2015

Trap-assisted transport and non-uniform charge distribution in sulphur-rich PbS colloidal quantum dot-based solar cells with selective contacts

Victor Malgras  
*University of Wollongong, vm961@uowmail.edu.au*

Guanran Zhang  
*University of Wollongong, gz943@uowmail.edu.au*

Andrew Nattestad  
*University of Wollongong, anattest@uow.edu.au*

Tracey M. Clarke  
*University of Wollongong, tclarke@uow.edu.au*

Attila J. Mozer  
*University of Wollongong, attila@uow.edu.au*

*See next page for additional authors*
Trap-assisted transport and non-uniform charge distribution in sulphur-rich PbS colloidal quantum dot-based solar cells with selective contacts

Abstract
This study reports evidences of dispersive transport in planar PbS colloidal quantum dots heterojunction-based devices as well as the effect of incorporating a MoO3 hole selective layer on the charge extraction behavior. Steady state and transient characterization techniques are employed to determine the complex recombination processes involved in such devices. The addition of a selective contact drastically improves the device efficiency up to 3.15 % (especially through the photocurrent and series resistance) and extends the overall charge lifetime by suppressing the main first-order recombination pathway observed in device without MoO3. The lifetime and mobility calculated for our sulphur-rich PbS-based devices are similar to previously reported values in lead-rich quantum dots-based solar cells. Nevertheless, strong Shockley-Read-Hall mechanisms appears to keep restricting charge transport, the equilibrium voltage taking more than 1 ms to be established.

Keywords
contacts, cells, solar, dot, quantum, colloidal, pbs, rich, sulphur, distribution, selective, charge, trap, uniform, non, transport, assisted

Disciplines
Engineering | Physical Sciences and Mathematics

Publication Details

Authors
Victor Malgras, Guanran Zhang, Andrew Nattestad, Tracey M. Clarke, Attila J. Mozer, Yusuke Yamauchi, and Jung Ho Kim

This journal article is available at Research Online: http://ro.uow.edu.au/aiimpapers/1747
SUPPORTING INFORMATION

Trap-Assisted Transport and Non-Uniform Charge Distribution in Sulphur-Rich PbS Colloidal Quantum Dot-based Solar Cells with Selective Contacts

Victor Malgras\textsuperscript{1,2}, Guanran Zhang\textsuperscript{3}, Andrew Nattestad\textsuperscript{3}, Tracey Clarke\textsuperscript{3}, Attila Mozer\textsuperscript{3}, Yusuke Yamauchi\textsuperscript{2}, Jung Ho Kim\textsuperscript{*1}

1. Institute for Superconducting and Electronic Materials (ISEM), Australian Institute for Innovative Materials, University of Wollongong, North Wollongong, NSW 2500, Australia
2. World Premier International (WPI) Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044 (Japan)
3. Intelligent Polymer Research Institute (IPRI), ARC Centre of Excellence for Electromaterials Science, University of Wollongong, North Wollongong, NSW 2500, Australia

Corresponding authors:

Yusuke Yamauchi: Yamauchi.Yusuke@nims.go.jp

Jung Ho Kim: jhk@uow.edu.au
Figure S1. Fill factor (FF) as a function of the light intensity.
Fitting of dispersive exponential

The fitting functions are:

- Geminate: $n(t) = n_0 e^{-\left(\frac{t}{\tau}\right)^{\alpha}}$
- Non-geminate: $n(t) = \frac{n_0}{1+n_0(\beta t)^{\alpha}}$

where $n_0$ is the initial concentration of free photogenerated charges and $\tau$, $\beta$, and $\alpha$ are the geminate recombination characteristic time, the non-geminate recombination coefficient and the dispersive factor. These two functions are obtained from solving the continuity reaction: $rac{dn}{dt} = \frac{dp}{dt} = -\beta n(t)p(t)$. In the case of geminate recombination, the equation can be simplified as the density of one charge carrier can be considered constant (e.g., $n(t) = n$ and $\beta n = \frac{1}{t}$), thus leading to a solution following a monoexponential decay. Typical non-geminate, assuming charge quasi-neutrality ($n(t) \approx p(t)$), results in a hyperbolic solution. The dispersive factor arises from taking into account the dispersive media where mobile charges have a high probability of encountering deep traps with a slow release time which compete with other characteristic transport times (diffusion, injection/transfer). For a non-dispersive random walk, the number of sites visited by the mobile charges is linear ($S(t) \propto t$). On the other, considering a dispersion in the spatial separation between the localized states and a dispersion in the potential barriers between these sites, Scher and Montroll demonstrated that the waiting time distribution function could be well-represented by a power law $S(t) \propto t^\alpha$ where $\alpha = T/T_0$ and $T_0$ is the characteristic temperature of the distribution.\(^1\,^2\) In the limit where $T_0 = T$ (or $\alpha = 1$), the distribution is inexistent and the solution is non-dispersive.

**Figure S2.** Experimental time-resolved charge density decay of the pixel without selective contact along with various fitting functions.
Figure S3. a) Comparison between normalized charge density in function of the light intensity, highlighting the fact that the decays are similar up to 20 μs. b) Current extraction curves for the device without selective contact under 40 μJ/cm² at various delay time (from 200 ns to 100 μs) highlighting the linear nature of the decay at fast delays.
Figure S4. Typical TPV signal fitted with various exponential functions. Inset: high resolution of the early stage of the decay, highlighting the difference between the double exponential and the stretched exponential.

References
