RESEARCH PROGRESS ON STRONTIUM TITANATE PHOTOELECTROCHEMICAL CATHODIC PROTECTION MATERIALS

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Abstract: Photoelectrochemical cathodic protection is a green, pollution-free metal cathodic protection method. In recent years, it has been favored by more and more scientific researchers in the field of corrosion protection. SrTiO3 has a relatively negative conduction band potential and has the advantages of good stability and non-toxicity. It is an excellent candidate material for photoelectrochemical cathodic protection. Firstly, the principle of photoelectrochemical cathodic protection is introduced. Secondly, the advantages and characteristics of SrTiO3 in this aspect are explained. Then the relevant research progress is reviewed around the preparation and modification methods of SrTiO3. Finally, the research direction and application of photoelectrochemical cathodic protection are summarized and prospected. **Keywords:** Corrosion protection; Photoelectrochemical cathode; Strontium titanate; Modification research

1 PHOTOELECTROCHEMICAL CATHODIC PROTECTION TECHNOLOGY

Developing the marine economy is one of the important measures to achieve the strategic goal of becoming a maritime power in my country. Factors such as the marine environment being rich in salt, oxygen, and strong sunlight will cause irreversible corrosion hazards to marine vessels and large metal facilities, directly or indirectly causing economic damage. The losses will greatly increase the capital investment in our country's marine engineering. At the same time, corrosion does not only exist in the ocean. Corrosion exists everywhere in daily life. Therefore, corrosion protection research is an issue that cannot be ignored and is progressive from small to large. research process. The most common corrosion protection method is electrochemical cathodic protection. Cathodic protection using electrochemistry is divided into two methods: impressed current and sacrificial anode [1, 2]: Broadly speaking, the essence of metal corrosion is the oxidation process of losing electrons. The current cathodic protection method is directly connected to the protected metal through the negative electrode of the external power supply. What really slows down or prevents metal corrosion is the electrons provided by the DC power supply; the sacrificial anode cathodic protection method uses the anode (the metal is more active) to protect the metal. The protected metal material provides a more negative electron (generated by anodization), which is transferred to the metal material to inhibit its oxidation reaction. Both have their own advantages and disadvantages. When combined with each other, they can be well applied to metal corrosion protection. However, they cannot avoid problems such as high pollution and high energy consumption. In the context of increasingly severe energy and environmental problems, the development of green and environmentally friendly metal protection technology is a top priority. With the continuous deepening of research on semiconductor materials, photoelectrochemical cathodic protection technology based on solar energy has emerged. In this technology, the energy source is taken from green and clean solar energy. The semiconductor plays the role of "catalyst", and the photogenerated electrons generated by stimulation are transferred to On metal materials, it replaces the DC power supply and the electrons provided by the anode in electrochemical cathodic protection.

Yuan et al. [3] first proposed the concept of photoelectrochemical cathodic protection, which combines sunlight with electrochemical corrosion protection through semiconductor materials, taking an important step towards green and environmentally friendly anti-corrosion technology. This technology relies on solar energy to achieve the goal of photoelectrochemical cathodic protection. It is an environmentally friendly, energy-saving, and environmentally friendly technology.

1.1 Theoretical Basis of Photoelectrochemical Cathodic Protection

In photoelectrochemical cathodic protection technology, the electrons provided to the protected metal materials come from photo-generated electrons in semiconductors, and the energy consumed comes from solar energy. The specific principle is: the semiconductor material is coated on the metal or the semiconductor is coupled to the metal. When photons with energy greater than the semiconductor bandgap width Eg illuminate the semiconductor material [4], the electrons in the semiconductor valence band transition to the conduction band and become photogenerated electrons. At the same time, photogenerated holes are left in the valence band. Theoretically, as long as the conduction band potential of the semiconductor material is more negative than the self-corrosion potential of the metal material, the photogenerated electrons can be transferred to the surface of the metal material to generate charge accumulation and put

it in an electron-rich state to protect the metal material. The macroscopic manifestation is a negative shift in the metal potential. In addition, photogenerated holes will migrate to the surface of the semiconductor material, react with electron donors in the environmental medium, and be consumed (such as water oxidation reaction). The above mechanism is shown in Figure 1.

1.2 Potential of SrTiO3 Photoelectrochemical Cathodic Protection

First of all, SrTiO3 is an n-type semiconductor material, which is perovskite type and has a stable structure. It is widely used in the fields of photocatalytic degradation of pollutants [5], photocatalytic hydrogen production [6-8], and nitrogen fixation in agricultural production. The research scope is extremely wide and can be referred to each other, especially when it comes to hydrogen production. Performance research, relevant personnel engaged in photoelectrochemical cathodic protection research can be greatly inspired by it. Secondly, the conduction band potential of SrTiO3 is -0. 49 V (vs. NHE), which is 0. 2 V less than the most studied conduction band potential of TiO2. This means that the photogenerated electrons generated by SrTiO3 have stronger reduction capabilities and are easier to migrate to the protected metal to achieve protection. the goal of. Finally, the band gap Eg of SrTiO3 is 3. 2 eV. It is a wide band gap semiconductor material and can only absorb 5% of the ultraviolet light of sunlight. This is obviously not conducive to photoelectrochemical cathode protection, but for SrTiO3, it is a means to solve this defect. There are many, and metal or non-metal doping can be done through doping and compounding to give it visible light response characteristics and expand the scope of SrTiO3's utilization of sunlight. Combining the above characteristics, SrTiO3 has broad research progress of SrTiO3 in recent years from the perspectives of preparation and modification.

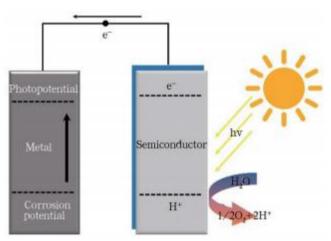


Fig. 1 Principle of photoelectrochemical cathodic protection

2 PREPARATION METHOD OF SRTIO3

The preparation methods of SrTiO3 mainly include sol-gel method, hydrothermal method, co-precipitation method, etc. There are very few preparation methods that directly apply SrTiO3 to photoelectrochemical cathodic protection, but the hydrogen production, degradation and other properties of SrTiO3 can reflect the photoelectric properties of SrTiO3. Chemical cathodic protection performance.

2.1 Sol-gel Method

The sol-gel method is widely used in the surface coating process of metal materials. Its advantages include simple process, accurate stoichiometric ratio, uniform film formation, and low film formation temperature. In 2011, there was clear domestic literature that classified SrTiO3 Introduced into the research of photoelectrochemical cathodic protection, that is, Bu Yuyu et al. [9] A nanometer SrTiO3 thin film photoelectrode was prepared by the sol-gel method, and the effect of cetyltrimethylammonium bromide (CTAB) surfactant on the photoelectrochemical cathodic protection performance of the SrTiO3 thin film was studied. The results showed that: at 300 W simulation Under sunlight irradiation, in 0. 5 mol/L NaCl solution, the SrTiO3 thin film photoelectrode synthesized with CTAB assistance can negatively shift the potential of the 304 stainless steel electrode by nearly 400 mV (vs. Ag/AgCl), which is more negative than the SrTiO3 photoelectrode without CTAB assistance under the same conditions. Negative shift of about 50 mV. This result shows that SrTiO3 has the potential ability to protect 304 stainless steel, and also provides ideas for further research by subsequent researchers.

2.2 Hydrothermal Method

The hydrothermal method artificially creates a high-temperature and high-pressure environment so that the reaction raw materials can fully react in the high-pressure reactor. The advantage of the hydrothermal method is that the prepared materials have high purity and high crystallinity. In the study of using hydrothermal method to prepare SrTiO3, researchers have continuously optimized the particle size of SrTiO3. Shao Yi [10] used Sr(NO3)2 and C16H36TiO4 as raw materials, and prepared SrTiO3 nanopowder with uniform particle size and a particle size of 100 nm by hydrothermal reaction at 175°C for 60 minutes. Kuang et al. [11] used a hydrothermal method to prepare SrTiO3 with a 3D hierarchical structure at 150°C. It was made of 60-80 nm highly oriented nanotubes with a specific surface area of 20. 83 m2·g-1. In the hydrogen evolution test Exhibited strong photocatalytic activity. The SrTiO3 nanopowder prepared by Zhang Na[12] using a one-step hydrothermal method has a regular morphology and a narrow particle size distribution of about 100 nm. A suitable particle size of SrTiO3 to a certain extent, which will help improve the separation efficiency of SrTiO3 photogenerated electron-hole pairs, the number of photogenerated electrons that migrate to the photocathode, and The speed is increased, thereby improving the photoelectrochemical cathodic protection capability of SrTiO3.

2.3 Co-precipitation Method

The co-precipitation method is to add a precipitant to the solution to precipitate the active ingredients in the solution. The precipitate is dried and calcined to obtain the required materials. Its advantages are that the preparation process is simple and the synthesis cycle is short. Lu Hongxia[13] selected Na2CO3 and NaOH as precipitants, SrCl2 and TiCl4 as reaction raw materials, and prepared spherical SrTiO3 with uniform size by co-precipitation method. The particle size is less than 1 μ m. This result is similar to the particle size of SrTiO3 nanomaterials prepared by the above hydrothermal method. It also shows that the same effect can be achieved through different processes. In addition, the co-precipitation method in a weak alkali system, with As the alkalinity of the system increases, the critical roasting temperature required to form strontium titanate crystal materials will increase. Li Wenxiu [14] applied the co-precipitation hard template method and concluded that when the optimal pH value is 8, the critical crystallization temperature of strontium titanate is only 420 °C, and has a large specific surface area of 160 m2·g-1. Overall, the co-precipitation method can still prepare SrTiO3 nanomaterials with a particle size of 100 nm, ensuring the same performance as the hydrothermal method to prepare SrTiO3 nanomaterials. That is, this method can also be applied in photoelectrochemical cathodic protection research.

2.4 Other Preparation Methods

In addition to the common preparation methods mentioned above, SrTiO3 also includes some other methods. In 2002, SrTiO3 was first published in international literature.

In current photoelectrochemical cathodic protection research, Tatsuma [15] and others used spray pyrolysis to prepare SrTiO3 films on ITO glass and applied them to the corrosion protection of carbon steel. The test results show that: SrTiO3 films at 10 mW /cm2 Under UV light irradiation, the corrosion potential of carbon steel in 3 wt% NaCl (pH=5, room temperature) solution is -770 mV (vs. Ag/AgCl). It is more negative than the self-corrosion potential of carbon steel (-500 mV), and the potential has been negatively shifted by 270 mV, indicating that the SrTiO3 film can protect carbon steel. This result has inspired subsequent research by researchers, which can extend the protection objects to stainless steel, copper and other materials. In 2014, Li et al. [16] used molecular beam epitaxy to grow an SrTiO3 film epitaxial layer on Si (001) and prepared a photoelectrode can reach 35 mA/cm2, the open circuit potential is 450 mV (vs. Ag/AgCl), and the performance has not declined after 35 hours of continuous operation. This result is that SrTiO3 in Si (001) provides guidance for growth and can be applied to photoelectrochemical cathodic protection research.

3 MODIFICATION METHODS OF SRTIO3 IN PHOTOELECTROCHEMICAL RESEARCH

Currently, in the research on photoelectrochemical cathodic protection, SrTiO3 needs to solve two main problems: it has a large bandgap and can only absorb 5% of the ultraviolet light of sunlight; it has a high recombination rate of photogenerated electrons and holes and a low utilization rate of photons. Solving these two problems can better promote SrTiO3 to photoelectrochemical cathode protection research. In addition, there are very few documents on SrTiO3 in photoelectrochemical cathode protection. The following description will start from the perspective of improving its own performance, and then analyze and extend it to photoelectrochemical cathodes. Conservation research in progress. The main modification methods include metal doping, non-metal doping, metal/non-metal co-doping and composite methods.

3.1 Doping Modification

3.1.1 Single element doping

In 2004, Wang[17] used cyclohexamethylenetetramine as the nitrogen source to study N-doped SrTiO3. The research results showed that N-doping can significantly improve the response ability of SrTiO3 to visible light. Under visible light irradiation, N-doping The removal rate of NO by doped SrTiO3 is 3. 5 times higher than that of pure SrTiO3. The photocatalytic activity of SrTiO3 is improved after doping with N. This result makes scientific researchers realize the importance of N doping SrTiO3, and then inspires subsequent scientific researchers to study N Element doping also introduces other elements, and the research progress of SrTiO3 is greatly accelerated.

In 2006, Wang[18] systematically studied Cr-doped SrTiO3 The influence of different positions in the structure on its photocatalytic activity finally confirmed that Cr ions occupy the Sr2+ position and have the highest photocatalytic activity. After that, Katya[19] studied the effect of Rh doping amount on the photoelectrochemical properties of SrTiO3 and found that when the Rh doping amount was 7%, SrTiO3 had the best effect at -0. 7 V (vs. Ag/AgCl) bias. Under the incident light wavelength of 420 nm, the photoelectric conversion efficiency is 0. 18%. Cr and Rh elements appear frequently in the research of SrTiO3, and many subsequent studies of SrTiO3 have introduced these two elements.

In 2014, Kawasaki[20] used pulsed laser deposition to prepare Ir-doped SrTiO3 films. By controlling the process conditions, Ir4+ and Ir3+-doped strontium titanate films were obtained respectively. The test results found that the SrTiO3 film formed by Ir4+ doping is active in water oxidation, while Ir3+ is inert. The Ir4+-doped SrTiO3 film exhibits visible light response at a wavelength of 600 nm. This result shows that the valence state of Ir needs to be controlled when Ir-doped SrTiO3 is used for photoelectrochemical cathodic protection.

In 2019, Kosaku et al. [21] doped Na element into SrTiO3. The results showed that the doping of Na element will introduce a new intermediate band gap, and the photogenerated electrons will fall into it. Not only the lifetime of these electrons is increased, but the reactivity still exists. , this result provides new ideas for doping.

3.1.2 Double element doping

In 2002, Kato[22] added Sb and Cr into SrTiO3 at the same time. SrTiO3 showed strong absorption in the visible light region, and the band gap was reduced to 2. 4 eV. In the same year, Ryan[23] and others used the molecular beam epitaxy method to simultaneously incorporate La and Cr into SrTiO3, and produced Sr1-xLaxTi1-xCrxO3 thin film material. After co-doping with La and Cr, the bandgap width of SrTiO3 decreased by 0. 9 eV. It is 2. 3 eV, which is reduced by 0. 1 eV based on the Kato research group. The visible light utilization rate of SrTiO3 has been improved again to a certain extent. In 2012, Yu et al. [24] doped N and Cr elements into SrTiO3. According to theoretical calculations, the defect formation energy of Cr/N co-doped SrTiO3 is much smaller than that of N-doped SrTiO3, indicating that Cr doping It can promote N doping into the O site of SrTiO3. Compared with Cr-doped SrTiO3, Cr/N co-doped SrTiO3 has higher photocatalytic activity, and the quantum efficiency at 420 nm reaches 3. 1%. This result once again proves that the N element can play an important role in the modification of SrTiO3.

3.1.3 Gradient doping

In 2019, Seiji et al. [25] proposed a method of gradient carrier doping into SrTiO3 in order to further improve the photoelectric conversion efficiency of the photoelectrode. The main idea is: grading the carrier density in the depth direction can improve the photoelectrode efficiency. In the critical surface layer, the lower carrier density leads to a longer photogenerated carrier lifetime and a wider space charge layer. , where the internal electric field drives the transmission of photogenerated carriers to the surface. In the deeper parts of the semiconductor photoelectrode, increasing the carrier density can reduce the photogenerated resistance loss. This method can better solve the problem of high recombination rate of photogenerated electron-hole pairs and improve the photoelectrochemical cathodic protection capability of SrTiO3.

3.2 Composite Modification

Compounding SrTiO3 with other semiconductors is an effective way to reduce the photogenerated electron-hole recombination rate. Two semiconductor materials with well-matched conduction and valence bands can ensure that carriers are effectively transferred from one semiconductor to another.

In 2011, Cao[26] used an in-situ growth method to prepare SrTiO3 composite TiO2 nanoarray materials, which were combined with pure TiO2 under UV light irradiation. Compared with nanoarray materials, SrTiO3 composite TiO2 array materials have stronger degradation ability of rhodamine B. This is due to the decrease in the photogenerated electron-hole recombination rate and the increase in photogenerated carrier density. Choudhary et al. [27] used sol-gel method and spin coating process to prepare nanostructured CuO/SrTiO3 double-layer materials and applied them to photoelectrochemical water splitting. The photocurrent density of the double-layer film reached 1. 85 mA/cm2 (at -0. 9 V vs. SCE potential), which is compared to single CuO and SrTiO3 The photocurrent density of the materials was increased by about 8 times and 30 times respectively. In 2017, Yao[28] prepared a double-layer CeO2/SrTiO3 composite photoelectrode material. After electrochemical testing, it was found that the composite photoelectrode has both electron storage capabilities and photoelectrochemical cathodic protection capabilities. The principle is: in the presence of light, external The inner layer of CeO2 will store the excess photogenerated electrons. In the absence of light, the inner layer of CeO2 will release the stored photogenerated electrons, and the released photogenerated electrons will be transferred to the cathode. Continue Protect the cathode.

In 2019, Wei et al. [29] designed a SrTiO3/TiO2 composite structure by hydrothermally growing SrTiO3 on the surface of TiO2 with a hollow Multi-shelled structure, achieving full coverage of SrTiO3 on the TiO2 surface and constructing a SrTiO3/TiO2 heterojunction. For the purpose, the hydrogen evolution rate of the composite material is 212 μ mol/h·g. After continuous exploration and summary by scientific researchers, the performance of SrTiO3 has been continuously improved after being combined with other materials. The electron storage performance of CeO2 has also been discovered, which provides theoretical guidance for SrTiO3 in future dark state research, realizing the goal from impossible to possible. development Goals.

3.3 Other Modification Methods

In addition to doping and compounding, the photoelectrochemical properties of SrTiO3 can also be improved through surface modification, dye sensitization and other methods.

In 2012, Ouyang et al. [30] changed the surface properties of SrTiO3 by constructing an alkaline environment to improve its photocatalytic performance. The surface alkalization caused by the highly alkaline environment made the conduction band become more negative, providing sufficient energy for water reduction. With the power of the photocathode, the hydrogen evolution efficiency of the photocathode is greatly improved.

In 2018, Miao Jijuan[31] loaded metal platinum (Pt) onto the SrTiO3/TiO2 composite structure. The test results showed that under ultraviolet illumination, when the Pt loading amount was 5%, the hydrogen evolution rate of the SrTiO3/TiO2 composite structure was 447 μ mol/h·g, which is 7. 7 times that of the unloaded Pt condition. Loading precious metal Pt significantly improves the photoelectrochemical performance of the SrTiO3/TiO2 composite structure. Li[32] loaded the size-adjustable noble metal palladium (Pd) onto SrTiO3 through in-situ growth. Compared with single -component materials, the photoelectrochemical performance was significantly improved, and the maximum hydrogen evolution rate was 440 μ mol/h·g.By loading precious metals onto the surface of the SrTiO3 material, it can be seen that the hydrogen production performance has been significantly improved. Compared with the previous section, it has increased from 212 μ mol/h·g to 447 μ mol/h·g. The improvement in hydrogen production performance can indirectly reflect the photoelectrochemical. The electron-hole pair separation ability is improved, and this method can be introduced into photoelectrochemical cathodic protection research.

In 2020, Robabeh[33] produced a SrTiO3@TiO2@Fe2O3 nanorod heterostructure film by hydrothermally growing a SrTiO3 film on rutile TiO2 nanorods with a template, and then using Fe2O3 for sensitization. By controlling the surface of the photocatalyst By increasing the ratio of Sr to Fe, the photocatalytic hydrogen production performance of this heterostructure is optimized, the photoelectron lifetime is improved, and the photoresistance is reduced. The photocatalytic hydrogen production capacity of this composite material has been significantly improved, providing a new idea for the research on SrTiO3 photoelectrochemical cathodic protection.

4 CONCLUSION AND OUTLOOK

Photoelectrochemical cathodic protection technology has attracted much attention from scientific researchers in metal anti-corrosion protection because of its green and environmentally friendly characteristics. Perovskite SrTiO3 has also been a research hotspot in recent years, but there are very few studies on SrTiO3 photoelectrochemical cathodic protection. For further research, it is more important to refer to the research progress of SrTiO3 in other fields. For example, the hydrogen production performance of SrTiO3 can indicate its photogenerated electron-hole separation ability, and the level of separation ability directly affects the photoelectrochemical cathodic protection ability. This article The research progress of SrTiO3 in recent years is also mainly reviewed from this aspect. For scientific researchers, the practical application of SrTiO3 in photoelectrochemical cathodic protection research is a long and arduous task. Future research directions and possible situations for the above phenomena include: (1) Controlling the conduction band potential of SrTiO3 is a key research idea, because the more negative the conduction band potential is, the greater the difference from the metal electrode potential will be, making it easier for photogenerated electrons to reach Metal material; at the same time, the valence band potential of SrTiO3 is adjusted to be sufficiently positive. In the absence of hole sacrificial agent and no bias, photogenerated holes can oxidize water to achieve depolarization and separate more photogenerated electrons. Complete the goal of polarizing the protected metal. Controlling the valence band and conduction band will inevitably involve visible light utilization, thereby raising the visible light utilization rate of SrTiO3 to a higher level of only 5%. These ideas will be hot research directions in the future.

(2) Photoelectrochemical cathodic protection technology involves multiple disciplines such as semiconductors, metals, interface chemistry, and corrosion research, and requires the cooperation and participation of different scientific researchers. The research content includes semiconductor/metal, semiconductor/solution interface, and metal/solution interface. Mechanism, etc., because during the actual experiment, only the condition that the conduction band potential of the semiconductor material is negative than the potential of the protected metal electrode is met, the metal is not necessarily well protected. For example, the difference between the two should also be considered. What To achieve the protection goal, the impact of corrosive media on semiconductor materials and other issues are also issues that researchers need to jointly solve in the future.

(3) The marine environment is complex and changeable, and the application of photoelectrochemical cathodic protection requires a long research process. Researchers must explore and summarize the reaction conditions of SrTiO3 in different environmental media in a laboratory environment, and then simulate Marine environment, research and improve the stability of SrTiO3 materials. In addition, it is necessary to realize that SrTiO3 still has photocathode protection capabilities in dark conditions, which requires further research in combination with energy storage materials, which also increases the difficulty of research.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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