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Crystallographic and optical properties of CuGa<sub>3</sub>S<sub>5</sub>, CuGa<sub>3</sub>Se<sub>5</sub> and CuIn<sub>3</sub>(S,Se)<sub>5</sub> and CuGa<sub>3</sub>(S,Se)<sub>5</sub> systems

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# Crystallographic and optical properties of $\text{CuGa}_3\text{S}_5$ , $\text{CuGa}_3\text{Se}_5$ and $\text{CuIn}_3(\text{S}, \text{Se})_5$ and $\text{CuGa}_3(\text{S}, \text{Se})_5$ systems

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## Abstract

To clarify the solid solution regions of  $\text{CuIn}_3(\text{S}_x\text{Se}_{1-x})_5$  and  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  systems and their optical properties, we prepared  $\text{CuIn}_3(\text{S}, \text{Se})_5$  and  $\text{CuGa}_3(\text{S}, \text{Se})_5$  samples by a mechanochemical process and post-heating. Single-phase solid solutions with a tetragonal stannite-type structure could not be obtained for  $\text{CuIn}_3(\text{S}_x\text{Se}_{1-x})_5$  with  $0 \leq x < 0.1$ . On the other hand, we successfully obtained single-phase solid solutions with a tetragonal stannite-type structure for  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  with  $0.0 \leq x \leq 1.0$ . The solid solution region of the  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  system is much wider than that of the  $\text{CuIn}_3(\text{S}_x\text{Se}_{1-x})_5$  system. The band gap energy of the  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  solid solution linearly increased from 1.85 eV of  $\text{CuGa}_3\text{Se}_5$  ( $x = 0.0$ ) to 2.58 eV of  $\text{CuGa}_3\text{S}_5$  ( $x = 1.0$ ). The energy levels of the valence band maxima (VBMs) were estimated from the ionization energies measured by photoemission yield spectroscopy (PYS). The ionization energy of stannite-type  $\text{CuGa}_3\text{Se}_5$  (5.69 eV) is approximately equal to that of  $\text{CuIn}_3\text{Se}_5$  (5.65 eV). The energy levels of the VBMs of the  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  solid solution decrease with increasing S content,  $x = \text{S}/(\text{Se} + \text{S})$  ratio. The conduction band minimum (CBM) levels of  $\text{CuGa}_3(\text{S}_x\text{Se}_{1-x})_5$  are almost constant with  $x = \text{S}/(\text{Se} + \text{S})$  ratio.  $\text{CuIn}_3\text{Se}_5$ ,  $\text{CuGa}_3\text{Se}_5$ ,  $\text{CuGa}_3\text{S}_5$  and  $\text{CuGa}_3(\text{S}, \text{Se})_5$  solid solution are expected to be useful for controlling the valence band offset ( $\Delta E_v$ ) and the conduction band offset ( $\Delta E_c$ ) at the interface between buffer layer and absorber layer in CIGS solar cells.

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