Strategic Review: The CZTS Thin-Film Using Tandem and Multi-junction Solar Cell

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Highlights

➢ Performing a strategic review of fabrication technology and the CZTS thin film solar cell
➢ The review tries to cover all the contents with a simple expression
➢ Currently, two technologies are dominant in making solar cells
➢ Suggestions for increasing the efficiency of $CZTS_xSe_{1-x}$ solar cells is discussed
➢ Third-generation solar cells are also in the research phase

Abstract

In this study, a strategic review of fabrication technology and the CZTS (Cu_2ZnSnS_4) thin film solar cell using tandem and Multijunction structures is discussed. This review is tried to cover all the contents with a simple expression and at the same time so that the reader with any scientific level of this research field can have a good understanding of the research field and the importance of solar cells. Currently, two technologies are dominant in making solar cells (first and second generation) and new technology is in the research phase (third generation). The first-generation technology is based on silicon wafers with a thickness of 300μm - 400μm. The second generation of technology or thin-film technology, based on the semiconductor coating layer on glass, metal, or polymer layers, is 1μm - 5μm thick. Third-generation solar cells are also in the research phase, which includes nanocrystalline solar cells (based on liquid crystals), polymer solar cells, organic solar cells, perovskite solar cells (lead-mineral-organic hybrids or materials based on tin halides), Color Sensitive Solar Cells, Photo Electro Chemical Solar Cells, Multijunction Cells, Quantum Well Solar Cells, and Quantum Point Solar Cells. Although research into production technologies, especially third-generation technology, has evolved rapidly, there is still no review of the structures and results of all-generation technology. Due to the content of CZTS thin cells, the density of defects is an important role in the number of photovoltaic properties of the solar cell, many of these defects reduce the efficiency of the cell, and suggestions for increasing the efficiency of $CZTS_xSe_{1-x}$ solar cells is introduced.

1. Introduction

Growing demand for energy due to improving living standards, declining conventional energy sources, global warming due to burning fossil fuels, and the consequences of climate change are challenges. That is what our society is facing today. Hence, there is a new tendency to use renewable energy sources such as water energy, wind energy, solar energy, etc. These resources are widely available and help reduce global warming by reducing carbon emissions and greenhouse effects. Renewable energies, including solar energy, play a key role in solving these problems [1, 2]. Solar energy as a source of clean and endless energy is one of the most important candidates for energy supply after non-renewable fuels. The solar cell is the most important way to harness this energy. Solar cells are devices that convert sunlight into electricity using certain semiconductors [3]. World electricity consumption is 12 to 13 tera and increases with population growth [4].

The International Energy Agency (IEA) estimates that global energy demand will increase by one-third from 2010 to 2035, given the 50% growth in China and India [5]. Today, the main sources of energy consumption are fossil fuels, which are harmful to the environment due to the excessive emission of greenhouse gases. A good alternative to fossil fuels is to use photovoltaics (PV) or solar cells. The energy of the sun, which reaches the Earth's atmosphere in one hour, is enough to supply energy to the world for one year [4]. Today, the use of photovoltaics is becoming a reality [6]. The biggest deterrent to the use of solar cells is its initial price, but in the long run, due to the durability of solar cells, the initial capital can be returned to the investor [7]. Despite the many benefits of solar energy, there are limitations, such as solar energy is not radiated at night, solar energy is almost constant at all times, and climate change can prevent sunlight from reaching the earth [8]. The sun transfers about 1000 joules of energy per square meter of the earth's surface per second, which can be collected to provide the energy needed for various tasks. The energy that reaches the earth through the sun is 10,000 times more than the energy needed by humans. Energy consumption in 2050 will be 50 to 300 percent higher than today. However, if only 0.1% of the earth's surface is covered with solar energy converters and they have only 10% efficiency, it is enough to supply human energy [9,10]. At the center of the sun, 700 tons of hydrogen are converted into energy every second. The temperature of the sun in its center is 15 million and at its surface is 6,000 degrees Celsius. The energy produced at the surface of the sun reaches the surface of the earth after 8 minutes. Sunlight that reaches the earth contains the following wavelengths: ultraviolet with wavelengths λ < 400 nm (frequencies above 750 terahertz), which contains 7% of the frequency spectrum, visible light with wavelengths 400 nm < λ < 700 nm (frequencies between 428 and 750 terahertz) which includes 46% of the spectrum and infrared with a wavelength of 700 nm < λ < 1000 µm (frequencies between 0.3 to 428 terahertz) which contains 47% of the frequency spectrum of solar radiation. Therefore, solar cells should have a high absorption of infrared and visible light [9,10]. The characteristics of solar cells include no need for a national electricity grid, no need for fuel, environmental friendliness, no noise pollution, no need for water to generate electricity, not take up much space, and moving parts They do not, their efficiency does not change much with changes in ambient temperature, they are relatively easy to install, they easily adapt to the systems used in the building, one of the most important advantages of photovoltaic systems is that they can be used in remote areas. In some off-grid locations, even half a kilometer from

power lines, using photovoltaic systems can be more cost-effective than power outages. These systems are especially suitable for remote and environmentally sensitive areas, such as national parks, cottages, and homes in remote areas. In many rural areas, small solar cell assemblies are used for lighting, charging electric fences, and pumping water. In some cases, combined solar wind or solar-diesel systems are used [9, 10]. Another advantage of photovoltaic technology is that it can be combined with building materials and embedded in the building itself and not just on the roof. In such buildings, photovoltaic systems become part of the building blocks. These systems are called integrated Building Integrated Photovoltaic (BIPV), which compared to certain types of solar cell technologies, has several construction methods and different shapes that can be based on crystalline silicon or thin layer. BIPV can include facades, ceilings, windows, Walls, and many other devices that are combined with photovoltaic material, but it is an expensive method [11]. The use of solar cells for energy supply is increasing, so researchers are looking to build low-cost, high-efficiency solar cells. For this purpose, they use thin-layer solar cells. There are several very important parameters to evaluate the performance of a solar cell, including J_{SC} short circuit current density, V_{OC} open-circuit voltage, maximum power P_{max}, power density, fill factor (FF), short circuit current I_{SC}, and efficiency η. Figure 1 shows the I-V curve of an ideal solar cell.
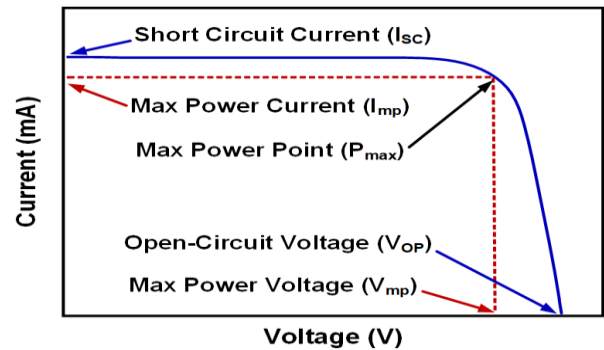

Fig. 1. Solar cell I-V characteristic curve.

When a short circuit occurs in a solar cell, the entire current is scattered throughout the p-n junction due to the intensity of the light rays without voltage dependence [11]. The amount of this total current can be expressed as I_{SC}. The maximum current that a cell can reach is called the short-circuit current density J_{SC}. Open circuit voltage V_{OC} is the voltage that is generated throughout the cell to delay the flow of current. In this paper, the strategic review of fabrication technology and CZTS thin-film solar cells using tandem and multi-junction structures is discussed with a simple expression.

The remainder of this paper is organized as follows. In sections 2 to 5, the topics of solar cell fabrication technology, thin film solar cells, multijunction solar cells, and CZTS thin film solar cells are discussed, respectively. In section 6, the results of research conducted to improve the performance of CZTS thin film solar cells are stated, and in section 7, conclusions and recommendations are presented.

2. Solar Cell Fabrication Technology

Currently, two technologies are dominant in the fabrication of solar cells: first-generation technology and second-generation technology. The first-generation technology is based on silicon wafers with a thickness of 300 μm – 400 μm that have a crystalline or polycrystalline structure that is either obtained by cutting ingots or by EFG method and are grown with the help of capillary properties. The second-generation technology or thin layer technology, based on the semiconductor coating layer on glass, metal, or polymer substrates, is in the thickness of 1 μm – 5 μm [13].   It is possible to achieve cells with efficiencies two to three times the current efficiencies. High efficiency can be achieved by using multi-bonded and tandem solar cells.

2.1. First-Generation Technology

The first generation is crystalline silicon(c-Si) solar cells, which is the dominant technology in commercial solar cell production, and in 2010 accounted for more than 87% of solar cell efficiency, which includes three categories: silicon solar cells Single crystal (Sc-Si), polycrystalline silicon solar cells (Si-Poly c), solar cells using Edge-defined Film-fed Growth (EFG) strip silicon and silicon sheet growth (EFG ribbon–sheet c-Si). The wide range of absorption and high mobility of 3 carriers are the advantages of this generation of solar cells. The high cost of fabrication of this type of solar cell is one of their disadvantages [1, 2, and 8].

2.1. Second-Generation Technology

The second generation is called thin-film solar cells. These cells are composed of successive thin layers 1 μm to 4 μm thick. Thin-film solar cells are deposited on large, inexpensive substrates such as glass, polymers, or laminated metals. As a result, construction costs are significantly reduced. Three types of thin-film solar cells have been commercially developed, [1, 8]. 1- Amorphous Silicon solar cells (A-Si), 2- Solar cells using cadmium telluride (CdTe), and 3- Solar cells using CIS and CIGS copper alloys. Amorphous silicon solar cells and cadmium telluride solar cells are the most developed and well-known thin-film solar cells. The main disadvantage of amorphous silicon solar cells is that they are not stable and over time there is a significant reduction in their output power. Thinner layers increase the electric field strength throughout the material, thus improving stability and reducing output powerlessness. However, it should be noted that thinner layers will reduce light absorption and also reduce the efficiency of the solar cell, so to increase the efficiency of thin film solar cells, thin film materials with high absorption coefficients such as CIGS and CZTS should be used. CIGS CIS solar cells are the most efficient thin-film solar cell technology. For them, the highest efficiency obtained in the laboratory is 21%. The second generation of solar cells is much cheaper to produce than the first generation, but they are also less efficient. CZTS solar cells are cheaper than CIGS and are made of abundant zinc and tin instead of the trace elements indium and gallium. But their efficiency is lower than CIGS solar cells, which needs more research to increase their efficiency [12]. The first CZTS solar cell was built in 1988 at Bell Labs with an efficiency of η=1.6% [12]. The main motivation for using thin film solar cells is to make them quickly and efficiently with the least number of raw materials, in which case the costs will be significantly reduced. Because the absorption coefficient of CZTS materials is high, we achieve high efficiency in low thickness [12].

2.3. Third-Generation Technology

This generation of solar cells is still in the research phase and includes nanocrystalline solar cells (based on liquid crystals), polymer solar cells, organic solar cells, perovskite solar cells (lead-mineral-organic hybrids or materials based on tin halides), and sensitive solar cells. In color Dye-sensitized solar cell (DSSC), photoelectrochemical (PEC) solar cells are multi-bonded cells, quantum well solar cells, and quantum dot solar cells. DS solar cells, sometimes called color-sensitive cells (CSCs), are the third generation of photovoltaic (solar) cells that convert any visible light into electrical energy. Photosynthesis is due to the way it imitates the absorption of light energy by nature. Color-sensitive solar cells (CSSCs) were invented in 1991 by Professor Michael Graetzel and Dr. Brian Oregon at the École Polytechnique Federal de Lausanne (EPFL), Switzerland, and are often referred to as the Gräetzel cells. Figure 2 shows a schematic of a Dye-sensitized solar cell device.
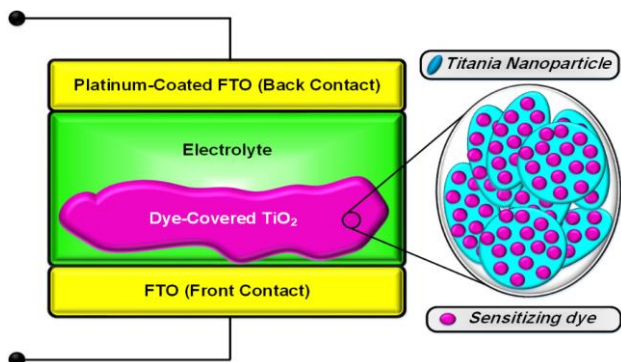
Fig. 2. Dye-sensitized solar cell device schematic.

Electrochemical cells (ECCs) are one of the third generations of solar cells, which is a promising method for producing hydrogen and is driven by solar energy, on the other hand, material limitations have significantly hindered their efficiency. Figure 3 shows how the PEC cell works. Sunlight stimulates the entry of free electrons near the surface of the silicon electrode. PEC cells use light energy (photons) to perform a chemical reaction to divide water into hydrogen (H_2) and oxygen (O_2) gases. They consist of an anode and a cathode immersed in an electrolyte and connected in an external circuit.
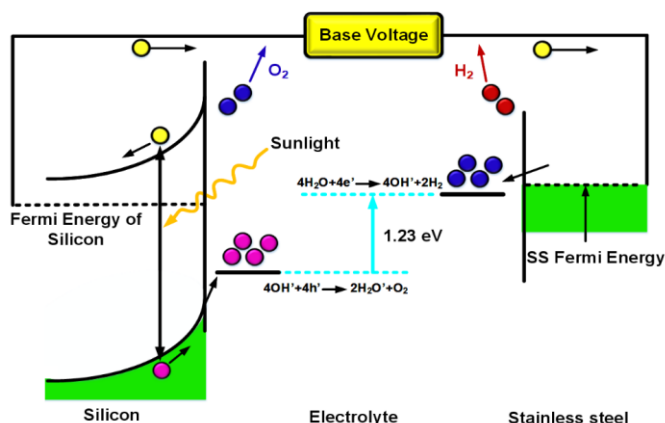

Fig. 3. Operation of a PEC cell.

Typically, the anode or cathode consists of a semiconductor that absorbs sunlight, and the other electrode is usually a metal. Photons with energy higher than the semiconductor bond distance can be absorbed by the semiconductor, creating pairs of electron cavities that are divided by an electric field in the charge region of the space between the semiconductor and the electrolyte. The electric field reflects the bending of the conduction edge band and the capacity at the semiconductor surface and is essential for the proper supply of free electrode carriers.

3. Thin-Film Solar Cells

In thin cells, thin films are formed by evaporation methods, a thin layer of about a few microns on surfaces such as glass, polymer, or, metal. Thin-film solar cells, in addition to making the manufacturing process easier and cheaper, also reduce raw material costs due to the use of a very thin layer of semiconductor materials. Several semiconductors have been proposed for use in thin-film solar cells; But several of them are most commonly used in industry: amorphous non-crystalline silicon, cadmium telluride, copper, indium, gallium and, selenium (CIGS), copper, zinc, tin, sulfide or, Selenium (called CZTS, CZTSe, CZTSSe, and CTS). The main advantages of thin film solar cells are flexible, low manufacturing cost, the ability to use large and continuous sublayers, high light absorption rate, reducing the amount of adsorbent required, and cheaper production [8, 12]. Thin-film solar cell technology emerged in the 1960s with the development of the Cu_2S/ cadmium sulfide (CdS) solar cell. This solar cell is the first flexible thin-film solar cell to be built on a layer of copper. The solar cell achieved an efficiency of 10% in 1981 [14]. From the 1970s to the 1980s, $CuInSe_2$, CdTe, and a-Si with efficiencies of approximately 10%, were the materials used to make solar cells. In the 1990s, thin-film solar cell performance improved, yielding more than 19% for $CuInGaSe_2$, more than 16% for CdTe, and more than 10% for a-Si. In the 1980s, flexible substrates were applied to a-Si solar cells. In the 1990s, $CuInSe_2$ solar cells were built on polymer and stainless-steel substrates [14]. The first thin-film solar cells were non-crystalline silicon cells called amorphous cells, which are still commercially produced and have the largest share of the thin-film cell market. To produce these cells, a thin layer of silicon several microns thick is deposited on the glass, plastic, or metal of the silane gas and hydrogen during the chemical deposition process in the vapor phase. Silicon consumption in this method is about one percent of crystal cells, which greatly reduces the cost of production [15, 16]. However, the non-crystalline structure of this type of cell has reduced its efficiency, so the best laboratory and industrial efficiency obtained from it is about 13% and 8%, respectively [15-17]. Cadmium telluride cells are the only thin-film cells available that have been able to outperform the industry's main competitor, silicon, in terms of production costs. The crystalline composite of cadmium telluride is a semiconductor with an energy gap of 1.44 eV, usually with cadmium sulfide (CdS) forming the p-n junction of a solar cell [18, 19]. The best efficiency of CdTe cells in industrial production, has been around 17% and its modules have been between 12% and 14%, and the best laboratory efficiency of these cells today has reached 19.5% [18, 19]. Another type of non-silicon material used

in thin film technology is a material composed of elements of groups one, three, and six of the periodic table, including copper, indium, gallium, and selenium, called CIGS. These cells are one of the most promising thin film technologies due to their high efficiency [20, 21]. These cells, like other thin-film cells, are made from deposits of glass or plastic. The best efficiency of CIGS cells is about 20% and the efficiency of its modules is between 13 to 15%, which is the highest value among thin-film cells [20, 21]. CZTS solar cells are cheaper than CIGS, and they are made from a lot of zinc and tin instead of the trace elements indium and gallium. But their efficiency is lower than CIGS solar cells, which needs more research to increase their efficiency. The highest laboratory yield obtained from single transplanted CZTS cells is 12.6% [22].

4. Multijunction Solar Cells

A photon with an energy greater than the energy gap of matter can only produce one electron-hole pair, and the excess energy of this photon relative to the energy gap becomes a phonon and is lost. Increasing the energy of the photon relative to the energy gap does not affect the cell flow rate and will only increase the losses [23]. Therefore, if this energy difference can be reduced in some way, energy loss will be reduced and efficiency will be increased. The solution is a multijunction solar cell. These cells are made up of a combination of several cells made of different materials so that by moving from the top (light entry point) down the energy gap, in which case the cell first absorbs the high-energy photons and then the low-energy photons The next cell arrives with a smaller energy gap, so in this structure, the difference between the photon energy and the energy gap will be less. The tunnel not only provides a low-resistance pathway for load carriers to flow but also acts as a communication path between the upper and lower cells [24]. The highest efficiency among solar cells belongs to multi-link cells. To achieve greater efficiency, there is a fundamental limitation for single-bonded solar cells. In these cells, photons with energy higher than the energy gap of the cells lose their excess energy in the form of heat, while photons with energy lower than the energy gap are not absorbed. The multijunction structure is an effective solution to this problem by dividing the spectrum into several different spectral regions, each region absorbing a cell [23]. The use of materials with different energy gaps will widely absorb more photon energy from the spectrum of sunlight and will reduce energy loss by using tunneling [23]. To achieve higher efficiency, the energy gap of the materials used in multi-bonded solar cells must be optimally selected to effectively absorb solar radiation. The energy gap of CZTS is equal to 1.5 eV, which is very close to

the conventional energy gap of 1.4 eV for the design of solar cells [12]; for this reason, we use CZTS for the top cell adsorbent layer.

4.1. LMJ/VMJ Multijunction Solar Cells

Tandem solar cells are promising photovoltaic systems that can absorb different wavelengths of the solar spectrum. There are many considerations in designing a multijunction solar cell, such as selecting materials with the appropriate energy gap, avoiding grid mismatch throughout the structure, and matching one cell with another. So, designing a tandem, the multijunction solar cell is a very complex process. Because in a multijunction solar cell, current flows from the top cell to the bottom cell, the layers of a multijunction cell are in series. Therefore, the current passes through each layer, and the output current is equal to the minimum current produced by the different layers of the whole cell. multijunction structures increase the efficiency of solar cells compared to single-junction cells. On the other hand, this expansion of layers and their improvement increases the price of solar cells, which makes their use in a wide range of space applications and with concentrators on the ground cost-effective. Inexpensive materials such as CZTS, CZTSe, and CTS can reduce the cost of making multi-transplant cells. Multijunction structures are more efficient, in the theory of using a multi-layer structure, when we increase the number of layers too much, we will be able to increase the absorption. To achieve high efficiency, we need to optimize the semiconductors used in the multilayer structure and their thickness [24]. Also, increasing the number of bonds to five or six bonds, to achieve an efficiency of nearly 50%, requires the use of materials that absorb the solar spectrum from 0.8 eV to 2 eV [25]. As shown in Figure 4, the Multijunction solar cell is divided into two groups based on the optical spectrum gap and mean junction angle methods. (a) Lateral Multijunction (LMJ), and (b) Vertical Multijunction (VMJ) [26].
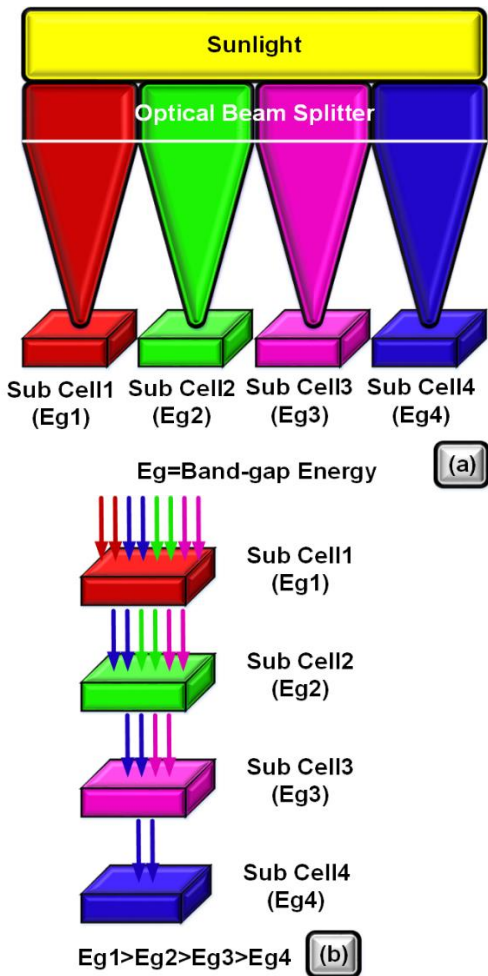
Fig. 4. Types of Multijunction solar cell structure, (a) LMJ, (b) VMJ.

Lateral Multijunction cells (LMJ), despite their advantages, are not yet as practical as VMJ. Vertical Multijunction solar cells are also made in two ways: Mechanically stacked and monolithically stacked. Figure 5 shows these two modes [26].
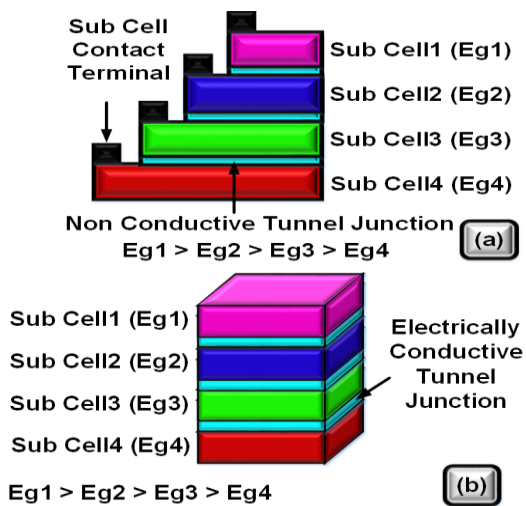

Fig. 5. The multijunction solar cell of VMJ, (a) Mechanically, (b)

monolithically.

In the construction of multijunction solar cells, different layers of different materials are placed in series with each other. So, the flow through them is the same. In this case, to plot the current-voltage curve of the whole cell, the value of the cell voltage in a given current is obtained from the sum of the voltages under the layers in the same current.

4.2. Main Layers of Multijunction Solar Cells

Figure 6 shows an example of a two-junction solar cell with its main layers. The top and bottom cells both contain the window layer, the emitter layer, the base layer, and the Back Surface Field (BSF) layer. Base and emitter can be either p or n. The window layer has the largest energy gap among all the layers, which is to pass maximum sunlight. The BSF layer is one of the most important layers in the cell, having a larger energy gap than the base layer. Due to the greater energy gap, this layer reduces the recombination of charge carriers and thus helps to increase the efficiency of the solar cell [24]. A high-energy n-type layer is located at the top of the cell as a transparent front-joint layer or window layer, usually made of high-density doped ZnO. This layer allows almost all the radiant light to pass easily to reach the absorbing layer. Also, the window layer with a large energy gap reduces the resistance of the cell series. The optically generated electrons can easily flow through the window layer to achieve a connection [6, 24]. This layer should provide a potential barrier for minority carriers of the emitter layer. The quality of the material must be very high so that the recombination rate is very low [6].
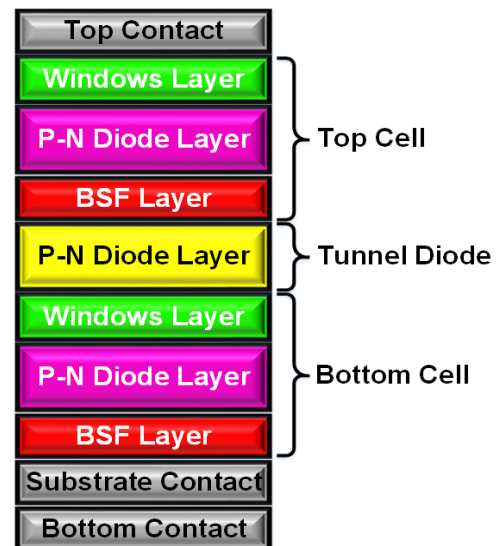

Fig. 6. Main Layers of Multijunction Solar Cells.

The n-type region in the p-n junction of a solar cell is called the emitter or buffer. Similarly, the p-type region is called the base or adsorbent. A schematic view of a solar cell

is shown in Figure 7, in which two regions, one described as an emitter and the other as a base, are described [27]. As shown in the figure, the presence of such a bond, provided there is an external short circuit, causes electrons to flow and create a non-zero electric current. If the n and p regions are reversed, the current generated is also reversed [27].
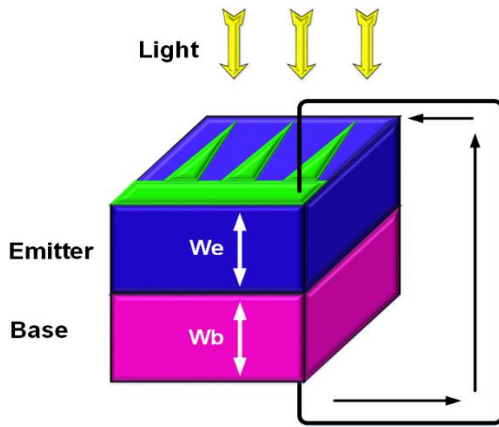

Fig. 7. Schematic of a simple solar cell.

The buffer layer is used to form a junction with the adsorbent layer so that the maximum amount of light enters the adsorbent layer, therefore the buffer layer should have the lowest absorption losses, for which the surface recombination rate should be the lowest, and have low electrical resistance So that the cargo carriers can be easily removed. To achieve the mentioned characteristics, the buffer layer should be as thin as possible and its energy gap should be more than the absorbent layer [24]. In CZTS solar cells, CdS is usually used as the buffer layer, with which the best efficiency can be achieved. However, CdS is classified as a toxic substance. A series of non-toxic materials with a high energy gap is proposed as an alternative to the buffer layer, including Zn (O, S), ZnSe, SnS_2, etc. [28]. ZnS on sulfide is a good choice for a buffer layer due to its desirable properties such as a high energy gap, high stability, and lower adsorption edge than CdS [28]. The BSF layer is usually used to eliminate recombination between the base and the tunnel junction of the top cell or the base and dorsal junction of the bottom cell. Therefore, to design the BSF layer, items 1, 2, and 3 must be observed [6,24]: 1- The energy gap of the BSF layer must be larger than the base layer, 2- This layer must create a potential barrier for minority carriers of the base layer, and 3- The BSF layer should provide the maximum light transparency for the photons to reach the bottom cell. The tunnel junction or the connection between two sub-cells is one of the most important parts of multi-junction solar cells. This bonding layer should provide a low-resistance path for the charge carriers to flow, and also act as a communication path between the upper and lower cells [6, 24]. CZTS solar cells are often built on a glass substrate, but it is generally possible to use any substrate. There are several reasons to choose a glass substrate. The glass substrate has temperature stability, is electrically insulated, is relatively inexpensive, and has a smooth surface. Stainless steel and polymer sheets are also among the materials used as substrates [12,14]. The positive electrode layer is layered on the substrate. The metal-bonding material (positive electrode) must be neutral, as the CZTS coating process is highly corrosive, acting as a barrier and preventing impurities from penetrating from the substrate into the adsorbent layer. Better electronic properties, ohmic connection formation for CZTS layer majority carriers, and low recombination rate are essential for minority carriers at the positive electrode/absorber layer boundary [23,24], which is why we use the SnS layer as BSF. High optical reflection is required to minimize optical losses. And for this reason, in some structures, it is also called the window layer [24]. In solar cell technology, various transparent conductive oxides have been used as electrodes, including ZnO, Indium doped tin oxide (ITO), Aluminum doped zinc oxide (AZO), Fluorine doped tin oxide (FTO), Graphene oxide (GrO) [22, 24].

5. CZTS Thin-Film Solar Cells

Environmental pollution due to the burning of fossil fuels and their scarcity of resources, as well as the growing demand for cheap energy, led to the use of renewable energy sources, including sunlight, to generate electricity. Therefore, the design of solar cells with low cost and high efficiency is important. CdTe, CIS, CIGS, CTS (Cu_2SnS_3), CZTS (Cu_2ZnSnS_4), and CZTSe ($Cu_2ZnSnSe_4$) are used to make thin film solar cells, which cost much less to make than silicon solar cells; because their rapid and efficient production is done using the least raw materials, these types of cells are also flexible and can be installed on non-flat surfaces. Among the mentioned materials, CZTS and CZTSe semiconductors are more desirable, because in CIS, CIGS, and CdTe the sources of indium (In), gallium (Ga), and tellurium (Te) are limited and cadmium (Cd) is toxic. While the constituent elements of the quaternary semiconductors CZTS and CZTSe belong to the elements of groups I, II, IV, and VI and are found in abundance on earth and are inexpensive and non-toxic [29,30]. Also, these two semiconductors have a direct energy gap in the range of 1.4eV - 4.1eV and 0.9eV - 1.12eV, and their optical absorption coefficient is above 10^4 cm^{-1}, so they are a very good option to replace CIGS [29, 30]. CZTS raw materials are approximately five times cheaper than CIGS raw materials. Estimates of global reserves of materials for

copper, zinc, tin, and sulfur show that we can produce enough energy to supply the world with only 0.1% of available primary resources [31]. The typical thickness of CZTS and CZTSe adsorbent layers is about 2µm-3µm. CZTS polycrystalline solar cells have high series specific resistivity $R_S=4/25\Omega$ small parallel resistivity $R_{Sh} = 370\Omega$ [32]. CZTS was first developed in 1966 and later in 1988 was shown to have a photovoltaic effect [33]. In 1997, a yield of 2.3% was reported for CZTS and CZTSe solar cells [33]. In 2005, with the optimization of the layering process, the efficiency of CZTS solar cells increased to 5.7% [33]. In 2014, researchers obtained a yield of 3.4% for a Bifacial cell using the CZTS absorbent layer and a transparent conductive electrode on the back of the cell, which can emit light to both sides of the cell (front and back surface), later in 2016 the efficiency for this cell based on double configuration increased to 5.8% [33]. In addition, sodium was shown to have an additive effect on the structural and electrical properties of the CZTS adsorbent layer. These developments, along with the start of commercial-scale CIGS production in the early 2000s, accelerated research interest in CZTS and related compounds [33]. Since 1988, CZTS has been considered as an alternative to CIGS in commercial solar systems. The advantage of CZTS is the absence of the relatively rare and expensive element indium. The British Geological Survey has assigned a relative supply risk index of 6.5 to Indium in the 2011 risk list, while the highest relative supply risk index was 8.5 [33]. In 2010, solar conversion efficiencies of about 10% were achieved for CZTS cells [33]. CZTS technology has now been developed by several private companies. In August 2012, IBM announced that it had developed the CZTS solar cell, which can convert 11.1% of solar energy into electricity [33]. In 2013, Rajshmon et al reported an efficiency of 1.85% for $CZTS/In_2S_3$ solar cells with a steam spray [33]. In November 2013, the Japanese thin-film solar cell company Solar Frontier announced that in a joint research project with IBM and Tokyo Ohka Kogyo (TOK), they had produced a world-record CZTSSe solar cell with an energy conversion efficiency of 12.6% [33]. David Mitzie is an American scientist who has been working on CZTS solar cells since working for IBM. He is the most well-known scientist to hold several CZTS solar cell efficiency records, including the latest achievement, which is CZTSSe with 12.6% efficiency for IBM [33]. The concentration of carriers and the adsorption coefficient of tin sulfide on copper (CZTS) is similar to CIGS. Other properties such as carrier lifetime (and associated diffusion duration) are low for CZTS (less than 9 nS). This short lifespan may be due to high defect density. Active or recombinant at grain boundaries [33]. The quaternary CZTS compound has a

kesterite and stannite structure. In the structure of CIGS chalcopyrite, by replacing the In/Ga trivalent atoms with a divalent Zn atom and a quaternary Sn atom, a CZTS with a kesterite structure is obtained [33]. Figure 8 shows the structure of the CZTS kesterite and stannite. The structure of CZTSe is obtained by replacing the sulfide atom (S) with the selenium atom (Se) [34]. CZTS and CZTSe are kesterite and stannite crystalline. kesterite and stannite are Body-centered tetragonal with c=2a (quasi-cubic) and can be thought of as two cubic lattices of surface center (FCC) of sulfur stacked on top of each other with half-empty space. The quadrilaterals in the FCC are occupied by copper (Cu), zinc (Zn), and tin (Sn). Two single cells for comparing the structure of kesterite and stannite are shown in Figure 8. Differences in the arrangement and accumulation of metal cations within the quadrilateral voids have created these two different structures [34].
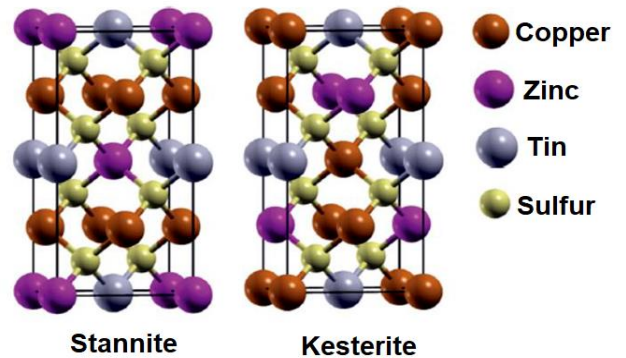

Fig. 8. Structure of (a) kesterite, (b) stannite, for CZTS.

The structure of kesterite is composed of two layers of periodic cations, so that each layer contains copper and zinc or copper and tin, while in the structure of stannite, a layer of copper with a layer of zinc atoms and tin is placed one on top of the other. In the structure of the stannite, the zinc and tin atoms on the same layer change position in each other layer. In addition, when half of the quadrilateral empty space is filled only by zinc atoms or only copper and tin atoms, ZnS or ZnSe and CTS or CTSe are formed, which are called secondary phases [34]. Figure 9 shows the phase diagram of the materials that may be formed during the crystallization of CZTS [35].
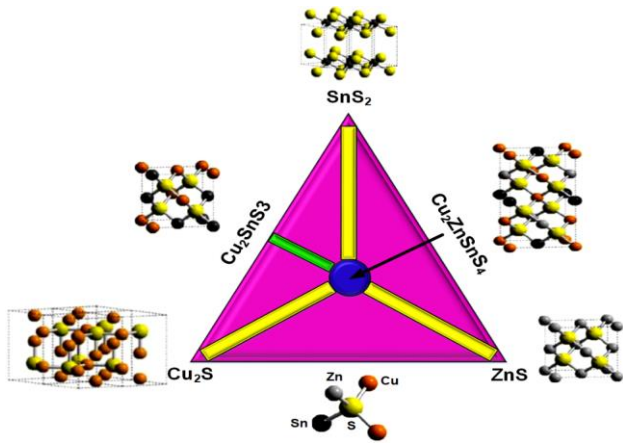
Fig. 9. Quasi-ternary phase diagram for CZTS phase formation and other secondary phases, along with their crystal structures.

CZTS is made with a variety of vacuum and non-vacuum methods. Researchers use most of the construction methods that have been successful for CIGS to build CZTS, although optimal construction conditions may vary. Manufacturing methods are broadly divided into two categories: 1- Vacuum deposition or non-vacuum coating layer-based methods, 2- One-step method or sulfurization/Selenization. Vacuum-based methods are prevalent in the current CIGS industry, but in the last decade, there has been an increasing interest in non-gap processes due to their potential cost reduction as well as their flexibility to cover large areas [33]. A particular challenge for CZTS and related alloys is the volatility of certain elements (such as Zn and SnS) that can evaporate under reaction conditions. After the formation of CZTS, the volatility of the elements is less problematic, but even then CZTS decomposes into binary and ternary compounds in a vacuum and at temperatures above 500 °C. This volatility and difficulty in preparing single-phase material has led to the success of many traditional vacuum methods. At present, the best CZTS cells have been obtained using special chemical methods that allow CZTS to form at low temperatures and prevent volatile problems [33]. An ongoing process has been developed using ethylene glycol as a solvent at Oregon State University, which may be suitable for mass production on an industrial scale [33]. The global record for CZTSSe solar cell efficiencies is now 12.6% [33]. But more work is needed to commercialize them.

The structure of a CZTS solar cell is heterogeneous, consisting of different semiconductor materials. A CZTS thin film solar cell has a base structure as shown in Figure 10. CZTS thin film solar cells are made up of several thin layers placed on a hard or flexible substrate. In this type of solar cell, the first layer is usually placed on the molybdenum metal substrate, which is used as a non-transparent backing layer. A layer of p-type kesterite, which is the same as CZTS and is used as an adsorbent, is coated on the molybdenum layer. This p-type absorbent layer absorbs most of the light emitted into the cell and produces a light current. Then the heterogeneous bond structure with the address layer of a thin layer of type n, which is usually made of CdS as a buffer layer and two layers of type n with a high energy gap as a window layer and a transparent front connection electrode, which is generally composed of ZnO and AZO with high-density doping [36].
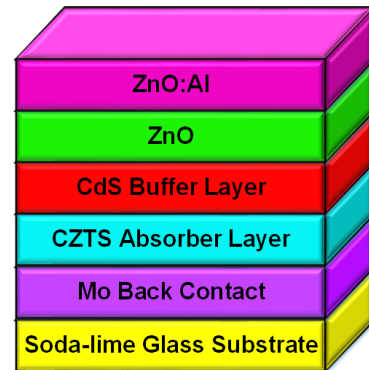

Fig. 10. Common structure of a CZTS solar cell.

The CZTS material has a typical direct energy gap of 1.5 eV. So far, researchers have been able to make very good cells with 12.6% efficiency with CZTSSe [37, 38].

6. Discussion and Review

Recently, a lot of research has been done to improve the performance of CZTS thin film solar cells, and the results of some of them have been discussed separately by layers and topics.

6.1. Absorbent Layer

CZTS is one of the most desirable materials for use as an adsorbent layer in thin-film solar cells. CZTS has a direct energy gap and a high absorption coefficient for the light emitted to it and also has good stability against external environmental conditions [12].

In 2016, Arbouz et al. conducted a study on the performance of the CZTS thin film solar cell, which aimed to find the optimal thickness of the absorber and buffer layers. By selecting a thickness of 40 nm for the buffer layer, they changed the thickness of the adsorbent layer (Wp) from 1μm to 4μm and examined the cell performance. By changing the thickness of the adsorbent layer from 1μm to 4μm, J_{SC} and cell efficiency Increased by 45% and 48.76%, respectively, because with increasing thickness, photons with longer wavelengths are absorbed and then more optical carriers are produced and collected, so J_{SC} and

efficiency increase. Maximum power for thickness 1μm and 4μm is 8.27mW/cm² and 12.33mW/cm², respectively, indicating that the power has increased by 49%, but the V_{OC} has increased by only 2.25% and the FF remained almost unchanged and increased by only 0.3 [29]. In another 2016 study, Zhang et al. Used Zn (O, S) instead of the CdS buffer layer. In the CZTS/Zn (O, S)/AZO cell structure, they changed the thickness of the CZTS adsorbent layer from 0.5μm to 4μm. It was observed that as the adsorbent layer thickness increased, all cell parameters increased. When the CZTS layer is thick, more photons accumulate in the adsorbent layer, which produces more electron-hole pairs and collects them, increasing the V_{OC} and J_{SC}, thus increasing the efficiency, so the optimal thickness for the CZTS adsorbent layer is between 2μm and 3μm. [39-42].

6.2. Buffer Layer

In heterogeneous bonded solar cells, the presence of a buffer layer is beneficial for component performance. The quality of the heterogeneous structure is improved by using an intermediate layer. In other words, the buffer layer regulates the energy gap between the absorber layer and the window layer. Also, one of its applications is network adaptation at the border. The buffer layer should be as thin as possible and the energy gap should be much larger than the adsorbent layer, which means that the buffer layer has the lowest absorption losses, low electrical resistance, and low surface recombination rate to extract the carriers produced by light [24]. So far, CZTS thin film solar cells have used more CdS for the buffer layer, however, cadmium is a toxic substance. However, other materials for use as a buffer layer in CZTS thin film solar cells were examined, including ZnS, ZnSe, In_2S_3, SnS_2, and Zn (O, S) [28]. In 2016, Arbouz et al. selected the optimal thickness of 3μm for the CZTS adsorbent layer to investigate the effect of changing the thickness of the CdS buffer layer from 10nm to 100nm on cell performance. The results showed that by changing the thickness of the buffer layer (W_n), J_{SC} and the efficiency decreased by 2.1% and 2.16%, respectively. But V_{OC} and FF do not change. The thinner the buffer layer, the more photons it allows to pass through the buffer layer and reach the adsorbent layer [29]. One of the factors that limit V_{OC} is how the Band alignment is arranged in the CZTS / CdS heterogeneous interface, which leads to recombination in the interface. In an experimental work done in 2016 by kaiwen et al., They used the $Zn_{1-x}Cd_xS$ layer, which has an adjustable energy gap, as a buffer layer instead of the CdS buffer layer. Because the conduction band minimum (CBM) of this material can be adjusted by controlling the ratio of cadmium to zinc (Cd/Zn). It was observed in the joint with CZTS that it formed a spike type CBO with a value

of 0.37 eV, which is very close to the edge of the optimal range (0.0 - 0.4Ve), so they are from $Zn_{0.35}Cd_{0.65}S$ with energy gap 2.7e V used CZTS for better cell function [43]. In another study in 2016, Zhang et al. achieved a yield of 14.9% using numerical simulation and using Zn (O, S) instead of the CdS buffer layer. Because the Zn energy gap (O, S) is larger than CdS, most photons pass through the buffer layer without adsorption and reach the adsorption layer, which increases cell efficiency. In terms of biocompatibility, because cadmium is toxic, it is replaced with the non-toxic element zinc [39].

6.3. A thin layer between the absorbent and the back contact

Sometimes CZTS decomposes at the CZTS/Mo interface, producing vacancy defects as well as the formation of MoS_2 and SnS_2, which increase the series resistance and decrease the F.F, thereby reducing the efficiency. To reduce the problems mentioned above, the researchers used intermediate layers such as TiN and ZnO to reduce the amount of MoS_2 and defects in the CZTS / Mo border and showed that the use of the intermediate layer significantly increases cell function [44]. Thus, in a 2014 experimental study, Hong Tao et al. used a 20 nm thick silver (Ag) layer as an intermediate layer to reduce MoS_2 formation and back-contact defects, as well as to prevent SnS_2 formation in the space charge region. Because with the use of Ag, the efficiency has increased from 2.3% to 4.4% [44]. In another experimental study conducted in 2014 by Fang Yang et al., The 2TiB interface layer was used to optimize the CZTS/Mo boundary. They showed that the TiB_2 layer significantly inhibited the formation of the MoS_2 layer at the common boundary of the backlink absorber layer, which reduced the series resistance and thus increased efficiency and improved cell performance. Precise control of the TiB_2 intermediate layer thickness is essential to ensure that the MoS_2 layer thickness is minimal and that the CZTS crystal structure has large grains during the growth process to have fewer defects and better crystalline properties. Which showed the best performance at a thickness of 30nm of the cell intermediate layer [45]. The researchers also used other materials such as carbon, TiN, TiB_2, ZnO, Bi, and Ag as intermediate layers to prevent the formation of MoS_2 at the adsorbent/junction interface and improve cell function. So far, maximum yields of 5.5% and 8.9% have been reported for CZTS and CZTSe solar cells, respectively, using the TiN intermediate layer [46].

6.4. Impurity Density of Adsorbent Layer

Due to the presence of Cu_{Zn} antisite defects and Cu_2 vacancy defects, it is inherently a p-type semiconductor. Due to the high stability of the faults in the wrong place, it is impossible to achieve the conductivity of type n in CZTS. For photovoltaic applications, Cu vacancy defects are superior to Cu_{Zn} antisite defects because acceptor impurities are formed at shallow depths. Therefore, with stoichiometric ratios of Cu/Zn and Cu/Sn, they control the density of the majority of cavities in CZTS [47]. In 2016, Samir et al. changed the density of the cavity in the CZTS adsorbent layer in the range of $10^{-17}cm^{-3}$-$10^{-13}cm^{-3}$ and examined the cell performance of the AZO/ZnO/CdS/CZTS/Mo structure. At a cavity density of 10^{16} cm^{-3}, yields and F.F. have the highest values. As the carrier density increases, recombination increases, leading to a decrease in J_{SC}. Therefore, at an optimum density of 10^{16} cm^{-3}, the adsorbent layer yields an F.F equal to 11.77% and 68.6%, respectively [47].

6.5. Nanostructures

In recent years, graphene nanotubes and nanotubes have been used for front and back electrodes in thin-film solar cells. In 2016, P. Darvishzadeh et al. proposed a new structure for the CZTS thin-film cell in which they used a thin layer of single-walled carbon nanotube (SWCNT) as the front electrode instead of the AZO transparent conductive electrode. The metal's excellent optical and electrical properties, including high and controllable work function, transparency over a wide range of wavelengths, and their high charge conductivity, and their good electrical and optical properties increase Jsc and quantum efficiency in CZTS cells. That quantum efficiency increases by about 30% [48, 49]. In 2016, Omar Abdul Rauf et al. used nanostructures to increase the uptake and collection of carriers in the CZTS solar cell. They simulated the CZTS adsorbent layer in the form of plates, nanorods, nanotubes, and nanotubes, which with CZTS nanotubes achieved the highest efficiency of 7.91% compared to other structures. Their results are listed in Table 1 [50].

Table 1. Specification of studied DGs.

Structure	JSC (mA/cm2)	VOC (mV)	FF	Efficiency (%)
Planar CZTS/CdS	19.19	590	0.56	6.59
Nanorods CZTS/CdS	21.11	590	0.56	7.25
Nanotexture CZTS/CdS	21.68	590	0.56	7.45
Nanotubes CZTS/CdS	23.03	590	0.56	7.91

In 2017, Ghosh et al. experimentally built a CZTS solar cell with a vertical ZnO nanorod window layer and a ZnS buffer layer with a yield of 3.63%. The efficiency of 3.63% is the highest efficiency among CZTS nanostructured cells that researchers have achieved by 2017 [51]. In 2018, Waliseri et al based their modeling work on a cell made by Ghosh. They first simulated a cell with the structure Au/CZTS/ZnS/ZnO NR/ITO, which yielded similar results by welding, and then the effect of layer thickness parameters, impurity density, and adsorption layer minority carrier life, and defect density on performance. Examined the cell. Taking into account the optimal values of all the obtained parameters, they reached efficiencies of 7.65%, V_{OC} = 0.85V, J_{SC} = 13.85 mA/cm^2, and F.F = 58.2%. The output parameters of the optimized cell were significantly increased compared to the primary cell [52].

6.6. Absorbent Layer with Graded Bandgap

In a cell with an ungraded structure, there is no thrust outside the discharge area to help move the carriers toward the graft to collect them. In contrast, in gradual structures, due to the potential difference in the conduction band, there is a quasi-electric field that conducts the conduction band electrons in the p-type adsorbent layer to the space charge-discharge region. Therefore, the probability of collecting minority carriers increases, which increases the production flow. In 2013, Kyoohee et al. Constructed a solar cell with a $Cu_2ZnSn(S_{1-x}, Se_x)_4$ absorbent layer, with a thickness of 0.78μm and gradual energy gap using non-toxic solvent-based methods and achieved an efficiency of 7.1%. [53]. In another experimental study in 2016, Yang et al. Were able to increase the efficiency of the CZTSSe cell by improving the fabrication process to increase the crystalline properties of CZTSSe, preventing the formation of secondary phases and controlling defects, and with a gradual energy gap. They created gradual energy by controlling the amount of S and Se in the gap absorber layer. They were able to achieve a return of 12.3% [54]. In 2016, Simya et al. From the gradual energy gap for CZT (S_{1-x}, Se_x), in the absorbent layer exponential and uniform from x=0 with energy gap 0.95eV to x= 1with energy gap 1.5 eV by modifying the Se/S + Se ratio, they modeled the gradual energy gap in the adsorbent layer. The higher the selenium content than the sulfur, the higher the yield. When the Se/S+S ratio was 0.8 and with a gradual energy gap, they achieved an efficiency of 14.97% [55]. In 2017, Adeyinka et al. achieved 8.33% efficiency in the cell with the basic structure of FTO/CdS/CZTS/Mo and AMPS software. They investigated the effect of a gradual energy gap at the front and back interface of the adsorbent layer on cell performance. The gradual energy gap in front of the

absorber layer weakened cell performance, while the gradual energy gap behind the absorber layer improved cell performance. Simultaneous use of a gradual energy gap in the front and back of the adsorbent layer achieved the best cell performance with an efficiency of 12.6% [32].

6.7. Two Absorbent Layers and a Back Surface Field (BSF) layer

In 2017, M Minbashi et al. Simulated a CZTSSe cell with a gradual energy gap and achieved a yield of 12.6% in the basal cell, which was in good agreement with the experimental data. To improve cell performance, they used a p-type silicon layer as the back surface field layer and achieved a yield of 16.59%, which significantly increased the yield [56]. In 2018, Cherouana et al. examined the effect of the silicon dorsal surface layer on CZTS cell performance. Using a silicon BSF layer, they increased the cell yield from 7.72% to 10.69% and showed that the thickness of the adsorbent layer decreased from 2μm to 1.3μm [57]. In 2018, Udi Saha et al. Simulated AZO/ZnO/CdS/CZTS/Mo cell structure using Lumerical software and achieved an efficiency of 8.4%, which was in good agreement with the reported experimental data. Using the structure CdS/ACZTS/CZTS (n/p/p+), they adjusted the doping density of the different layers to extend the discharge area over the entire ACZTS layer, taking into account the common recombination effects of the optimal ACZTS thickness (550nm). And CZTS (200nm) is obtained with a maximum efficiency of 17.59% [58].

6.8. Multijunction Structure

Work on CZTS-based multi-link solar cells has only recently begun. In 2014, Theodore et al., In an experimental study, built a tandem solar cell and monolithic solar cell with the perovskite adsorbent layers $NH_3CH_3PbI_3$[6] and CZTSSe for the top and bottom cell, with a yield of 16%. This cell has a large V_{OC} of 1.40 V, which is close to the sum of the V_{OC} of the top and bottom monoclonal cells. Perovskite and CZTSSe have good optoelectronic properties and are also compatible for integration into integrated cell structures [59]. However, high resistance, low optical conductivity of Al electrode, low perovskite stability, high manufacturing cost due to high-temperature annealing, low cell life, reduced cell performance under environmental conditions, use of lead toxin (Pb) are among the factors that yield Limits perovskite/CZTSSe dual cells [60]. Due to the limitations mentioned for the

perovskite/CZTSSe tandem cell, it is better to use CZTS and CZTSe absorbent layers that have a direct energy gap for the top and bottom cells. CZTS, CTS, CZTSe, and CZTSSe-based multijunction cells are biocompatible and low-cost, they can be implemented and fabricated with current technologies. In addition, they overcome the problems and limitations of perovskite and are composed of cheap and non-toxic materials. Therefore, in another study in 2017, Udi Saha et al. Simulated a CZTS/CZTSe dual-junction cell with an ITO binding layer using Lumerical software. They used CZTS with eV = 1.5 for the high cell adsorbent layer and CZTSe with Eg=1.04eV for the bottom cell adsorbent layer, investigating the effect of changing the thickness of the adsorbent layers on cell performance so that with an optimal thickness of nm 200 and 850 nm for CZTS and CZTSe, the maximum efficiency was 19.87%. Then, the energy gap change of the adsorbent layers on the cell parameters was investigated, for which in the optimal case, the efficiency was 21.74%. They also replaced the toxic CdS buffer layer with ZnS and achieved a yield of 21.44% [60]. The researchers also replaced the toxic CdS buffer layer with the non-toxic substance ZnS. Because the ZnS energy gap (3.58 eV) is larger than the CdS energy gap (2.42 eV), most photons pass through the buffer layer and reach the adsorbent layer. With ZnS, the efficiency increased by about 8% [60, 61].

6.9. CTS Cells

The Cu_2SnS_3 (CTS) ternary semiconductor is composed of abundant non-toxic materials in the earth's crust and inexpensive elements. The CTS also has an absorption coefficient of 10^4 cm^{-1} and an energy gap of 1.77 eV - 0.93eV. CTS is a good material for p-type adsorbents in thin-film solar cells [62]. In another experimental study in 2015, Nakashim et al. Constructed CTS cells with efficiencies of 4.63%, V_{OC} = 0.283V, J_{SC} = 37.3 mA/Cm^2, and F.F = 43.9% [63]. In 2020, Ashebir et al. planar heterojunction solar cells configured as FTO/TiO$_2$/CdS/Cu$_2$SnS$_3$/P3HT/MoO$_3$/Ag are fabricated, in which poly (3-hexylthiophene) (P3HT), an organic composite polymer, acts primarily as a carrier material for cavities. An efficiency of 2.03% is obtained in the solar cells with a 60-nm thick Cu$_2$SnS$_3$ thin film prepared at 300 ºC [64]. An overview of the technology status and values of important parameters in CTS thin film cells is shown in Table 2.

Table 2. The technological status of CTS thin films in recent years.

Process	Reference	Deposition Method	η (%)	V_{OC} (mV)	J_{SC}(mA/cm2)	FF (%)
	65	Sputtering	3.05	243	26.2	47.9

Vacuum	66	Sputtering	0.76	215	13.8	25.5
	67	EBE	1.20	134	27.9	31.2
	68	EBE	2.70	244	29.0	38.5
	69	EBE	2.56	211	28.0	43.0
	70	Evaporation	3.66	248	33.5	43.9
	71	Evaporation	4.29	258	35.6	46.7
	72	Evaporation	4.63	283	37.3	43.9
	73	PLD	0.82	260	11.90	24.0
Non-vacuum	74	ED	2.84	249	29.3	39.0
	75	ED	0.54	104	17.1	30.4
	76	Ball milling	1.92	320	23.4	28.0

7. Conclusion and Recommendations

In this paper, the strategic review of fabrication technology and CZTS thin-film solar cells using tandem and multi-junction structures was discussed with a simple expression. According to the contents of the CZTS thin film solar cell, the density of defects plays a Kesterite defect will reduce cell efficiency. The main factor in determining the density of defects in the method of making the adsorbent layer material. As mentioned, there are two structures, Kesterite and Stannite, for CZTS. According to previous research, it was found that the structure of Kesterite is more stable, so the density of defects in the material is lower. Therefore, improving manufacturing methods is effective in reducing the density of defects, one of which is fabricating the material in a vacuum. In addition, as the amount of sulfur in the CZTS material increases, the energy gap will increase and will be more in line with the spectrum of the sun. In other words, it is expected that by selecting more energy gaps, higher efficiency will be provided for the cell. On the other hand, as the number of sulfur increases, the density of defects also increases, so there is a limit to increasing the amount of sulfur. And no large energy gap can be allocated to matter. By adding germanium to the system during the manufacture of $Cu_2Zn (Sn, Ge) S4$ material is formed, which allows the optimal adjustment of the energy gap in the range 2eV - 0.96eV and can be used in the structure of a four-junction cell. Another point that can be used to improve the performance of CZTS thin-film solar cells is the use of buffer layers with more energy gaps. The buffer layer should be as thin as possible and its energy gap should be much larger than the adsorbent layer, which means that the buffer layer has the lowest absorption losses, low electrical resistance, and low surface recombination rate. Apart from CdS, which is a toxic substance, ZnS, $ZnSe$, TiO_2, In_2S_3 and SnS_2 have been investigated for use as a buffer layer in CZTS thin-film solar cells, we can use these buffer layers in multijunction structures to increase cell efficiency. By using quantum dots, nanostructures, and the application of perovskite and polymer adsorbent layers in multi-bonded cells, the efficiency of CZTS thin film solar cells can also be increased.

As future work, a review of the research conducted on the performance of CZTGS thin film solar cells can be done.

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