Compositional engineering of highly emissive and widely tunable I-III-VI based quantum dots (QDs) for photovoltaic applications

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Abstract

In quantum dot sensitized solar cells (QDSSCs), replacing heavy metal based binary II-VI QDs such as Pb (S, Se or Te) or Cd (Se or Te) with I-III-VI counterparts is of high interest for environmental considerations while ensuring optimal device performance [1]. In this context, it becomes imperative to extensively investigate the chemical pathways, composition and structure of Cu-In-Zn-Se QDs to achieve competitive photovoltaic characteristics [2].

The present study reports a robust, quick and inexpensive aqueous phase-based approach to produce Cd-free, bright, and highly emissive core/shell alloyed Zn-Cu-In-Se/ZnS QDs of small size (~ 2.20 nm). The prepared core/shell structured nanocrystals show wide tunable photoluminescence (PL) emission from 618 to 765 nm by varying feed molar ratio of precursors or the capping ligand. Being in line with the systematic PL tunability, their energy gap could be enlarged from 1.73 to 2.12 eV. By leveraging the off-stochiometric effect on PL, QDs with Zn:Cu molar ratio of 1:2 exhibited the highest PL quantum yield of 54%. Further, the QDs were characterized with UV-visible, PL spectroscopy, DLS, FT-IR in combination with XRD and TEM analysis of the purified samples to investigate the role of cation deficiency on their structure. Finally, QDs-sensitized electrodes were fabricated via magnetron sputtering and spin coating techniques to effectively immobilize Cu-In-Zn-Se/ZnS QDs on rutile TiO_2 film electrodes. Preliminary attempts to perform photoelectrochemical measurements on tailored photoanodes were conducted. The presence of a small and tight semi-circular shape of Nyquist plot in comparison to unsensitized rutile TiO_2 film electrodes indicates an improved charge transfer with minimal interfacial effects. These results demonstrate the high potential of Cu-In-Zn-Se/ZnS QDs sensitized TiO_2 film electrodes as potential photoanode in QDSSCs.

Keywords: Quantum dots, Aqueous synthesis, Surface functionalization, Optoelectronic properties, Photoanode, Photovoltaics

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