Research Status of Recovery of Tellurium from Cadmium Telluride Photovoltaic Modules

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Abstract. Cadmium telluride is a direct band gap semiconductor with high optical absorption coefficient and excellent photoelectric conversion efficiency. Therefore, the proportion of cadmium telluride thin film solar cells in photovoltaic emerging technologies is increasing. The shortage of metal tellurium is the main factor restricting the development of cadmium telluride thin film photovoltaic cell technology. Therefore, it is necessary to recover tellurium from decommissioned cadmium telluride photovoltaic modules. In this paper, the treatment status of cadmium telluride photovoltaic decommissioning components is studied, and the characteristics of various recycling methods are analyzed and compared. Based on recent research progress and important findings, the challenges and future prospects of commercialization are forecasted.

1. Introduction
As a rare metal, tellurium has a very low content in the earth's crust. The estimated average crust abundance is 3 ppb and the distribution is extremely dispersed. The main use of tellurium is in the development of cadmium telluride thin films in photovoltaic solar cells, which are considered emerging applications, especially in developing economies in China and India. The European Photovoltaic Industry Association (EPIA) predicts that by 2040, the contribution of photovoltaic energy to the global electricity supply may be as high as 12.6% [1-4].

The cost of photovoltaic production of cadmium telluride film is very low, and the current component cost can be around $ 0.64 /W [5]. Therefore, in emerging technologies, the proportion of cadmium telluride thin film photovoltaic continues to increase. However, there are two main problems in cadmium telluride photovoltaic technology: on the one hand, the impact of cadmium pollution; on the other hand, tellurium may be in short supply. The estimated life of cadmium telluride thin film modules is about 30 years. When they reach the end of their service life, the brackets used on the panels may damage the environment if they are not recycled or handled improperly. The heavy metals in them are toxic and may cause cancer or teratogenicity [6-8].
Therefore, the recovery of tellurium from tellurium-containing waste has become an indispensable link in the sustainable development of the tellurium industry, and its significance to resources, environment and economy is increasingly important. Production-scale recycling technologies have been fully developed in the 1990s. The recovery and recycling of the scrapped battery components not only avoids the environmental hazards of heavy metals such as cadmium and lead, but also reduces the environmental liability cost of the enterprise. The reuse of scarce metals such as Te, In, Ga reduces the supply pressure of these metals. The cross section of the cadmium telluride thin film solar cell is schematically shown in Fig. 1.

2. Recovery of Tellurium Resources from CdTe Photovoltaic Modules

2.1. Hydrometallurgical Recovery Process for CdTe Photovoltaic Modules.

An evaluation of the Photovoltaic recycling strategy shows that there is currently only one process on the market that operates on an industrial scale. The CdTe thin film module is recovered by a combination of mechanical and chemical processes. This technology was developed by First Solar Corporation [10-12]. The process of other technologies is evolving, mainly on a laboratory scale.

The work of First Solar and Brookhaven National Laboratory (BNL) in the United States is representative. The usual method is to separate the semiconductor film by acid dissolution and then precipitate in an alkaline environment. Because most metals form precipitates in an alkaline environment. Fig. 2 shows the technical process diagram of First Solar in 1997. The assembly is first disassembled and a lead or copper common conductor is recovered, and then the two layers of the encapsulated semiconductor film are pulverized by a hammer crusher. CdTe and CdS films, glass cullet, EVA, etc. are placed together in a mixed solution of sulfuric acid and hydrogen peroxide, and the CdTe and CdS semiconductors are dissolved in the acid together with the back electrode metal.

![Diagram of CdTe photovoltaic module recycling](image_url)

**Figure 2.** Flow chart of recycling of CdTe photovoltaic module via acid-dissolution/precipitation method [13].
Its main reaction formula is as follows:

\[
CdTe(s)+3H_2SO_4+3H_2O_2=CdSO_4(aq)+Te(SO_4)_2(aq)+6H_2O \tag{1}
\]

\[
CdS(s)+H_2SO_4+H_2O_2=CdSO_4(aq)+S(s)+2H_2O \tag{2}
\]

The filtered solution was added to Na\(_2\)CO\(_3\)(s) to obtain sediment containing Cd and Te by precipitation:

\[
CdSO_4(aq)+Na_2CO_3(s)=CdCO_3(s)+Na_2SO_4(aq) \tag{3}
\]

\[
Te(SO_4)_2(aq)+2Na_2CO_3(s)=TeO_2(s)+2Na_2SO_4(aq)+CO_2(g) \tag{4}
\]

Precipitated CdCO\(_3\), mixed with TeO\(_2\) sediment, can recover economically valuable products in two ways. The first method is to reduce CdCO\(_3\) and TeO\(_2\) with hydrogen to obtain a mixture of Cd and Te, and then heat the mixture to synthesize cadmium telluride. Its reaction formula is as follows. The disadvantage of this method is that various metal impurities cannot be removed, and the recovered CdTe cannot be directly used for battery assembly production.

\[
CdCO_3+TeO_2+3H_2=Cd_0+Te_0+CO_2+3H_2O \tag{5}
\]

\[
Cd_0+Te_0=CdTe(N_2,500^\circ C) \tag{6}
\]

Another way is to dissolve the mixed sediment in a NaOH or KOH solution. This will redissolve the tellurium and separate it from other metals, including cadmium. High purity tellurium ingots can be obtained from the obtained K\(_2\)TeO\(_3\) (dissolved) electrolysis:

\[
TeO_2+2KOH = K_2TeO_3(aq)+H_2O \tag{7}
\]

\[
K_2TeO_3(aq)+H_2O = Te(s)+2KOH+O_2 \tag{8}
\]

This recycling process has a recovery rate of over 67%. First Solar's improved method can produce high purity (99.7%) elemental Te and achieve a recovery rate of 80% or higher. More than 99% of the cadmium is recovered as a precipitate. After precipitating the ions in the solution with a NaOH solvent, a small amount of cadmium ions are still dissolved in the solution. This portion of the cadmium remaining in the solution can be completely recovered by ion exchange treatment to a residual concentration of 0.1 ppb. However, considering the cost-effectiveness of large-scale treatment, the Cd residue obtained by using low-cost and high-efficiency ion exchange treatment waste liquid can generally be maintained at a level below 0.1 ppm, meeting the requirement of the highest emission standard of industrial wastewater in China <0.1 mg/L[14]. Treatment of 2 MWp components directly produced approximately 333t of wastewater containing 0.1 ppm of Cd, 346.9 mg Cd GW\(^{-1}\) h\(^{-1}\). We assume that even if less than 1 ton of wastewater is discharged using less stringent water recycling technology, the cadmium emission rate is only 0.005 mg Cd m\(^2\), which is 1.04 mg Cd GW\(^{-1}\) h\(^{-1}\) [15]. Therefore, the recycling process is highly dependent on the water recycling technology.

Based on the First solar company recycling process, US researchers have developed a series of leaching methods: (1) Leaching with solutions of ferric chloride/hydrochloric acid [16]; (2) oxygen-pressure leaching using sulfuric acid; (3) nitric-acid leaching [17-19]; and (4) leaching with sulfuric acid/hydrogen peroxide [20-22]. The first method failed to separate Cd from Fe (tellurium was not leached into solution). Oxygen-pressure leaching separated them to a significant degree, but the residues contained 4%~7% of cadmium mixed with tellurium and sulfur. Further, due to the high pressure and temperature needed, this process is capital intensive, and thus, unattractive for low-cost recycling. Nitric-acid leaching recovered 96% of the tellurium, leaving Cd in solution. However, the subsequent
recovery of Cd by electrolysis was based on artificially high Cd concentrations (e.g. 10000 ppm instead of ~1000 ppm obtained from leaching). Also, leaching with highly concentrated nitric acid requires elevated temperatures between 60°C and 80°C that inevitably create nitric-acid fumes.

Wolfgang Berger proposed mechanical flotation to treat cadmium telluride decommissioning components. The method mainly includes semiconductor layer enrichment, flotation, and separation. The recycling process is shown in Fig. 3. The semiconductor layer was enriched by vacuum blasting and mechanical stirring, and the flotation agent was potassium amyl xanthate (KAX). The enriched semiconductor layer is separated and purified by H2SO4 and H2O2. The experimental results show that the optimum process time is 30 min at 900 rpm Container speed and 25 rpm agitator speed. The material is further shredded due to the powerful forces that exist during the mixing process. After the abrasion treatment, the ratio of less than 5 mm is about 22%. Material, and plastic foil was rinsed and sieved in fractions of >500um, 500–150um, and <150um. The output material of the attrition processes contained 2260 mg CdTe/kg (1201 mg Te/kg). An advantage of the wet mechanical processing in comparison to the conventional procedure consists in the fact that during the attrition no chemicals are used, and during the flotation process only small amounts of flotation reagents are necessary. However, the obtained tellurium product has impurities and requires secondary processing.

Vasilis Fthenakis and Paul Duby et al. used ion exchange to recover tellurium [24-28]. First, leaching tellurium and cadmium with sulfuric acid and hydrogen peroxide solution; The cadmium in the adsorption solution is adsorbed by a cation exchange resin, and tellurium remains in the solution. The tellurium-containing solution and the cadmium-rich cation exchange resin are obtained to realize the separation of tellurium and cadmium. The experiment found that low sulfuric acid strength is the most effective way to remove cadmium in solution; the cation exchange rate of cadmium is faster in the first 20 min and reaches equilibrium within 2 h. The tellurium and cadmium were separated by an ion exchange resin method, and the removal rate of cadmium was 99.99% or more. The cation exchange column was eluted with a mixed sulfuric acid/sodium sulfate solution to form a cadmium-rich solution, and cadmium was recovered by electrowinning. In order to recover tellurium from solution, we have studied several precipitation methods. To recover the tellurium from the leach solution, we explored several precipitation methods. They included (1) precipitating tellurium as tellurium dioxide using sodium hydroxide or sodium carbonate, (2) precipitating tellurium as tellurium sulfide using sodium sulfide, (3) reducing/precipitating tellurium as metallic tellurium using zinc or iron metal, and, (4) reducing/precipitating tellurium as a metallic salt using sodium metabisulfite. In all these processes, the recovered tellurium was in the form of fine powder. Elemental analysis of the aqueous solutions using an ICP spectrometer showed that the removal efficiencies ranged from 60% to 100% (Table. 1).

Figure 3. Flow chart of recycling of CdTe photovoltaic module via flotation method [23].
Table 1. Recovery of tellurium by reactive precipitation

|          | Na₂S₂O₅ | Zn | Fe | Na₂CO₃ | Na₂S |
|----------|---------|----|----|--------|------|
| Removal efficiencies (%) | 99.6  | 100 | 99.4 | 60    | 99.5 |

Zhang and Sun proposed a method of electrodeposition extraction to recover tellurium. The electrolyte solution is divided into a first electrolyte solution and a second electrolyte solution, and is separated by an anion exchange membrane. The cadmium telluride component is contacted with the first electrolyte solution. A voltage difference is applied between the electrodes to remove the tellurium from the cadmium telluride-containing assembly and deposit it on the opposite electrode. The viscous material between the cadmium telluride film and the glass encapsulation layer is melted by heating before deposition, and the glass layer is removed to expose the semiconductor layer. In the electrodeposition process, tellurium and cadmium are present in the first electrolyte solution in the form of Cd²⁺ and HTeO₃⁻. Because of the selectivity of the anion exchange membrane, Cd²⁺ remains in the first electrolyte and HTeO₃⁻ enters the second electrolyte.

Caelen D. Anderson [29] studied the recovery of cadmium telluride by ammonia precipitation. The method mainly uses H₂SO₄ and H₂O₂ leaching, and ammonia water is added to precipitate TeO₂. Leaching results illustrated that it is possible to produce a 98% pure TeO₂ product while recovering 74% of the tellurium. The most favorable conditions for leaching were determined: pH = 1, pulp density of 40 g/L, hydrogen peroxide: tellurium ratio of 6:1, temperature of 50°C, agitation rate of 330 rpm and a residence time of one hour. Precipitation results demonstrated that 98% of the aqueous tellurium species present can be recovered from solution using ammoniacal precipitation. The most favorable conditions for both TeO₂ grade and tellurium recovery were determined. To produce a low-grade (46%) TeO₂ precipitate, a precipitation pH of 4.0 and temperature of 25°C are required. To recover 98% of the aqueous tellurium present, a precipitation pH of 5.6 and a temperature of 25°C are needed.

Sulfuric acid can be used as an electrolyte for electrochemical cells to remove cadmium telluride from cadmium telluride waste batteries. By adjusting the voltage used, the stirring speed, and the concentration of cadmium in the electrolyte, a cadmium telluride film having a ratio of cadmium to tellurium of 1:1 to 1:6 can be regained on the electrode. However, simulation experiments have shown that it is difficult to obtain cadmium telluride which can be directly used to produce cadmium telluride films. In addition, when the simulation experiment attempts to recover tellurium alone, pure tellurium can only be obtained at a very low pH, which may be because tellurium and cadmium are present in the form of ions in the sulfuric acid electrolyte. They all deposit on the electrodes under similar conditions and contaminate each other. In particular, the concentration of ions in the electrolyte changes more significantly as the deposition process changes. Industrial applications are often difficult to maintain the electrolyte pH in a narrow range, so it is difficult to obtain reliable quality defects.

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**Figure 4.** Flow chart of recycling of CdTe photovoltaic module via vulcanization-vacuum distillation method.
2.2. Pyrometallurgical Recovery Process for CdTe Photovoltaic Modules.

The hydrometallurgical recycling technology for the treatment of cadmium telluride waste is complicated and cumbersome, consumes a large amount of chemical reagents, and generates a large amount of waste liquid. Liu and Zhang [30] et al. proposed a vulcanization-vacuum distillation method to treat cadmium telluride waste (Fig. 4). Compared with the wet method, the vacuum distillation process has the advantages of short process flow, good economic benefit and small environmental pollution. This method is widely used in the fields of metallurgy and secondary resource recovery [31-40].

First, the cadmium telluride waste is broken into powder; the obtained cadmium telluride waste powder is uniformly mixed with sulfur, and further densified according to a stoichiometric ratio of cadmium telluride and sulfur at a ratio of 1:1.1 and under a pressure of 8 MPa; The small cylinder is sealed and sealed with a protective gas at a temperature of 400°C for 30 min to obtain a vulcanized product; the obtained vulcanized product is broken into small pieces, and then vacuum distilled (600°C, 15 Pa) to obtain Te. Fig. 5 is a graphical representation of the XRD of the sulfurized product and condensate.

![Figure 5](image_url)

Figure 5. (a) X-ray diffraction patterns of sulfurized products in 400°C; (b) X-ray diffraction patterns of vacuum distillation condensate products in 600°C.

The vulcanization-vacuum distillation process was used to achieve the separation of cadmium and tellurium. The feasibility of this process was verified on the laboratory scale and semi-technical scale, thus providing theoretical guidance for the recycling of cadmium telluride photovoltaic modules. Smelting of cadmium telluride and sulfur can produce Te and CdS. With a temperature of 550 °C - 800°C, a vacuum distillation time of 30 min and a residual gas pressure of 15 Pa, Te was enriched to 99.93%, and the recovery rate was 94.14%. The contents of Cd and S in the Te were 0.034% and 0.037%, respectively. The purity of cadmium sulfide in the vacuum distillation crucible can reach 99.5%. Meanwhile, the CdS collected by vulcanization-vacuum distillation can be used as a raw material for semiconductor production after purification. Compared with the existing methods, the vulcanization-vacuum distillation approach features no waste gas or wastewater, high resource utilization rate, short refining process, good working conditions, and meets the requirements of energy saving and clean production.

3. Main Problems and Suggestion

By analyzing the current status of recycling and recycling of used photovoltaic modules, it is known that the following problems still exist in this aspect of research:

(1) Technical level: The process of hydrometallurgical recycling technology for treating cadmium telluride waste is complicated and cumbersome, and the consumption of chemical reagents is large, and a large amount of waste liquid is generated at the same time. At the same time, the waste liquid and waste gas generated during the production process need to be further processed to avoid secondary
pollution to the environment. It is recommended to set up a professional research institution to conduct in-depth research.

(2) Economic level: The economic benefits of recycling used PV modules are low, and the market has little driving force for recycling PV modules. Although the cost-benefit of the cadmium telluride thin-film photovoltaic module material cycle is relatively high, its production is small, and its recycling demand is correspondingly small.

The value of material recovery has certain economic benefits, but the process of centralized collection of waste materials will also generate certain transportation costs. It is suggested that the development of centralized power plants has advantages and is convenient for centralized on-site treatment. After the preliminary crushing and sorting of the decommissioned components, the waste glass can be recycled as a building material, and the weight of the recovered semiconductor material accounts for only a small part of the weight of the component, which greatly saves the transportation cost.

The battery pack is integrated with the building or other electrical equipment. Even if it is designed to be easy to separate and disassemble, it is still necessary to recycle the entire assembly before centralized processing. In this process, the difficulty of recycling is greatly increased. Therefore, it is recommended that the government set up a special photovoltaic module recycling agency to recycle and reuse used PV modules.

(3) Legal perspective: It is recommended that the state can pay attention to research in this area, issue relevant standards, and regulate the handling behavior of waste photovoltaic modules on the market.

Research in the global photovoltaic industry has never stopped, technology is changing with each passing day, and the efficiency of battery cells is constantly improving. Of course, the technology and business model of recycling are also on the agenda. It is believed that the technology of photovoltaic module recycling will be rapidly upgraded and developed under the policy and profit, and the cost of recycling will inevitably decrease. The most important thing is that the contribution of component recycling to the environment is more significant and far-reaching.

4. Conclusion

(1) Cadmium telluride thin film batteries are the main consumption products of tellurium, and the recycling of decommissioned photovoltaic modules is an indispensable component of renewable tellurium resources.

(2) At present, the recovery of tellurium metallurgy is not only costly, but also produces a large amount of waste liquid pollutants. Most of the obtained tellurium does not meet industrial requirements and needs further purification, thereby increasing production costs.

(3) The vulcanization-vacuum distillation method developed by Kunming University of Science and Technology, which recycles and treats cadmium telluride photovoltaic modules with short cycle, no pollution, low raw material cost and high recovery rate. This method has broad development space and potential application prospects.

(4) Today, with the rapid development of the photovoltaic industry, the demand for component recycling and reuse is imminent. There are not many laws and standards for cadmium telluride photovoltaic modules. Therefore, in the context of imperfect recycling mechanisms, organizations need to look forward to the economics and technology of component recycling from a social and environmental perspective with a forward-looking perspective. At the same time, the lack of technology in this area also means a huge potential market. It is believed that more enterprises and research institutions will invest in this area under the stimulation of this part of the market and the progress of recycling technology.

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