Structural study about cation disorder in kesterite based compound semiconductor

Shaham Quadir[1,2,4], Cheng-Ying Chen[1,5], Shin-ichiro Yano[3], Wei-Tin Chen[1], Chin-Wei Wang[3], Chun-Ming Wu[4], Szu-Yuan Chen[2,4], Kuei-Hsien Chen[1,2], Li-Chyong Chen[1,5]

[1]Center for Condensed Matter Sciences, National Taiwan University, Taipei, Taiwan, [2]Academia Sinica, Institute of Atomic and Molecular Sciences, Taipei, Taiwan, [3]Neutron Group, National Synchrotron Radiation Research Center, Hsinchu, Taiwan, [4]Department of Physics, National Central University, Taoyuan, Taiwan, [5]Center of Atomic Initiative for New Materials, National Taiwan University, Taipei, Taiwan.
chen.chengying.cyc@gmail.com

Abstract

The photovoltaic absorber $Cu_2ZnSn(S_xSe_{1-x})_4$ (CZTSSe) has attracted interest due to the earth abundance of its elements & the realization of high performance (12.6% power conversion efficiency). To raise the performance of CZTSSe based solar cells, much effort has been applied to improve the quality of absorbers, band alignments/passivation at p-n junction, front, and back interfaces/contacts [1,2,3,4]. The open-circuit voltage in kesterite based CZTSSe solar cell is limited due to absorber band tailing caused by the high density of Cu/Zn antisites. Due to the similarity between the covalent radii of Cu & Zn, the most prevalent Cu/Zn antisite defects will limit the open-circuit voltage (Voc). By replacing Cu in CZTSe with Ag, whose ionic radius is 15% larger than that of Cu, the density of I–II antisite defect is expected to reduce and hence enhance the photovoltaic performance. We introduced Ag into $Cu_2ZnSnSe_4$ (CZTSe) powder sample to replace Cu by 0%, 10%, 20% and form $(Ag_xCu_{1-x})_2ZnSnSe_4$ (ACZTSe) to characterize the structural property of the mixed Ag–Cu kesterite compound as a function of the Ag / (Ag + Cu) ratio. The study of defect property of (ACZTSe) has been done by measuring structural disorder with the help of high resolution synchrotron and neutron powder diffractometer. The diffraction patterns from both instruments were analysed using full pattern fitting combined Rietveld refinement method. It has been found that powder CZTSe adopts the kesterite type structure (space group $I\bar{4}$) with a partial disorder of copper and zinc on the two Wyckoff position 2c and 2d. Sn has been found on Wyckoff position 2b (0, 0, ½), whereas Cu/Zn is located on 2a (0, 0, 0), 2c (0, ½ , 1/4) and 2d (0, ½ , ¾) sites. With the presences silver doping, various cation antisite defect density (Cu_{Zn}, Zn_{Cu}, Zn_{Sn} and $Sn_{Zn...}$) has been quantified.

Keywords – $Cu_2ZnSNSe_4$, Combined refinement, Neutron diffraction, Ionic radius

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