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The Advancement of Silicon as A Photovoltaic Material: Historical Accomplishments, Present Efficiency and Future Development

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Abstract: In today's society where energy and environmental issues are increasingly gaining attention, research, development, and utilization of renewable and clean energy continuously attract the interest of academia, environmental protection, and business alike. Its application has already entered people's lives and is gradually expanding its market share. As a result, there is a growing demand for more efficiently designed, environmentally friendly and less costly new energy devices. Among various energy resources such as tidal, wind or geothermal, solar energy stands out as the most widely used clean and renewable resource. Given that solar energy is virtually limitless for humans, the function of solar cells in converting solar energy to electricity demonstrates significant social, environmental, and economic benefits. For the development of solar energy and new research, photovoltaic materials are crucial. Non-toxic and with excellent photovoltaic properties, silicon is the second most abundant element in the Earth's crust and the most abundant in water. Since crystalline silicon solar cells produced for the first time in 1954, silicon has held the most important material position in photovoltaic cell manufacturing and maintained market dominance. The importance of silicon solar cell research, its development and current status are briefly outlined in this paper. It reviews the advantages and disadvantages of existing silicon-based photovoltaic materials and potential future developments. As a photovoltaic material, silicon holds significant advantages compared to others, suggesting a broad scope for both research and commercial prospects in the long term.

Keywords: Silicon, Photovoltaic material, Solar cell, Photovoltaic cell.

1. Introduction

1.1. Research significance of photovoltaic materials

In today's energy and environmental issues are increasingly concerned, although mineral fossil energy still occupies a key position in modern energy use, how to develop new clean energy and renewable energy has become a top priority. Of all the various sources of renewable energy, the sun's energy is the most inexhaustible for human beings. At the same time, compared with all kinds of traditional non-renewable energy and other clean energy, solar energy has significant environmental benefits and utilization feasibility, conducive to the realization of sustainable development, so it has become the top priority of energy research. It is clear that photovoltaic (PV) technology - the direct conversion of the sun's energy into electricity for use - has become a highly sought-after and promising area of research.

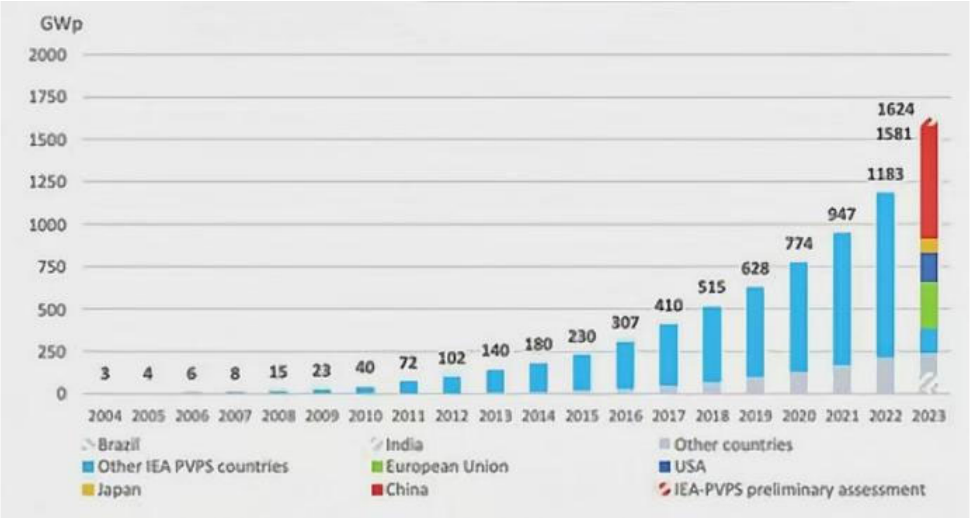

Figure 1. Cumulative installed PV system capacity (GW) from 2004 to 2023

In the context of photovoltaic systems, solar cells occupy a pivotal role, underscoring the significance of photovoltaic materials in the pursuit of novel energy sources. The efficiency of solar cells has been on the rise in recent years as a result of continuous research and improvements in photovoltaic materials. It is exciting that in 2011, the global

PV market has reached a high installation of 27.4GW.[1] Since 2018, the global rooftop PV market has continued to grow gradually, while the reduction in installation costs and the rise in electricity costs have prompted investors to accelerate investment in rooftop PV systems. According to the "Global PV Market Snapshot 2024" report released by the IEA-PVPS, the installed capacity of photovoltaic systems worldwide has grown from 1.2TW in 2022 to 1.6TW in 2023. Specific values and trends can be seen in Figure 1. The market demand for more efficient and lower-cost photovoltaic materials is still expanding, which also means broader research prospects.

1.2. The importance of silicon in photovoltaic materials

Due to its superior photoelectric performance and high reliability, but also because of its relatively low preparation cost, inorganic photovoltaic materials have always been concerned. Silicon is one of the most prevalent inorganic photovoltaic materials, comprising approximately 28% of the Earth's crust. Consequently, silicon possesses a substantial reserve that can be exploited as a vital material for an extended period. Silicon not only has the advantages of inorganic photovoltaic materials, but also has the advantages of superior photovoltaic properties and mature preparation methods, and can be used as a key material by the electronics and photovoltaic industry for a long time. The data indicate that in excess of 90% of solar cells are manufactured on a silicon crystal basis on an annual basis.[2] Because of this, silicon wafer technology is the most important technology in today's photovoltaic technology. At the same time, the development of silicon materials continues to attract significant research and commercial attention and efforts in various countries to improve their conversion efficiency and reduce manufacturing costs.

2. The History of Silicon as Photovoltaic Materials

Over the past century, silicon has always been considered the most important material for manufacturing photovoltaic (PV) cells (panels). At the same time, silicon has been shown over the past 30 years to be a degraded photovoltaic material with high reliability and limited efficiency. Bell LABS in the United States first produced 6% crystalline silicon solar cells in 1954, since this milestone achievement, photovoltaic power generation technology has become a worldwide concern and reached a leap in the development process. Martin Green once felt that "the development of solar cells has gone through three generations." Following decades of development since the initial introduction of solar cells, silicon-based solar cells have demonstrated considerable advancement. The advancement of silicon solar cells has proved advantageous for numerous countries, enabling them to attain grid parity. Consequently, silicon-based photovoltaic modules have become the dominant technology in this field.

The photovoltaic industry is characterised by a diverse range of technologies, with silicon-based solar cells occupying a dominant position in the market. In the past two decades, crystalline silicon cell manufacturing has experienced three major technological iterations, and solar cells have developed from the first generation to the fourth generation. During the Al - BSF (Aluminium Diffused Back Surface Field) era, PV cell efficiencies were less than 20%, while at the PERC (Passivated Emitter Back Contact) stage, efficiencies have improved, but are still below 25%. Since last year, the TOPCon (Tunneled Oxide Passivated Contacts) technology upgrade has been carried out, resulting in a great improvement in PV cell efficiency, directly exceeding the 25% value. In May of this year, LONGi Green Energy Technology Co., Ltd. announced that its self-developed back-contact crystalline silicon heterojunction solar cells (HBC), with a photoelectric conversion efficiency of up to 27.30 per cent, which once again broke the world record for monocrystalline silicon photovoltaic cell conversion efficiency. It is worth mentioning that this is another major breakthrough after LONGi set the world record of 27.09% conversion efficiency of HBC cells in December 2023, which demonstrates LONGi's excellent strength and continuous innovation ability in the field of photovoltaic technology research and development.[3]

Figure 1 is a compilation by the National Renewable Energy Laboratory (NREL) of the highest proven research cell conversion efficiencies for solar cells prepared by different photovoltaic technologies from 1976 to 2022.

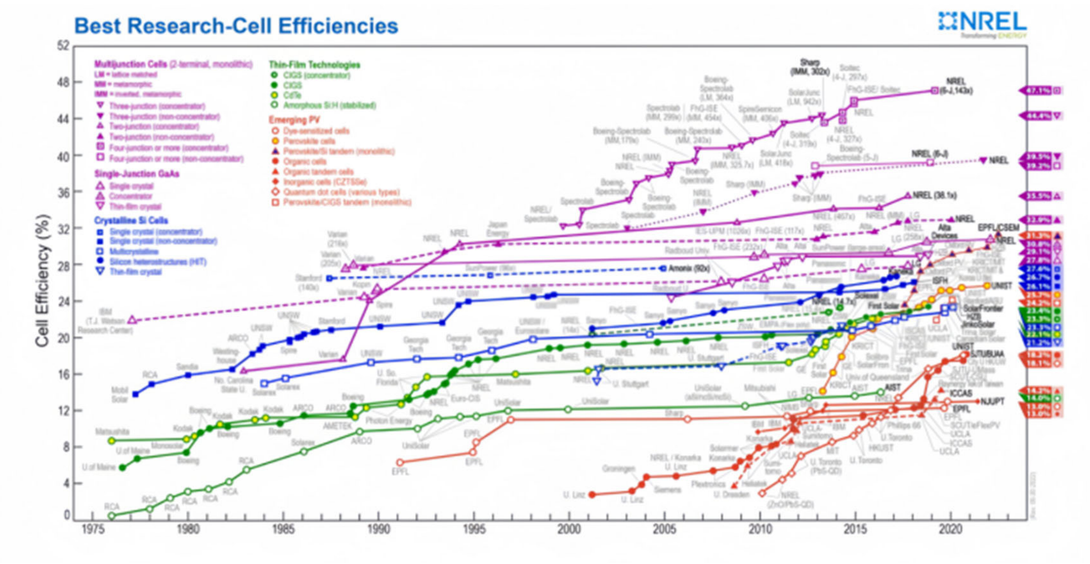

Figure 2. NREL Best Research-Cell Efficiencies chart

3. Current Typical Types of Silicon as Photovoltaic Materials

3.1. Introduction to Silicon

Silicon, an indirect semiconductor, has a high degree of stability and diffusion length due to its band gap of 1.12 eV at the temperature of the room. This bandgap is perfectly aligned with the AM1.5 standard solar spectrum. The theoretical efficiency of single-junction silicon solar cells can reach 29.4% when direct recombination, Auger recombination and defect-assisted recombination are taken into account and calculated using the Shockley-Queisser formula.[4] Additionally, the indirect band-gap absorption coefficient of silicon is relatively low, allowing for only gradual change around the band-gap energy. Consequently, a relatively thick chip is necessary to absorb all light above the band-gap photon energy. The relatively thick chip also means that the material cost increases, and this may also lead to more bulk recombination, and the open circuit voltage will decline.

A number of key factors can impact the fundamental efficiency of a system: bandgap, capturing all the photons above the bandgap and collecting all the generated charge carriers. The quality of the material, especially when it is related to volume, interface, and charge carrier trapping caused by interface defects, plays a vital role in determining the final photovoltaic conversion efficiency. The efficiency of solar cells can be enhanced by optimising the structure of high-quality silicon materials.[5] Table 1 provides the performance parameters of different silicon solar cells.

Table 1. Performance parameters of different silicon solar cells.

Device	Eff. (%)	V_{OC} (mV)	J_{SC} (mA cm^{-2})	FF(%)	Description
Ideal, 110μm	29.4	761	43.3	89.3	Modeled[6]
n-type mono-Si	26.3	744	42.3	83.8	Kaneka[7]
n-type mc-Si TOPCon	22.6	678	40.8	81.9	FhG-ISE[8]
n-type UMG-Si PERL	21.1	666	35.7	79.9	ANU[9]
p-type mono-Si PERC	22.61	684	40.5	81.5	Trina solar[10]
p-type mc-Si PERC	21.3	667	398	80	Trina solar[11]

In contrast, composite silicon solar cells have lower technical requirements and lower production costs, and therefore have a larger market share compared with single-crystal silicon solar cells. Nevertheless, the conversion efficiency of composite silicon photovoltaic cells remains inferior to that of single-crystal silicon cells, with an average difference of approximately 1% to 2%.[5] Therefore, the research and development of battery structures that require better cell performance are all based on single-crystal silicon, while composite silicon is the basis for experiments aimed at improving the quality of the material.

3.2. Crystalline silicon in conventional solar cells

3.2.1. The manufacture of silicon solar cells

The most popular crystalline silicon photovoltaic modules on the market are typically prepared through the following process. The production of silicon metal begins with the use of high-purity silica as a raw material. This is processed through a high-quality carbon in an arc furnace, undergoing a reduction reaction to obtain approximately 99% purity of metallurgical silicon. However, this process also results in the presence of minor impurities, including aluminium, iron, and gallium. Then it is further purified by chemical means such as the Simenzi process to obtain solar grade silicon (purity of about 99.9999%). Here Czochralski or directional solidification can be used to achieve silicon ingot growth and obtain monocrystalline silicon and polysilicon.[12] Finally, the wafer can be obtained by wire-cut silicon ingot.

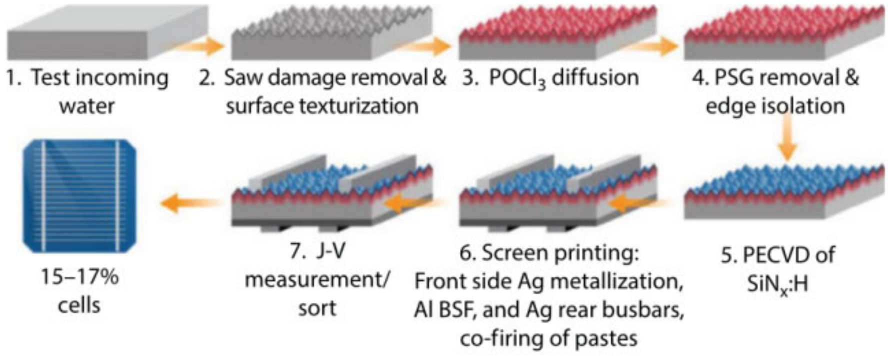

Figure 3. Production process of P-type silicon solar cell.[5]

Figure 3 illustrates the manufacturing process of a silicon solar cell, which involves the diffusion of phosphorus and the formation of the p-n junction of the silicon wafer through the Al-back surface.

The silicon solar cell is, in fact, a p-n junction device, which makes the fabrication process of the junction a matter of great importance. The following section will present a detailed description of the production process of a p-n junction. After the wafer is annealed at high temperatures, under normal circumstances, the doping will form an n-type layer a few microns thick (the doping here is usually phosphorus). In this case an doped region is formed and the n-doped region is defined as an "emitter". The electrons and hole pairs created by the light are separated and effectively collected in this region. In the environment of nitrogen or argon, phosphorus diffusion usually occurs in the temperature zone of about 850°C to 900°C, and the phosphorus source is phospho-oxychloride ($POCl_3$).[5] The reaction of O_2 with $POCl_3$ to produce phosphorus pentoxide is very important in this process. Here is the reaction:

$$4POCl_3 + 5O_2 = 2P_2O_5 + 6Cl_2 \uparrow \tag{1}$$

$$2P_2O_5 + 5Si = 4P + 5SiO_2 \tag{2}$$

The phosphor diffusion stage has another important benefit for solar cells, especially polysilicon cells. During phosphorus diffusion, contamination due to metal impurities in the thicker parts of the material (e.g. gelling effects) can be minimised. Transition metals (e.g. iron, nickel, chromium and copper) can be efficiently transported to the surface region, thus extending the recombination lifetime of the minority carriers.[13] [14]

Following the diffusion of phosphorus, a phosphosilicate glass layer (PSG) is formed on the surface of the wafer. The complete removal of the PSG layer and edge isolation can be achieved through the application of wet chemistry, whereby the wafer is immersed in a hydrofluoric acid solution. This process ensures that only one p-n junction forms on top of the solar cell.[5]

3.2.2. Efficiency loss of silicon-based solar cells

The band gap of silicon is 1.2 electronvolts, which is consistent with the standard solar spectrum. Based on the detailed balance, it can be seen that the mobility is infinite, the carrier generated anywhere can be collected, and as long as the photon is above the band gap, it can be fully absorbed. According to the findings of Shockley and Queisser (1961), the maximum efficiency of a semi-infinitely thick silicon photovoltaic device is 33.5% when both room temp. and AM1.5 are met.[8] A theoretical efficiency of 29.4% is obtained by recalculation when the effects of radiation and auger recombination are taken into account.

The following is how silicon solar cells operate: First light generates electron-hole pairs, then p-n junctions separate electrons and holes, and the solar cell outputs voltage, thereby supplying power to the external circuit load.

The efficiency of photovoltaic cells is divided into three different types, thermodynamic efficiency, conductive efficiency and carrier separation efficiency. The total efficiency is calculated by adding the individual efficiencies.

The proportion of the incident power converted into electricity determines the efficiency of the solar cell, which is defined as:

$$\eta = \frac{V_{OC} I_{SC} FF}{P_{in}} \tag{3}$$

In this context, the term V_{OC} refers to the open-circuit voltage, while I_{SC} represents the short-circuit current, FF denotes the fill factor, and finally, η signifies the efficiency.

Figure 4 shows the efficiency loss of a silicon solar cell at different steps.

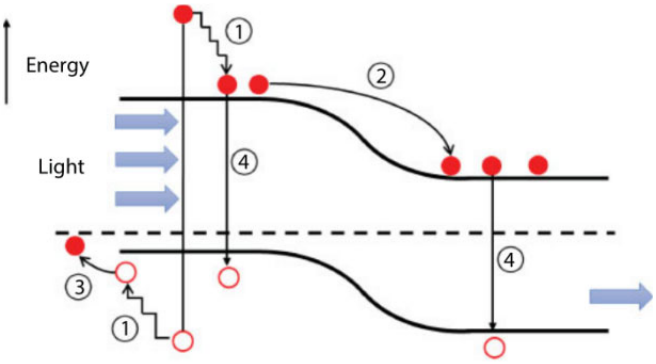

Figure 4. Efficiency loss of silicon solar cells at different steps.[5]

The efficiency of silicon-based solar cells is contingent upon a multitude of factors, including the materials utilized, the manufacturing techniques employed, and the structural configuration of the device. These factors can give rise to a number of potential losses, including optical loss, recombination loss, and contact resistance loss, which collectively serve to diminish the overall efficiency of the solar cells. Generally speaking, these losses are divided into two categories, one is the electrical loss, the other is the optical loss.

3.2.3. A way to increase the efficiency of solar cells

In order to boost the efficacy of silicon-based solar cells, a multitude of measures have been implemented in the past to mitigate losses by optimising manufacturing processes, utilising superior-grade raw materials, and enhancing equipment design.

At present, there are several ways to control the loss of solar cells: (1) the use of superior quality materials, such as n-type silicon or high-performance silicon, be employed; (2) Reduce the recombination of carriers in the front and rear surface areas by improving passivation technology, for example by enhancing the passivation film; (3) Enhancing the preparation technology of the p-n junction in order to obtain an improved emitter, while simultaneously increasing the separation of effective electrons and holes within the junction region; (4) Reduce contact loss by improving the contact performance of the metal.

It has been shown that the ultimate efficiency of silicon-based solar cells depends on three key factors: short circuit current, fill factor and open circuit voltage. Higher efficiency can be reached by increasing V_{OC}, FF and I_{SC} to further improve wafer quality, light trapping, emitter, metal contact and front and back surface passivation. Figure 5 illustrates potential avenues for boosting the efficiency of silicon solar cells.

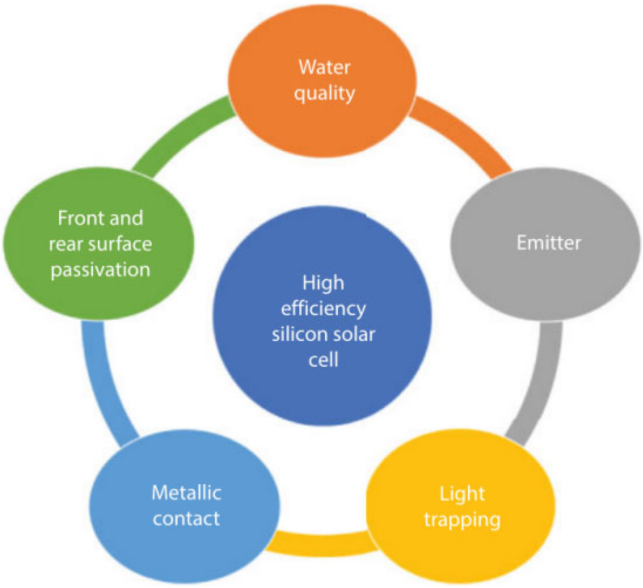

Figure 5. New strategy for high-performance silicon solar cells. [5]

3.3. Low cost crystalline silicon

Silicon wafers account for a large proportion of the cost of silicon photovoltaic systems, and even if the price of crystalline silicon has declined in recent years, this part of the cost cannot be ignored. In order to save the cost of silicon photovoltaic systems while pursuing more efficient silicon photovoltaic systems, we can achieve this goal by improving crystal growth and crystal purity.

3.3.1. Ordinary metallurgical silicon

The general method of production of crystalline silicon ingot is to carry out carbothermal reduction of high-purity raw material SiO_2 at about 1700°C, but it is accompanied by serious adverse environmental side effects, such as carbon release, high energy consumption and heat release. And the resulting metallurgical silicon is not pure enough to be used in photovoltaic systems. The good news, however, is that *Chen et al.* and *Nhoira et al.* have reported a new method that can be used to produce silicon more quickly and completely, while using less energy. Silica powder or a mixture of other metal oxide powder can be transformed into porous particles, and pure silicon or a silicon alloy with the same properties can be produced by electric reduction in molten $CaCl_2$ at 900°C.[5]

3.3.2. Upgraded metallurgical silicon

In contrast to the utilisation of chemical methodologies (e.g. the Modified Siemens Method), the production of SOG-Si (solar grade silicon) is conducted via a metallurgical processing route, resulting in the generation of upgraded metallurgical grade silicon (UMG-Si).[5] Compared to the production of SOG-Si requires high silicon materials, complex technology and generated exhaust pollution, UMG-Si production costs are lower. Because the production process has low energy consumption, low infrastructure costs, and the process only needs low temperature, the cost of high energy consumption and high temperature process is avoided. However, compared with SOG-Si, the impurity concentration in UMG-Si is higher, about ppm, and contains metal impurities such as B, Al and P.

3.3.2.1. Characteristics of upgraded metallurgical silicon

UMG-Si contains higher concentrations of aluminium, boron and phosphorus than conventional solar cell grade silicon wafers. Although the metal impurity content of UMG-Si is marginally greater than that of polycrystalline silicon (mc-Si), it is still within the electric performance range of silicon wafers.[15]

Upgraded metallurgical grade silicon has a 30% to 70% higher concentration of substitutional carbon and interstice oxygen than common polysilicon chips. The content of crystal defects (mainly dislocation) of the two is the same. Therefore, for the final battery performance, only a few impurities with higher concentrations may increase the activity of defect recombination in UMG wafers due to enhanced defects, while the adverse effects of carbon and oxygen concentrations and defect content are almost zero.

While metallurgical methods are relatively effective in removing metal impurities like iron and copper, the high segregation coefficients of B and P impurities (B segregation coefficient: 0.8 to 0.9, P segregation coefficient: 0.36) make their removal challenging. Consequently, UMG silicon exhibits a diminished carrier lifetime, which is consistently offset by the elevated residual density of dopant atoms. Consequently, it is challenging to either mitigate or resolve the issue of low carrier mobility and pronounced photodegradation of p-type silicon or even n-type silicon in B-O complexes.[16]

The life of a few carriers determines the quality of the silicon, and the effective life of a few carriers in UMG silicon wafers is only one-third or less of that of traditional mc-Si. In this way, the efficiency of the solar cells can be reduced due to their poor electrical performance.[17]

3.3.2.2. Development of upgraded metallurgical grade silicon

UMG-Si has developed rapidly over the past few years, helping to increase the PCE used in UMG-Si solar cells. It can be used alone in the photovoltaic industry and can be mixed with electronic grade silicon characteristics so that its research has been concerned by researchers. As early as 2010, the efficiency of p-type silicon solar cells was a notable development, with polycrystalline UMG-Si solar cells reaching 17% efficiency and single-crystal UMG-Si solar cells achieving 17.6% efficiency. Following years of research, *Zheng et al.* achieved an efficiency of 20.96% for 100% n-type Cz UMG-Si solar cells in 2016. They subsequently enhanced the battery efficiency to 21.1% around 2018.[5]

3.4. High performance multicrystalline silicon

Although traditional mc-Si solar cells occupy a part of the market with shorter crystal growth technology and lower cost, its defects and impurities can adversely impact the effectiveness of solar cells. Compared to traditional mc-Si, the quality of new high performance mc-Si (HP mc-Si) is significantly improved by fine crystallisation of dislocation clusters and reduction of density. As a result, HP mc-Si has demonstrated superior conversion performance and energy output power in the solar cell and photovoltaic industry.

3.4.1. Study on properties of high performance polysilicon materials

Crystal defects contain dislocations, grain boundaries, and impurities in the silicon material, and these defects have the probability to cause additional energy states in the energy gap and become SPH recombination centers.[5] If the carrier recombines at the defect, it may shorten the life of a few carriers and lead to solar cell performance to degrade. Since impurities can easily accumulate at the dislocation and grain boundaries, impurity modification defects exhibit increased recombination activity. Compared with traditional mc-Si, high-performance mc-Si has significantly reduced dislocation cluster density and lower random Angle grain boundary recombination activity.[5]

The minority carrier lifetime in HP mc-Si is both longer and more homogeneous than in traditional mc-Si. In HP mc-Si, the short life region caused by dislocation clusters is markedly diminished in comparison to traditional mc-Si, and the carrier lifetime in the wafer exhibits minimal fluctuation.

The formation of the emitter is a step in the procedure of solar cell preparation to absorb impurities. In order to mitigate the detrimental effects of metal impurities on silicon material defects, phosphorus diffusion quenching is typically the preferred approach. The difficulty in removing impurities at the defect site results in a significantly reduced operational lifetime for traditional mc-Si compared to HP mc-Si after phosphorus removal.[18]

3.4.2. Solar cells based on high performance polysilicon research

The advancement of crystallisation technology has facilitated the enhancement of material quality, thereby facilitating the advancement and proliferation of mc-Si solar cells, making it the primary material used in silicon photovoltaics. The efficiency distribution of HP mc-Si solar cells is higher and narrower. In a recent announcement, Trina Solar revealed an industrial manufacturing process efficiency of 21.25% for the p-type HP mc-Si. Additionally, *Schindler et al.* reported in 2017 a TOPCon battery with an efficiency of 21.9% based on the n-type HP mc-Si. In 2018, the utilisation of a boron diffusion front emitter and full area passivation contacts resulted in an efficiency value of 22.6%.[8]

4. The Future of Silicon as Photovoltaic Materials

In comparison with traditional silicon solar cells, the development of upgraded metallurgical grade silicon and high-performance polysilicon has demonstrated satisfactory solar cell efficiency in recent years. Concurrently, the financial outlay required for the procurement of the requisite silicon material, the costs associated with the requisite preparation technology and the complexity of the manufacturing process have all been reduced, which is highly conducive to the commercialisation of these products. Nevertheless, there is considerable scope for enhancement in the efficiency loss of solar cells resulting from crystal defects, including impurities, displacement and grain boundaries. It is anticipated that in the near future, scientists will either overcome the efficiency loss caused by defects or discover new, more efficient and low-cost silicon-based solar cells.

The efficiency of these cells has been steadily improving, thanks to ongoing research and development in materials science. Innovations in this area, for instance Passive-Emitter-Rear-Cell (PERC) technology, have played a key role in improving the conversion efficiency of silicon solar cells.

As the technology matures, the cost per watt of crystalline silicon photovoltaic systems continues to decline. Economies of scale, along with improvements in manufacturing processes, have significantly reduced the price of solar modules. This trend is likely to continue, with the result that solar energy will become more competitive with traditional power generation based on fossil fuels.

The environmental impact of silicon-based solar cells is another area where progress has been made. The lifecycle assessment of these cells indicates a lower carbon footprint compared to other forms of energy production. As a contribution to global effort to lower greenhouse gas emissions and mitigate climate change, the sustainability of solar energy will continue to improve with improved recycling technologies for silicon modules.

In the past few decades, crystalline silicon has undergone significant advancements in the production chain, establishing itself as a cost-effective power source with immense potential. There are compelling indications that by 2040-2050, carbon-silicon photovoltaic cells could emerge as the dominant source of electricity globally.[20]

5. Summary

Over the past few decades, the global photovoltaic (PV) market has exhibited a remarkable growth rate of approximately 50% per year. Silicon has consistently been the predominant material in the photovoltaic industry, with a global PV market share reaching 90%. In response to the growing demand for enhanced photoelectric conversion efficiency, silicon solar cells have been rapidly developed through improvements in material quality, the introduction of novel device structures, enhanced performance, and reduced costs. The ultimate conversion efficiency is contingent upon a multitude of factors, the most pivotal of which are recombination within the silicon material, utilisation of light, separation of carriers at the junction, and transport of carriers between the semiconductor and the metal electrode. Through changes in the manufacturing technology, low-cost UMG silicon and high-performance mc-Si exhibit material features, that include a higher minority carrier lifetime, a more uniform defect distribution, and a lower impurity concentration. The enhanced electrical performance has resulted in an improvement in the performance of solar cells, with only 54 emerging photovoltaic materials undergoing advancement. The introduction of new device structures, including PERC, IBC and SHJ, has effectively reduced contact loss and emitter recombination, while increasing the probability of photon collection. This has resulted in a notable enhancement in efficiency in comparison to conventional technology. Silicon photovoltaic modules are still too expensive and inefficient for large-scale global deployment. However, they remain the most promising photovoltaic industry.

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