Colloidal Quantum Dot Photovoltaics

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In the last decade, colloidal quantum dots (CQDs) have aroused great interest due to their promising applications in optoelectronic devices such as light emitting diodes, field effect transistors, photo detectors, and solar cells.[1] In recent years, CQD based photovoltaics received attention due to rapid improvement in their efficiencies, as well as their promise for low-cost, high-efficiency photovoltaics deriving from solution-phase processibility and quantum-size-effect tunability for tandem and multijunction devices.[2,3]

Since the first report of IR-capturing CQD solar cells in 2005,[4] the device structure has quickly evolved from Schottky[5] into the depleted-heterojunction device combines a wide-bandgap n-type oxide with lower-bandgap p-type CQDs.[6] To match the energy level of the CQDs layer, band-edge engineering of the wide bandgap metal oxide layer was carried out.[7] Meanwhile, the doping density of the wide bandgap oxide was increased to extend the depletion region of the CQD film, which helped to reduce the series resistance of the device.[8]

To further extend the depletion region and thus aid in charge carrier drift, bulk heterojunction[9] and ordered nanopillar structures[10] were developed. A quantum funnel device – built using multiple different-bandgap CQDs - was explored to create an inner electric field to enhance carrier extraction from the quasi-neutral region on the back side of the device.[11]

To fully utilize the quantum-size-effect tuning character and increase the light harvesting of the broadband solar spectrum, CQD-based tandem solar cells were developed.[12] The use of a graded recombination layer can be further optimized to reduce barriers to charge transport inside the intervening layer that connects the junctions of a tandem cell.[13]

Recently, quantum junction solar cells have been reported. These rely on a junction between p-type and n-type CQDs (Fig. 1).[14] Simulations indicate that these inherently band-aligned homojunction solar cells have the potential to reach high efficiencies even when the bandgap is widely tuned. The n-type CQDs film was prepared by using an all-inorganic strategy relying on halide atomic ligands. The n-type films were fabricated in an inert atmosphere. The doping density of the film was modulated by changing the halide ligands and the film fabrication process.[15] A charge-orbital balance theory was developed to explain the doping type and density of the CQDs film.[16]

On the materials processing front, the field recently saw the development of a solution-phase quantum dot passivation strategy that achieves better charge-balance and thus lower trap state density.[17] Improved passivation in the solution phase was found to be critical to reduce midgap traps, confirmed by trap spectroscopy achieved via photovoltage transient decay. The hybrid passivation strategy, which combined solution-phase cadmium chloride treatment and solid-state thiol ligand exchange, led to a record certified efficiency of 7%.

Solution-phase halide passivation was also explored for all-inorganic homojunction devices. We found that the halide passivation in the solution phase is more efficient than solid state treatment alone.[18,19] The scheme of the solution halide ligand exchange process is shown in Fig. 1. By reducing midgap traps, a record carriers mobility of 0.24 cm^2 V^{-1}s^{-1} for PbS CQDs was achieved, and the performance of all-inorganic homojunction device was further improved.

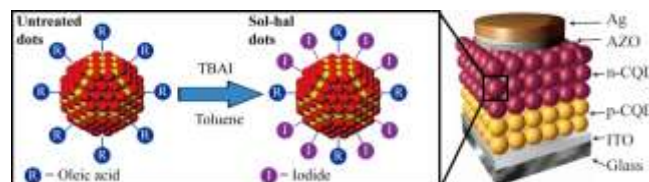**Fig. 1** The ligand exchange procedure for the solution phase halide passivation.

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