Beyond 4% photo conversion efficiency achieved by low temperature phase selective solvothermally synthesized CZTS (Cu_2ZnSnS_4) quantum dot solar cell

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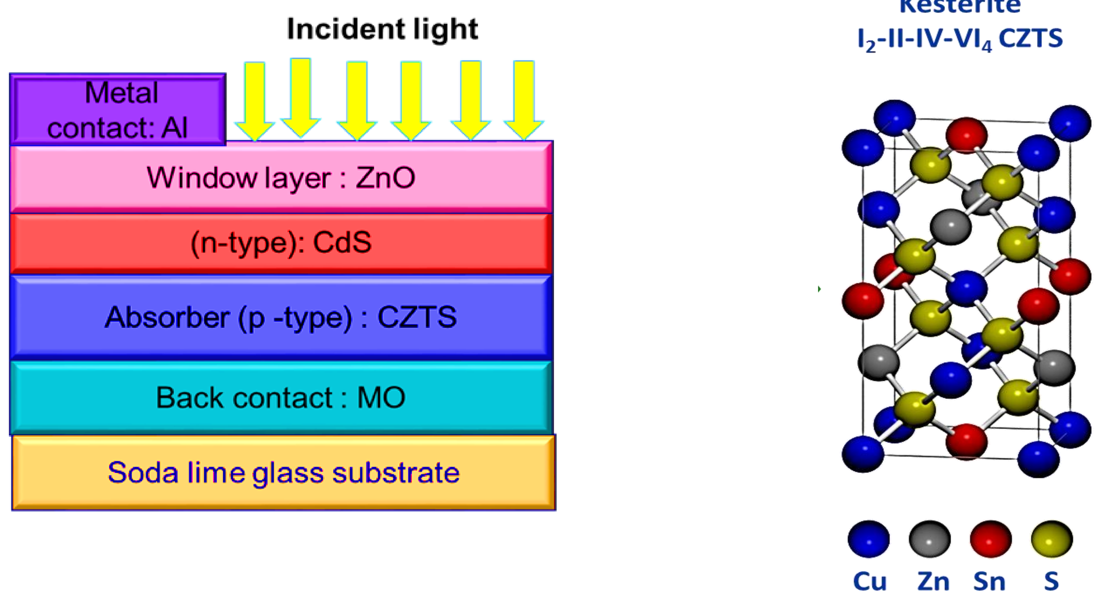
Abstract: CZTS (Cu_2ZnSnS_4) nanoparticle/quantum dot ink prepared by solution based method offers an alternative effective route for the fabrication of earth abundant CZTS photovoltaic absorber layer. Herein, we report an efficient kesterite CZTS solar cell with device configuration of SLG/Mo/CZTS QDs/CdS/ZnO/Al beyond 4% photo conversion efficiency (PCE), fabricated by a modest solution processed non-toxic, environment friendly and low cost method with absorber thickness less than 1µm. The fabricated CZTS quantum dot absorber based photovoltaic device showed active area PCE of 4.26% under AM1.5 illuminations with current density (J_{sc}) of 18.2 mA/cm².

Introduction: CZTS Photovoltaic cell

- Cu_2ZnSnS_4 (CZTS) similar to CIGS structure (I_2-II-IV-VI_4)
- earth abundant, nontoxic material nature
- high absorption coefficient (~$10^4 cm^{-1}$)
- direct band gap in the optimal range (1-1.65) eV

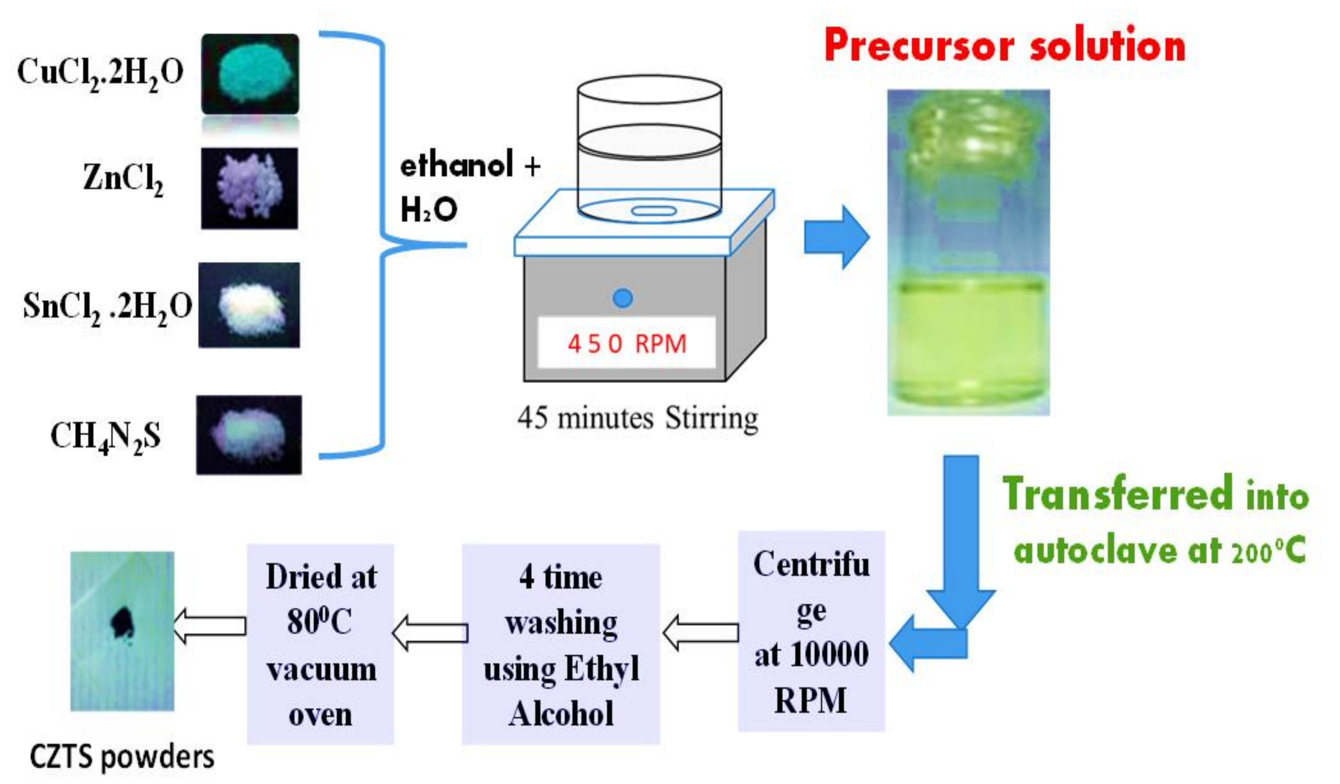


- Shockley Queisser (SQ) photon balance limit: *PCE* for CZTS thin film PVC is *32.2%* theoretically, but practically *12.6%* is achieved.

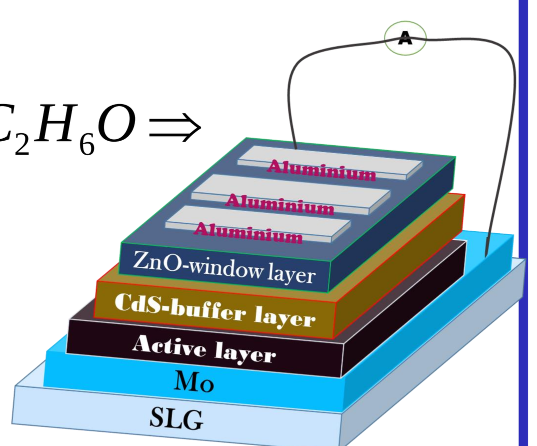
Use the UV range of solar spectrum (high energy photons)

- CZTS quantum dots offers: tuneable band gap, efficient optical absorption, multiple e-h pair generation capability, hot-electron extraction (ideal *PCE ~ 60%* in QD PVC)

Experimental



$$2CuCl_2 + ZnCl_2 + SnCl_2 + 8H_2O + 2C_2H_6O \Rightarrow Cu_2ZnSnS_4 + 8NH_4Cl + 8CO_2 + 6H_2$$



- CZTS QDs dispersed in DMF (~10mg/mL) spin coated on Mo coated SLG substrate.
- CdS buffer layer deposited by chemical bath deposition technique ($CdSO_4$ (2mM) and ammonium (1.5M)).
- Window layer (ZnO) deposited by spin coating technique, prepared by solution method (zinc acetate dihydrate (0.1M) and KOH (1M) in methanol solvent).

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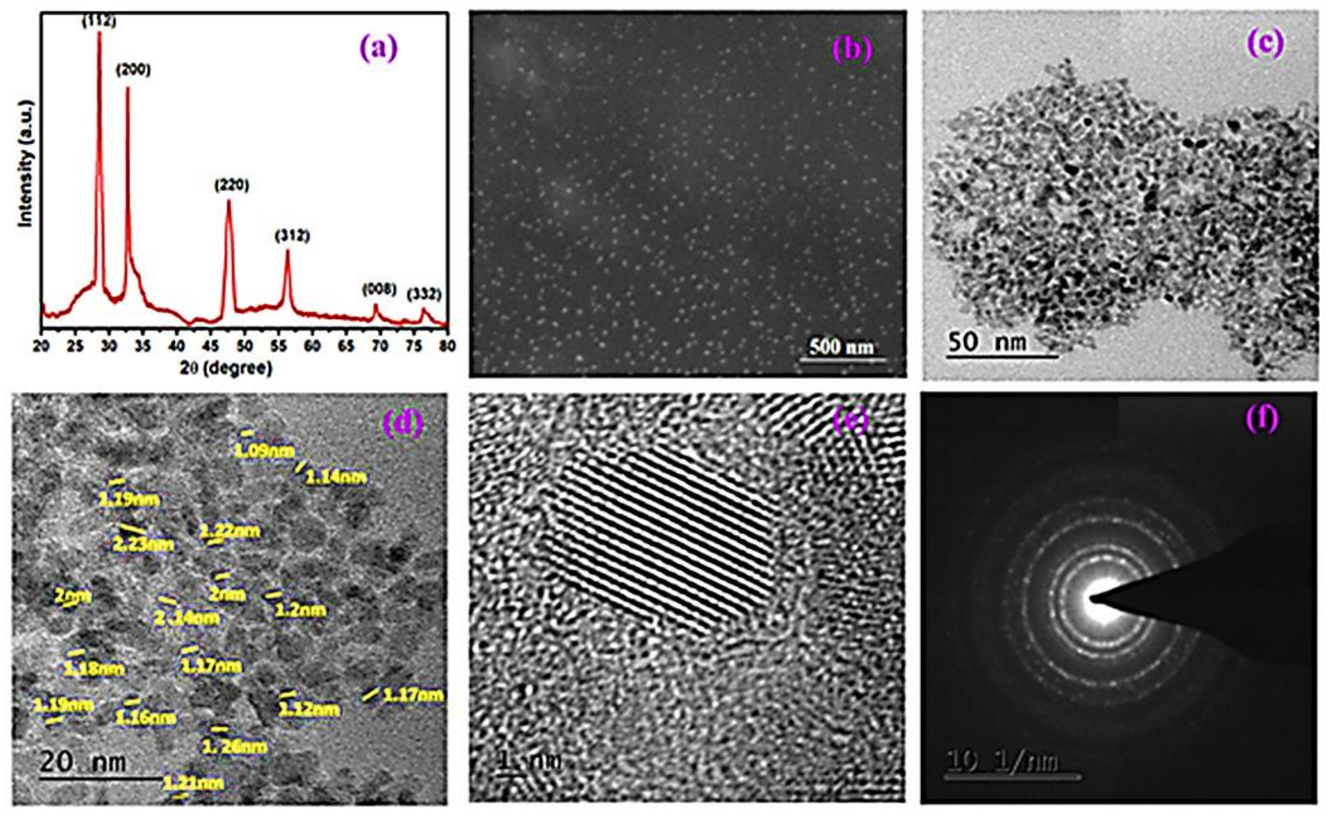
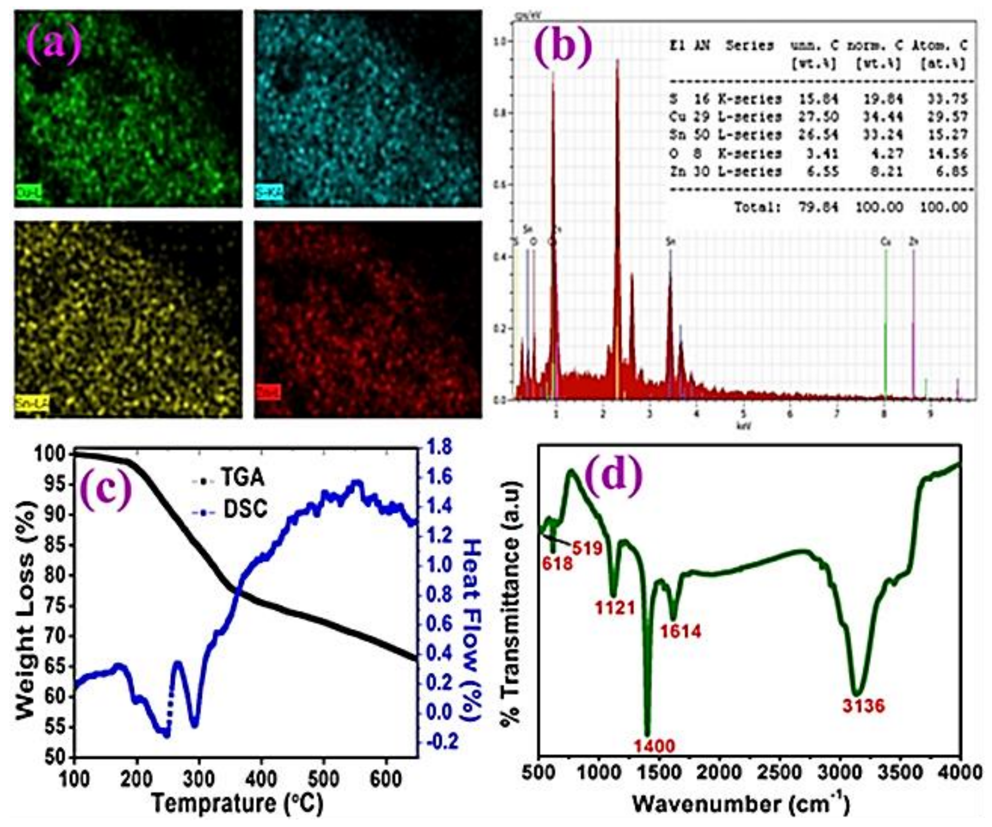
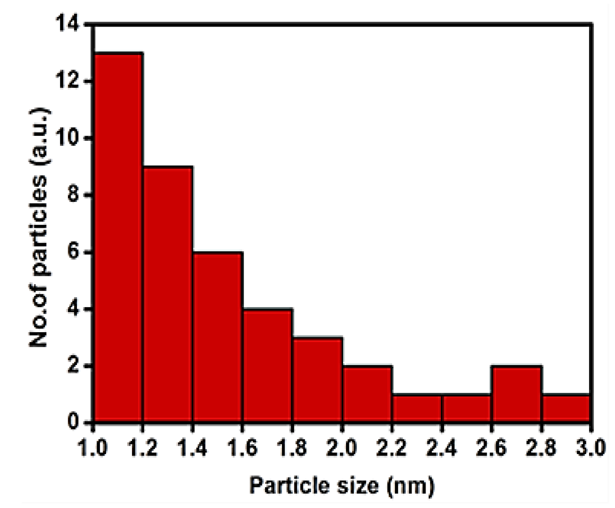
Results and discussions

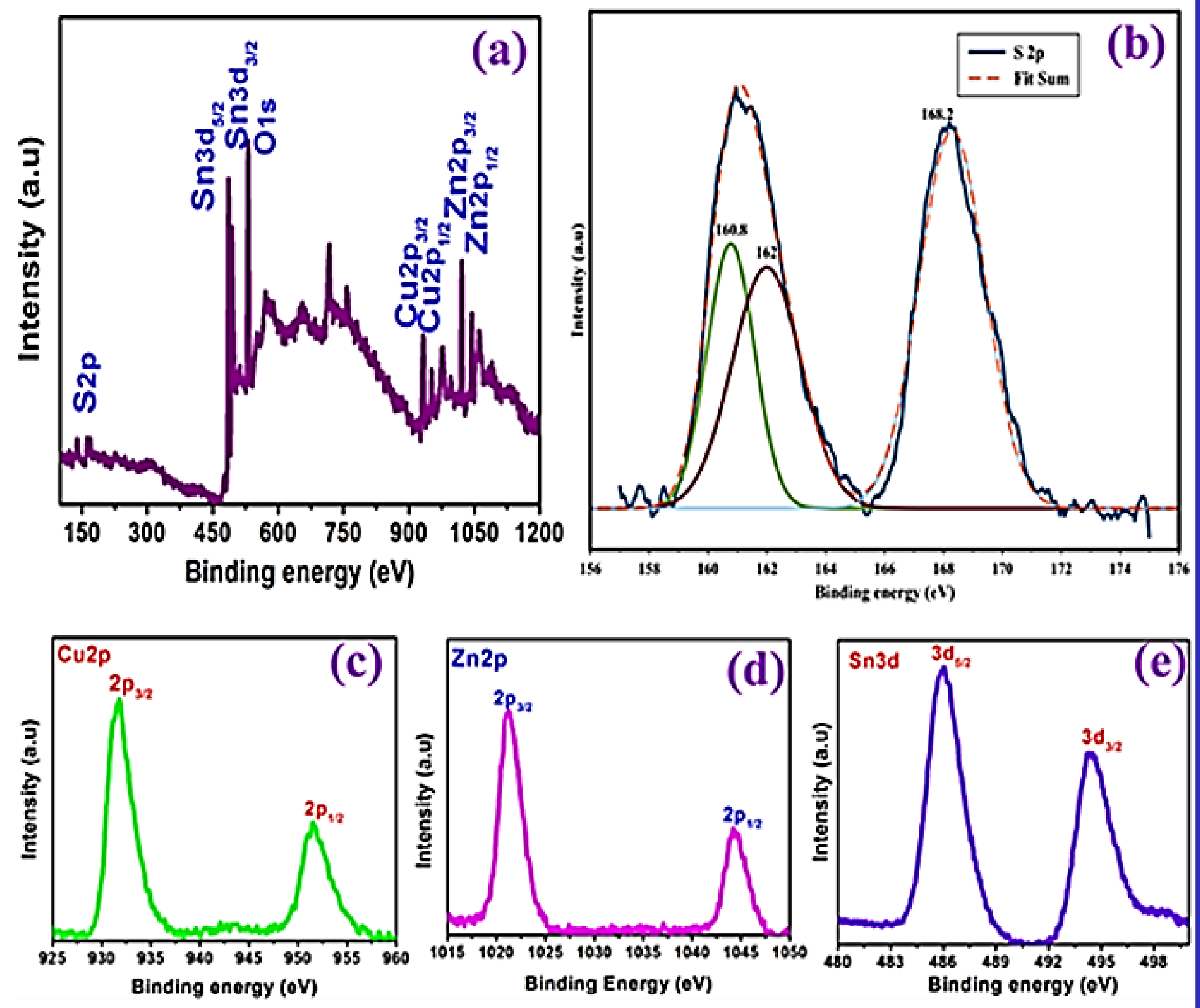
Structural, morphological, compositional Analysis



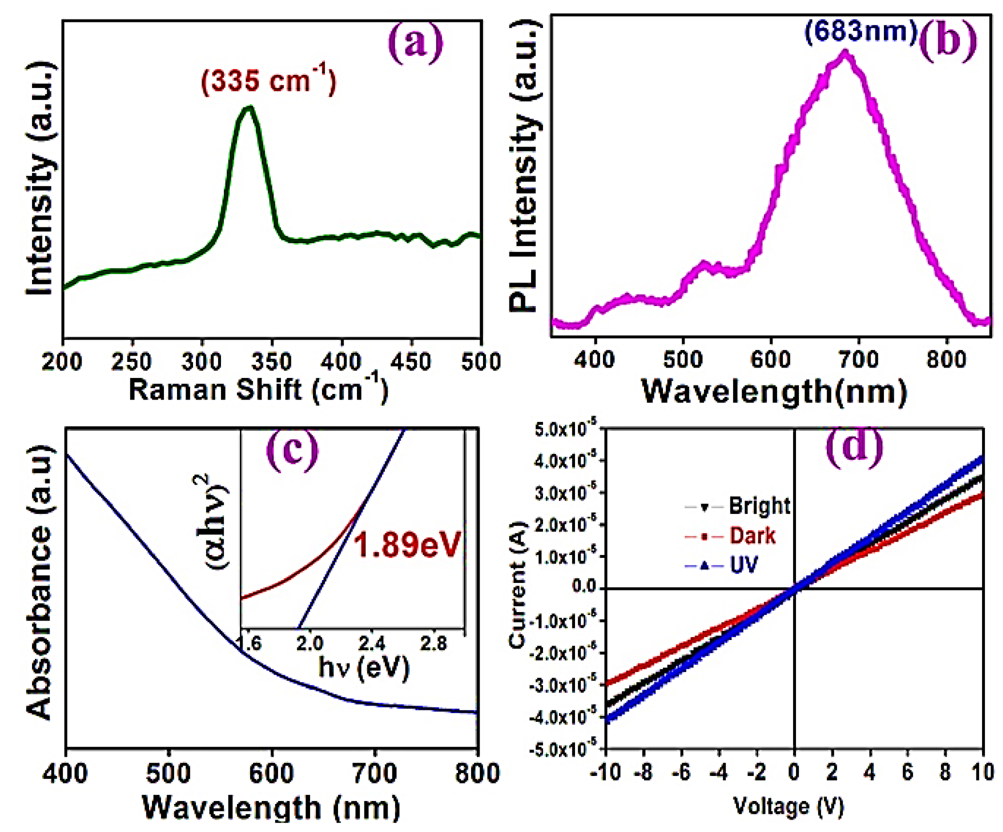


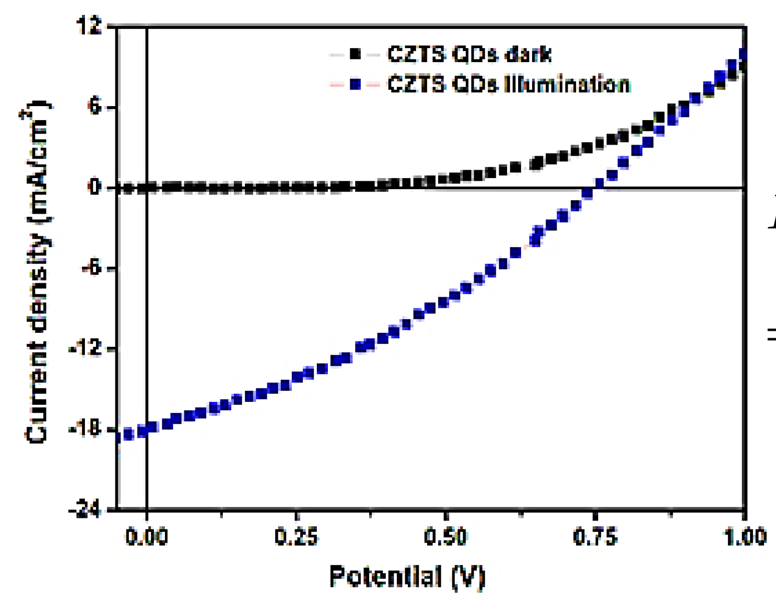


- from XRD, CZTS in kesterite phase (JCPDS card no-260575)
- average crystallite size was found to be 2.3 nm by using Scherer's equation.
- particle size ranges from 1-3 nm
- significant weight loss (~ 20wt. %) T=200 °C (thiourea decomposition temperature)
- sharp dip peak near 230°C in DSC graph (decomposition of excess amount of thiourea)
- peaks at 1614, 1400, 1121, 618 and 519 cm^{-1} are attributed to the metal-thiourea intermediate complexes.
- from core level XPS, formation of Cu_2ZnSnS_4 compound is confirmed.



Optical, electrical and device characteristic Analysis





$$E_{total} = E_g(bulk) + E_g(confinement) + E_g(exciton)$$

$$= 1.00 - 1.65eV + \frac{\hbar^2\pi^2}{2\mu d^2} - \frac{1}{\varepsilon_r^2}\frac{\mu}{m_e}R_y$$

$$E_g(confinement) = \frac{\hbar^2\pi^2}{2\mu d^2}$$

$$FF = \frac{I_{MPP}\cdot V_{MPP}}{I_{SC}\cdot V_{OC}} \quad \eta = \frac{V_{oc}J_{sc}FF}{P_{in}}$$

Conclusion

- Phase pure kesterite CZTS quantum dots of controable size (~1-3 nm) are successfully prepared by a facile, environment friendly solvothermal synthetic approach.
- The comparative higher band gap energy than bulk CZTS gives the confirmation of quantum confinement of synthesized CZTS QDs.
- From photo response analysis, it confirms that the obtained QDs can utilize hot photo-generated carriers to produce higher photocurrents.
- The fabricated solar cell by using prepared CZTS QDs as absorber, the device shows an V_{oc} *of 0.73V,* J_{sc} of *18.2 mA/cm²* and *FF* of *24.8%* with *PCE* of *4.26%.*

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