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Review

# Development of antimony sulfide–selenide $Sb_2(S, Se)_3$-based solar cells

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ABSTRACT

Antimony sulfide–selenide $Sb_2(S, Se)_3$, including $Sb_2S_3$ and $Sb_2Se_3$, can be regarded as binary metal chalcogenides semiconductors since $Sb_2S_3$ and $Sb_2Se_3$ are isomorphous. They possess abundant elemental storage, nontoxicity, good stability with regard to moisture at elevated temperatures and suitable physical parameters for light absorption materials in solar cells. To date, quite a few attempts have been conducted in the materials synthesis, photovoltaic property investigation and device fabrication. Benefiting from previous investigation in thin film solar cells and new generation nanostructured solar cells, this class of materials has been applied in either sensitized-architecture or planar heterojunction solar cells. Decent power conversion efficiencies from 5% to 7.5% have been achieved. Apparently, further improvement on the efficiency is required for future practical applications. To give an overview of this research field, this paper displays some typical researches regarding the methodologies toward the antimony sulfide–selenide synthesis, development of interfacial materials and device fabrications, during which we highlight some critical findings that promote the efficiency enhancement. Finally, this paper proposes some outstanding issue regarding fundamental understanding of the materials, some viewpoints for the efficiency improvement and their future challenges in solar cell applications.



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## 1. Introduction

Solar energy exists in irradiant form. Converting the irradiant energy into electricity by solar cell is one of the significant practical methods to utilize solar energy. Since the first demonstration of silicon solar cell in 1954 at Bell laboratories by Chapin, Souther and Pearson, researchers have exploited many kinds of active materials for solar energy conversion. The ultimate goal is to develop a solar cell that can simultaneously meet the so-called golden-triangle requirements: low cost, high power conversion efficiency (PCE) and long-term stability. Although the production cost of silicon solar cell gradually decreased year by year, the price is still higher than the electricity generation by conventional fossil fuels. The second generation thin-film solar cells, such as those based on $Cu(In,Ga)Se_2$ (CIGS) and CdTe, are able to reduce the cost, they also achieved PCE of around 22% [1,2], however, much more efforts are still needed in the technological development with regard to large area production. In addition, the scarcity of indium and toxic Cd are potential problems in the large area practical applications.

The third generation emerging solar cells adopt low cost functional materials that are promising for grid parity application, while they are still under fundamental investigation. For instance, substitution of In and Ga in CIGS for Zn and Sn leads to the formation of $Cu_2ZnSn(S,Se)_4$ (CZTS) light absorbing material, which contains only earth-abundant elements. The abundant storage of Zn and Sn guarantees low materials cost; this kind of inorganic semiconducting compound is also stable. With several years of intense investigation, the PCE has reached 12.6% in 2012 [3]. However, due to the difficulty in defect manipulation, further improving PCE poses grant challenge. Dye-sensitized solar cells and polymer solar cells possess unique properties such as colorful and flexible characteristics [4–9]. They can be applied in the building-integrated photovoltaics or flexible devices. However, the long-term stability and attainable PCEs require significant enhancement. Most recently, organometal trihalide perovskite, e.g. $CH_3NH_3PbI_3$, based solar cells have achieved unprecedented progress with PCE increasing from 3.8% in 2009 to 22.1% in 2016 [10,11]. This PCE is on par with those of second generation thin-film solar cells. Nonetheless, it considerably degrades in days in moderate moisture. The sensitivity to moisture is believed to be an obstacle in the practical application of perovskite solar cell. Engineering the crystal structure as well as the device configuration is able to remarkably improve the lifetime of perovskite solar cells [12–16], while essential breakthrough in extending the lifetime is critical for the transition of this technology into industrial applications.

In addition to the above widely investigated light absorption materials and device configurations, some researchers have attempted to utilize unclassical materials for solar cell construction, such as $Cu_2SnS_3$ [17–19], $Cu_2GeS_3$ [20], $Cu_2(Sn,Ge)S_3$ [21], GeSe [22], $Sb_2(S,Se)_3$ [23,24], etc. These materials contain earth-abundant elements and have high stability. Solar cells based on these classes of semiconducting materials have delivered PCEs of 4.63%, 2.67%, 6.7%, 1.48% and >6.0%, respectively for $Cu_2SnS_3$ [17–19], $Cu_2GeS_3$ [20], $Cu_2(Sn,Ge)S_3$ [21], GeSe [22], $Sb_2(S,Se)_3$ [23,24]. Compared with the extensively studied light absorption materials and well-established device structures, these materials did not receive much attention. As a result, many problems associated with the materials synthesis, physical properties, interfacial engineering and device construction remain unresolved. In this paper, we focus on $Sb_2(S,Se)_3$, including $Sb_2S_3$ and $Sb_2Se_3$ unless otherwise stated, based solar cells with special attention to the fundamental understanding, materials synthesis, device construction and performance enhancement strategies. Previously, an excellent review described the $Sb_2Se_3$ solar cell [25]. Here, we attempt to formulate the $Sb_2S_3$, $Sb_2Se_3$ and $Sb_2(S_{1-x}Se_x)_3$ together as light absorber materials and make comparisons.

## 2. Materials properties of $Sb_2S_3$ and $Sb_2Se_3$

In specific, the elemental storage of Sb, S and Se in earth crust are 0.2, 260 and 0.05 ppm, respectively. The storage of Sb and S are considerably higher than those of In (0.049 ppm) and Te (0.005 ppm). This abundant storage set the basement for solar panel production in large scale at low cost, the relevant price and abundance of the corresponding elements are summarized in Fig. 1(a). In addition, all of the three elements are not included in the list of highly toxic or carcinogenic materials by Chinese, American as well as European Union regulation authorities. They can be regarded as environmental benign materials. In addition, they are binary compound. The controlling over the phase formation is not that rigorous. Furthermore, the melting points of $Sb_2S_3$ and $Sb_2Se_3$ are 500 and 608 °C, respectively, indicating low phase formation

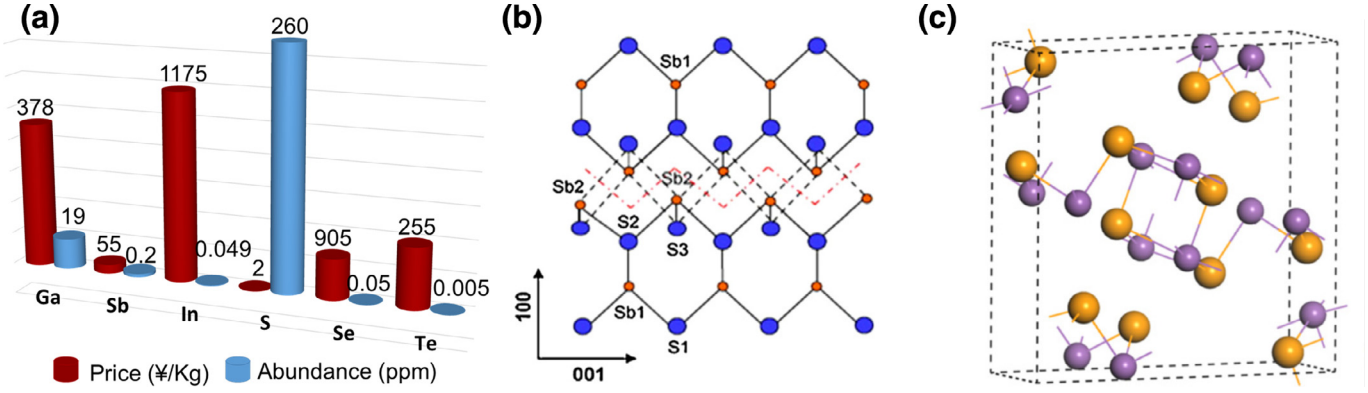


**Fig. 1.** (a) Price and earth abundance of Ga, Sb, In, S, Se, and Te; (b) Crystal structure of $Sb_2S_3$ viewed along the [010] direction. The Sb–S covalent bonds are indicated by solid lines and the weak van der Waals bonds by the black dashed lines. The cleavage trace is demarcated by the red dashed line. Reprinted with permission from Ref. [29] Copyright 2010 Elsevier Ltd; (c) Crystal structure of $Sb_2Se_3$, the violet balls stand for antimony and the yellow balls stand for selenium. Reprinted with permission from Ref. [30]. Copyright 2016 IOP Publishing Ltd.

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