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ACCEPTED MANUSCRIPT

Chemical crystallographic investigation on Cu₂S-In₂S₃-Ga₂S₃ ternary system

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Abstract

Sulfide chalcopyrites Cu(In,Ga)S₂ (CIGS) have been proposed as top cell for tandem solar devices with silicon as bottom cell. In that context, the knowledge of the Cu₂S-In₂S₃-Ga₂S₃ pseudo-ternary system is of prime importance. This study presents an overall investigation of this system on the basis of a chemical crystallographic approach on bulk materials. Particular attention was paid on the copper-poor side of the so-called stoichiometric CuInS₂-CuGaS₂ line. Based on our investigations, it turns out that the sulfide CIGS system is much more structurally complex than the selenide Cu(In,Ga)Se₂ one (CIGSe). Especially, the adaptability of the chalcopyrite structure towards Cu deficiency is significantly lower for the sulfides than for the selenide counterparts. Correlatively, a large 2-phase domain exists between the CuInS₂-CuGaS₂ line and the very copper-poor region (Cu(In,Ga)₃S₅ and Cu(In,Ga)₅S₈). Additionally, the crystal structures of these very copper-poor compounds contain [InS₆] octahedra and then are totally different than the chalcopyrite structure. These features could explain why CIGS-based-solar cells are, up to now, less efficient than the CIGSe ones.

1. Introduction

In the last decades, photovoltaic industry has grown rapidly and power conversion efficiency of several technologies approach now the Shockley Queisser (SQ) limit (e.g. over 26 % for silicon solar cell [1]). To overpass the SQ limit, tandem solar cells in which a wide

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