistance (Rₛ) of glass-ITO/TiO₂/Sb₂S₃/ZnPc/Au had been improved as FF from 0.415 to 0.471, and Rₛ from 33 ohm cm² to 20 ohm cm² after 7 d, respectively. Therefore, using TiO₂ is effective to adjust the interface of the device under the conditions with UV cut filter at 63°C and 50% RH under 1 sun. On the other hand, it was found by TEM-EDX observations of the cross-sectional views of glass-ITO/TiO₂/Sb₂S₃/ZnPc/Au (Figure 5) that initial distribution of S was well-corresponding with that of Sb, however, S diffused wider than Sb and migrated toward Zn after the durability test and presumably substitution of S of Sb₂S₃ for O of ZnO might occur and result in the formation of ZnS.

Effect of UV light irradiation on stability.—In order to investigate the effect of UV light irradiation, another durability test of glass-ITO/TiO₂/Sb₂S₃/ZnPc/Au without UV cut filter was performed at 63°C and 50% RH under 1 sun. The J-V curves of the initial photovoltaic performance (solid) and after 3 d (broken) were indicated in Figure 6. The relative PCE of 90% was retained for 1,500 h with UV cut filter, however the photovoltaic ability has almost been lost without the cut filter after 72 h. Decolorization was observed after the irradiation without the UV cut filter though the decolorization did not occur with the filter. Once the TiO₂ layer was removed from the cell, described as glass-ITO/Sb₂S₃/ZnPc/Au, there was no decolorization even when without using the UV cut filter. In this context, anatase TiO₂ nanoparticles in this case act as the photocatalyst during the UV light irradiation to oxidize and decompose Sb₂S₃, which resulted in the above decolorization of the cells. TEM-EDX observations of the cross-sectional views of colored (being not decolorized) site and decolorized site (Figure 7) indicated that the existence of S and O was confirmed at the colored site though only O was confirmed (viz. Figure 7).
1,500 h. Effective suppression of the photoactivation of TiO₂ through oxidative decomposition of Sb₂S₃ by photoactivated TiO₂. S was not detected) at the decolorized site. This is also supporting the elemental maps of colored site (left) and decolorized site (right) of ITO/TiO₂/ZnPc/Au without UV cut filter after 1 d at 63°C at a relative humidity of 50% for 1,500 h.

Summary

We prepared Sb₂S₃-based solar cells with ETL of TiO₂ or ZnO in addition to HTL of P3HT/PEDOT: PSS, ZnPc, or MoO₃ and compared their photovoltaic performance in terms of durability with encapsulation by using glass and UV cutoff films. Combination with ZnO and TiO₂ has the highest durability with retaining the relative PCE of 90% under the conditions of 1 sun at 63°C at a relative humidity of 50% for 1,500 h. Effective suppression of the photoactivation of TiO₂ through UV light irradiation by using cut filter is essential to realize the long term stability and promising further extension for optimization of the photovoltaic performance.

Acknowledgments

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