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Recent progress and challenges in CIGS and CZTS thin film photovoltaic technologies. Review

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This review provides a comprehensive analysis of the current state, progress, and challenges of thin-film solar cells based on Cu(In,Ga)Se_2 (CIGS) and $Cu_2ZnSn(S,Se)_4$ (CZTS(Se)) absorber materials. The study highlights the global transition toward renewable energy, emphasizing the advantages of thin-film photovoltaic technologies as cost-effective, material-efficient, and flexible alternatives to conventional crystalline silicon solar cells. CIGS-based devices demonstrate high power conversion efficiencies exceeding 23%, wide tunability of the bandgap, and excellent long-term stability, making them among the most mature thin-film technologies. However, their dependence on scarce and costly elements such as indium and gallium motivates research into earth-abundant alternatives. CZTS(Se) materials, composed of non-toxic and widely available elements (Cu, Zn, Sn, S, Se), offer a sustainable substitute with comparable optical properties and tunable bandgaps (1.0–1.5 eV). Despite lower efficiencies (currently up to 15.8%), ongoing progress in controlling phase purity, defect passivation, and band alignment is rapidly improving device performance. Key research directions include the optimization of absorber synthesis, defect engineering, alkali-metal and cation doping, and development of environmentally benign, cadmium-free device architectures. The review outlines both the technological maturity of CIGS and the emerging potential of CZTS(Se), identifying critical challenges and future pathways toward high-efficiency, sustainable thin-film photovoltaics.

Keywords: CIGS; CZTS(Se); thin-film solar cells; photovoltaics; absorber materials; bandgap engineering; defect passivation; renewable energy; efficiency improvement; sustainable technology.

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Content

Introduction

The modern world faces global challenges related to climate change caused by greenhouse gas emissions from fossil fuel combustion and the depletion of non-renewable energy resources. The urgent need to transition to a sustainable energy future drives the intensive development and deployment of renewable energy sources (RES) worldwide. Among the various RES, solar photovoltaics (PV), which provides direct conversion of solar radiation into electrical energy, is recognised as one of the most promising and rapidly growing technologies [1, 2]. Over the past decades, the PV industry has demonstrated exponential growth in installed capacity, a significant reduction in the cost of electricity generated, and significant technological improvements [3]. This makes solar energy a key element in the decarbonisation and energy security strategies of many countries.

Crystalline silicon (c-Si) solar cells characterised by high efficiency and proven long-term stability remain the dominant technology in the modern PV market [4]. However, the production of high-purity silicon is an energy-intensive process, and the modules, although they have become significantly cheaper, still have certain limitations in terms of flexibility and integration into various architectural forms. In this context, thin-film solar cells (TFSCs) represent an important alternative and complement to silicon technologies, offering a number of unique advantages [5]. Historically, the development of TFSCs began in the 1970s, in parallel with the improvement of silicon technologies, with the aim of creating cheaper and less material-intensive photovoltaic converters [6].

A key feature of TFSCs is the use of extremely thin layers of photoactive materials, typically ranging in thickness from a few hundred nanometres to a few micrometres. This is hundreds of times less than the thickness of silicon wafers (about 150-200 micrometres). Such a small thickness of the absorbing layer has become possible thanks to the use of materials with a high optical absorption coefficient, which allows sunlight to be effectively absorbed in a thin layer. As a result, TFSCs have significant advantages. First, reduced material consumption not only lowers the cost of raw materials, but also reduces the "embodied energy" (energy payback time – EPBT), i.e. the time it takes for a solar module to generate an amount of energy equivalent to that spent on its production [7]; for some thin-film technologies, EPBT can be significantly shorter than for traditional silicon modules [8]. Secondly, many TFSCs can be manufactured by using large-scale deposition methods with the potential for low-cost production, such as vacuum deposition or chemical methods over large areas, including roll-to-roll technologies. Thirdly, the use of thin layers enables their deposition on flexible substrates (polymer or metal), paving the way for the development of lightweight, flexible and portable solar modules for a wide range of applications, from mobile devices to integration into textiles or curved building surfaces (BIPV – building-integrated photovoltaics) [7-9]. In addition, some types of TPE demonstrate better temperature coefficients and more efficient performance in diffuse illumination compared to c-Si cells.

The main types of PV cells that have achieved varying degrees of technological maturity and commercialisation include solar cells based on amorphous silicon (a-Si), cadmium telluride (CdTe), copper indium gallium diselenide (CIGS) and, more recently, perovskite solar cells (PSC), which demonstrate rapid increase in efficiency [10]. CdTe technology is one of the leading TFSC technologies in the market in terms of production volume, demonstrating high efficiency and low cost, but the use of toxic cadmium raises certain environmental concerns, although modern production and recycling technologies mitigate these risks [11].

In this review article, we will focus on two promising types of chalcogenide thin-film absorbers: $Cu(In,Ga)Se_2$ (CIGS) and Cu_2ZnSnS_4 (CZTS) (or its sulphur-selenium analogues $Cu_2ZnSn(S,Se)_4$, hereinafter referred to as CZTS(Se)). These two classes of materials were selected due to their unique properties and potential for further development in photovoltaics.

CIGS-based solar cells belong to the second generation of TFSCs and have already achieved significant commercial maturity. Laboratory samples of CIGS cells demonstrate energy conversion efficiency exceeding 23%, which is the highest among all polycrystalline thin-film technologies and comparable to the efficiency of multicrystalline silicon. High absorption coefficient ($>10^4 cm^{-1}$), the ability to adjust the bandgap width (from ~1.0 eV for $CuInSe_2$ to ~1.7 eV for $CuGaSe_2$) by changing the [Ga]/ ([In]+[Ga]) ratio, as well as demonstrated long-term stability make CIGS extremely attractive for a wide range of applications [12]. Research in the field of CIGS continues to focus on further improving efficiency, reducing manufacturing costs (for example, by reducing layer thickness or using alternative deposition methods) and developing flexible modules. However, the use of relatively rare and expensive elements such as indium and gallium, as well as selenium, in CIGS drives the search for alternative materials [13].

This is where the CZTS(Se) material family comes to the forefront. These compounds, whose crystal structure (kesterite or stannite) is derived from the chalcopyrite structure of CIGS, consist of elements that are widely spread in the Earth's crust and are non-toxic (or low-toxic): copper, zinc, tin, sulphur and selenium [14]. This makes them potentially much cheaper and more environmentally sustainable in the long term. CZTS(Se) possess an optimal band gap (about 1.0-1.5 eV, depending on the S/Se ratio) and a high absorption coefficient similar to CIGS. Although the record efficiency of CZTS(Se)-based solar cells (about 13-14.9% for CZTSSe and 10-11% for pure sulphide CZTS under standard conditions [15]) is still inferior to CIGS and CdTe, significant progress has been achieved over the last decade. The main challenges for further improving CZTS(Se) efficiency include the large open-circuit voltage (Voc) deficit, the difficulty in controlling the phase composition and avoiding of secondary phase formation (e.g., ZnS(Se), $Cu_xS(Se)$, $SnS_y(Se)_y$), as well as the high concentration of intrinsic defects and recombination centres (CuZn and ZnCu) and their clusters.

The purpose of this review article is to conduct a comprehensive analysis of the current state of research and development in the field of thin-film solar cells based on CIGS and CZTS(Se) compounds, highlighting their fundamental properties, technological aspects, achievements and existing problems. The main objectives of the article are: to systematise current data on the crystal structure, optical and electrical properties of the absorbing layers of CIGS and CZTS(Se); to analyse modern vacuum and non-vacuum methods for obtaining high-quality CIGS and CZTS(Se) films; to consider typical architectures of solar cells based on them, the performance of individual layers and heterojunctions; to evaluate the achieved level of efficiency and stability of devices, as well as the factors that limit them; conduct a comparative analysis of the advantages, disadvantages, economic feasibility and environmental aspects of CIGS and CZTS(Se) technologies; identify key scientific and technical challenges and outline promising areas for future research for further progress in this field. This work is expected to be useful for scientists, engineers, and students working in the field of photovoltaics and materials science.

I. Thin films solar cells based on CIGS

1.1. Structure and properties of CIGS materials

The CIGS material family, which form the basis of high-efficiency thin-film solar cells, are solid solutions based on copper-indium diselenide ($CuInSe_2$, CIS) with partial substitution of indium with gallium ($CuIn_{1-x}GaxSe_2$, CIGS) or selenium with sulphur ($CuIn_{1-x}Ga_x(Se_{1-y}S_y)_2$, CIGS(S) or CIGSS). These compounds belong to I-III-VI semiconductors and crystallize predominantly in a tetragonal chalcopyrite structure (space group I$\bar{4}$2d) [13, 14]. The structure of chalcopyrite is a superstructure of the cubic structure of sphalerite (ZnS), in which the cation positions are occupied by Cu and In (or Ga) atoms in an orderly manner, and each selenium (or sulphur) atom is coordinated by two Cu atoms and two In/Ga atoms. The crystal lattice parameters vary slightly depending on the ratio of Ga/(In+Ga) and S/(S+Se), and an increase in the content of gallium or sulphur leads to a decrease in the a and c parameters due to the smaller atomic radii of Ga compared to In, and S compared to Se (Fig. 1) [15, 13].

One of the key advantages of CIGS materials is their optical properties. They are semiconductors with direct optical transitions, which ensure a high probability of photon absorption. The band gap (Eg) of CIGS materials can vary widely by changing their chemical composition. For pure $CuInSe_2$ (CIS, x=0), Eg is about 1.00-1.04 eV, while for $CuGaSe_2$ (CGS, x=1), Eg increases to about 1.68-1.70 eV. Thus, by changing the ratio x=[Ga]/([In]+[Ga]) from 0 to 1, it is possible to smoothly adjust Eg in the range which is optimal for absorbing the solar spectrum and creating tandem structures [16]. Typically, a composition of $x \approx 0.3$, corresponding to $Eg \approx 1.15-1.20$ eV, is used for high-efficiency single-crystal solar cells. A further increase in Eg is possible by replacing selenium with sulphur; for example, $CuInS_2$ has $Eg \approx 1.5$ eV, and $CuGaS_2$ has about 2.4-2.5 eV. It is very important that CIGS materials have a very high optical absorption coefficient (α) in the visible and near-infrared regions of the spectrum, reaching $10^4–10^5$ cm^{-1}. This makes it possible to effectively absorb most of the solar radiation in a layer with a thickness of a mere 1-2 μm, resulting in a significant reduction in material consumption.

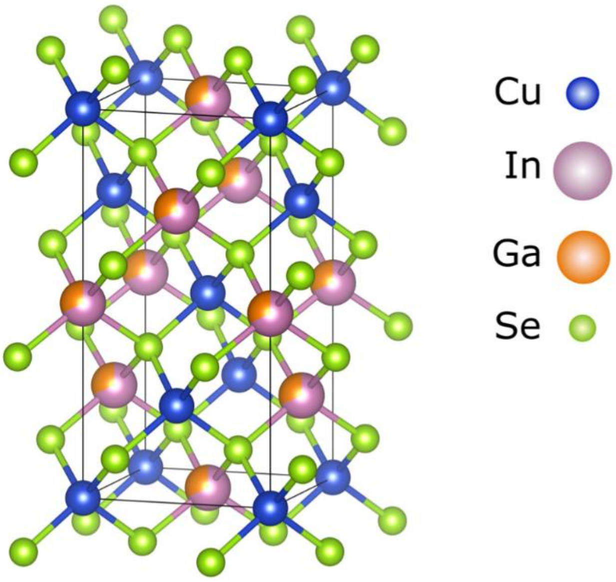


Fig. 1. Tetragonal structure of a CIGS-type compound [15].

The electrical properties of CIGS make it highly suitable for photovoltaic applications. Undoped CIGS materials typically exhibit p-type conductivity [17]. This type of conductivity is generally attributed to intrinsic defects in the crystal structure, particularly copper vacancies (VCu), which act as shallow acceptors. Other important intrinsic defects include (InCu, GaCu – donors), selenium vacancies (VSe – donor), and defect complexes, that can significantly influence the concentration and mobility of charge carriers, as well as the lifetime of minority carriers. The hole concentration in CIGS films used in solar cells usually ranges from 10^{15} to 10^{17} cm^{-3}. Hole mobility can vary, but typical values are within the range of 1–25 $cm^2V^{-1}s^{-1}$, depending on the composition, fabrication technique, and film microstructure [18].

The influence of the In/Ga and S/Se ratios on the properties is essential for optimising the parameters of solar cells. It was mentioned that increasing the Ga/(In+Ga) ratio leads to an increase in the width of the Eg band gap. This allows not only to adapt the Eg of the absorbing layer for maximum utilisation of the solar spectrum, but also to generate an Eg gradient across the thickness of the CIGS film (Fig. 2) [19]. For example, gallium enrichment of the back contact can form a quasi-electric field, which improves the collection of charge carriers (electrons) and reduces recombination at the back contact, while gallium enrichment of the surface layer (closer to the p-n junction) can increase the open-circuit voltage (Voc).

However, an excessively high gallium content can lead to a reduction in carrier lifetime and degradation of device performance due to the formation of deep defect levels or problems with the formation of a high-quality p-n junction. Replacement of selenium with sulfur also increases Eg, that can be useful for the upper cell in tandem solar cells or for devices operating at elevated temperatures. In addition, sulfur can affect the chemical stability and surface properties of CIGS, that is important for the formation of a high-quality heterojunction with a buffer layer. The optimal combination of components and their distribution throughout the film thickness is the subject of active research and optimization for each specific solar cell manufacturing process.

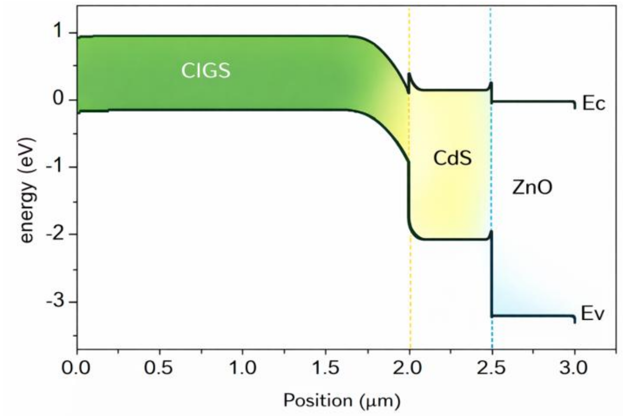


Fig. 2. Zone diagram of the CIGS structure.

1.2. Methods for the deposition of CIGS absorber layers

The quality of the Cu(In,Ga)Se_2 (CIGS) absorber layer is critical for achieving high solar cell efficiency. The properties of the CIGS film, such as stoichiometry, crystal structure, grain size, presence of defects, and distribution of elements (especially Ga and In) across the thickness, are largely determined by the method used to obtain it [20]. At present, a significant number of methods for depositing CIGS layers have been developed and are being studied, which can be conditionally divided into two large groups: vacuum and non-vacuum (chemical) methods.

1.2.1. Vacuum based methods

Despite the higher cost of equipment, vacuum technologies make it possible to obtain CIGS films with the best characteristics and the highest energy conversion efficiency.

The co-evaporation method is considered the benchmark technique for obtaining the highest quality CIGS films, and this method allows record efficiency values to be achieved for laboratory samples. The process consists of simultaneous or sequential thermal evaporation of constituent elements – copper (Cu), indium (In), gallium (Ga) and selenium (Se) (or sulphur (S) in the case of CIGS(S)) – from separate sources onto a heated substrate under high vacuum ($<10^{-3}$ Pa) (Fig. 3) [20]. A distinctive feature of this method is the ability to precisely control the evaporation rate of each element, which allows the formation of films with a predetermined chemical composition and the creation of a concentration gradient of Ga and/or In across the film thickness [21]. The most common approach is the “three-stage co-evaporation process”, which enables the formation of a high-quality microstructure and film composition. This technique delivers very high solar cell efficiency, excellent film quality, precise composition control with the possibility of compositional profiling, as well as the ability to “in-situ” film growth. However, its disadvantages include the high cost of equipment, the complexity of the process and its control, the relatively low deposition rate for large-scale production, and certain difficulties in scaling to very large areas while maintaining uniformity.

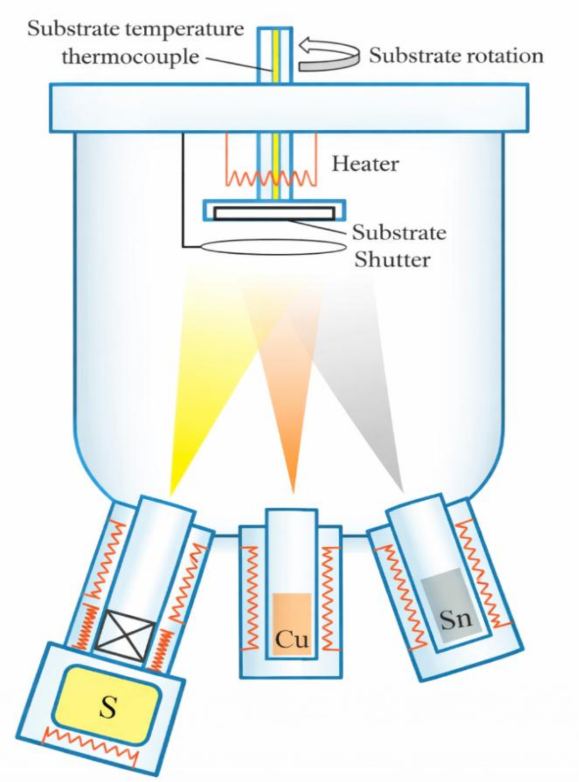


Fig. 3. Schematic representation of the co-evaporation method.

Another widely used vacuum-based technique is magnetron sputtering followed by selenisation/sulphurisation. This two- step approach is more prevalent in industrial production due to its better scalability and potentially lower cost. In the first step, metal precursors (individual metals or alloys) are deposited onto the substrate by magnetron sputtering. In the second step, the formed precursor is annealed at high temperatures (400-600 °C) in an atmosphere containing vapours of selenium, sulphur or their compounds (H_2S, H_2Se), leading to the formation of a CIGS(Se,S) film (Fig. 4) [20].

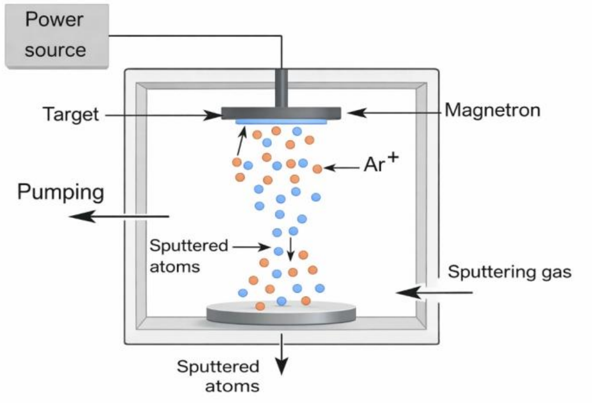


Fig. 4. Magnetron sputtering method.

This method is well suited for large-scale production, offering higher precursor deposition rates, potentially lower equipment and process costs, and good film adhesion. On the other hand, it is an “ex-situ” process, where control of stoichiometry and element distribution can be more challenging; secondary phases may form, and films often exhibit a higher defect density or smaller grain size compared to the best films obtained by co-evaporation, which may limit efficiency [22].

1.2.2. Non-vacuum (chemical) methods

Non-vacuum methods are considered a promising alternative as they have the potential to significantly reduce production costs and enable the use of continuous deposition technologies. However, the efficiency of solar cells fabricated by these methods is still generally lower.

Solution-based methods include electrodeposition, in which Cu, In, and Ga precursors are deposited onto a conductive substrate from an electrolyte under the influence of an electric current, followed by selenisation [23]. This method is characterised by low equipment costs and the ability to deposit over large areas, but it faces challenges in controlling composition, film uniformity, and possible incorporation of impurities [24]. Sol-gel methods involve preparing a precursor solution, applying it to the substrate, drying and heat treatment with selenisation. These methods allow good control of the stoichiometry in the solution, however, the resulting films may exhibit high porosity and contain residual organic contaminants.

Considerable attention is being directed towards nanoparticle-based methods or printing technologies that involve the synthesis of CIGS nanoparticles or precursors, the formation of “inks” from them, and their deposition using various printing techniques, followed by sintering and/or selenisation [25]. These methods are highly attractive due to their potential for high-throughput and low-temperature manufacturing, particularly on flexible substrates. However, the main challenges remain low film density, the presence of organic residues, small grain size and, as a result, the lower device efficiency.

In overall, non-vacuum methods offer advantages such as potentially low cost, high throughput, the possibility of processing under atmospheric conditions for certain steps, and compatibility with flexible substrates and roll-to-roll technologies [26]. Nevertheless, they are generally inferior to vacuum-based methods in terms of solar cell efficiency due to challenges in controlling microstructure, phase purity, potential impurities incorporation, and the challenge of obtaining films with high density and large grains.

The choice of a specific method for obtaining CIGS layers depends on many factors, including the desired efficiency, production scale, cost, and the specific application of solar cells. Research continues both in improving existing vacuum technologies to reduce their cost and in enhancing the quality of films and the efficiency of devices fabricated using non-vacuum methods [27].

1.3. Device structure and operating principles of CIGS based solar cells

The effective performance of a CIGS-based solar cell depends not only on the quality of the absorbing layer itself, but also on the optimal multilayer design of the entire device and the proper functioning of each of its components. A typical CIGS solar cell has a heterostructure comprising several sequentially deposited layers, each of which performs a specific function to ensure efficient light absorption, as well as the generation, separation and collection of charge carriers [22, 28].

The standard structure of such a device begins with a substrate that serves as a mechanical support (Fig. 5). Most commonly, sodium-calcium glass (SLG) is used for this purpose, as it is relatively inexpensive, thermally stable, and an important source of sodium (Na) diffusion into the CIGS layer during high-temperature processes, which helps to improve the overall cell efficiency [29]. Alternatively, polyimide films or thin metal foil are used for flexible solar cells [13]. A back contact is deposited onto the substrate, typically molybdenum (Mo) layer with a thickness of 0.5-1 μm, deposited by magnetron sputtering. Molybdenum provides a good ohmic contact with p-type CIGS, exhibits high electrical conductivity and chemical stability to selenium at high temperatures. A thin $MoSe_2$ layer often forms at the Mo/CIGS interface, which can improve the contact and reduce recombination [30].

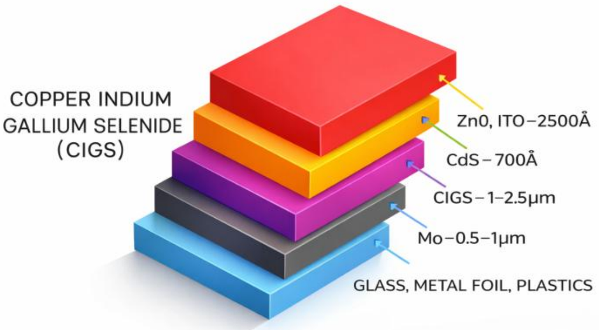


Fig. 5. Diagram of thin-film converters based on CIGS compounds.

The next layer is an absorbing layer of CIGS ($Cu(In,Ga)Se_2$) of p-type conductivity, with a thickness of 1-2.5 μm, where the absorption of sunlight and the generation of electron-hole pairs occur. It is followed by a buffer layer, usually a thin (30-100 nm) layer of cadmium sulphide (CdS) of n-type, deposited by chemical bath deposition (CBD method) [31]. This layer forms a p-n heterojunction with CIGS, protects its surface and optimises the energy band alignment to minimise recombination at the heterojunction. Due to environmental considerations and to improve optical properties, cadmium-free buffer layers such as Zn(O,S), ZnMgO or In_xS_y are being actively developed.

A transparent conductive oxide (TCO) serves as the front contact. It must be both highly transparent to sunlight and possess high electrical conductivity. A double-layer structure is often used: a thin layer of intrinsic zinc oxide (i-ZnO) to protect the buffer layer, and on top of it a thicker layer of aluminium-doped ZnO (AZO) or boron-doped ZnO (BZO), or indium tin oxide (ITO) [32]. As a rear contact, a metallic contact grid (typically made of nickel/aluminium (Ni/Al) or silver (Ag)) is applied onto the TCO to ensure efficient current collection and reduce the series resistance of the solar cell.

The mechanisms of charge carrier generation and recombination are key to understanding how a solar cell operates. When photons with energy exceeding the band gap width Eg of the CIGS material are absorbed in it, electron-hole pairs are generated. The built-in electric field of the p-n heterojunction formed at the interface between CIGS (p-type) and the buffer layer (n-type) separates these pairs: electrons move towards the n-region (buffer layer and TCO), while holes move towards the p-region (back contact). This directed movement of charges creates a photocurrent in the external circuit.

However, not all generated charge carriers contribute to the photocurrent. Some of them recombine, that is, the electron and hole "annihilate", giving up their energy as heat or light. Recombination processes are the main mechanism of losses in CIGS solar cells and may occur in different regions of the device. These include recombination within the absorber bulk through defects or at grain boundaries, recombination at the CIGS/buffer interface, often dominant, recombination at the back contact, and recombination within the space charge region of the heterojunction. Minimising these recombination losses by improving material quality, optimising device structure, and passivating defects is a key objective for further increasing the efficiency of CIGS-based solar cells [33].

1.4. Efficiency and stability of CIGS solar cells

$Cu(In,Ga)Se_2$ (CIGS) solar cells have firmly established themselves as one of the leading thin-film photovoltaic technologies, largely due to their high energy conversion efficiency (efficiency) and potential for further improvement. To date, laboratory-scale CIGS solar cells of small area have demonstrated efficiencies exceeding 23%, for example, 23.35% was achieved by the research group Solar Frontier, and 23.6% has been reported for devices incorporating silver (Ag) doping [34]. For commercial modules, the efficiency is typically ranges from 15 to 19% (Table 1)), which is a competitive level in the photovoltaic market. Such high efficiencies result from the optimisation of key photovoltaic parameters: open-circuit voltage (Voc), short-circuit current density (Jsc) and the fill factor (FF).

The high short-circuit current density (Jsc) in CIGS cells (often exceeding 35-40 mA/cm^2) is achieved due to the broad spectral absorption of CIGS material, which arises from its direct band gap and high absorption coefficient, as well as the efficient collection of generated charge carriers [35]. The open-circuit voltage (Voc), which exceeds 0.70-0.75 V in the best devices, depends on the bandgap width Eg of the absorber, and the level of recombination losses, particularly in the CIGS/buffer heterointerface and within the absorber bulk [18]. Optimisation of the Ga/(In+Ga) composition gradient across the CIGS layer thickness, which enables the creation of a favourable Eg profile, plays a crucial role in maximising both Jsc (through improved carrier collection) and Voc (through reduced recombination at the back contact and optimisation of Eg near the heterojunction). The fill factor (FF), which characterises the "squareness" of the current-voltage characteristic and exceeds 75–80% in high-quality CIGS cells, depends on the series and parallel (shunt) resistances of the solar cell, as well as the diode characteristics of the p-n junction [36]. Reducing the defect density in the CIGS material, improving heterojunction quality, optimising contacts and minimising the resistance of the transparent conductive oxide are key factors for achieving a high FF. Controlled dopant incorporation, particularly of sodium (Na) either from the substrate or by targeted doping, also plays a crucial role in enhancing efficiency by promoting grain boundary defect passivation and improving the electronic quality of the absorber [37].

Along with high efficiency, stability and long-term reliability are critically important parameters for any photovoltaic technology aiming for large-scale commercial application. CIGS solar cells generally exhibit good stability, however, certain degradation mechanisms

Table 1.

Record efficiencies of CIGS-based solar cells

Cell Type / Configuration	Record Efficiency (%)	Notes	References
Single-junction CIGS	23.64	Laboratory-scale, small-area; current certified world record	Uppsala Univ., Nature Energy / Fraunhofer ISE (SciDaily, 2024)
Previous top CIGS	23.35	Lab element by Solar Frontier	PV Magazine, 2024 (pv-magazine.com)
Perovskite-CIGS tandem (monolithic, certified)	24.6	Two-terminal tandem; bottom CIGS, top perovskite	Fraunhofer ISE / SciDaily, 2025 (sciencedaily.com)
Perovskite-CIGS flexible tandem	23.64	Flexible substrate (polyimide / metal foil)	KIER, Joule / PV Magazine, 2025 (pv-magazine.com)
Perovskite-CIGS 2-terminal tandem	24.6	Lab-scale, two-terminal design	Chinese research / PV Magazine, 2025 (pv-magazine.com)
Perovskite-CIGS 4-terminal tandem	26.2	Lab-scale, four-terminal architecture	University of Tokyo / Perovskite-Info, 2025 (perovskite-info.com)
Commercial CIGS module	20.3	Certified large-area module (~30×30 cm)	Avancis / PV Magazine, 2023 (pv-magazine.com)

can limit their lifetime or performance under real operating conditions. One of the major issues is sensitivity to moisture and oxygen, especially in the presence of ultraviolet (UV) radiation and elevated temperatures [20]. Moisture ingress can lead to corrosion of metal contacts (particularly the molybdenum back contact), degradation of the TCO layer, as well as chemical changes within the CIGS absorber or at its surface/heterojunction. For instance, moisture may react with CIGS components, causing the formation of oxides or hydroxides, which deteriorates electronic properties. Oxygen exposure can also affect the defect structure and surface states of CIGS [27].

To enhance the stability of CIGS solar cells, several approaches are employed. The most important of these is ensuring proper encapsulation of the module which prevents the ingress of moisture and oxygen into the active layers. This involves the use of special barrier films, sealants, and structural design solutions. In addition, research is being conducted to develop chemically more stable buffer layers and TCOs, as well as to modify the CIGS layer itself to improve its intrinsic stability [38]. For example, the use of dopant elements or specific surface treatments can enhance resistance to oxidation. Contact stability is also important, in particular, alternative materials for the back contact or protective coatings for molybdenum are being investigated [39]. Although light-induced degradation is not as pronounced in CIGS as in some amorphous silicon elements, for example, prolonged exposure to light and elevated temperature can cause gradual changes in properties related to atomic diffusion or defect state alteration, which remain subjects of ongoing research [40]. Overall, with appropriate encapsulation and design, CIGS modules exhibit long-term stability, as confirmed by field test results and commercial operation over many years.

1.5. Recent advances and challenges for CIGS-based solar cells.

The technology of thin-film solar cells based on $Cu(In,Ga)Se_2$ (CIGS) continues to advance actively, demonstrating significant recent achievements while facing certain challenges that need to be overcome for its wider adoption and enhanced competitiveness. Research efforts are focused on further improving efficiency, reducing manufacturing costs, enhancing long-term stability, and expanding application areas, including through the development of flexible photovoltaic devices.

One of the key directions remains the improvement of energy conversion efficiency. Recent breakthroughs include achieving laboratory efficiencies exceeding 23%, made possible through improved manufacturing processes and a deeper understanding of the material and device physics [34].

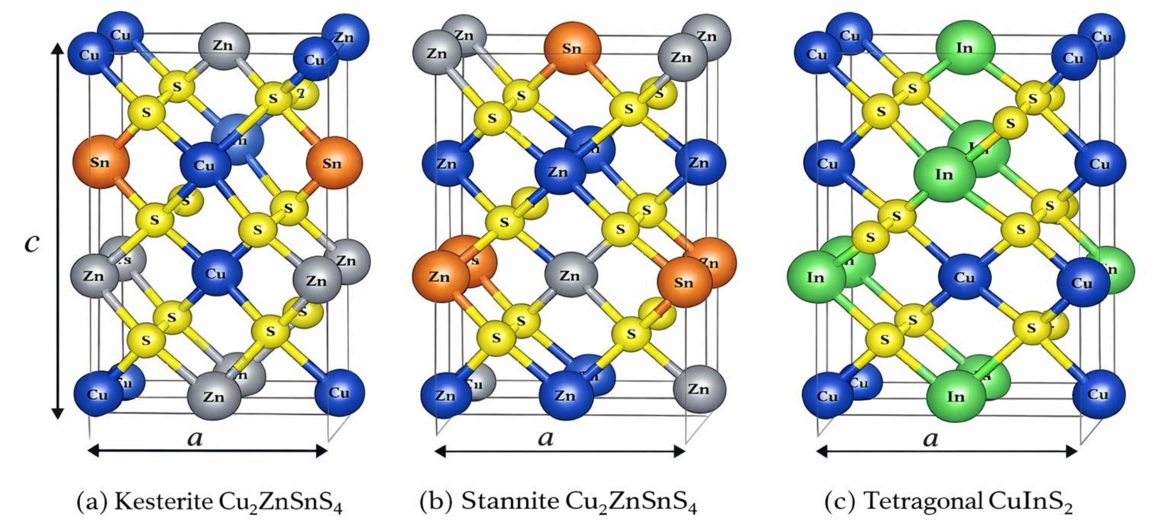
This includes the optimization of Ga/(In+Ga) composition gradients across the thickness of the absorber layer to improve carrier collection and increase open-circuit voltage, the use of post-deposition treatments (e.g. treatment with alkali metals such as potassium (K) or caesium (Cs) in addition to sodium (Na)), that passivate defects on the surface and within the bulk of CIGS, as well as the development and implementation of new or modified buffer layers for better band alignment and reduced recombination at the heterojunction [41]. Deposition methods themselves are also being improved, for instance, through the development of faster co-evaporation processes or modified approaches to the selenisation of sprayed precursors to enhance film quality while simultaneously increasing production throughput [42].

Despite the high efficiency, a significant challenge for CIGS technology is the cost and availability of some of its components, particularly indium (In) and gallium (Ga). These metals are relatively rare and their extraction is often associated with the production of other metals, making their prices unstable and creating potential risks for sustainable large-scale manufacturing of solar cells. Consequently, considerable efforts are being directed towards reducing the use of these materials. One approach is to develop ultra-thin CIGS absorber layers (thickness below 1 μm) without substantial loss of efficiency, which requires very high material quality and optimized light absorption strategies. Other directions include exploring the recycling of indium and gallium from end-of-life modules or production waste, as well as investigating ways to partially substitute indium and gallium with more abundant elements, although the latter is extremely challenging due to the unique role of these elements in achieving the desired electronic properties of CIGS [20].

Significant progress has been made in the development of flexible CIGS solar cells, which open new opportunities for integrating photovoltaics into various structures where rigid glass modules are unsuitable [13]. Flexible CIGS cells are fabricated on polymer substrates (e.g., polyimide) or thin metal foil (e.g., stainless steel) (Fig. 6). The efficiency of flexible CIGS solar cells has already reached values close to those achieved on glass substrates, exceeding 20-22% under laboratory conditions [28]. However, there are specific challenges associated with flexible substrates, such as ensuring reliable layer adhesion, managing mechanical stresses during bending, differences in thermal expansion coefficients between the layers and the substrate, as well as the need for effective barrier layers to protect against moisture and oxygen ingress through the flexible substrate, which may be less hermetic than glass. Successful solutions to these challenges would enable the expansion of CIGS applications in portable electronics, transportation, aerospace, and building-integrated photovoltaics (BIPV).

Fig. 6. Appearance of a flexible CIGS-based solar cell.

Despite the remarkable progress achieved, the main challenges for CIGS technology remain further reduction



(a) Kesterite Cu_2ZnSnS_4 (b) Stannite Cu_2ZnSnS_4 (c) Tetragonal $CuInS_2$

Fig. 7. Crystal structures of (a) kesterite-CZTS, (b) stannite-CZTS, and (c) tetragonal-CIS.

of module production costs to successfully compete with the mass production of silicon-based solar cells, the scaling up of the most efficient laboratory processes to industrial levels without quality loss, and the ensuring of guaranteed long-term stability and reliability of modules under various climatic conditions throughout their service life [43]. Overcoming these challenges requires integrated efforts in materials science, device engineering, and manufacturing technologies.

II. Thin films solar cells based on CZTS

2.1. Structure and properties of CZTS materials

Materials of the CZTS type, which include Cu_2ZnSnS_4 (CZTS), $Cu_2ZnSnSe_4$ (CZTSe) and their solid solutions $Cu_2ZnSn(S,Se)_4$ (CZTSSe), have attracted significant attention as promising absorber layers for thin-film solar cells due to their composition of abundant and non-toxic elements [44]. These compounds belong to the class of complex chalcogenides and are structural analogues of CIGS, in which the In/Ga atomic pairs are replaced by Zn/Sn.

The most stable and photoelectrically active Crystalline structure for CZTS(Se) is considered to be the kesterite structure, which belongs to the tetragonal crystal system (space group $I\bar{4}$) (Fig. 7). It is derived from the chalcopyrite structure, where two cation sublattices of chalcopyrite are further divided into sublattices of Zn and Sn atoms. There also exists another energetically close crystalline structure – the stannite structure (space group $I\bar{4}2m$), which differs in the arrangement of Cu and Zn atoms within the cation sublattice [45]. The energy difference between these two phases is very small, making it difficult to distinguish them experimentally, using techniques such as X-ray diffraction, especially in the presence of cation disorder between Cu and Zn sites (Cu↔Zn antisite defects). However, theoretical calculations indicate that the kesterite phase is more stable for CZTS [46].

One of the most significant challenges in synthesising high-quality CZTS(Se) films is ensuring phase purity. These materials have a very narrow homogeneity (phase stability) range in the phase diagram (Fig. 8). Small deviations from stoichiometry or suboptimal synthesis conditions can easily lead to the formation of various secondary phases, such as binary sulphides/selenides (ZnS(Se), $Cu_xS(Se)$, $SnS_y(Se)_y$) or ternary compounds (e.g., Cu_2SnS_3 (Se_3) – CTSE). The presence of these secondary phases, even in small amounts, can significantly degrade the photovoltaic performance of solar cells, as they may act as recombination centres, create shunting paths, or deteriorate the properties of the heterojunction [47]. Therefore, precise control over the composition and synthesis conditions is critical for obtaining single-phase CZTS(Se) films.

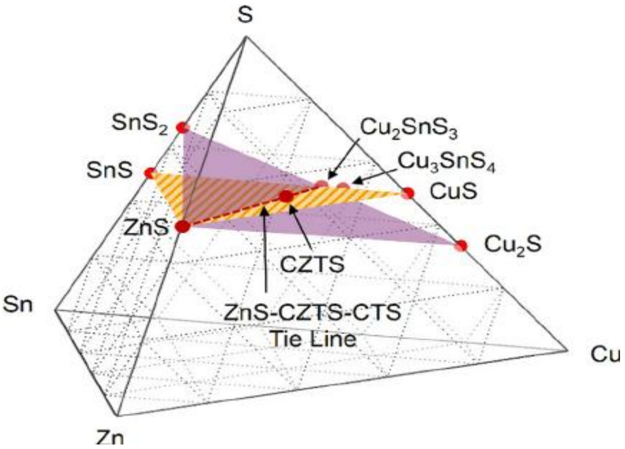


Fig. 8. Quaternary phase diagram of CZTS, showing known phases. Two pseudo- ternary planes, each containing CZTS, are depicted as intersecting striped yellow and solid purple planes. The planes intersect along the connecting line ZnS-Cu2ZnSnS4-Cu2SnS3, shown as a dashed red line [48].

From the perspective of optical properties, CZTS(Se) materials are semiconductors with a direct type of optical transition, which is favourable for efficient light absorption. An important advantage is the possibility to tune the band gap (Eg) by varying the sulfur and selenium ratio in the solid solution $Cu_2ZnSn(S_xSe_{1-x})_4$. For pure sulphide CZTS (x=1), Eg is approximately 1.45-1.55 eV, which is close to the optimal value for a single-junction solar cell, whereas for pure selenide CZTSe (x = 0), Eg decreases to approximately 0.95-1.05 eV [48]. This allows for optimisation of solar spectrum absorption. Similar to CIGS, CZTS(Se) materials exhibit a high optical absorption coefficient ($\alpha > 10^4$ cm^{-1}) in the visible spectral range, enabling the use of thin absorbing layers (1-2 μm) [18].

The electrical properties of CZTS(Se) are determined by a high concentration of intrinsic point defects. Undoped materials typically exhibit p-type conductivity, which is believed to originate mainly from copper vacancies (V_{Cu}) and CuZn substitution defects (antisite defects) CuZn (a copper atom occupying a zinc site), both acting as acceptors [49]. However, donor-type defects such as ZnCu (a zinc atom on a copper site) are also readily formed in CZTS(Se), leading to conductivity compensation. A high concentration of various intrinsic defects and their complexes (e.g., neutral defect clusters [CuZn + ZnCu]) is a characteristic feature of these materials, leading to pronounced band tailing near the band edges and a high density of deep defect states. This, in turn, is considered the primary cause of the large open-circuit voltage deficit (V_{oc} deficit = $E_g/q - V_{oc}$), which remains a major factor limiting the efficiency of CZTS(Se)-based solar cells compared to CIGS. Typical charge carrier (hole) concentrations in CZTS(Se) films range from 10^{15} to 10^{17} cm^{-3}, and the carrier mobility is generally lower than that of CIGS, about 1-10 cm^{2}V^{-1}s^{-1} [50].

Despite the mentioned challenges, the main driving force behind research on CZTS(Se) is the advantages associated with the abundance and non-toxicity of its constituent elements. Copper, zinc, tin, and sulphur are widely available and relatively inexpensive elements in the Earth's crust. Selenium is less abundant than sulphur, but significantly more available and cheaper than indium and gallium, which are used in CIGS [36]. In addition, the components of CZTS(Se) are significantly less toxic than cadmium (Cd), which is used in CdTe solar cells and as a standard CdS buffer layer for many thin-film technologies, or tellurium (Te) [51]. These factors make CZTS(Se) materials highly attractive in terms of potentially reducing the cost of solar energy and ensuring sustainable, environmentally safe large-scale production of photovoltaic modules in the future [7].

2.2. Methods for the deposition of CZTS absorber layers

The synthesis of high-quality $Cu_2ZnSn(S,Se)_4$ (CZTSSe) absorbing layers is considerably more complex task compared to CIGS due to the multicomponent nature of the system (four or five elements) and the narrow phase stability region of the kesterite phase. The choice of deposition method and precise control of process parameters play a crucial role in achieving the desired stoichiometry, phase purity, microstructure and, ultimately, efficiency of solar cells. Similar to CIGS, the methods for obtaining CZTSSe layers can be divided into vacuum-based and non-vacuum-based techniques.

2.2.1. Vacuum-based methods

Vacuum technologies make it possible to obtain CZTSSe films with relatively high efficiency, although they are associated with certain technological challenges.

One of the main vacuum methods is co-evaporation, in which the elements Cu, Zn, Sn and S/Se are evaporated simultaneously or sequentially from separate sources onto a heated substrate under high vacuum conditions (Fig. 9). This method potentially allows precise control of the fluxes of each element and the formation of films with a predetermined composition profile. However, for the CZTS(Se) system, it is particularly challenging due to the need for simultaneous control of four or five components with different volatilities, especially the high volatility of tin (in the form of SnS or SnSe) and chalcogens (S, Se) at synthesis temperatures. The loss of tin and chalcogens may lead to the formation of secondary phases and deviations from the desired stoichiometry. Despite these difficulties, methods based on co-evaporation or its combination with other approaches often enable the achievement of record efficiency values for CZTSSe solar cells [52].

A more common vacuum-based method is magnetron sputtering of metal precursors followed by sulphurisation or/and selenisation [53]. In the first stage, metallic layers are deposited onto the substrate by magnetron sputtering in a specific sequence (e.g., Cu/Sn/Zn or Zn/Sn/Cu) or as alloy layers (e.g., Cu-Zn, Cu-Sn) (Fig. 10). Afterwards, the metallic precursors are annealed at high temperatures (typically 500-600°C) in an atmosphere of sulphur and/or

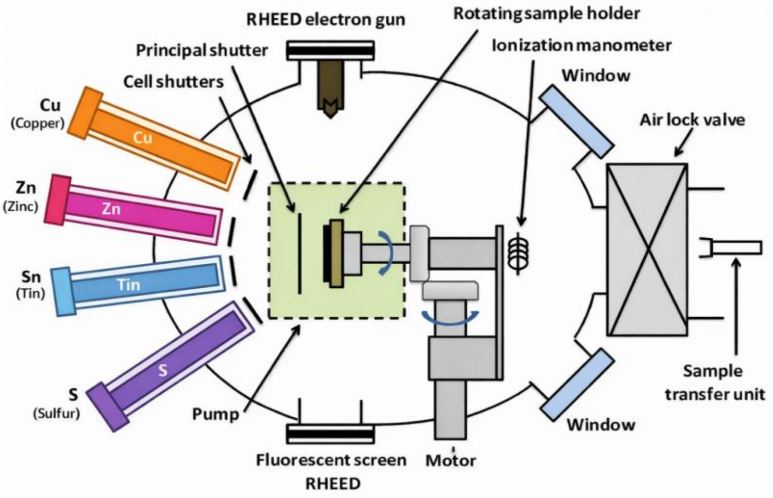


Fig. 9. Schematic representation of the co-evaporation method for CZTS-type heterostructures.

selenium (or their compounds, such as H_2S, H_2Se) to form the CZTS(Se) kesterite phase [54]. This two-step approach is more attractive for industrial application due to its better scalability and potentially lower cost of the sputtering stage. However, there are still significant challenges associated with controlling the annealing process: ensuring complete and uniform reaction between all components, preventing the loss of volatile tin compounds (e.g., SnS), controlling the S/Se ratio in the film, and avoiding the formation of undesirable secondary phases such as ZnS(Se) on the surface or at the back contact interface, as well as copper and tin sulphides/selenides.

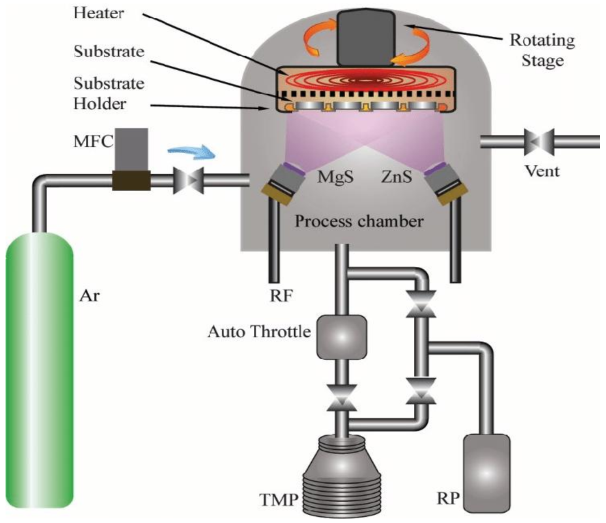


Fig. 10. Schematic illustration of the magnetron sputtering process for the deposition of a CZTS thin film. Argon ions (Ar^+) generated by a plasma discharge bombard Cu, ZnS and SnS targets, causing the release of atoms that condense on a heated substrate. After sputtering, the film is thermally annealed in a sulphur atmosphere to form the crystalline kesterite phase.

2.2.2. Non-vacuum (chemical) methods

Non-vacuum methods are being actively developed to significantly reduce the production cost of CZTS(Se) solar cells by eliminating expensive vacuum equipment and employing potentially high-throughput solution or suspension - based deposition processes [55].

Among these, sol-gel methods (Fig. 11) are notable, that involve the preparation of a solution containing metal salts or alkoxides (Cu, Zn, Sn), depositing this solution onto a substrate (for example, by spin -coating or dip coating), drying it, performing pyrolysis to remove organic components, and subsequently carrying out high-temperature treatment in a sulfur/selenium atmosphere. This method allows for good control of stoichiometry at the solution preparation stage and is relatively simple to implement. However, challenges remain including possible crack formation, high porosity and incomplete removal of carbon residues from organic precursors, which can negatively affect the properties of the material [56].

Other solution-based approaches include chemical bath deposition or electrodeposition of metallic precursors followed by annealing in a chalcogen-containing atmosphere. Electrodeposition enables the formation of Cu-Zn-Sn alloy layers directly on a conductive substrate, which is economically attractive. However, it is difficult to precisely control the composition of the multicomponent alloy due to the difference in the electrochemical potentials of the metals and possible contamination of the films by electrolyte components [57].

Particularly popular are nanoparticle-based (ink technologies) methods. They involve the preliminary synthesis of CZTS(Se) nanoparticles or binary/ternary

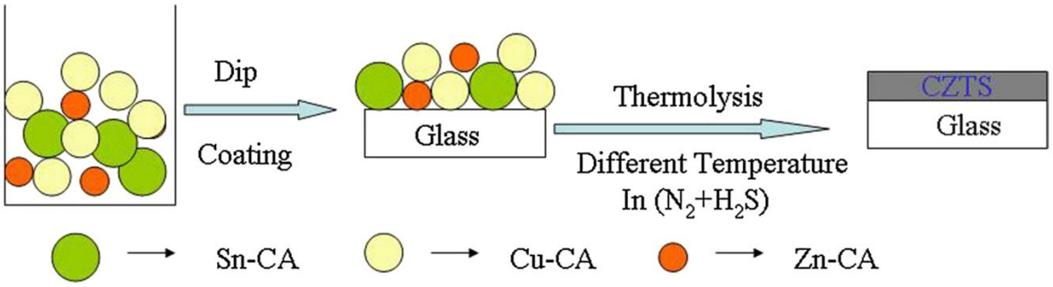


Fig. 11. Schematic diagram of sol–gel sulfurisation process of CZTS thin films [57].

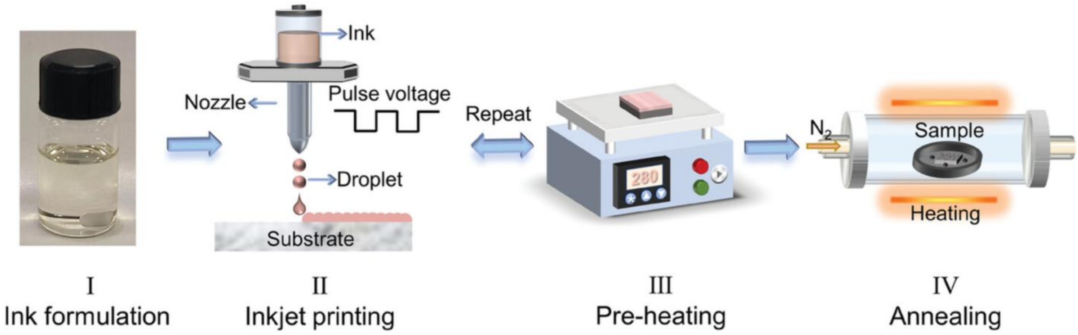


Fig. 12. Schematic illustration of inkjet printing process for thin-film nanostructures [58].

chalcogenide precursors, the preparation of stable suspensions ("inks") from them, and the deposition of these inks onto a substrate using various printing techniques (inkjet printing, screen printing, spray coating, the Dr. Blade method) (Fig. 12) [58]. After the nanoparticle layer is deposited, it is subjected to sintering and, if necessary, additional sulphurisation/selenisation to improve crystallinity and phase composition. These methods are considered promising for roll-to-roll production and the fabrication of flexible solar cells. The main challenges include achieving high film density, ensuring good inter-grain contact, a complete removal of organic ligands and solvents used for nanoparticle stabilisation, and controlling grain growth during sintering [59].

Precise control of stoichiometry and phase composition is critically important for all methods of CZTS(Se) deposition methods. Studies have shown that to achieve better photovoltaic performance, CZTS(Se) films should exhibit a slightly copper-poor and zinc-rich composition (i.e., $[Cu]/([Zn]+[Sn]) < 1$ and $[Zn]/[Sn] > 1$) [60]. Such a composition helps to suppress the formation of undesirable copper-rich secondary phases (e.g., $Cu_xS(Se)$), which are conductive and can shunt the p-n junction, and also promotes the growth of larger grains. In furthermore, precise control of the partial pressure of sulphur/selenium and temperature during high-temperature treatment is also essential to ensure the complete conversion of precursors into the kesterite phase, to regulate the S/Se ratio, and prevent the decomposition of CZTS(Se) or the loss of volatile components, particularly SnS(Se). Multi-step annealing processes or special conditions (for example, the use of SnS vapour retaining caps) are often applied to improve the phase purity and morphology of the films [57].

2.3. Device structure and operating principles of CZTS-based solar cells

The structure of solar cells based on $Cu_2ZnSn(S,Se)_4$ (CZTSSe) is largely inherited from the more mature CIGS technology, given the similarity of these materials as p-type absorbers in thin-film heterostructures. A typical device features a multilayer configuration, in which each layer performs a specific function to ensure efficient solar energy conversion. However, due to differences in the material properties of CZTSSe compared to CIGS, specific challenges arise, particularly at the heterojunction with the buffer layer, which significantly affect the operation and overall efficiency of the device [61].

The standard layer sequence in a CZTSSe solar cell is shown in Figure 13. The layer sequence is formed on a substrate similar to that used in CIGS devices. The process begins with a substrate, most commonly sodium-calcium glass (SLG), which serves not only as a mechanical support but also as a source of sodium, positively influencing the properties of the CZTSSe absorber layer by promoting grain growth and defect passivation. A back contact, traditionally made of molybdenum (Mo), is deposited onto the substrate to provide an ohmic contact to the p-type CZTSSe layer. However, during the high-temperature synthesis of CZTSSe, the interaction between Mo and sulphur/selenium can lead to the formation of a MoS_xSe_y interfacial layer. While a thin $MoSe_2$/MoS_2 layer may be beneficial for improving contact quality, excessive formation, especially in purely sulphide-based CZTS, can increase series resistance and deteriorate adhesion [62].

The central element is the p-type CZTS(Se) absorber layer, where electron-hole pairs are generated under illumination. As discussed earlier, achieving a high quality of this layer in terms of phase purity, stoichiometry, and defect minimisation is a key but challenging task [60, 63]. Above the absorbing, an n-type buffer layer is formed, which together with CZTS(Se) creates the main p-n heterojunction. Historically, as with CIGS, cadmium sulphide (CdS) deposited by chemical bath deposition (CBD) has been most commonly used, providing the best efficiency results for CZTSSe solar cells [64].

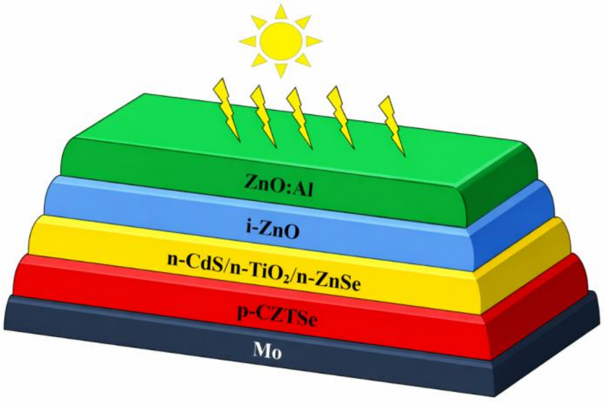


Fig. 13. Structure of a CZTSSe-based photovoltaic device [97].

However, the CZTS(Se)/buffer layer heterojunction remains one of the most problematic regions in these solar cells and is considered a primary source of losses that limit the open-circuit voltage (Voc). The main issue lies in the suboptimal alignment of energy bands, particularly the position of the conduction band offset (CBO). For pure sulphide CZTS, which has a relatively high electron affinity, the heterojunction with CdS is often characterised by a so-called 'cliff-like' conduction band offset (Ec, CZTS < Ec, CdS), contributing to a high recombination rate at the heterojunction. For CZTSe or selenium-rich CZTSSe, which have lower electron affinity, the offset may be small spike-like or nearly flat, which is more favourable but still not ideal. Improper band alignment enhances electron -hole recombination at the interface, directly reducing Voc and overall efficiency. Therefore, considerable research efforts focus on finding and developing alternative cadmium-free buffer layers with better conducting band alignment with CZTS(Se), such as ZnS, Zn(O, S), In_2S_3, $Zn_{1-x}Sn_xO_y$ (ZTO), TiO_2, and others. However, the design of such buffers is challenging because it requires not only optimal CBO, but also good coverage of the CZTS(Se) surface (which can be quite rough), chemical stability, sufficient n-type conductivity, and minimal light absorption.

Above the buffer layer lies the transparent conductive oxide (TCO), typically bilayer, consisting of a thin undoped i-ZnO layer and a thicker ZnO layer doped with aluminium (AZO) or boron (BZO), or indium tin oxide (ITO) (Fig. 13) [65]. This layer serves as the front electrode, collecting electrons and allowing light to pass

Table 2.

Reported best efficiencies of CZTS- and CZTSSe-based thin-film solar cells

Absorber Material	Best Reported Efficiency (PCE)	Cell Type / Area	Key Remarks	Reference
CZTS (Cu_2ZnSnS_4)	~7.6 %	Rigid substrate, ~1 cm²	Sulfide-only absorber, strong recombination losses	Wang et al., *Prog. Photovolt.*, 2014
CZTS	~10.0 %	Laboratory cell	First CZTS device exceeding 10 % efficiency	Shin et al., *Adv. Energy Mater.*, 2013
CZTS (flexible)	~11.4 %	Flexible substrate	Demonstrated on polyimide foil	Kim et al., *Prog. Photovolt.*, 2019
CZTSSe ($Cu_2ZnSn(S,Se)_4$)	12.6 %	Lab-scale (<1 cm²)	Partial Se substitution improves grain growth	Wang et al., *Energy Environ. Sci.*, 2014
CZTSSe	12.7 %	Record laboratory device	Reported by Solar Frontier	NREL Best Research-Cell Chart
CZTSSe	13.0–13.5 %	Small-area research cells	Interface and defect engineering	Giraldo et al., *Prog. Photovolt.*, 2019
CZTSSe	~14.0–14.9 % (reported)	Experimental reports	Advanced passivation & bandgap grading	Haight et al., *Appl. Phys. Lett.*, 2016; recent reviews
CZTSSe (large-area)	~10–11 %	>1 cm²	Scalability remains challenging	Siebentritt & Schorr, *Prog. Photovolt.*, 2012

through to the absorber. The device is completed by a metallic contact grid (e.g., Ni/Al or Ag) deposited on the TCO to reduce series resistance and efficiently extract current.

The operating principles of a CZTS(Se)-based solar cell are similar to those of other photovoltaic devices. Photon absorption in the CZTS(Se) layer generates electron-hole pairs. The built-in electric field of the p-n heterojunction separates these carriers: electrons move through the buffer layer to the TCO, and holes move to the back contact. This directed flow of charges produces a photocurrent. The efficiency of this process depends on the balance between the generation, transport, and recombination of charge carriers. As mentioned, minimising recombination losses, especially at the CZTS(Se)/buffer heterojunction, is a key task for further improving these solar cells [66]. Therefore, heterojunction engineering and surface passivation of CZTS(Se) play a crucial role in enhancing the performance of devices based on these promising materials.

2.4. Efficiency and stability of CZTS solar cells

Despite significant interest due to the abundance and environmental safety of components, the energy conversion efficiency of solar cells based on $Cu_2ZnSn(S,Se)_4$ (CZTSSe) is still inferior to more mature thin-film technologies such as CIGS or CdTe. However, substantial progress has been made in recent years. Record efficiency for laboratory -scale solar cells based on pure CZTS sulphide has reached approximately 10-12% [57], while for CZTSSe, where part of the sulphur is substituted with selenium, the efficiency is significantly higher, approaching 10-14.9% (Table II). Substituting sulphur with selenium generally improves crystallinity of the film, reduces defect concentration and provides better energy band alignment at the heterojunction, all of which contribute to the higher efficiency [69].

Despite these achievements, the efficiency of CZTSSe solar cells is still significantly limited by a number of fundamental and technological factors. The primary limitation is the large open-circuit voltage deficit ($V_{oc\ deficit} = E_g/q - V_{oc}$), which is substantially higher for CZTSSe than for CIGS with a comparable bandgap [56]. This large Voc deficit arises from several factors. First, it is due to the high concentration of intrinsic point defects and their clusters within the absorber layer, such as CuZn (copper on a zinc site) and ZnCu (zinc on a copper site) antisite defects, copper vacancies (V_{Cu}), chalcogen vacancies (V_S, V_{Se}) and more complex defect complexes, e.g., [CuZn + ZnCu]. These defects create deep energy levels within the band gap and induce a significant density-of-states tailing at the band edges, which act as efficient non-radiative recombination centers for electron-hole pairs. Secondly, a significant factor limiting the open-circuit voltage (Voc) and overall efficacy is recombination at the CZTS (Se)/buffer layer heterojunction [60]. As discussed in the previous subsection, a non-optimal conduction band offset (CBO) between CZTS (Se) and the conventional CdS buffer layer often results in an increased recombination rate at this interface. Thirdly, recombination at grain boundaries can also contribute to losses, although some studies indicate that grain boundaries in CZTS may be less recombinantion active than those in CIGS, or can be effectively passivated, especially in the presence of sodium. However, when the grain size is small, which is often observed in films obtained by non-vacuum methods, the effect of intergranular recombination can be significant.

Another important limiting factor is the presence of secondary phases in the absorbing layer, such as ZnS (Se), Cu_xS (Se), SnS_y $(Se)_y$ or Cu_2SnS_3 (Se_3) (CTSE). These phases may form due to slight deviations from the optimal stoichiometry or uncontrolled synthesis conditions and can act as recombination centers, create shunting paths or degrade the quality of the p-n junction, thereby negatively affecting all photovoltaic parameters: Voc, short-circuit current density (Jsc) and fill factor (FF) [67]. A low fill factor (FF) is also commonly observed in CZTS (Se) solar cells and may be associated with high series resistance (arising from contacts, the bulk resistivity of the film, or poorly conductive secondary phases) or low shunt resistance (due to defects or conductive secondary phases). Precise control of film stoichiometry, particularly maintaining a slightly copper-poor and zinc-rich composition, is critical for minimizing the formation of secondary phases and optimizing material properties [68].

Regarding the stability of CZTS(Se)-based solar cells, they still require detailed investigation, although the kesterite structure is considered to be sufficiently chemically stable. As in other thin film technologies, potential degradation mechanisms may involve interactions with environmental components such as moisture and oxygen, especially at elevated temperatures and under UV radiation. These factors can lead to surface oxidation of CZTS(Se), degradation of contacts, or changes at the heterojunction interface. There are also concerns about possible long-term phase instability or material decomposition, particularly if the initial film contains metastable phases or significant deviations from stoichiometry [68]. The loss of volatile components such as tin (in the form of SnS/SnSe) or chalcogens during prolonged operation at elevated temperatures may also pose a problem if the device is poorly encapsulated. Interaction with the back contact, for example, excessive formation of the MoS_xSe_y layer, can also affect long-term stability [64]. Efforts to enhance the stability of CZTS(Se) solar cells include the development of effective encapsulation methods, optimisation of interfacial boundaries, use of protective coatings, and synthesis of more stable and phase-pure absorber layers. Further improvement of efficiency and ensuring long-term reliability remain key research directions for enabling CZTS(Se) technology to realise its potential as a cost-effective and environmentally friendly solution for solar energy applications.

2.5. Recent advances and challenges in CZTS-based solar cells

Despite existing efficiency limitations, $CuZnSn(S,Se)_4$ (CZTSSe)-based solar cells remain the subject of intensive research due to their potential as an environmentally friendly and cost-effective alternative to other photovoltaic technologies. Current scientific efforts are focused on overcoming key issues limiting the performance of these devices, particularly the large open-circuit voltage deficit (Voc), and on developing technologies for obtaining high-quality materials and efficient device architectures [70].

One of the most important directions is improving the quality of CZTS(Se) absorber layers. This is achieved through several strategies. Doping is one of the most effective approaches. Special attention has been given to doping with alkali metals such as sodium (Na), potassium (K), rubidium (Rb) and caesium (Cs). These elements, similar to their role in CIGS, can contribute to the passivation of defects at grain boundaries and within the bulk, enhance grain growth, increase carrier concentration, and overall improve efficiency. For example, post-deposition treatment of CZTSSe films with potassium fluoride (KF PDT) has been shown to significantly enhance Voc and FF [71]. The effect of doping with other elements, such as silver (Ag), lithium (Li), germanium (Ge) or antimony (Sb), on the optical, electrical and structural properties of CZTS(Se) is also being investigated [60].

An integral part of improving the quality of the layers is precise control of the chemical composition and stoichiometry. Maintaining an optimal copper-poor and zinc-rich composition ($[Cu]/([Zn]+[Sn]) < 1$ and $[Zn]/[Sn] > 1$) is critical for reducing the formation of secondary phases [56]. Advanced synthesis methods and precursor designs are being developed to achieve better composition homogeneity throughout the film and to control the S/Se ratio for optimal bandgap tuning and defect minimisation. Optimisation of annealing processes also plays a key role. This includes the development of multi-stage temperature profiles, precise control of the partial pressure of sulphur and/or selenium to prevent the loss of volatile components (especially SnS/SnSe and chalcogens), the use of special protective capping layers (e.g., made of SnO_2 or carbon) during annealing, as well as the application of rapid thermal processing (RTP) techniques to improve crystallinity and reduce defect formation.

The central challenge remains overcoming the significant open-circuit voltage deficit (Voc deficit). Since the main causes of this deficit are associated with a high concentration of bulk defects (point defects, their clusters and band tail states) and recombination at the heterojunction (Fig. 14), strategies to reduce it are focused on these aspects. To reduce bulk recombination, in addition to the previously mentioned doping and composition/annealing optimisation, researches explore defect passivation methods, grain boundary engineering, and approaches that suppress the formation of particularly harmful defect complexes, such as electrostatically neutral clusters [CuZn + ZnCu] [72]. Surface treatment of the CZTS(Se) absorber layer prior to buffer layer deposition, such as chemical etching or application of thin passivation layers, can also help reduce recombination at the heterojunction.

In this context, the development of effective buffer layers as an alternative to traditional CdS is extremely important. The main goal is to find a material that provides better conduction band alignment (CBO) with CZTS(Se), exhibits lower absorption in the short-wavelength region of the spectrum, and is more environmentally friendly. Significant progress has been achieved with Zn(O,S)-based buffer layers, where varying the O/S ratio allows tuning the band gap width and the conduction band position, thereby optimising the CBO for different CZTS(Se) compositions [73]. Other promising candidates include In_2S_3, $Zn_{1-x}Sn_xO_y$ (ZTO), TiO_2, SnO_2 and their combinations. However, these alternative buffers still face challenges related to ensuring uniform conformal

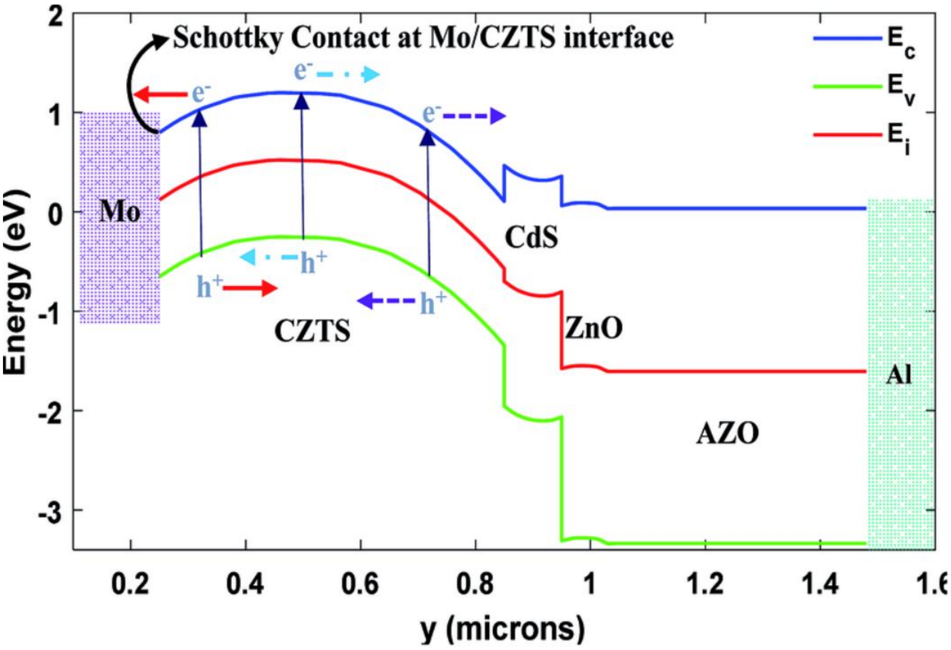


Fig. 14. Band diagram of a CZTS-based heterostructure [72]

coverage of the rough CZTS(Se) surface, achieving sufficient n-type conductivity, chemical stability during subsequent fabrication stages, and ensuring the long-term stability of the heterojunction itself.

Among other important challenges and research directions, further development and optimisation of non-vacuum, potentially cheaper and scalable methods for obtaining CZTS(Se) films with controlled properties should be noted [62, 67]. The development of flexible CZTS(Se)-based solar cells is also a relevant area that could expand their range of applications. Further studies are required to gain a deeper understanding of the degradation mechanisms and ensure the long-term stability of devices under real operating conditions. In addition, the potential use of CZTS(Se) in tandem solar cells is being explored, for example, as an top cell with an optimally tuned bandgap that absorbs the high-energy portion of the solar spectrum, combined with a bottom cell based on silicon or CIGS [74].

Although many challenges on the path to commercialising CZTS(Se) technology remain unresolved, active research and the progress achieved inspire optimism about its future. Successfull overcoming of existing challenges, particularly those related to Voc deficiency and defect control, could make CZTS(Se) solar cells a significant contributor to the development of affordable and sustainable solar energy.

Conclusions

This review article presented a comprehensive analysis of the current state of research and development in the field of thin-film solar cells based on chalcogenide semiconductor compounds $Cu(In,Ga)Se_2$ (CIGS) and $Cu_2ZnSn(S,Se)_4$ (CZTSSe). Their fundamental properties, fabrication methods, device structure and operation, achieved efficiency and stability levels were examined, along with a comparative analysis outlining future prospects and challenges.

The comparative analysis shows that CIGS currently offers higher performance and greater technological maturity, while CZTS(Se) demonstrates substantial long-term potential due to its material advantages. Both technologies face competition from the market-dominating crystalline silicon, which requires continuous improvement and the finding of specific market niches or cost reduction pathways.

Further research in this field remains highly promising and necessary. For CIGS technology, further efforts should focus on further reducing production costs, in particularly through the development of methods for utilising ultra-thin absorber layers, effective recycling of expensive components, and improvement of more cost-efficient deposition techniques. It is also important to increase the module efficiency, bringing it closer to theoretical limits, and to develop new products, such as high-efficiency flexible and transparent solar cells, as well as to promote the wider adoption of cadmium-free buffer layers.

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Сучасний стан розвитку та виклики тонкоплівкових фотоелектричних технологій на основі CIGS і CZTS. Огляд

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У цій оглядовій статті подано комплексний аналіз сучасного стану, досягнень і ключових викликів розвитку тонкоплівкових сонячних елементів на основі поглинальних матеріалів $Cu(In,Ga)Se_2$ (CIGS) та $Cu_2ZnSn(S,Se)_4$ (CZTS(Se)). Дослідження акцентує увагу на глобальному переході до відновлюваних джерел енергії та підкреслює переваги тонкоплівкових фотоелектричних технологій як економічно ефективної, матеріалоощадної та гнучкої альтернативи традиційним кремнієвим сонячним елементам. Пристрої на основі CIGS демонструють високі коефіцієнти перетворення потужності, що перевищують 23 %, широкий діапазон регулювання ширини забороненої зони та відмінну довготривалу стабільність, що робить їх одними з найбільш технологічно зрілих тонкоплівкових рішень. Водночас залежність від дефіцитних і дорогих елементів, зокрема індію та галію, стимулює активний пошук альтернативних матеріалів на основі хімічно доступних елементів. Матеріали CZTS(Se), до складу яких входять нетоксичні та поширені елементи (Cu, Zn, Sn, S, Se), розглядаються як перспективна й екологічно стала́ заміна, що характеризується порівнянними оптичними властивостями та регульованою шириною забороненої зони в діапазоні 1,0–1,5 еВ. Незважаючи на нижчі значення ефективності (на сьогодні — до 15,8 %), постійний прогрес у контролі фазової чистоти, пасивації дефектів і оптимізації зонного вирівнювання сприяє швидкому покращенню характеристик приладів. Основні напрями подальших досліджень охоплюють оптимізацію методів синтезу поглинального шару, інженерію дефектів, легування лужними металами та катіонами, а також розробку екологічно безпечних безкадмієвих архітектур сонячних елементів. У статті узагальнено рівень технологічної зрілості CIGS і потенціал розвитку CZTS(Se), окреслено критичні проблеми та можливі шляхи створення високоефективних і сталих тонкоплівкових фотоелектричних систем.

Ключові слова: CIGS; CZTS(Se); тонкоплівкові сонячні елементи; фотоелектричні системи; абсорбційні матеріали; інженерія забороненої зони; пасивація дефектів; відновлювана енергія; підвищення ефективності; стійкі технології.