Solvent Engineering of Liquid-phase Ligand Exchanged PbS Quantum Dot Inks for Infrared Photovoltaics

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Colloidal PbS quantum dots (QDs) have received significant attention as promising candidate materials toward infrared photovoltaics due to their size-dependent-bandgap and solution-processability. PbS QDs bear long alkyl chains as ligands to maintain a uniform dispersion in solution. Therefore, to construct solar cells, alkyl chains must be replaced with small ligands such as halide ions. In doing so, a liquid-phase ligand exchange process has been widely used to prepare ligand-exchanged PbS QD solution (QD ink), which enables to avoid a disruptive solid-phase ligand exchange process. So far QD inks have been synthesized using near-infrared absorbing QDs (exciton peak: ~950 nm)[1]. However, there are few reports on infrared PbS QD. In the previous paper, a mixed solution of butylamine (BA, 78°C) and a high boiling point solvent such as DMF (153°C) or DMSO (189°C) was used to prepare infrared absorbing QD ink[2]. However, it is difficult to obtain QD layers with stable film quality by using mixed solvents because of the influence of the difference in boiling points. In this study, we synthesized infrared PbS inks exhibiting the first exciton peak near 1280 nm, and investigated how the dispersion solvent affected the performance of the infrared PbS QD/ZnO solar cells (the inset figure in Fig. 1).

With reference to previous studies[2], a solution of BA or hexylamine (HA, 123°C) mixed with DMF (9:1 v/v) was used to obtain a QD ink in which the I-PbS QDs were uniformly dissolved. However, the QD film formed by the spin coating method had a rough surface and non-uniform film thickness, indicating the difficulty in controlling a mixed solvent system with large difference in boiling points. Then we focused on a BA or HA single solvent to uniformly dissolve the I-PbS QDs. By adjusting the dissolution time, we have succeeded to prepare QD inks with uniformly dissolved I-PbS QDs. From these QD inks, it was possible to deposit homogeneous solid films of QDs. After depositing the QDs on the ZnO layer, the QD layer was annealed at 80°C for 20 min to fabricate the solar cells[3]. Although homogeneous QD films were deposited using HA, the film thickness was only about 200 nm. This is probably because the high boiling point of HA caused insufficient evaporation of the solvent during spin coating, resulting in less QD ink remaining on the ZnO layer. On the other hand, when BA was used, a QD layer of about 500 nm could be fabricated with a single spin-coat deposition. The corresponding solar cell exhibited a power conversion efficiency of 8% (J_{sc}=31.1mA/cm^2, V_{oc}=0.45V, FF=0.568). In the case of QD inks prepared with a single amine solvent, homogeneous QD films could be formed with high reproducibility. The formation of high-quality QD films and further improvement of solar cell efficiency can be expected by exploring the preparation method and annealing conditions of QD inks.

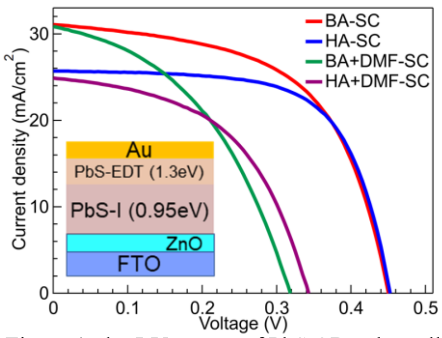

Figure 1. the J-V curves of PbS QD solar cells fabricated with different solvents.

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