Improving Carrier Transport Properties of CZTS by Mg Incorporation with Spray Pyrolysis

Stener Lie^{\perp}, Shin Woei Leow^{\perp}, Douglas M. Bishop[§], Maxim Guc⁴, Victor Izquierdo-Roca⁴, Oki Gunawan[§], and Lydia Helena Wong^{\perp}*

[⊥]School of Materials Science & Engineering, Nanyang Technological University, 50 Nanyang Ave, 639798, Singapore

[§]IBM T.J. Watson Research Center, 1101 Kitchawan Rd, Yorktown Heights, NY 10598, USA.

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

Corresponding Author

*LydiaWong@ntu.edu.sg







Figure S1. Cross section SEM images of $Cu_2Mg_xZn_{1-x}SnS_4$ thin films with their respective EDS line scans at **a**) x =

0, **b**) x = 0.05, **c**) x = 0.1, **d**) x = 0.2, **e**) x = 0.4, **f**) x = 0.6, **g**) x = 0.8 and **h**) x = 1.0



Figure S2. Plan-view SEM images of $Cu_2Mg_xZn_{1-x}SnS_4$ thin films with **a**) x = 0, **b**) x = 0.05, **c**) x = 0.1, **d**) x = 0.05, **c**) x = 0.05, **c**) x = 0.1, **d**) x = 0.05, **e**) x = 0.05, **e** x = 0.05,

0.2, e) x = 0.4, f) x = 0.6, g) x = 0.8 and h) x = 1.0



Figure S3. Grain size distribution for Cu₂Mg_xZn_{1-x}SnS₄ thin films based on area (top) and diameter (bottom)



Figure S4. X-ray diffraction pattern of $Cu_2Mg_xZn_{1-x}SnS_4$ thin film (x = 0 -1.0). In the bottom is list of the signature peaks of possible quaternary, ternary and secondary phases from ICDD database.



Figure S5. Position and FWHM of the A1-like peak in Cu₂Mg_xZn_{1-x}SnS₄ thin films.



Figure S6. Raman scattering spectra of $Cu_2Mg_xZn_{1-x}SnS_4$ thin films measured under 785 (left) and 325 (right) nm excitation wavelength. In the right spectra the first (LO), second (2LO) and third (3LO) order of main ZnS peak are indicated.



Figure S7. External Quantum Efficiency of Cu₂Mg_xZn_{1-x}SnS₄



Figure S8. a) Voc vs temperature (T) and b) Jsc vs T under 1 sun measurement of $Cu_2Mg_xZn_{1-x}SnS_4$ for x = 0.0

-0.2