

Supporting Information

Comprehensive Physicochemical and Photovoltaic analysis of Different Zn Substitutes (Mn, Mg, Fe, Ni, Co, Ba, Sr) in CZTS-inspired Thin Films Solar Cells

Stener Lie^{a,b}, Maxim Guc^c, Venky Tunuguntla^d, Victor Izquierdo-Roca^c, Susanne Siebentritt^d, and Lydia Helena Wong^{a,b}

^aSchool of Materials Science & Engineering, Nanyang Technological University, 50 Nanyang Ave, 639798, Singapore.

^bSingapore-HUJ Alliance for Research and Enterprise (SHARE), Nanomaterials for Energy and Energy-Water Nexus (NEW), Campus for Research Excellence and Technological Enterprise (CREATE), Singapore, 138602 Singapore

^bCatalonia Institute for Energy Research (IREC), Jardins de les Dones de Negre, 1, 2^a pl., 08930 Sant Adrià de Besòs, Barcelona, Spain

^cLaboratory for Photovoltaics, University of Luxembourg, 42, rue du Brill, 4422 Belvaux, Luxembourg.

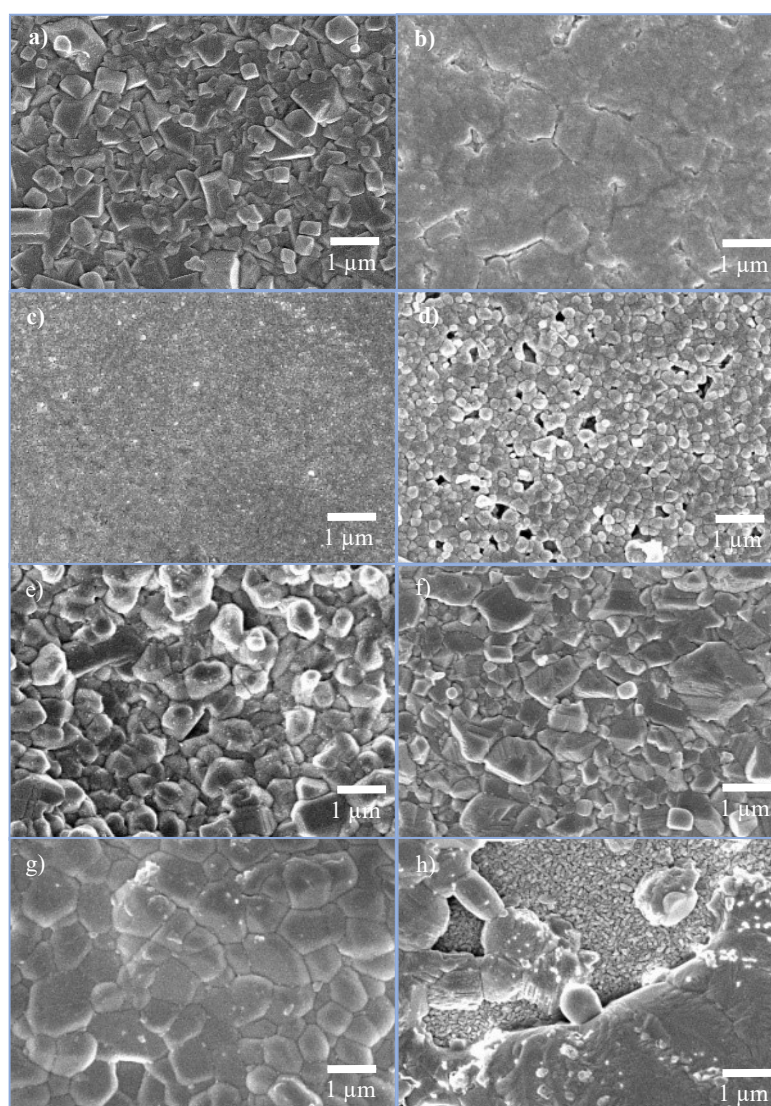


Figure S1 Plan-view SEM images of CZTS films where a) CZTS, b) CMnTS, c) Mg + CTS, d) Ni + CTS, e) CFeTS, f) CCoTS, g) CBaTS and h) CSrTS.

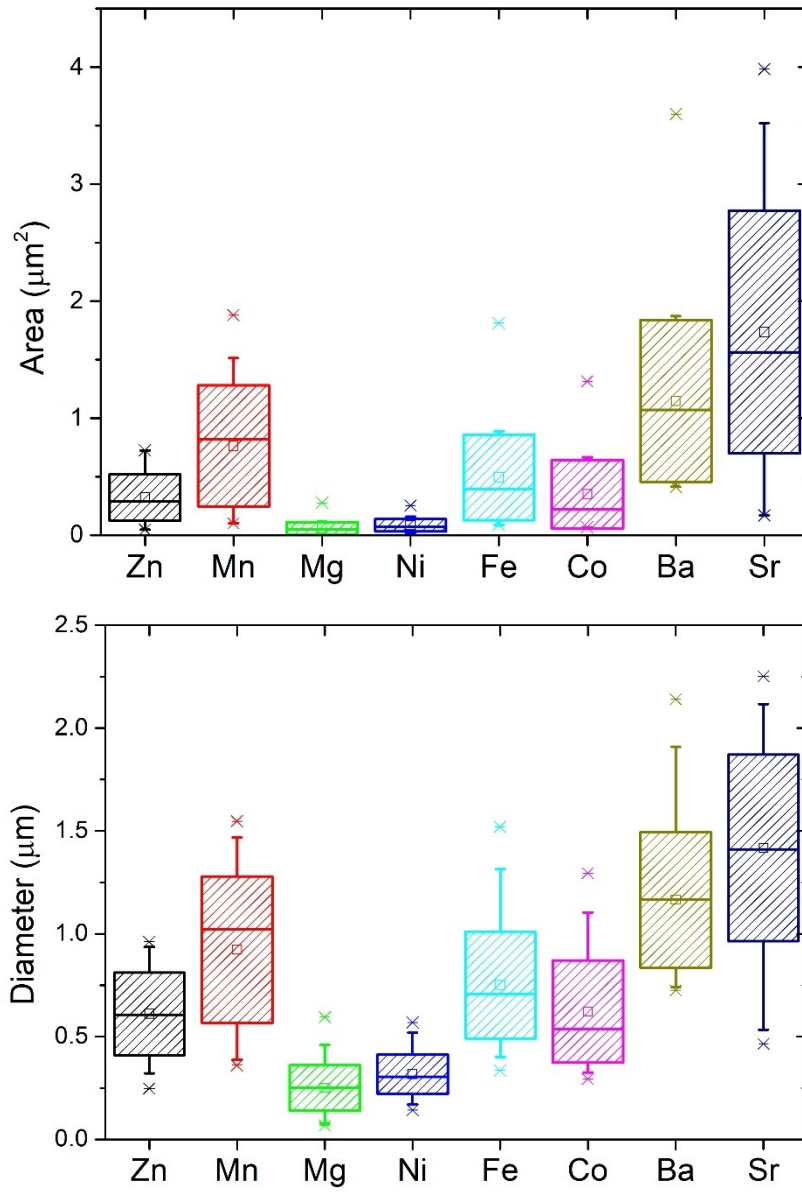


Figure S2 Grain size distribution for CXTS thin films based on area (top) and diameter (bottom)

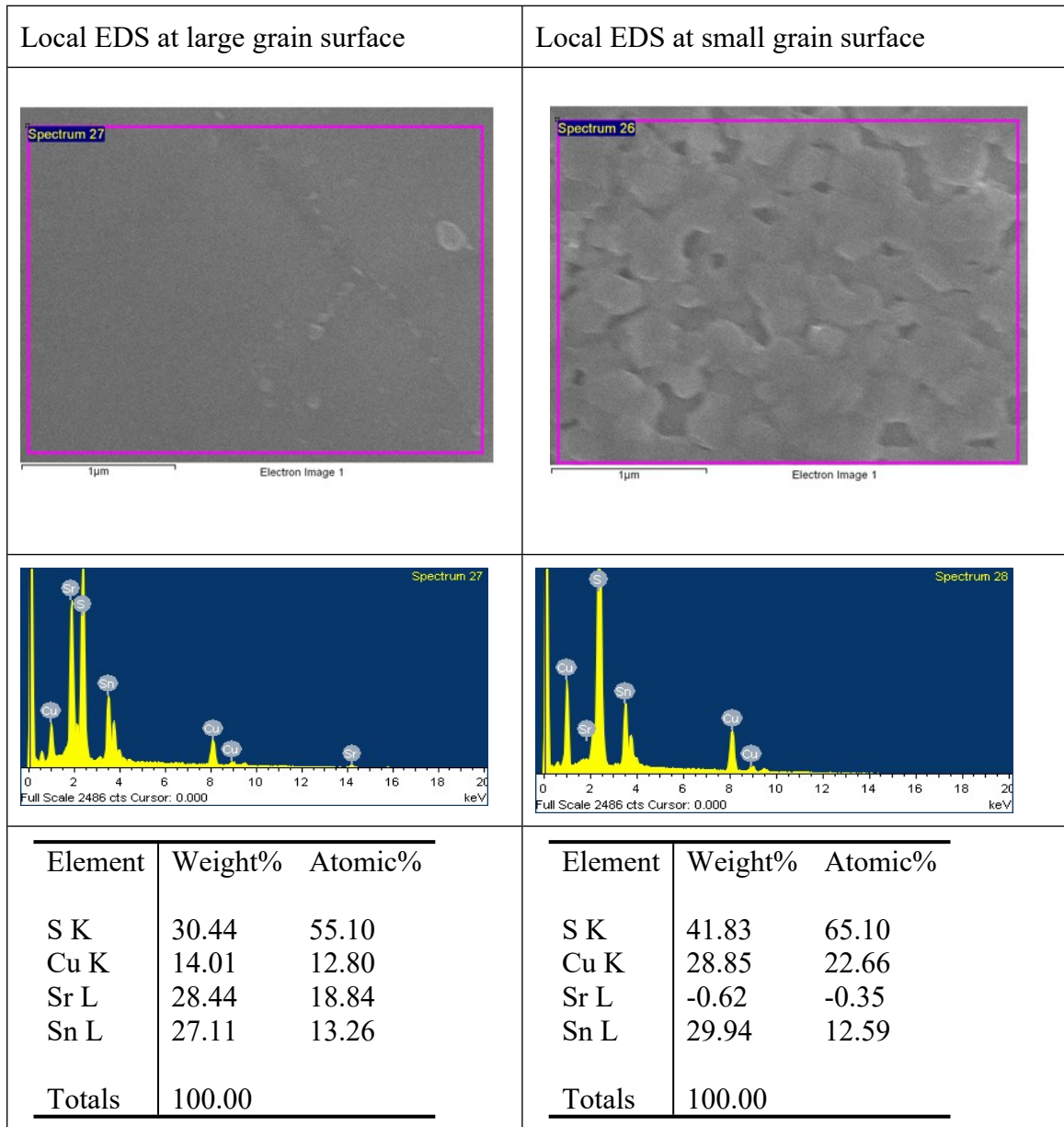
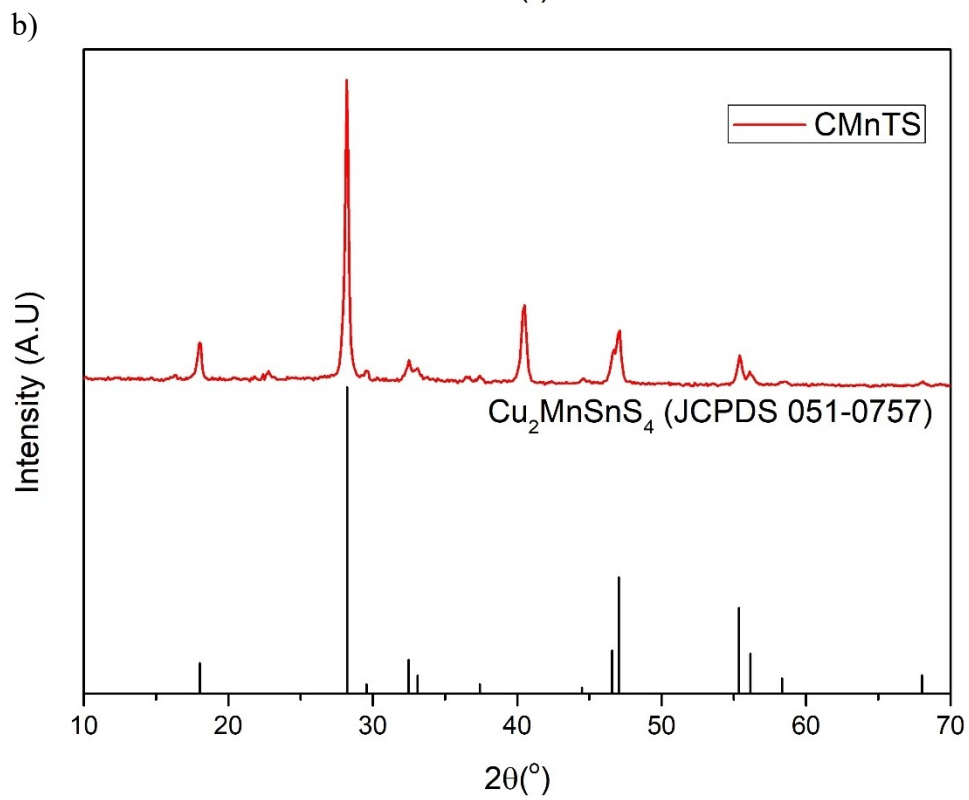
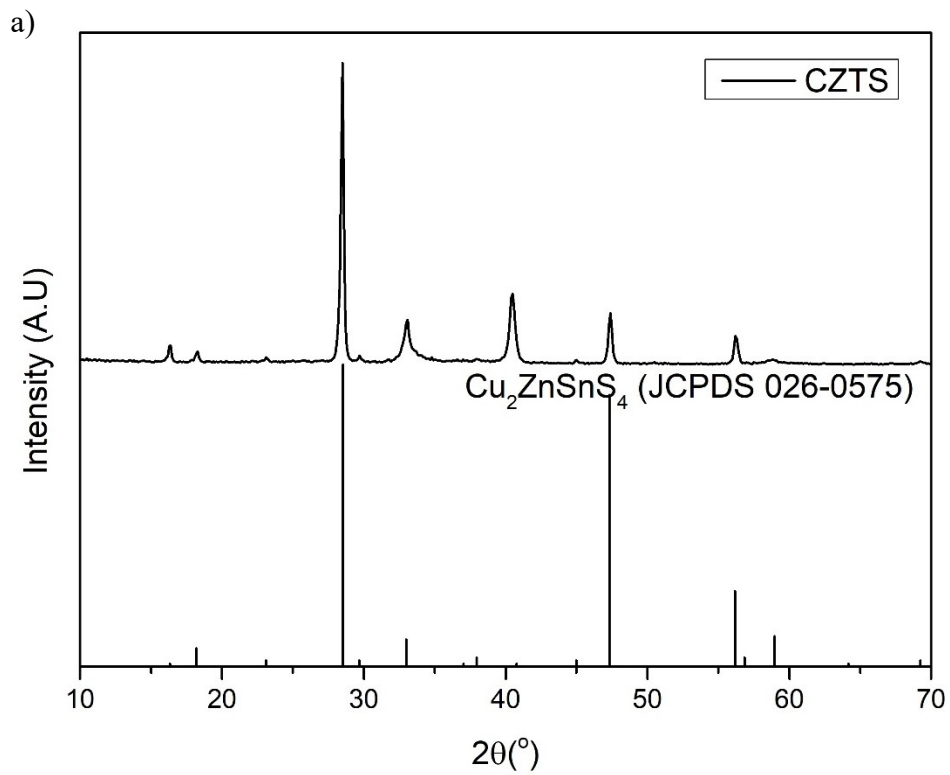
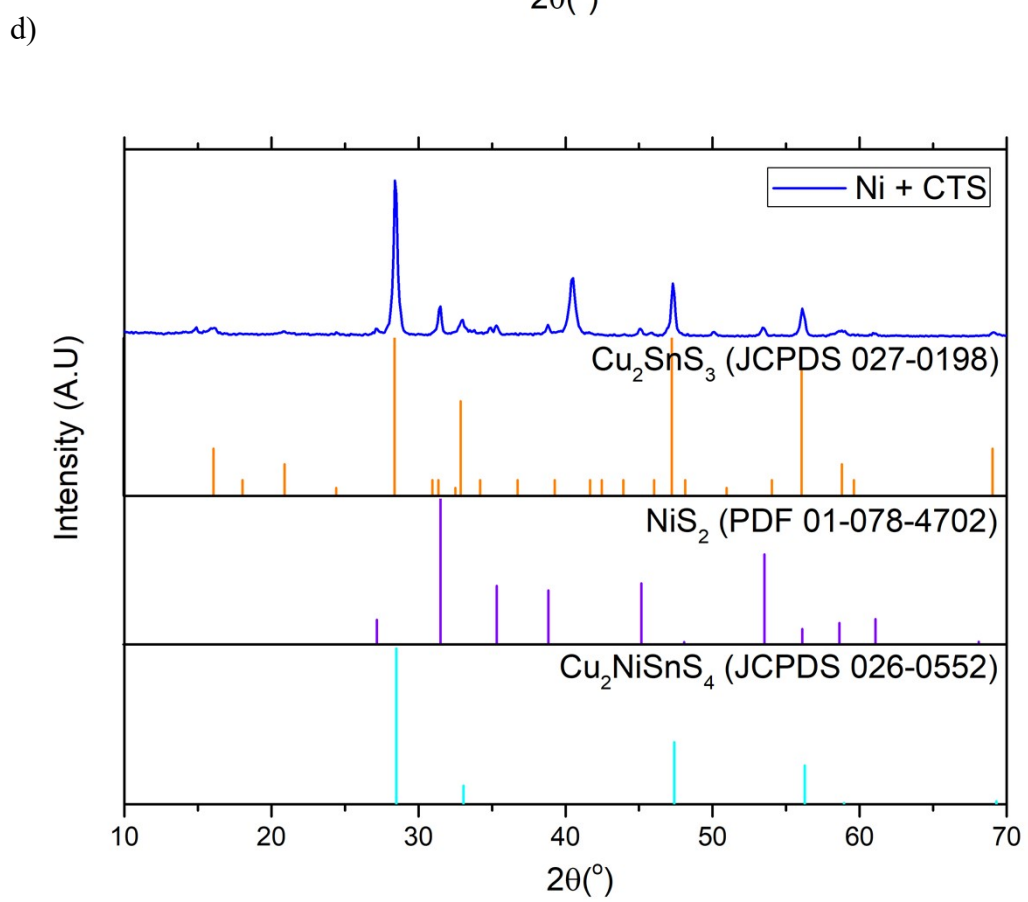
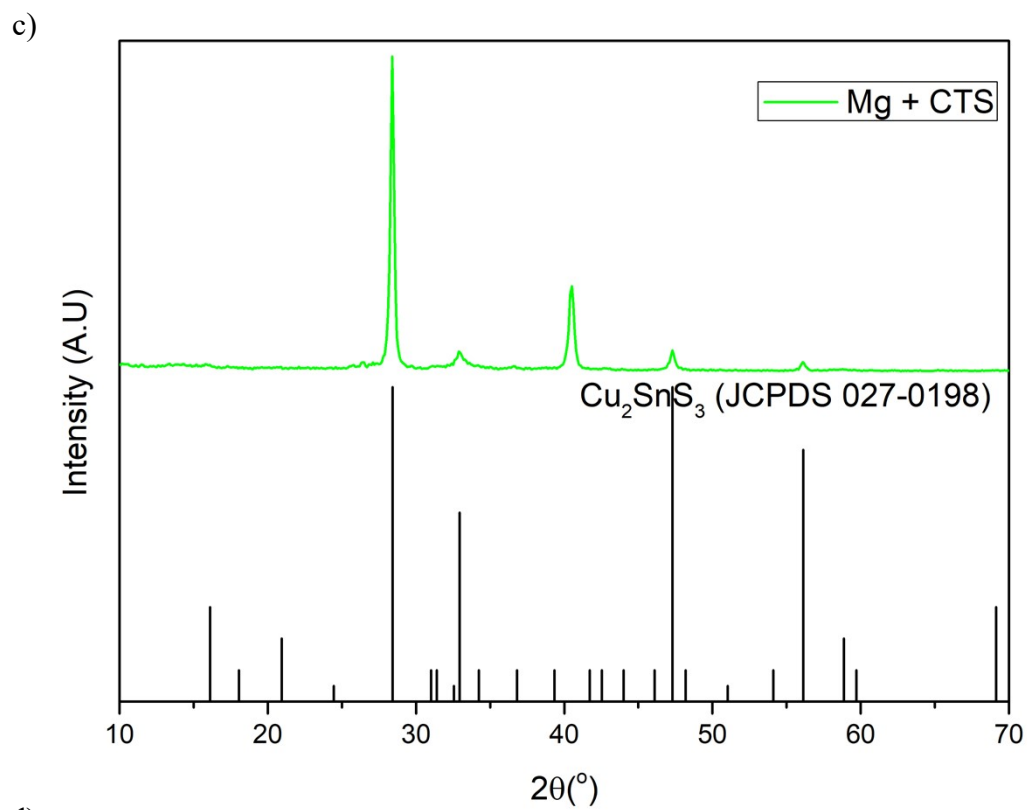
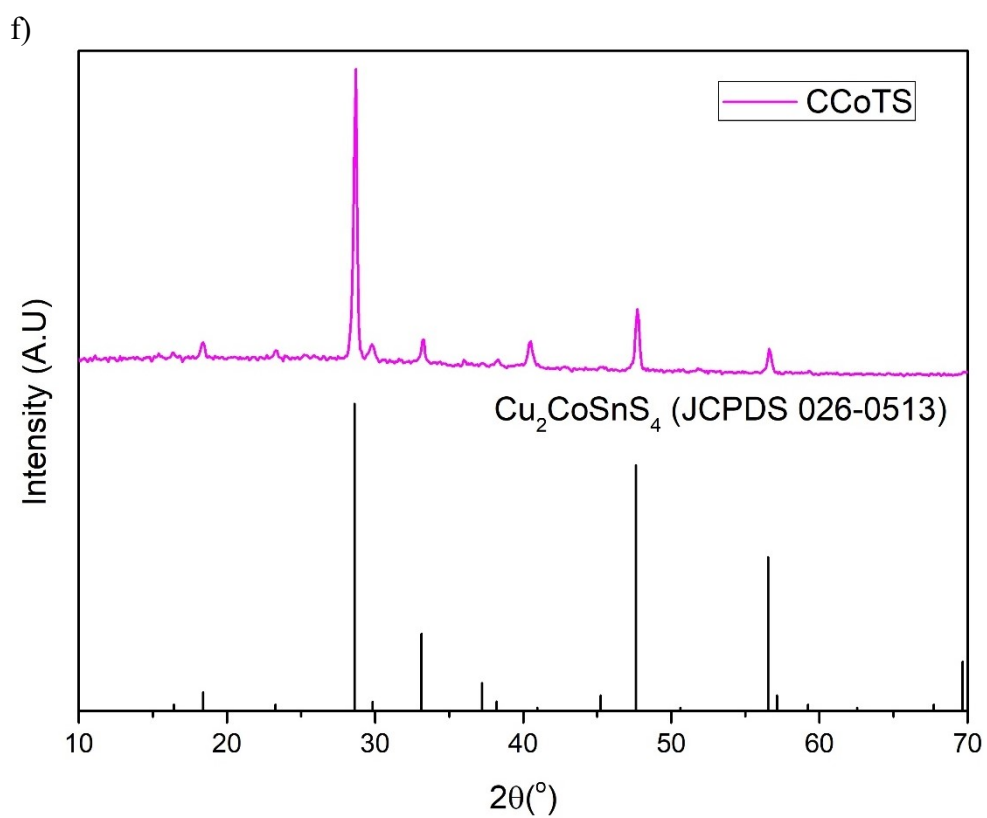
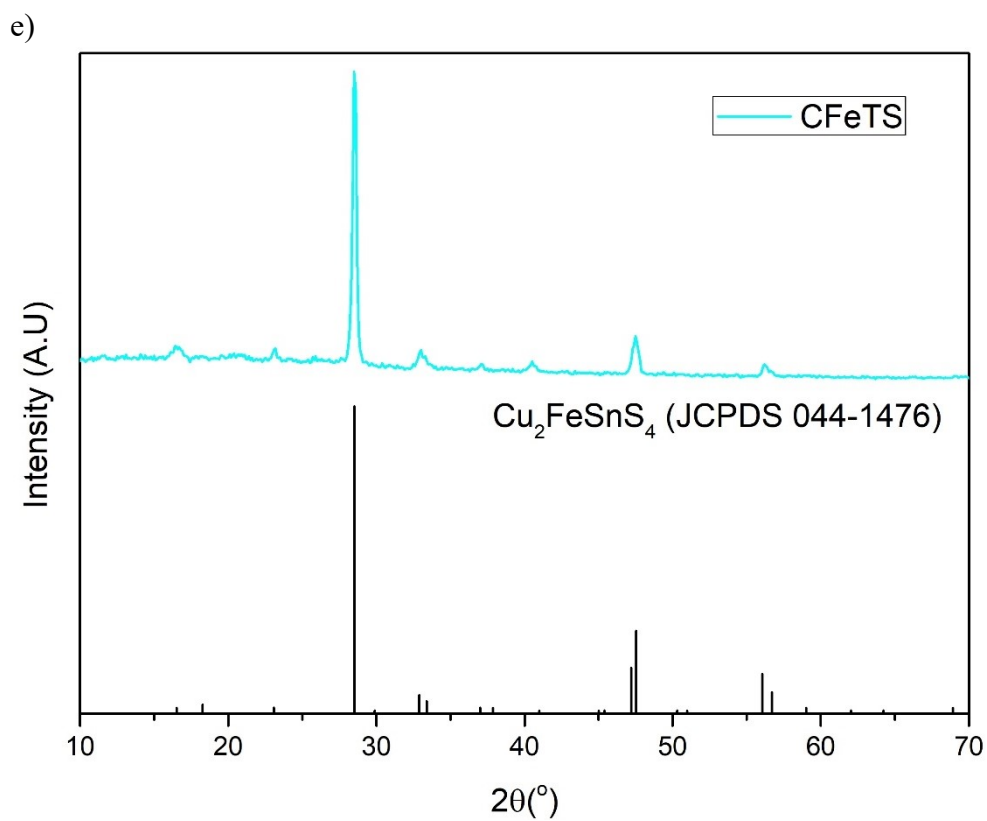


Figure S3 Local EDS measurement for CSrTS at large grains area (left) and small grains area (right)







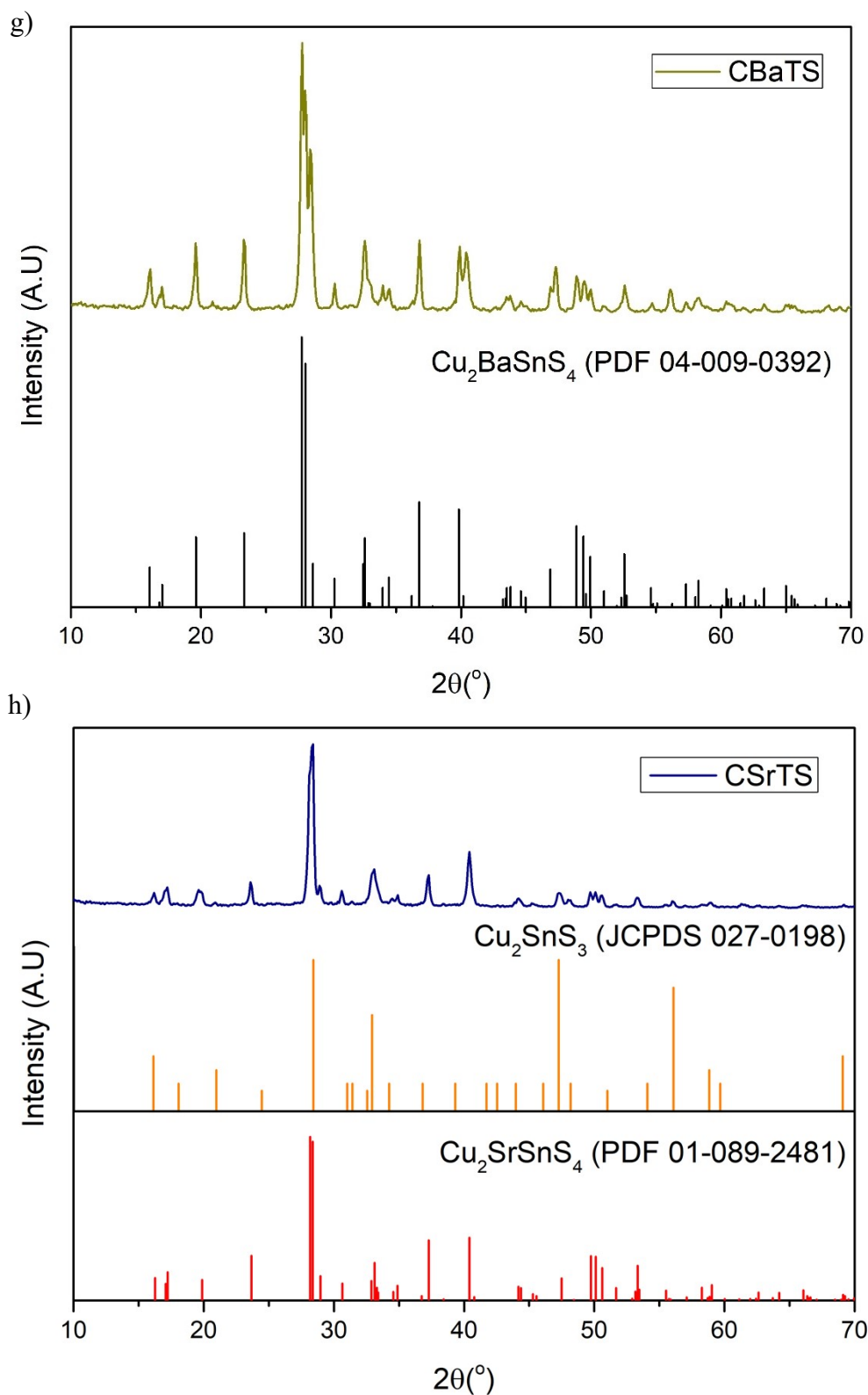


Figure S4 XRD pattern of CXTS films with each respective reference patterns (excluding CMgTS, no reference available).

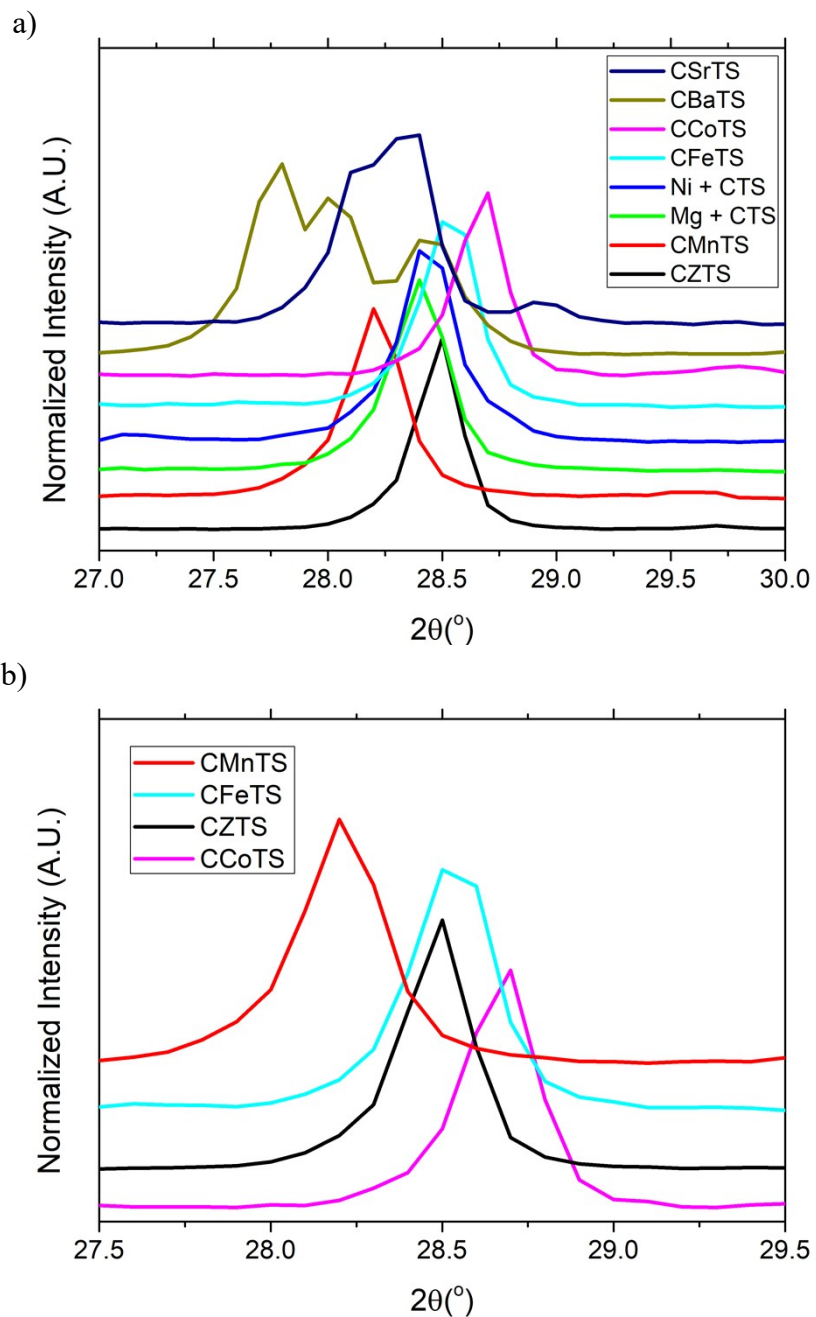


Figure S5 a) Normalized XRD peaks for CXTS compounds at 27° - 30°, b) Peak shift toward lower angle for tetragonal phase from smallest to largest ionic radius; Co²⁺ (0.72 Å), Zn²⁺ (0.74 Å), Fe²⁺ (0.77 Å), and Mn (0.80 Å)

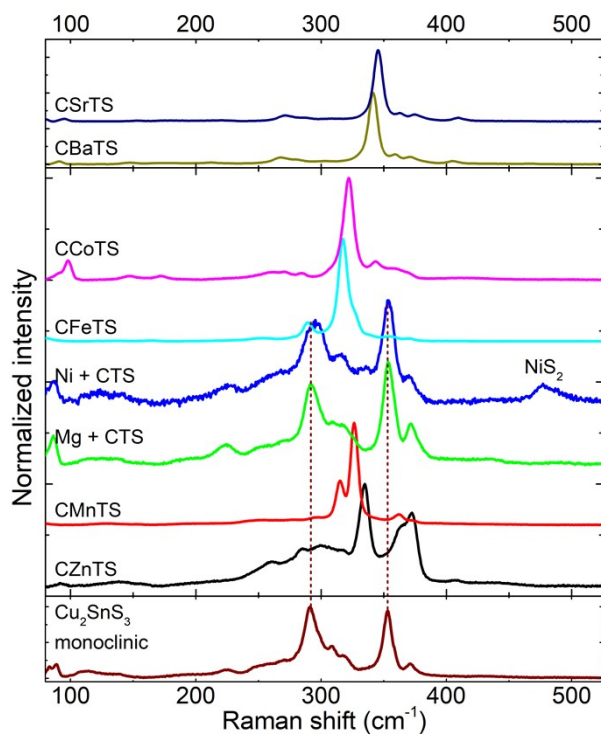


Figure S6 Raman scattering spectra of CXTS compounds measured under 785 nm excitation. The reference Raman spectra of monoclinic Cu_2SnS_3 compound is added for convenience.

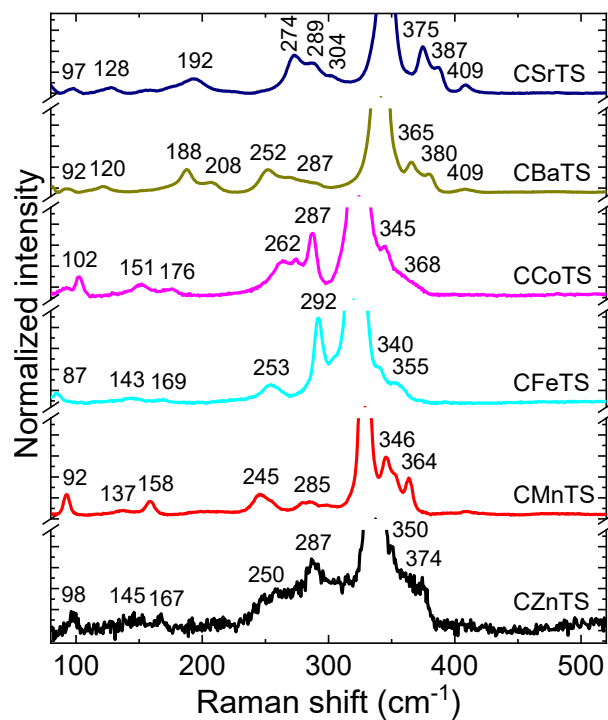


Figure S7 Raman scattering spectra of CXTS compounds with dominant quaternary phase measured under 532 nm show in the way to maximize the low intensity peaks. Numbers indicate the positions of the well pronounced peaks in the spectra.

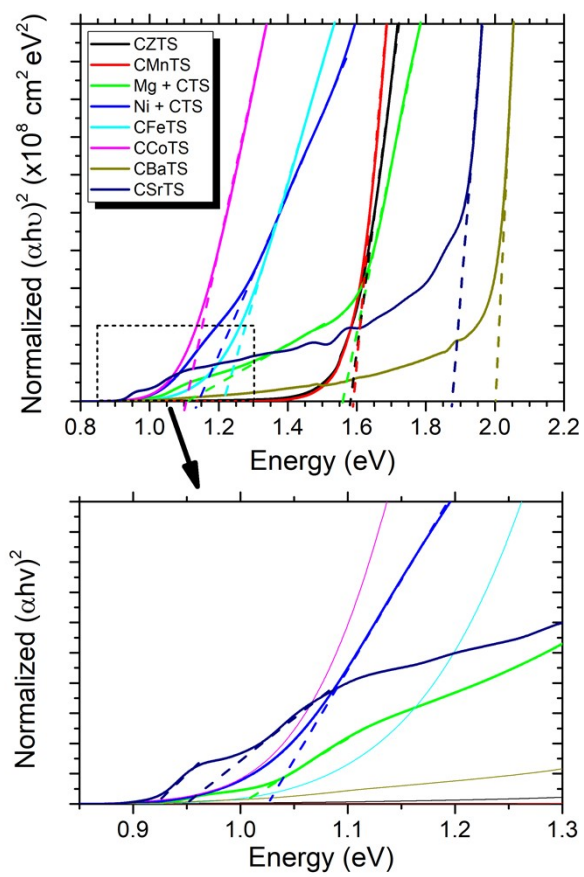


Figure S8 Tauc plot for band gap measurements based on UV-Vis spectroscopy

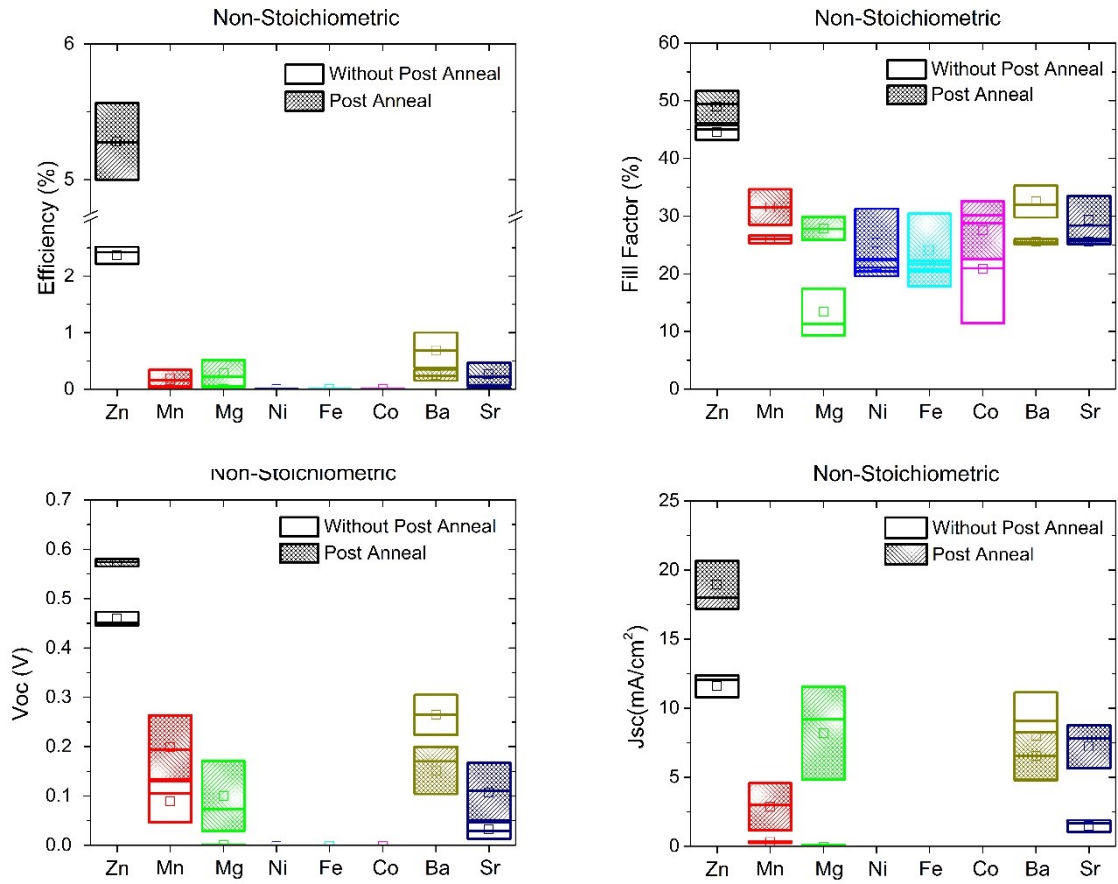


Figure S9 Device parameters for non-stoichiometric (Cu-poor and X-rich) CXTS solar cells as a function of X cations with respect to CZTS reference. The square box inside the box indicates the mean value.

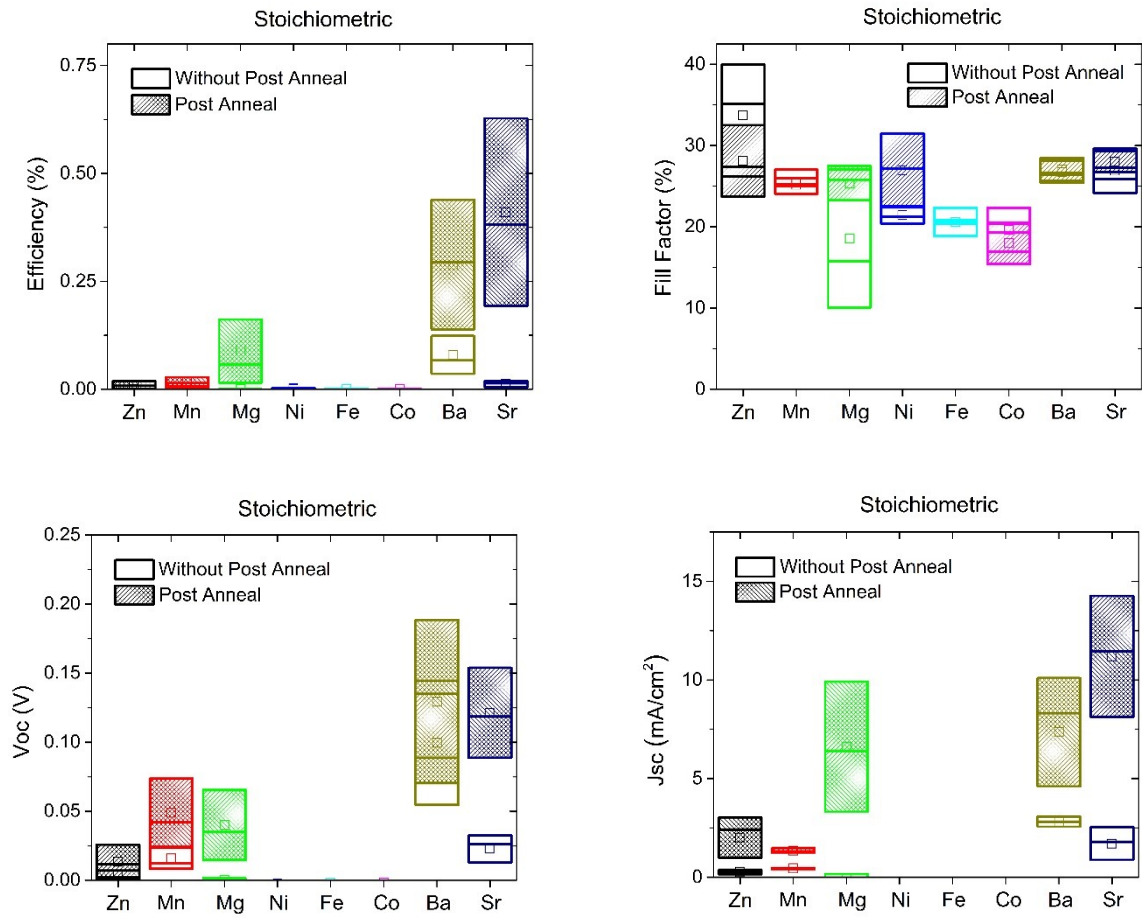


Figure S10 Device parameters for stoichiometry CXTS solar cell as a function of X cations. The square inside box indicate mean value.

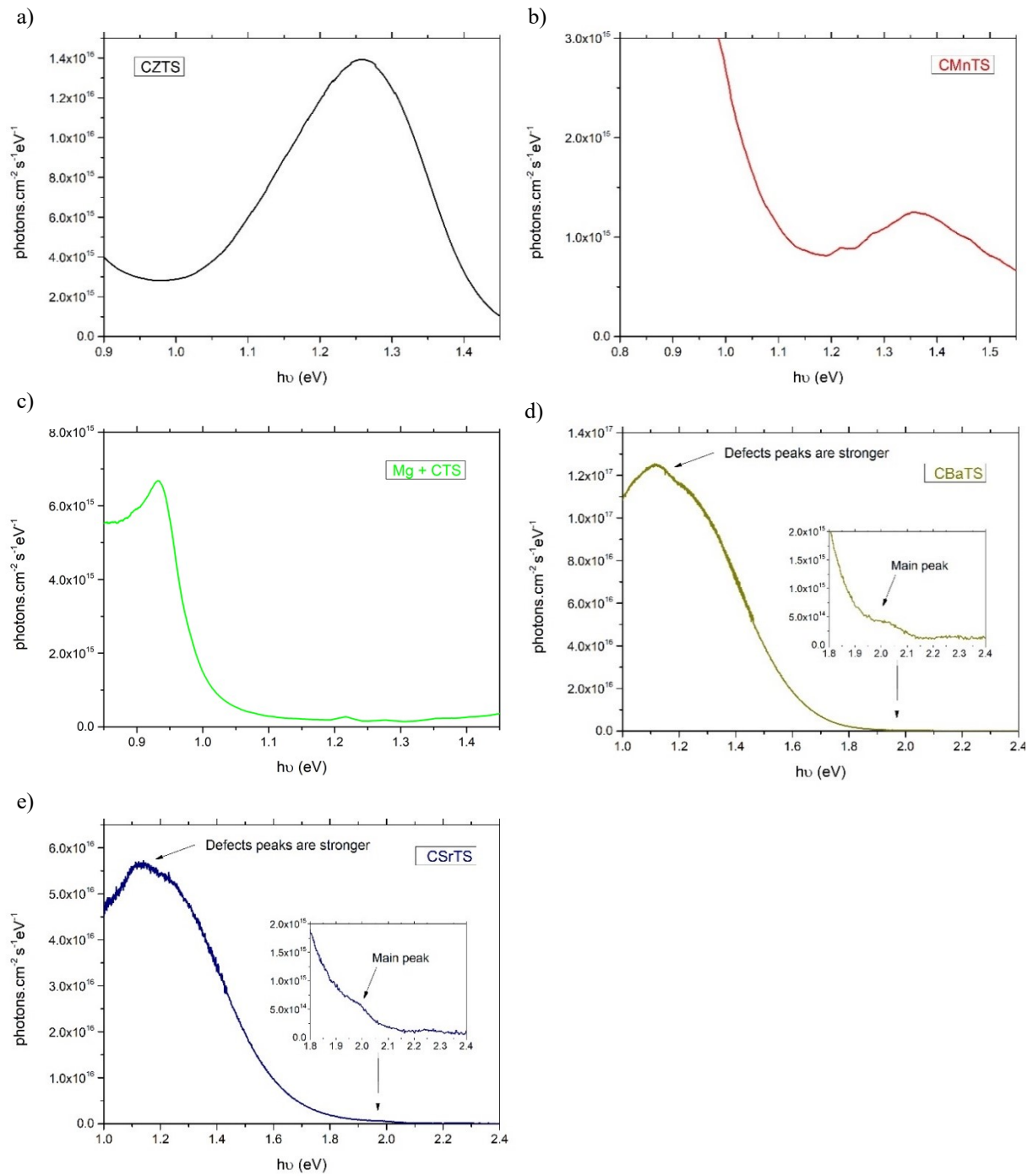


Figure S11 Photoluminescence spectra of a) CZTS, b) CMnTS, c) Mg + CTS, d) CBaTS and e) CSrTS