DOI: 10.54254/2755-2721/24/20230693

Design and process of perovskite/silicon tandem solar cells

AnXu Ge

College of Materials, Beijing Institute of Technology, Beijing, 102401, China

1120200524@bit.edu.cn

Abstract. At present, the solar cells that occupy most market are still silicon solar cells. However, the power conversion efficiencies (PCEs) of the devices made of silicon have achieved an extreme value. Therefore, a new type of solar cells to get higher power conversion efficiencies are in great need. Since the 21st century, perovskite/silicon tandem solar cells have gained great attention because of their potential to offer higher PCE compared with other traditional solar cells. This article elaborates the inevitability of the development of perovskite/silicon crystal tandem solar cells from the perspectives of development history, cell structure. This article also discusses the different materials of both perovskite top cell layers and silicon crystal bottom cell layers in this new type of solar cell, including methylammonium lead iodide, formamidinium lead iodide, cesium lead iodide, crystalline silicon, thin-film silicon and heterojunction with intrinsic thin layer. This article mainly presents the characteristics and advantages and disadvantages of different materials when they are used in this new type device.

Keywords: tandem solar cell, photovoltaics, perovskite, Si solar cell, PCE.

1. Introduction

In the 19th century, Perovskii and Rose discovered a mineral material $CaTiO_3$ with an ABX_3 crystal structure and named it perovskite. Perovskite refers to a specific crystal structure whose chemical formula is like ABX_3. The letter A and B represent cations (positively charged ions) and letter X is an anion (negatively charged ion). In addition, perovskite materials must meet the Goldschmidt tolerance factor formula:

$$t = \frac{r_A + r_B}{r_B + r_x} \cdot \frac{\sqrt{2}}{2} \quad (1)$$

r_A, r_B, and r_x represent the effective radius of A, B, and X ions respectively, $0.8<t<1.15$. Equation (1) describes that when t value is too high or too low, stable perovskite structure cannot be formed.

Perovskite materials have got significant attention in recent years because of their unique optoelectronic properties, including high absorption coefficients and long carrier lifetimes, which make them promising for a range of applications. When manufacturing different kinds of solar cells, perovskite materials have great potential for application.

However, the PCE of the devices made of perovskite nowadays still can not satisfied scientists. Therefore, scientists developed perovskite/silicon tandem solar cells. These cells have a higher efficiency, lower cost, lower power consumption and Versatility. The history of perovskite/silicon tandem solar cells is as follows.

DOI: 10.54254/2755-2721/24/20230693

In 2012, Jin Young Kim et al. demonstrated a proof-of-concept tandem solar cell that combined a thin-film silicon solar cell with a perovskite solar cell. The device achieved an PCE of over 6% [1]. In 2014, Norman Pellet et al. made this concept come true by developing a device with a PCE of 14.9% [2]. They used a perovskite top cell with a mixed-cation formulation ($MA_{0.6}FA_{0.4}PbI_3$) and a silicon heterojunction bottom cell. In 2016, Jérémie Werner et al. achieved great success through combining monolithic perovskite with silicon heterojunction. The new device achieved a PEC of up to 21.2% on a cell whose area was only 0.17 cm^2. Its efficiency can also be over 19.2% when the cell areas was 1.22 cm^2 [3]. In 2017, Catchpole et al. combined back electrode contact crystalline silicon batteries with perovskite batteries, resulting in an increase in the efficiency of four terminal perovskite/silicon tandem cells to 26.4% [4]. In 2020, Amran Al-Ashouri et al. reported a monolithic perovskite/silicon tandem solar cell with a certified PCE of 29.1% by enhanced hole extraction [5].

Perovskite solar cells become a star as an excellent potential alternative to silicon-based solar cells. However, whether it is silicon single crystal solar cells or perovskite solar cells, their PCE are not high enough. For instance, currently, the efficiency of silicon based solar cells are around 25.6%, close to the Shockley-Queisser limit efficiency (29.4%) [6].

Although crystal silicon solar cells occupy the vast majority of the solar cells market, it can be predicted that in the future perovskite/silicon tandem solar cells have broad application prospects due to the wonderful properties. Therefore, this paper will describe the structure of the new devices, characteristics, advantages and disadvantages of several perovskite materials and silicon single crystal that are most widely used in perovskite/silicon tandem solar cells.

2. The structure of perovskite/silicon tandem solar cells

Perovskite/silicon tandem solar cell is a kind of series tandem cells. From a structural perspective, they include four terminal perovskite/silicon tandem solar cell and two terminal perovskite/silicon tandem solar cell.

Four-terminal perovskite/silicon tandem solar cell are a type of tandem solar cell architecture that consists of two separate solar cells, which are made of perovskite and silicon separately and connected in series through a four-terminal interconnection scheme. These two cells are stacked together and connected in series through a transparent interconnection layer. In a four-terminal perovskite/silicon stacked battery, each cell has its own set of two electrical contacts, resulting in four terminals in total. The two contacts of the top cell are connected to the top terminals, and the two contacts of the bottom cell are connected to the bottom terminals. The top and bottom terminals are connected to an external circuit. The advantage of the four-terminal architecture is that each cell is independent. Each cell can be changed without considering other cells, which reduces the effects of current mismatch and allows each cell to use different materials and designs, enabling greater flexibility in the design of the cells.

Two-terminal perovskite/silicon tandem solar cell has a similar structure with four-terminal perovskite/silicon tandem solar cell. The only difference is that the two separate solar cells are connected in series through a two-terminal interconnection scheme. Therefore, in a two-end perovskite/silicon stacked battery, the two cells share a common set of electrical contacts, resulting in only two terminals. The top and bottom of the device are connected to external electrical contacts. The advantage of the two-end architecture is that it simplifies the device structure and reduces the complexity of the fabrication process, which can help to lower the cost of manufacturing. However, the perovskite and silicon cells must be carefully designed to avoid bad current matching and reduce optical loss.

3. Perovskite top cells

3.1. Methylammonium lead iodide ($MAPbI_3$)

$MAPbI_3$ is one of the most widely studied materials for use as the perovskite top cell. It has a bandgap of around 1.6 eV, which makes it well-suited for use as the top cell in a tandem device with a silicon bottom cell. The absorption coefficient of this material is very high, no matter in visible region or near-infrared region of the spectrum. It was first used as a material for solar cells in 2009. A Japanese scientist

DOI: 10.54254/2755-2721/24/20230693

added two materials with perovskite crystal forms $CH_3NH_3PbBr_3$ and $CH_3NH_3PbI_3$ as dyes to a dye sensitized battery with mesoporous TiO_2, achieving a battery efficiency of 3.8% [7]. Since then, solar cells using it as a material have developed rapidly, and as of 2017, the highest conversion efficiency value in its laboratory was 19.3%, which is very close to the conversion efficiency using Si as a material [8].

Methylammonium lead iodide has many obvious advantages. It has a high absorption coefficient and good carrier transport properties, which can greatly improve its PCE. Moreover, the bandgap of it is changeable. The exact value of the bandgap is decided to halide composition, making it possible to optimize the bandgap for tandem solar cell applications. Compared to other kinds of solar cells, the solar cells made of $MAPbI_3$ cost less.

However, there are still some problems with this material. First, $MAPbI_3$ is tend to degradation when the devices are exposed to moisture, heat, and light, which can lead to a decrease in device performance over time. Second, Pb is a toxic material, and the use of $MAPbI_3$ in large-scale production may raise environmental and health concerns. Therefore, perovskite materials have also begun to try pure inorganic materials and pure lead-free materials.

3.2. *Formamidinium lead iodide ($FAPbI_3$)*

$FAPbI_3$ ($HC(NH_2)_2PbI_3$) is another widely studied material. It has a bandgap of around 1.5 eV in the visible region of the spectrum. In 2015, Seok et al. first introduced FA^+ organic cations in perovskite materials to replace organic cations MA + and proposed a molecular exchange method to improve the crystal quality of perovskite thin films. In this method, the PCE of the devices exceeded 20% for the first time [9]. In 2017, Seok et al. successfully increase the PCE of the devices to 22.1% by improving the deep level defect state. This achievement was based on introducing additional iodine ions into the precursor liquid [10]. In 2020, Haizhou Lu et al. successfully achieved room temperature stable a-$FAPbI_3$ perovskite material through a gas-assisted growth method, and prepared high-efficiency stable solar cells with a photoelectric conversion efficiency greater than 23% [11]. In 2022, You Jingbi et al. published a paper in Science, which states that a certified PCE of 25.6% was obtained for $FAPbI_3$ perovskite solar cells, and 80% efficiency was still maintained after 500 hours of operation at 85 ℃ [12].

In terms of PCE and absorption spectrum, $FAPbI_3$ has similar performance to $MAPbI_3$. Compared to $MAPbI_3$, $FAPbI_3$ has been shown to be more stable and have better resistance to moisture, heat, and light-induced degradation. Compared to $MAPbI_3$, $FAPbI_3$ contains less toxic lead, making it a potentially more environmentally friendly and safer material for large-scale production.

However, the disadvantages are that $FAPbI_3$ is more expensive to produce than $MAPbI_3$ and it also has lower carrier mobility than $MAPbI_3$. $FAPbI_3$ is more sensitive to processing conditions, such as temperature and pressure. Therefore, the stability of tandem devices made by FAPbI3 is lower when the temperature is high.

3.3. *Cesium lead iodide ($CsPbI_3$)*

$CsPbI_3$ is also an excellent and promising potential material. $CsPbI_3$ is a promising perovskite material that has a wider bandgap than $MAPbI_3$ and $FAPbI_3$ (around 1.8 eV). In 2019, Zhao Yixin et al. successfully made a kind of solar cells based on β-$CsPbI_3$ whose PCE is over 18% [13]. In 2022, Luo Jingshan et al. fabricated a kind of perovskite solar cells made of γ-$CsPbI_3$. The PCE of this cell exceeds 18%, and its stability is also improved [14].

The difference between $CsPbI_3$ and the first two materials is significant. $CsPbI_3$ is an all inorganic perovskite material, while the first two materials are both organic perovskite materials. Therefore, $CsPbI_3$ exhibits much higher stability than the first two organic perovskite materials in high temperature and light environments. Meanwhile, $CsPbI_3$ has a narrower bandgap. Because of this unique property, the devices made of $CsPbI_3$ can absorb light in the near-infrared region. Therefore it can be a better candidate for use as the top cell, compared to the first two organic perovskite materials.

Unfortunately, although $CsPbI_3$ has several advantages and higher PCE than $MAPbI_3$ and $FAPbI_3$, it

DOI: 10.54254/2755-2721/24/20230693

still has two obvious disadvantages. First, while $CsPbI_3$ is more stable than some other perovskite materials, it is still vulnerable to degradation under moisture, oxygen, and light exposure. Second, the process of producing $CsPbI_3$ is very complicate so the material quality and properties of $CsPbI_3$ cannot be ensured

4. Silicon based cells

4.1. Crystalline silicon (C-Si)

This is the most widely used material for solar cells. The technology of silicon single crystal solar cells has become very mature, accounting for 90% of the current solar cell market because of its high photoelectric conversion efficiency, good stability and abundant availability. But it can be relatively expensive to manufacture and is only able to absorb a narrow range of wavelengths of sunlight, limiting its ability to efficiently capture all of the available energy from the sun. Its structure is rigid, which means that it cannot be easily shaped or molded and is hard to be integrate into flexible or portable solar devices.

According to a research published by Zhao Peng et al., since the bandgap of crystalline silicon is 1.12 eV, the perovskite cells whose bandgap are around 1.60~1.75 eV have the highest matching degree with crystalline silicon cell, and the obtained laminated cell has the best conversion efficiency [15]. However, the bandgap of $MAPbI_3$, $FAPbI_3$ and $CsPbI_3$ is around 1.6 eV, 1.5 eV and 1.8 eV respectively. In a word, the biggest problem of using crystalline silicon is that the matching degree between crystalline silicon and current mainstream perovskite solar cell materials is not high enough.

4.2. Thin-film silicon (TF-Si)

Thin-film silicon is another option for the material of silicon bottom batteries in perovskite/silicon series solar cells. The cost of thin-film silicon cells in the process of manufacture is very low. More importantly, thin-film silicon has a higher absorption coefficient than crystalline silicon, which allows it to absorb more light and generate more electricity per unit of material. Its structure is more flexible than crystalline silicon, which enable it to some portable solar devices. The manufacture method of thin-film silicon solar cells are various. They are typically made through deposition techniques. Among them, the most classic and widely used method is plasma-enhanced chemical vapor deposition (PECVD) and sputtering. In this way, its cost can be greatly reduced.

However, the disadvantages of thin-film silicon solar cells are obvious too. First, these devices typically have lower power conversion efficiencies than the ones made of crystalline silicon. The main reason is that the bandgap of thin-film silicon is decided by the composition of thin-film silicon, surface roughness, film quality and so on. The bandgap of thin-film silicon is uncertain in the range from 1.1 eV to 1.7 eV. If the bandgap is low, for example, close to 1.1 eV, the PEC of thin-film silicon is certainly low. While the bandgap is close to 1.7 eV, the PEC of thin-film silicon is high. Therefore, if the high PEC of thin-film silicon is required, the important factors including the composition, surface roughness, film quality and etc. must be controlled strictly in the process of manufacturing, which raises the cost and reduced the qualified rate. Second, their stability is low because of the defects and impurities that appear in the process of manufacture, which means that the cells made of thin-film silicon has a shorter lifespan compared to the ones made of crystalline silicon. Third, thin-film silicon is more sensitive to high temperatures than crystalline silicon, which makes the cells made of thin-film silicon sometimes limited or even invalid in some high temperature environment.

4.3. Heterojunction with intrinsic thin (HIT) layer (Si)

HIT layer is a type of c-Si cell that have a thin layer of intrinsic amorphous silicon (a-Si) between the p-type and n-type layers. The heterojunction solar cell was first developed in 1990s by Sanyo Company [16]. The PCE of it was 12%. In 2017, the PCE of HIT-Si devices made by Kaneka Corporation in Japan was over 26.7% [17].

HIT-Si cells have an extremely high PCE because of its unique and high-quality intrinsic amorphous

DOI: 10.54254/2755-2721/24/20230693

silicon thin layer. Through controlling the composition of this high-quality a-Si thin layer, the bandgap of it can be adjustable. The bandgap can be adjusted to match the one of perovskite top cells. The stability of HIT-Si devices is also high due to the stable output voltage and low degradation rate. In addition, compared to thin-film silicon cells, HIT-Si cells are much less sensitive to high temperature. But HIT cells are more expensive to manufacture than the other two types of cells. The manufacturing process of HIT-Si cells is complex because the quality of the a-Si thin layer is the key point and hard to control.

5. Conclusion

The development and progress of perovskite/silicon tandem solar cells are very optimistic. Through this technology, the manufacturing cost of solar cells can be significantly reduced, more range of solar spectrum can be used so the PCE of solar cells can be hugely improved. It can be predicted that the theoretical limit efficiency of solar cells can be broken through. But there are still many unresolved issues. For example, the stability of hybrid organic perovskite materials needs to be improved, and the toxicity issues during mass production need to be addressed. Currently, researchers are attempting to replace hybrid organic perovskite materials with fully inorganic perovskite materials or tin based perovskite materials. Mixing different types of perovskite materials is also a good solution. Through this method, it is possible to leverage strengths and avoid weaknesses, further improving the PCE and stability of solar cells. Meanwhile, when improving perovskite materials, attention should be paid to the degree of compatibility between the top and bottom cells. In short, the technology of this new type solar cells is still in the laboratory stage and cannot be produced in industrial scale.

References

[1] Melnick J G, et al. 2007. MerB. Science, 317 225-227
[2] Pellet N, et al. 2014. Angewandte Chem Int Ed, 53 3151-3157
[3] Werner J, et al. 2016. J. Phys Chem Lett, 7 161-166
[4] Duong T, et al. 2017. Adv. Energy Mater, 7 1700228
[5] Al-Ashouri A, et al. 2020. Science, 370 1300-1309
[6] Li C, et al. 2018. Physics, 47 367-375
[7] Kojima A, et al. 2009. J Ame Chem Soc, 131 6050-6051
[8] Zhou H, et al. 2014. Science, 345 542-546
[9] Yang W S, et al. 2015. Science, 348 1234-1237
[10] Yang W S, et al. 2017. Science, 356 376-1379
[11] Lu H, et al. 2020. Science, 370 eabb8985
[12] Zhao Y, et al. 2022. Science, 377 531-534
[13] Wang Y, et al. 2019. Science, 365 591-595
[14] Duan L, et al. 2022. ACS Energy Lett, 7 2911-2918
[15] Zhao P. 2022. Xidian University, PhD dissertation
[16] Tanaka M, et al. 1992. Japanese J Appl Phys, 31 3518
[17] Green M A, et al. 2022. Prog. Photovolt. Res. Appl, 30 687-701