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Research on Surface Nano-modification of High Voltage Electrical Corona-resistant Nanocomposite

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Abstract

Materials with good electrical and mechanical properties such as corona resistance and tensile resistance are needed in various electrical engineering. Polyimide composite material is a kind of electrical materials with excellent performance and a wide range of applications, especially the polyimide material after the composite modification of nanomaterials, which has superior performance. This paper introduces a method to further optimize the properties of polyimide materials through surface nano-modification treatment in the composite modification. That is, by modifying the surface of the nanocomposite material with functional groups (hydroxyl groups), the dispersibility and connection strength of the noble metal particles and the nanomaterial itself in the polyimide matrix are improved. We have selected titanium dioxide nanotubes (TiNT) and silver nanoparticles as representatives of inorganic nanoparticles and precious metal nanoparticles, and prepared a new type of polyamide with corona resistance and tensile resistance far beyond that of pure polyimide. Our research provides a reference for further research on the composite of other inorganic nanoparticles and precious metal nanoparticles in polymer materials.

Keywords

Electrical insulation; corona resistance; functional groups; nano-modification.

1. Introduction

Polyimide is currently one of the most widely used corona-resistant electrical materials. With the technological development of the electrical industry, more and more electrical engineering and electronic equipment require polyimide materials with better corona resistance, stretch resistance and stability [1,2]. To this end, there are continuous new research works to improve and enhance the various properties of polyimide [3]. Through the application of the most popular nanotechnology, a variety of inorganic nanomaterials are doped into polyimide, and composite materials with superior corona resistance can be obtained [4-7]. Among the commonly used inorganic nanomaterials in current research, there is a type of inorganic nanomaterials containing titanic acid structure represented by titanium dioxide, such as barium titanate, copper calcium titanate, etc. [8-10]. The corresponding research work of these materials can improve the corona resistance of polyimide to a certain extent. However, as in the case of other inorganic nanomaterials, the doping amount of these titanic acid structured nanomaterials also has different upper limits. Moreover, as the amount of doping increases, the dielectric loss of the composite material will also increase, which will have an adverse effect on the electrical properties of the composite material.

In order to overcome the shortcomings of this composite material, researchers began to study the composite and modification treatment of precious metal nanoparticles and titanic acid inorganic nanoparticles, and the use of precious metal nanoparticles to eliminate undesirable dielectric loss. Related research work includes: compounding silver nanoparticles with copper calcium titanate to obtain a polyimide composite material with high dielectric and low loss;

compounding titanium dioxide nanowhiskers with silver nanoparticles to obtain a greater amount of doping And dielectric properties, etc. [11-13]. However, these researches on the composite of titanic acid nanomaterials and precious metal nanoparticles mostly use physical blending and ion adsorption methods to achieve the composite of precious metal nanoparticles and titanic acid particles. This kind of preparation process is often difficult to finely control, and noble metal nanoparticles often form random agglomerates on the surface of titanic acid particles due to the nano-size effect, the dispersion is uneven, and the connection effect is also poor. The clustering of a large number of precious metal nanoparticles leads to percolation effect, which will increase the dielectric loss of composite materials [14,15]. And it will also increase the amount and waste of precious metal nanoparticles.

The root cause of this problem is that the connection mechanism between noble metal nanoparticles and titanic acid-based materials is not considered. Relying only on the direct physical and chemical adsorption of noble metal particles and titanic acid materials cannot solve this problem. Moreover, previous research only paid attention to improving the dispersibility of inorganic particles through graft modification of inorganic particles, and ignored the connection and dispersion effect between the inorganic particles themselves and the precious metal particles. In view of this, this thesis aims at this problem, selects a representative titanium dioxide nanotube material, first modifies its surface with functional groups with dopamine, and establishes a hydroxyl group that is beneficial to silver nanoparticles. Subsequently, a more regular and stronger linking group is established through the hydroxyl group and the titanium atom in the titanium dioxide nanotube. Then, the other end of the dopamine is modified with silver nanoparticles, so as to obtain evenly distributed noble metal nanoparticles modified with titanium dioxide nanotubes. Finally, it is combined with the polyimide matrix material to obtain a composite material with better corona resistance and dielectric loss performance. Our research can also be applied to other nanomaterials with titanic acid structure and other precious metal nanomaterials such as gold and platinum.

2. Sample preparation and testing

2.1. Samples preparation

(1) Using a 10M concentration of NaOH and anatase titanium dioxide powder mixture, the titanium dioxide nanotube material is prepared by hydrothermal reaction in an autoclave. The hydrothermal reaction temperature is 140°C, and the reaction time is 72 hours.

(2) In a round bottom flask, mix 0.5 g of the titanium dioxide nanotubes obtained in the previous step and 0.05 g of dopamine hydrochloride. Add 30 ml of deionized water to the flask, the mixture will turn bright brown and then the mixture will be sonicated for 15 minutes. The mixture was heated at 70°C while stirring overnight. The brown solid was filtered, washed with 250 ml of deionized water and 50 ml of ethanol; dried under vacuum and ground into a fine powder.

(3) Mix 0.1 g of dopamine-modified titanium dioxide nanotubes with 0.5 ml of 0.1M silver nitrate solution and stir overnight. 1 ml of methanol was added to the mixture, and the mixture solution was placed under ultraviolet light for 20 minutes. Filter and wash the photoreduction reaction product with deionized water and ethanol.

(4) Add the titanium dioxide nanotubes with silver nanoparticles modified on the surface to the ethanol solution of the silane coupling agent, ultrasonically disperse them for 15 minutes, and then treat them in an oil bath at 400°C for 4 hours. Finally, the reaction product is repeatedly washed with deionized water and ethanol, filtered, and naturally dried for later use.

(5) Add the surface-grafted titanium dioxide nanotube material to the DMF solution of pyromellitic dianhydride (solid content 10%). 80°C water bath while stirring for 30 minutes. A

5% concentration of BPADA was added, and the reaction was stirred for 4 hours to obtain a polyimide precursor solution.

(6) Using the polyimide precursor solution obtained in the previous step, a polyimide conforming film with a thickness of about 25 microns is prepared by a knife coating process. The content of titanium dioxide nanotubes in the finally obtained polyimide composite film is 10%.

(7) Prepare sample 2 using basically the same preparation steps as sample 1, but without adding dopamine hydrochloride.

(8) Prepare sample 3 using basically the same preparation steps as sample 1, and the content of titanium dioxide nanotubes is 15%.

(9) Prepare sample 4 using basically the same preparation steps as sample 1, and the content of titanium dioxide nanotubes is 20%.

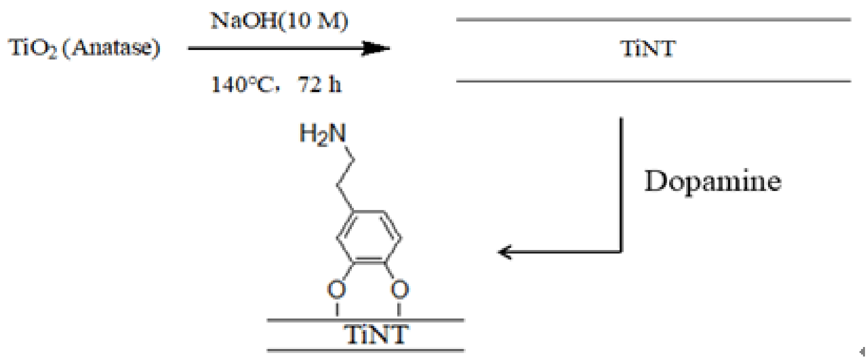

Fig. 1 The basic process of dopamine modification of titanium dioxide nanotubes.

2.2. Testing methodes.

The FTIR test of the experimental samples was done on the Nicolet 6700 FTIR spectrometer. The UV-Vis absorption spectrum of the sample was completed on a Shimadzu UV-2550 UV/Vis spectrometer. The projection electron micrograph of the sample was measured on the Tecnai G2 S-Twin projection electron microscope. The energy spectrometer (EDS) analysis of the samples was performed on a Hitachi SU8010 field emission scanning electron microscope (SEM).

3. Literature References

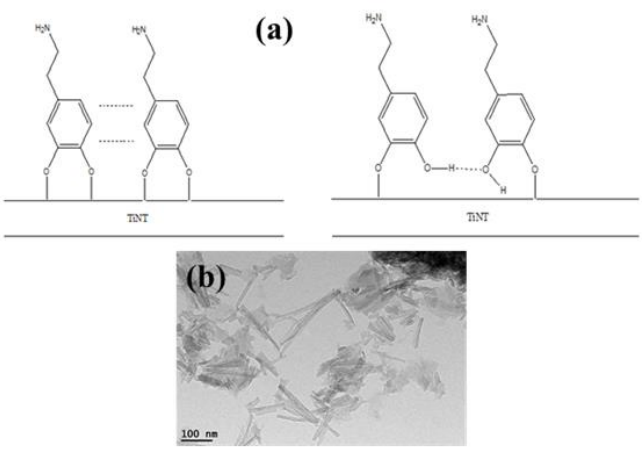

Fig. 2 (a) Possible connection methods between dopamine and titanium dioxide nanotubes and (b) Transmission electron micrographs.

Figure 2(a) shows a schematic diagram of possible connections between dopamine and titanium dioxide nanotubes. After the stirring and heating reaction, dopamine establishes a strong functional group connection with the titanium atom in the titanium dioxide nanotube by virtue of the hydroxyl structure that is attached to the benzene ring in its structure. And the distribution of dopamine obtained by our reaction on the surface of titanium dioxide nanotubes is relatively regular and uniform. Compared with general physical dispersion and physical and chemical adsorption, the distribution effect is much better. These are evenly distributed on the surface of the titanium dioxide nanotubes, and the strong functional groups can also play a stronger role in the subsequent modification of the silver nanoparticles. In order to observe the effect of dopamine forming functional groups on the surface of titanium dioxide nanotubes more intuitively, we conducted transmission electron microscopy tests and analysis on the samples. From Figure 2(b), we can see the structured titanium dioxide nanotubes prepared by the hydrothermal reaction method. However, because the molecular structure we modified on the surface of titanium dioxide is relatively small, we can only observe the titanium dioxide nanotubes with smooth inner and outer walls from the transmission electron microscope picture in Figure 2(b), and we cannot visually see the dopamine in the nanotubes. On the other hand, we can see from the projection electron microscope picture in Figure 2(b) that the dopamine modification process engineering did not damage the structure of the titanium dioxide nanotubes, indicating that the modification process we used is appropriate. We further speculate that the dopamine modification process we use will also not damage the structure of other titanate nanomaterials such as barium titanate, copper calcium titanate, and so on.

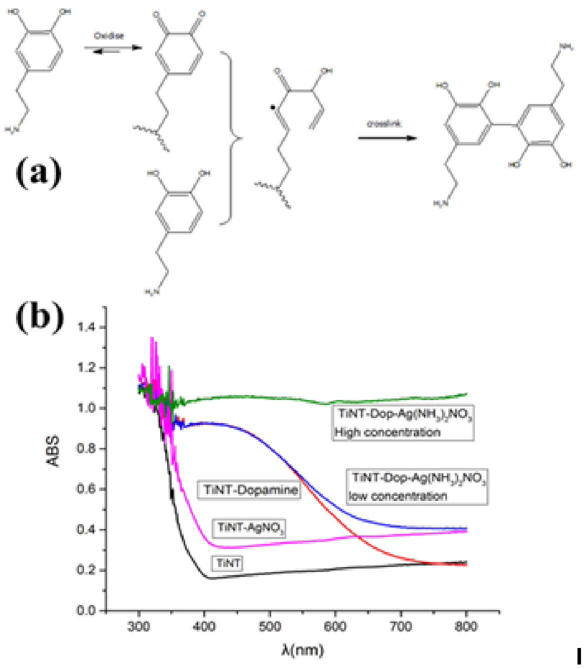

Fig. 3 (a) Cross-linking process of dopamine and (b) UV-Vis spectra of samples before and after the modification of silver nanoparticles.

Figure 3(a) shows a schematic diagram of the cross-linking process of dopamine we used. After the dopamine is modified on the surface of the titanium dioxide nanotubes, it can be further cross-linked to form a more stable structure. Such a stable structure can further improve the connection effect between the silver nanoparticles and the titanium dioxide nanotubes. The noble metal nano-particle modification effect with more uniform distribution and stronger connection is obtained. However, we also need to perform corresponding physical and chemical analysis to verify whether dopamine and silver nanoparticles are actually modified on the surface of titanium dioxide nanotubes. Therefore, we carried out UV-Vis spectrophotometer test (UV-Vis) on titanium dioxide nanotubes, dopamine-modified titanium dioxide nanotubes,

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and silver nanoparticles modified samples with different amounts of silver nitrate. The results are shown in Figure 3(b). Show. From the comparison of the UV-Vis spectra of the samples before and after the dopamine modification, we can see that the sample after the dopamine modification process has an obvious absorption peak between 400 and 600 nm, and this absorption peak does correspond to the absorption peak introduced by dopamine. This test result shows that the dopamine modification process we used does indeed establish well-connected functional groups on the surface of the titanium dioxide nanotubes. It can also be seen in Figure 3(b) that after the silver nanoparticles are further modified, the absorption of the dopamine-modified titanium dioxide nanotubes in the region above 600 nm has further increased. This phenomenon is due to the precious metals brought by the silver nanoparticles Local plasma effect [16]. At the same time, it should be noted that the amount of silver nitrate used in the modification process is not as much as possible. In fact, the agglomeration of silver particles caused by excessive silver nitrate not only can not improve the performance of the composite material, but also leads to disadvantages such as dielectric loss.

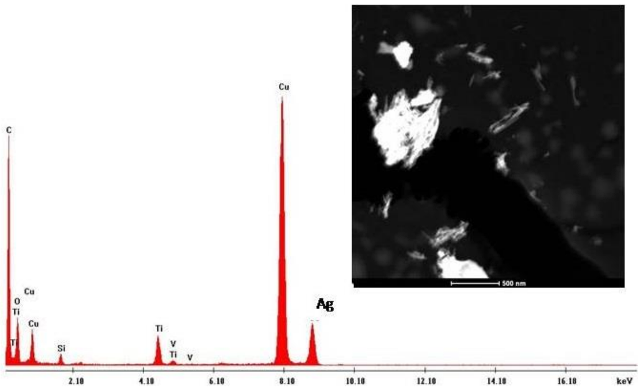

Fig. 4 EDS element analysis of samples modified with surface dopamine and silver nanoparticles.

Through the UV-visible spectrophotometer test and transmission electron microscope photos, only the topography and absorption spectrum analysis results can be obtained. If we want to get more intuitive results on elemental analysis, we also need to perform EDS elemental analysis tests on the samples. Figure 4 shows the EDS element analysis of samples modified with surface dopamine and silver nanoparticles. Our EDS element analysis is based on transmission electron microscopy testing. In the transmission electron microscopy testing process, first select a suitable sample microscopic area, and then perform elemental analysis on this area. The analysis result is shown in Figure 4. . From the analysis results of EDS, it can be seen that the silver nanoparticles are indeed modified on the surface of the titanium dioxide nanotubes through the modification process of silver nanoparticles.

Through the analysis and test of Fourier Infrared Spectrophotometer (FTIR), the situation of various chemical bonds or groups on the surface of the sample can be well analyzed [17]. We tested and analyzed the FTIR spectrum of samples modified with surface dopamine and silver nanoparticles, and the results are shown in Figure 5(a). From the comparison of the absorption peak curve of the titanium dioxide nanotubes before and after modification in the figure, it can be seen that after the surface dopamine and silver nanoparticles are modified, the titanium dioxide nanotubes increase the absorption between 1000 and 2000 nanometers corresponding to the absorption caused by the modification of dopamine. The FTIR test results again verified the results of the previous transmission electron microscope and UV-Vis tests. It shows that our modification process has indeed got a good modification effect. Next, we analyze and study the internal composite effect of the final silver nanoparticles and dopamine-modified titanium dioxide nanotube composite polyimide materials. Figure 5(b) shows a scanning electron micrograph of the inside of the cross-section of the final polyimide composite material. It can be seen from the figure that silver nanoparticles and dopamine-modified titanium dioxide nanotubes form a uniform and dense structure inside the polyimide. Such a uniform and dense internal structure also brings stable mechanical and electrical properties of the composite material.

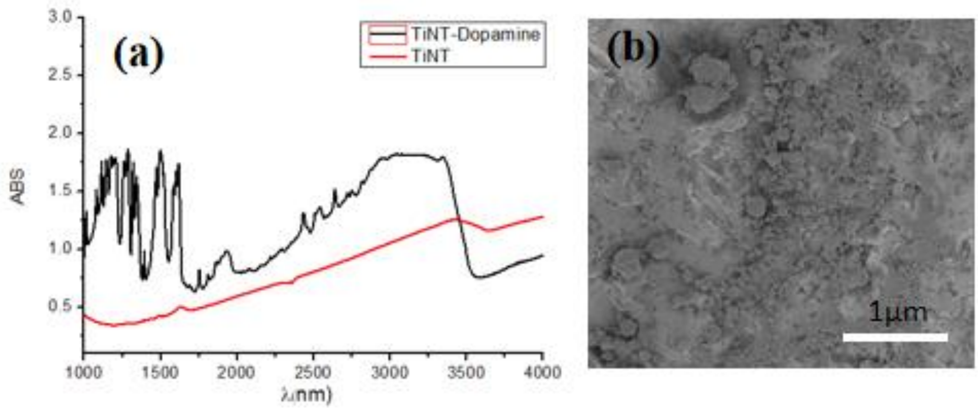

Fig. 5 FTIR spectra of samples modified by surface dopamine and silver nanoparticles and (b) scanning electron micrograph of composite film cross-section

Table 1 Comparison of corona resistance and mechanical properties of different samples

Numble	Film thickness (μm)	Corona resistance time (hours)	Tensile strength (MPa)	Elongation at break (%)
1	25.0	501	144	30
2	25.1	421	122	20
3	25.1	507	149	32
4	25.0	497	139	27

Next, we will test the corona and tensile properties of different samples in the sample preparation process. We perform corona resistance tests on all four groups of samples in accordance with the technical requirements of GB/T22689-2008/IEC60304:1991 (test parameters: 20kHz, 1kv); in accordance with the technical requirements of GB/T 13542.2-2009, 50 mm The tensile speed per minute is used to test the mechanical properties of the sample. The results are shown in Table 1. From the performance comparison of different samples in Table 1, it can be seen that the corona resistance and mechanical properties of the polyimide composite film modified with dopamine and silver nanoparticles have been significantly improved. At the same time, it is also seen that it is not that the more dopamine and silver nanoparticles modified titanium dioxide nanotubes are used, the better. The sample with 15% doping has the best performance.

4. Summary

This study uses dopamine to modify the surface of titanic acid nanomaterials represented by titanium dioxide nanotubes, and uses the hydroxyl group of dopamine to connect the titanium atoms in the titanium dioxide nanotubes, thereby forming a uniform distribution and strong connection on the surface of the titanium dioxide nanotubes. Then the silver nanoparticles are connected through the modified functional groups. Finally, uniformly distributed and stable silver nanoparticles are modified on the surface of the titanium dioxide nanotubes. And through the corona resistance and mechanical performance test and comparison of samples with different doping content, the best doping ratio samples were obtained. Our research findings can also be applied to other nano-material composite polyimide films with titanic acid structure, which has certain reference value for the development of new corona-resistant materials.

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