

Research on recycling and disassembly of waste crystalline silicon solar cells

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Abstract. Past two decades have witnessed a rapid increase in the application of photovoltaics which is being vastly produced considering the fact that they are the crucial elements that contribute to the transition from conventional finite fossil energy to new-generation renewable energy. In order to offset the undesirable environmental impact attached to the mass production of Photovoltaics and also recover valuable metals, a comprehensive and environmentally friendly strategy of solar cells disposal and recycling handling are indispensable. Systematical analysis of different contemporary distinctive methods involved in end-of-life solar cell handling is needed for gaining a deep insight into the recycling process. This article mainly focuses on summarizing and comparing three highly effective methods for solar cells recycling and disassembly: physical treatment, chemical treatment, thermal treatment. Our results show that the physical method is mostly seen as a preliminary process that is indispensable before chemical or thermal treatment and that despite chemical treatments using solvents are crucial for the elimination and recovery process of valuable metals, there are several drawbacks, like harmful waste and damage to recovered silicon bulk, all of which are not contained by thermal treatment.

Keywords: solar cell recycling, physical disassembly, chemical treatment, thermal treatment, metal recovery.

1. Introduction

Solar energy, as a form of renewable energy, has developed rapidly in the past few decades and presents a huge market potential. At most times, aging is a significant factor that affects the performance of PV cells. Therefore, sustainable ways of end-of-life PV cell handling are essential for both economic and environmental perspectives. Solar cells contain valuable substances and materials, such as aluminum, crystalline silicon, etc. This paper mainly manipulates how to separate and recycle various materials in waste solar cells without destroying the integrity of various materials. The specific methods of materializing the goal, after consulting and sorting out a large number of papers and publications, can be divided into physical disassembly, chemical treatment, and heat treatment by different characteristics. This paper also makes a detailed description of the steps and principles of these methods. Among them, physical methods are generally used to separate aluminum and other materials of waste solar cells before chemical methods and heat treatment methods. Except chemical treatment is suitable for the recovery of metal, the mission of the chemical method and heat treatment method is similar, which are both applied to reach a clear elimination EVA film which is the outer layer of crystalline silicon that can only be recovered after the elimination of EVA film. The application of these two methods in modern industries reflects their advantages, disadvantages, and applicable scenarios. This article mainly introduces and compares the previous researches and findings of scholars and aims to encourage future research in the sense of providing a simple

understanding of the previous research and current situation and offering inspiration and direction for future research.

2. physical method

2.1. Introduction of the physical method

Physical methods are the basis for the recycling process of galvanic cells. To recycle solar cells, we first separate them into different parts and then recycle them according to the different properties of each part. Crystal silicon solar cell structure from top to bottom includes glass plate, EVA, cell, EVA, back-plane, and TPT [1].

2.2. Physical methods as the basis for solar cell recycling

One of the most important parts of the successful division is the battery piece. This requires the complete removal of EVA, and the extraction of crystalline silicon, as well as aluminum, silver, and other precious metals in solar cells.

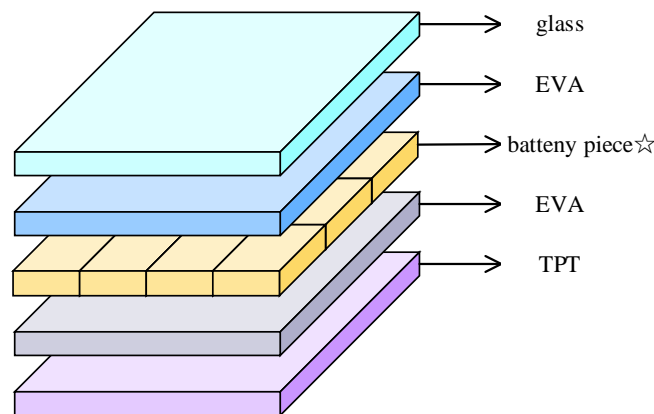


Fig. 1 The structure of the solar cell

Firstly, the cells in crystalline silicon solar panels are separated by physical methods, and then the metals in the cells are recovered and extracted by chemical or heat treatment methods. Finally, the solar level crystalline silicon can be directly used by cleaning and drying.

The battery is separated from the components first, and then the metal in the battery is separated from the battery, and finally, crystal silicon, aluminum, silver, and other precious metals are obtained.

The physical separation will just include separating the broken panels by mechanical separation but does not encapsulate material and inner materials like Ag/Al metallization.[19] The battery needs to be separated from the panel first while recycling aluminum frame, low iron tempered glass, and other usable auxiliary materials. The aluminum frame can be separated directly by manual disassembly.

2.3. Specific application of physical methods

Part B describes that the physical method can be the first step in the whole battery recycling process. This means that first, we use physical methods to separate the large pieces of cells to get to the cell plates, which are the most important part of the solar cells to be recycled. Then, we need to continue with other methods - chemical or thermal - to follow up and remove the EVA adhering to the plates.

Today, with the advent of technology, it is possible to obtain clean, high-quality solar panels with no EVA residue on the surface using only physical methods. These methods are also practical and effective.

2.3.1 Recycling of thin-film solar cells

Generally, we are using mainly chemical methods to recycle thin-film solar panels, which will use many steps. The first step requires the solar cell plate to be crushed first, which allows all the

structures of the solar cell plate to be destroyed, and the internal structure can thus be exposed. Then the second part is to use a chemical solvent, generally, we use an acidic solvent to dissolve the solid metal material, at this time the solution will contain tellurium ions and cadmium ions. The third step is to filter the EVA residue out of the second part of the solution, at which point we get the second stage of the solution. In the fourth step, we add alkaline substances to the second stage solution to obtain the third stage solution. The fifth step is to add polyaluminium chloride solution and activated carbon powder to the third stage solution, the concentration of polyaluminium chloride should be 8-12%, and the ratio of the volume of the third stage solution to the amount added should be 333:0.8-1.2. After the addition of polyaluminium chloride, activated carbon is added at the same time, and the mass ratio of activated carbon powder to the third solution is 0.5-2kg:60T, at this time we form the fourth stage solution. of the solution. In the sixth step, the tellurium compounds and cadmium compounds in the fourth solution are filtered out by DF membrane filtration. [20]

This approach breaks with the traditional notion of using chemical recovery methods, such as vapor phase and liquid phase methods to recover the tellurium and cadmium fractions of the cell. Instead, a purely physical method is used to recover thin-film solar cells.

The steps to recover thin-film solar cells by physical methods includes:

Crushing: The purpose of this step is to break the tempered glass layer inside the solar cell and form a web of cracks. The operation carried out is the knocking treatment, knocking the solar cell and up to make the internal glass layer broken effect.

Freezing: The freezing treatment is to freeze the thin-film solar cell that underwent the knocking process in the previous step, placing it in liquid nitrogen and using liquid nitrogen for freezing treatment. It should be noted here that the amount of liquid nitrogen should be sufficient to at least submerge the solar cell, and the freezing treatment time should be 10-60s, preferably controlled at 20-40s.

Separation: To separate the solar cell version with the first two steps after crushing, freezing treatment of thin-film solar cells. Here just use the method of tapping that can be separated at the solar panel.[21]

We can notice that only three steps are needed here to get solar panels, and the solar cells used in this recycling method should be multi-compound solar cells. If you want to achieve the best application of this method, you should choose GaAs, CdS, CdS, or CIGS thin-film solar cells, and the better choice is CdS or CIGS thin-film solar cells.

There are many advantages of this method, such as short processing time and simple operation (only physical methods are used). And, then without to the subsequent other methods at the same time, it can be separated to get a clean surface, rice with other impurities of solar panels. And, these solar panels are reusable.

As shown below, Figure 2 is a Structure diagram of toughened glass layer and metal layer before freezing treatment; Figure.3 is the Structure diagram of toughened glass layer and metal layer after freezing treatment;



Fig. 2 Structural diagram of tempered glass layer and metal layer before freezing treatment.

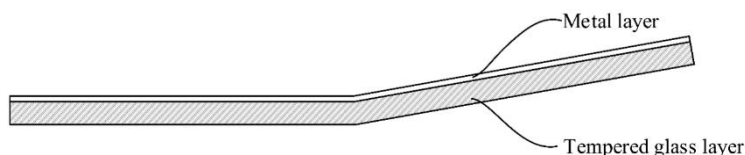


Fig. 3 structural diagram of tempered glass and metal layer after freezing treatment.

2.3.2 Solar panel recycling and processing device

Nowadays, the main problem that exists with solar cell recycling is the treatment and removal of EVA. As mentioned in B, the result of our solar cell is a layer of EVA plus the solar cell plate, plus a layer of EVA, so this structure makes it easy for EVA to stick to the solar cell plate. The existing treatment methods are generally chemical methods mostly.

There is another method to remove EVA by metal electric wire, but this method uses a high temperature, and EVA will melt and re-adhere under the high-temperature condition. This can make the subsequent operation more complicated and may damage the cell, causing more damage.

In order to solve the problem of EVA re-adhesion at high temperatures, there is a new solar recycling device that can be used. This is mainly achieved by a new device: the solar panel is placed in a slot on the carrier table and it has to be placed vertically. Then the solar panel is fixed with limit plates on both sides of the fixing mechanism. A drive device is provided in the two limit plates to control the position of the plates so that they can be exactly the length of the solar panel. The driving device can make the limit knives align with EVA vertically, and the limit knives are provided with driving devices that can make them move closer or farther away. There is a heating device on the limit knife. And there is a tractor device with suction cups on it, and there is a drive device on the suction cups, and this drive device can control the two suction cups to move closer or further away.

The usefulness of each part is: the carrier table is used to place the solar panel, the limit knife is used to limit the position of the solar panel so as to ensure the accuracy of the subsequent processing steps, the suction cups can play the role of supporting the solar panel, and the cutting knife can cut off the EVA stuck on the solar panel.

When cutting, the suction cups can well stop the glass panel and the back panel from re-fusing together, because two suction cups are sucked on the glass panel and the back panel, pulling the two panels apart as much as possible, so that the two panels in the EVA state will not be stuck together again after the high temperature melting.

Figure 4 shows the structure of the device.

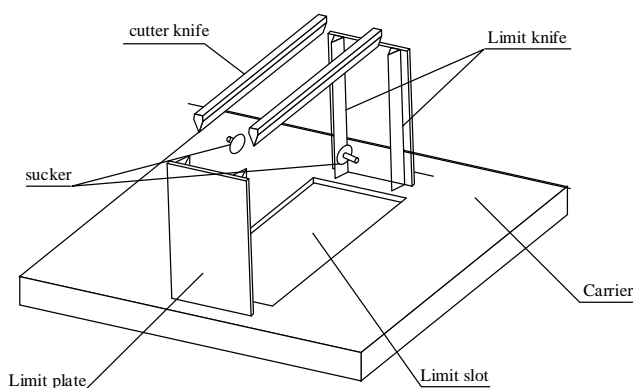


Fig. 4 Solar panel recycling and processing device

3. Chemical Method Application

After the physical removal process of the glass on the superficial surface of the PV modules, encapsulating material and inner materials like Ag/Al metallization, Anti-reflective coating, n-p junction can be removed by thermal or chemical, or mechanical delamination [1] (Deng et al., 2019) This section mainly focusses on the application of chemical methods.

[3] (Ewa) developed a chemical process in order to remove different layers, including EVA, electric connectors, anti-reflective coating, and n-p junction, of PV modules. The composition, temperature, and alkali concentration of the solvent for different layers were tested in order to formulate a universal composition. Figure 5 shows the flow of the chart for the general chemical process. From this chart, the separate analysis of effective removal of different layers is carried out in the following sections.

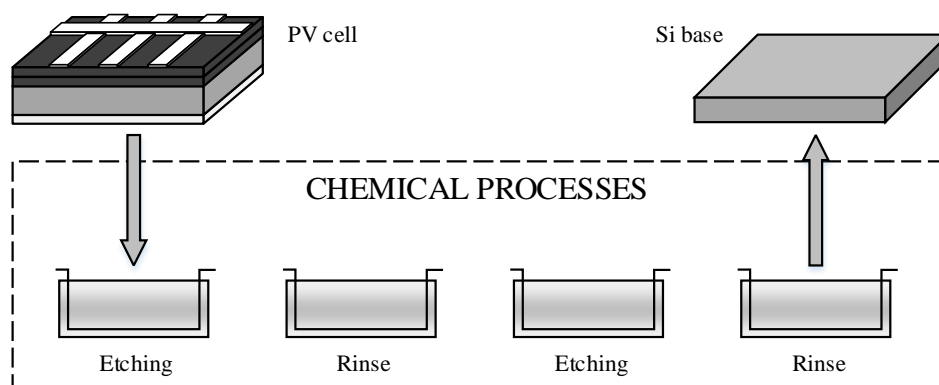


Fig. 5 General chemical process.

3.1. Removing of EVA.

One major problem in the chemical treatment removal of EVA is that the swelling of the EVA layer. In many research papers, this drawback has been reported. Some attempts to remove EVA were conducted using an organic solvent with the assist of ultrasonic process, Kim and Lee [7] conducted experiments at different solvent concentrations, temperatures, ultrasonic powers, and irradiation times. EVA can completely melt in 3M toluene at 70oC in the presence of 450W ultrasonic radiation. However, one problem was identified with this process: PV cells were damaged as a result of EVA swelling. This disadvantage was reported and has been taken care of by a Japanese scientist Doi. The safe removal of EVA was achieved by Doi et al. [16] Who applied multiple organic solvents to immerse PV cells to test the effectiveness and feasibility of various solvents. EVA layer is found to be dissolved most effectively in trichloroethylene solvent. The PV cells were treated by mechanical pressure, which was an essential method to prevent the swelling of the EVA layer when it was soaked in trichloroethylene for 10 days at 80 C. After the recycling process, the recovered Si cells could function validly.

P. M. Tembo et al [14] experimentally tested several solvents including acetone Hexane, Iso-propanol, Toluene, PTA, and acidified versions of them. It is concluded that the 5 M of pure Hexane solvents best detaches the EVA layer at 70oC after 24 hours of solving, with a detachment rate of 92.4%. Their experiment applied organic solvents to remove the EVA encapsulant layer and, therefore, demonstrated the predominant behavior and effectiveness of the Hexane amongst organic solvents.

3.2. Removal and recovery of the metal coating.

PV cells contain various precious metals including silver, copper, tin, and hazardous lead. (Park, J. et al.) [14]. The recovery of those metals always has been discussed and favored by many scientists around the globe.

Ewa et al. [3] compared the effectiveness of KOH and NaOH aqueous solutions regarding Al metal layer removal, which was found can be dissolved by using 30% KOH (aq) solvent at a temperature of 60-80 oC for 2-3 minutes.

Rong et al. [1] summarized and compared three fundamental approaches of Ag metal extraction by immersing the cell into a 6N HNO₃ leaching solution. These three methods involve electrolysis, metal replacement, and precipitation. The result of comparison gives a maximum Ag recovery rate of 89.7%, 87.4%, and 99.5% via precipitation, electrolysis, and zinc powder replacement, respectively. [17] Lee, C.-H. also reported the clear removal of Al using a NaOH solvent as Ewa did.

Jung, B. et al. [18] introduced a method for the removal of the Pb in the process of Ag metal recovery. After the HNO₃ solution is added to extract Ag, Pb is also dissolved in the acid leaching solution and takes the form of Pb²⁺. 5M Na (OH) is added to this leaching solution to get Pb (OH)₂ precipitate. Sodium sulfide (NaS) is also added after precipitation in order to react with the remaining Pb in the solution to form lead sulfide (PbS) which can be removed by filtration.

As reported by Jung, B. et al. [18], copper can be successfully recycled at an efficiency of 90%, by adding 20% LIX84-I (2- hydroxy-5-nonylacetophenone oxime) to extract Cu and 150g/LH₂SO₄ to form Cu²⁺, which will eventually be solidified with the method of electrowinning.

Figure 6 illustrates the general procedures for metal recovery and disposal manipulation

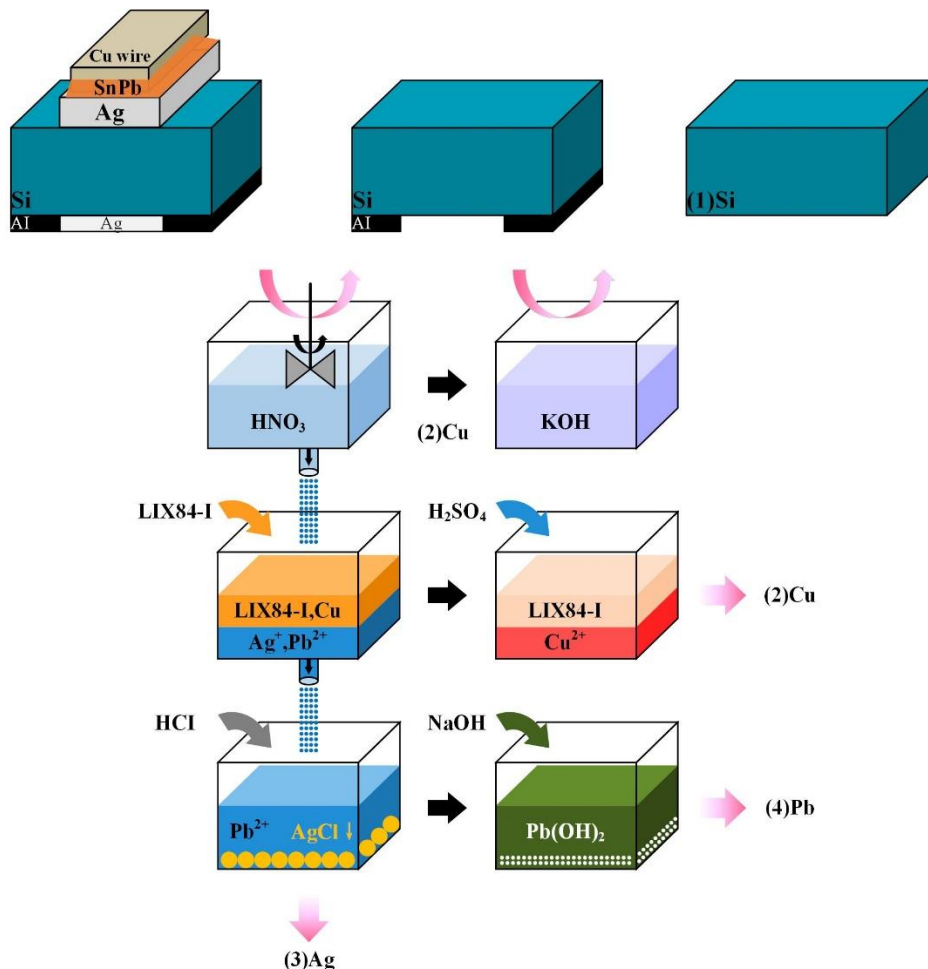


Fig. 6 the general procedures for metal recovery and disposal manipulation

3.3. Removing of AR coating and n-p junction.

By precisely adjusting the chemical treatment conditions, incorporating temperature, reaction time, component for solution mixtures, Ewa et al. [3] derived the universal mixture for removal of anti-reflective coating and n-p junction as: 250 ml HNO₃ (65%): 150 ml HF (40%): 150 ml CH₃COOH (99.5%) 3 ml Br₂ for removing AR coatings and n-p junctions (temperature 40 C, time 9 s).

3.4. Summary of chemical solvent treatment of end-of-live PV cells.

Chemical treatment of end-of-life PV cells are feasible and paramount to the understanding of the concept and principle of the PV cells recovery process. Chemical treatment is also found essential for metal recovery and harmful metal discarding. Three distinct and effective methods were introduced with respect to the recovery of valuable metal Ag which is highly desirable and Pb can be thoroughly removed with the addition of 5M Na(OH) and Sodium sulfide (NaS). However, many researchers have reported various disadvantages of chemical treatments. P. M. Tembo et al[14] experiments revealed an undesirable disadvantage of the application of chemical solvents: the harmful waste and emissions could be produced. Though, Doi et al.[16] successfully prevented the swelling effect from damaging silicon substrate by incorporating the method of mechanical pressure, it's reported that the quality of the recovered silicon would be affected. Therefore, the method of thermal treatment which generates more precise and environmentally responsible results of end-of-life solar cell recycling.

4. Thermal Treatment

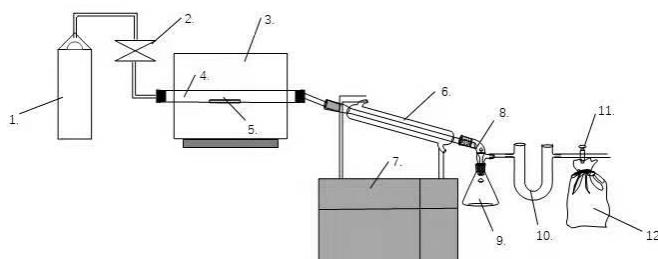


Fig. 7 Laboratory device for the heat treatment process

Use the device shown above to inject nitrogen and air at 600 °C.[22] Meaning of numbers: 1. Gas cylinder 2. Flowmeter 3. Tubular heating tube 4. Quartz tube 5. sample 6. Condensing tube 7. Thermostatic water-cooled condenser 8. Horn tube 9. Liquid collection bottle 10. Silica gel drying tube 11. tee valve 12. Gas sampling bag

In this process, EVA will be degraded. The degradation process of EVA is mainly divided into three steps.[23] The first step is deacetylation to form multi dilution, the second step is to form multidilution under inert conditions, and the third step is the formation and degradation of carbonization under aerobic conditions.

4.1. Deacetylation

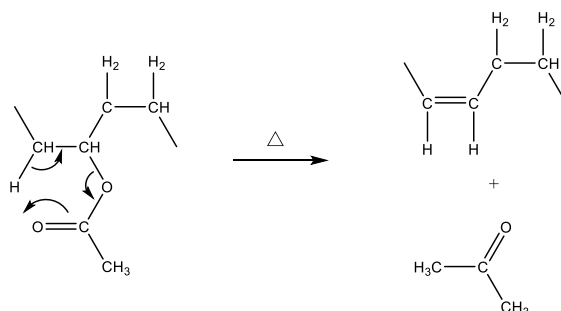


Fig. 8 Chemical reaction equation of deacetylation of EVA

Under the condition of the heating, the ester bond of the side group is fragile and easy to break. In EVA, the content of VA is 33%, and the average number of monomers of VAC in the main chain is between 1.14 and 1.21. Therefore, there is no autocatalytic deacetylation reaction[24]-[25]

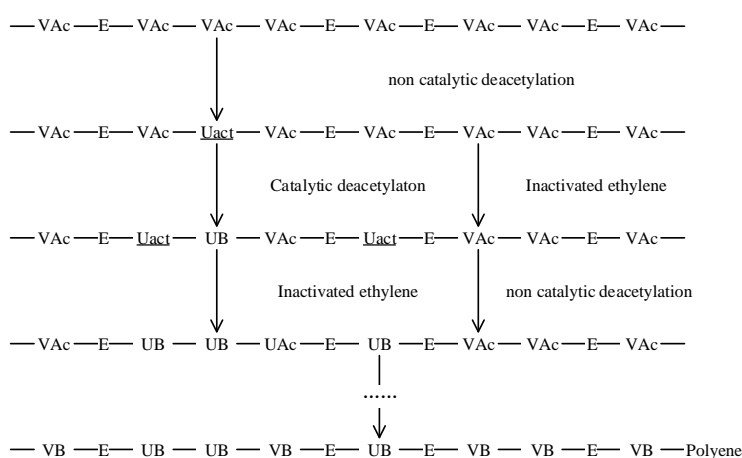


Fig. 9 Deacetylation schematic diagram

Among them, VAC is the vinyl acetate monomer, e is the ethylene monomer, UB represents the inactive double bond, and UACT represents the active double bond catalyzing the adjacent deacetylation of vinyl acetate.

4.2. Inert degradation of polyene

Conjugated polyenes were formed after deacetylation of EVA. This is formed by deacetylated VAC, unsaturated and inactive double bonds, and in ethylene monomers, which mainly depends on the initial composition of the polymer. Two parallel reaction steps can explain the bond-breaking process of conjugated polyenes in an inert atmosphere. The first is that the uncomfortable double bond is degraded into aromatic substances, and the second is that ethylene becomes aliphatic substances. The number of aromatic substances and fatty inflammatory substances depends on the initial content of vinyl acetate in the copolymer. Because the polyenes obtained after deacetylation of vinyl acetate are inactive double bonds, only aryl compounds can be formed.

4.3. Oxidative degradation of polyene

Under aerobic conditions, after deacetylation of vinyl acetate, the reaction will be divided into two steps: one is the formation stage of carbonization and the other is the oxidation stage. In the carbonization stage, aromatic solid residues and coke substances are formed due to inactive double bonds, while ethylene breaks to form fatty substances; In the oxidation stage, aromatic solid residues or coke substances are formed in the carbonization stage are oxidized to produce CO₂, etc. If the VA content in EVA is small, there is no carbonization step, and aliphatic substances and oxidation reactions will occur at the same time.

4.4. Summary of thermal treatment

EVA film is mainly decomposed in the heat treatment process with little impact on other components. The effect of aerobic heat treatment is similar to that of anaerobic heat treatment, and the products contain methane, ethane, ethylene, and carbon dioxide, but the content of each substance in the products is different. The glass panels and batteries obtained in this way are free from the risk of breakage, the cost is low and can be processed on a large scale. The disadvantages are the need for additional treatment of exhaust gas and exhaust liquid and the inability to recover monocrySTALLINE and polysilicon.

5. Comparison of these methods

EVA is the major concern for comparison. Inorganic acid can dissolve the crosslinked EVA in the middle of the cell and glass, but at the same time, a composition such as a line silver bar and aluminum paste on the cell are separated. The acid consumption of the method is tremendous, which will produce a large amount of waste liquid and gas.

The organic solvent solution Hexane-like method can dissolve EVA so that the glass and the battery can be separated. However, this method requires the panel to be pressurized, and the treatment cycle is long, the treatment of organic waste liquid is difficult, the glass expansion may break, and the recovery efficiency is low.

Heat treatment can effectively, with a shorter time compared to organic chemical treatment, treat the EVA in the panel and keep the glass intact. This method requires the treatment of exhaust gas and liquid products. Through the comprehensive comparison of the three process methods, the heat treatment process can be selected to remove the EVA layer in the panel. The crystalline silicon cells produced by manual disassembling and heat treatment can be chemically decomposed into recyclable materials such as silicon, silver, aluminum, and copper.

5.1. Comparing the thermal with the chemical treatment in an economic sense

When comparing chemical treatment with heat treatment, it is found that heat treatment is a more convenient method for the stratification process. It is possible to obtain better process results at a moderate energy cost. On the other hand, the effect of chemical treatment is low and the process lasts longer, which reduces the effect. Because of the variety and quantity of compounds, they are expensive. The additional cost of liquid waste treatment must also be considered in the CA [19].

6. Conclusion

The treatment and recycling of solar cells are gaining traction around the globe. Large cooperations around the globe are devoted to reducing carbon footprint. With that being said, the significance of developing a comprehensive method for solar cells recycling is obvious. This paper mainly introduced and summarized the previous research, and compared their methods. Three basic concepts related to the recycling of end-of-life solar cells were elaborated in detail. In conclusion, Compared with chemical methods, heat treatment methods are both economical and environmentally friendly and can be manipulated on a massive scale. Chemical methods can make the metal cycling process, where most chemical treatments happen, more efficient under different conditions. This article makes a simple summary of the previous research and current situation, which lays a foundation for future research in this direction. It aims to provide help for recycling solar panel systems in the future and an easy understanding of previous research progress and status for later researchers.

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