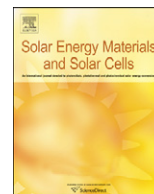




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Progress in chalcopyrite compound semiconductor research for photovoltaic applications and transfer of results into actual solar cell production

Arnulf Jäger-Waldau

European Commission, DG JRC, Institute for Energy Renewable Energies Unit, 21027 Ispra, Italy

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ABSTRACT

This paper gives an overview of the main research directions in chalcopyrite material research and the application of results for the improvement and fabrication of solar cells. So far the copper indium gallium sulphur selenide material family is the base for the highest efficiency thin-film solar cells and the most advanced in terms of actual commercialisation. The transfer of research results into actual production from its early stage and the development of the chalcopyrite thin-film solar cell industry are sketched. The last part of the review shortly describes a number of current industrial players involved in the manufacturing of chalcopyrite solar cells.

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1. Introduction

Since a number of years photovoltaics continues to be one of the fastest growing industries with growth rates well beyond 40% per annum. This growth is driven not only by the progress in materials and processing technology, but by market support programmes in a growing number of countries around the world [1].

Since 1988, world wide solar cell production has increased from about 35 MW to 11.5 GW in 2009 or more than 340 times (Figs. 1 and 2). Between 1988 and the mid 1990s, PV production exhibited a moderate growth rate of about 15%. With the introduction of the Japanese 70,000 roof programme in 1997, the growth rate more than doubled for the end of the decade. Since the introduction of the German feed-in tariff scheme for PV generated electricity in 2000, total PV production increased more than 30 fold, with annual growth rates between 40% and 80%.

Wafer-based silicon solar cells is still the main technology and had around 80% market shares in 2009, but thin-film solar cells are continuously increasing their share since 2005. Back then, production of thin-film solar modules reached for the first time more than 100 MW per annum. Since then, the *Compound Annual Growth Rate* (CAGR) of thin-film solar module production was even beyond that of the overall industry, increasing the market share of thin-film products from 6% in 2005 to 10% in 2007 and 16–20% in 2009.

Amongst the different thin film technologies, the family of chalcopyrites has so far demonstrated the highest efficiency for solar cells in the laboratory as well as in production. This review gives an overview about the long term R&D efforts to realise this as

well as the industry activities to commercialise chalcopyrite solar cells. The specific details of research for chalcopyrite solar cells will be left to the specialised article in this special edition.

2. Research directions

The chalcopyrite compound family has two subclasses, which are based on the use of elements from group I, III and VI or II, IV and V of the periodic table and are derived from the III–V and II–VI compounds, respectively. The first ternary semiconductor device was already made in the 1920s. It was a point-contact diode made out of a mineral chalcopyrite (CuFeS_2) [6]. In the early 1950s the search for new semiconductor materials led to investigations of the first III–V materials focused on finding materials with a high carrier mobility and an amphoteric doping [7,8]. Soon the search was broadened to ternary compounds and the artificial synthesis of the two subgroup families led to intensive studies of their physical properties [9–12]. In the 1960s and 70's electrical, optical and structural properties of the $(\text{Cu,Ag})(\text{Al,Ga,In})(\text{S,Se,Te})_2$ as well as the $(\text{Zn,Cd})(\text{Si,Ge,Sn})(\text{P,As})_2$ family were investigated [13–15].

The large number of the ternary compounds and their corresponding variety of properties like the variation of bandgaps ranging from 0.26 eV (CdSnAs_2) to 3.5 eV (CuAlS_2), the possibility to substitute certain elements like In by Ga or Se by S, as well as the possibility to synthesise the material n- or p-type resulted in the development of a number of novel devices at the end of the 1960s and early 70's [14,16,17].

Initially they were explored for their non-linear optical properties [14]. Chalcopyrites were investigated with respect to their use as frequency mixing devices like the parametric oscillator, harmonic

E-mail address: arnulf.jaeger-waldau@ec.europa.eu

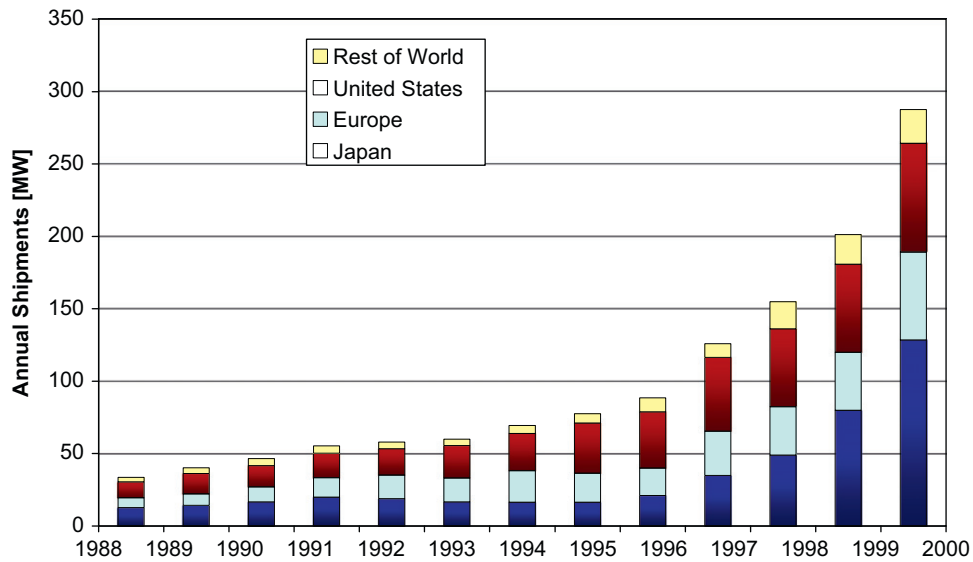


Fig. 1. World solar cell production from 1988 to 2000 [2].

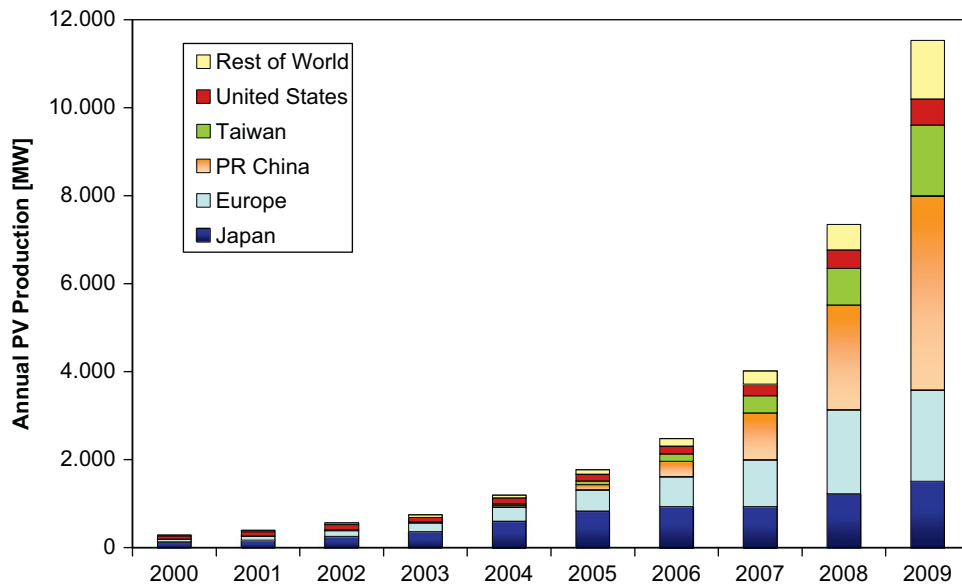


Fig. 2. World solar cell production from 2000 to 2009 [1,3–5].

generator as well as up and down converters. Frequency mixing materials are: ZnGeP_2 , CdGeAs_2 , AgGaS_2 , AgGaSe_2 , ZnSiAs_2 , AgGaTe_2 and Proustite (Ag_3AsS_3) [18]. Another non-linear optical device application investigated was the use of chalcopyrites as electro-optic modulators.

The fact, that a large number of chalcopyrites exhibit direct bandgaps and relatively narrow luminescent bands (sometimes at low temperatures only) made them an interesting candidate to obtain stimulated emission/laser action. It could be demonstrated, that stimulated emission could be achieved in six of the ternary compounds by brute force pumping with either a high voltage electron beam [19,20] or laser pumping [21–23]. The chalcopyrites in question were CdSiAs_2 , CdSnP_2 , $\text{CdSnP}_2\text{:Ag}$, CuGaS_2 , AgGaS_2 and AgGaSe_2 .

The ability of some of the ternary compounds to be synthesised with both n- and p-type conduction and a low resistivity was used to form the first homojunctions in CdSnP_2 [24], CuInS_2 [23] and CuInSe_2 [25] by dopant diffusion or stoichiometry variation and vapour annealing. The stoichiometry variation and vapour anneal

technique was used to produce the first ternary homodiode, which could be characterised in detail [26]. A n-type CuInSe_2 crystal was treated with a short annealing in Se vapour to make a junction a short distance below the crystal surface. Then the crystal was etched to a mesa structure. For ± 1 V a rectification ratio of 300:1 was obtained. Electroluminescence could be obtained with a forward bias centred on $1.34 \mu\text{m}$ and with 10% internal quantum efficiency at 77 K.

The first ternary heterodiode was reported in 1970 based on a n-type CdSnP_2 single crystal combined with p-type Cu_2S [27]. Despite a lattice mismatch of around 5% the device exhibited a large photovoltaic response under white light illumination. The focus of investigations changed after the first $\text{CuInSe}_2/\text{CdS}$ solar cell device with 12.5% efficiency was produced at the Bell Laboratories in 1974 [28]. This device was based on a p-type CuInSe_2 single crystal onto which a thin n-type CdS layer was deposited. The result was outstanding at the time and by far better than the results achieved with thin film devices, which at that time reached around 6%

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