

GROWTH OF $\text{Cu}_2\text{CdGeSe}_4$ MONOGRAIN POWDERS IN MOLTEN SALTS

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As we all know, energy demand in the world increases with the development of science and technology. Nowadays majority of energy produced by fossil fuels, not only it has some bad impacts to our environments but also fossil energy is limited on the earth. Therefore, developing sustainable energy has become necessary and indispensable. Currently, photovoltaics, which directly convert solar energy into electricity, offer a practical and sustainable solution to the challenge of meeting the increasing global energy demand. The implementation of solar energy is a fast growing sector due to its abundant and inexhaustible energy resource, less greenhouse gases emission and an effective renewable alternative to conventional fossil fuels.

The large group of quaternary and ternary copper chalcogenides has attracted considerable attention due to their suitable properties for thin film solar cell absorbers. Among them the $\text{Cu}_2\text{CdGeSe}_4$ (CCGSe) compound is maybe less studied, therefore, it motivates researcher to do research on the quaternary $\text{Cu}_2\text{CdGeSe}_4$ due to its suitable physical and electronic properties.

The quaternary $\text{Cu}_2\text{CdGeSe}_4$ has suitable properties for photovoltaic applications - p-type conductivity, high absorption coefficient ($> 10^4 \text{ cm}^{-1}$) and direct energy band gap (1.2 - 1.29 eV). However, the formation conditions and properties of $\text{Cu}_2\text{CdGeSe}_4$ monograin powders have not been studied.

In this study, new quaternary compound $\text{Cu}_2\text{CdGeSe}_4$ was synthesized by molten salt method for photovoltaic applications. $\text{Cu}_2\text{CdGeSe}_4$ powder materials were synthesized from binary compounds (CuSe, CdSe) and elemental metal powder (Ge) and Se at temperatures - 500, 600, and 700 °C in the liquid phase of KI and CdI_2 as the flux materials. The effect of salt material (CdI_2 and KI), synthesis temperature and synthesis time on the structural, morphological, compositional and optoelectronic properties was investigated.



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The sieving analysis revealed that median particle size of produced powder crystals increased with increasing synthesis temperature and duration. Median sizes of particles are 25, 58 and 139 μm for powders synthesized in CdI_2 at 500, 600 and 700 $^\circ\text{C}$, respectively. Median sizes of particles are 112 and 132 μm for powders synthesized in KI flux for 60 and 120 hours, respectively.

Structural analysis by Raman spectroscopy and X-ray diffraction showed that $\text{Cu}_2\text{CdGeSe}_4$ powder synthesized at 500 $^\circ\text{C}$ had a tetragonal structure and powders synthesized at temperatures 600 and 700 $^\circ\text{C}$ had orthorhombic structure.

Compositional analysis by EDX showed that increasing the synthesis temperature from 500 to 700 $^\circ\text{C}$, the bulk composition becomes Cd-rich. The powders synthesized in CdI_2 at 500 and 700 $^\circ\text{C}$ had stoichiometric composition ($[\text{Cu}]/([\text{Cd}]+[\text{Ge}]) \sim 1.0$), but powders synthesized at 600 $^\circ\text{C}$ had Cu-poor composition ($[\text{Cu}]/([\text{Cd}]+[\text{Ge}]) = 0.93$). Bulk composition of the $\text{Cu}_2\text{CdGeSe}_4$ powders synthesized in KI flux for different times was Cd-rich and did not depend on synthesis time, but the ratio of $[\text{Cu}]/([\text{Cd}]+[\text{Ge}])$ decreased from 1.0 to 0.96 by increasing synthesis time.

All monograin powders exhibited p-type conductivity regardless of the composition, but powders with Cu-poor and Cd-rich compositions were produced at 600 $^\circ\text{C}$ in CdI_2 or 700 $^\circ\text{C}$ in KI flux.

The band gap obtained from EQE measurements was found to be 1.27 eV for orthorhombic structured powders and 1.14 eV for tetragonal structured powders. The best solar cell fabricated from the *o*- $\text{Cu}_2\text{CdGeSe}_4$ powder showed the power conversion efficiency of 4.21 % (active area), with an open-circuit voltage of 0.46 V, a short-circuit current density of 23.3 mA/cm^2 and fill factor of 39 %.



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