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Research Progress of Polyetherimide Dielectric Materials

Jiaqi Su[1,a,*], Kun Lu[1]，Shidong Qin[1]

[1]school of Materials Science and Engineering ,Shijiazhuang Tie dao University，Shijiazhuang City，China，050043

[a] 1927658246@qq.com
**corresponding author*

Abstract:*Polyetherimide (PEI) is a high-performance thermoplastic engineering plastic. With excellent thermal stability, mechanical strength and chemical stability, it shows wide application potential in many fields. However, its stability at high temperatures and relatively low dielectric constant limit its application. In recent years, researchers have modified PEI by adding conductive fillers, inorganic nano-fillers and developing all-organic composite materials, effectively improving its dielectric properties. However, the balance between the thermal stability and dielectric properties of PEI, as well as the selection and optimization of fillers, remain urgent problems to be solved. In the future, in-depth research can be conducted in the directions of new matrix selection, optimization of filler types and forms, and structural improvement, in order to prepare dielectric materials with high temperature resistance and high energy storage density.*

Keywords: *Polyetherimide ;Dielectric properties; Composite material ;Modification ;Research Progress*

Polyetherimide (PEI) is widely used in aerospace, electronics, automobiles and other fields due to its excellent high-temperature resistance, chemical stability and mechanical properties. Since its first synthesis in the 1960s, PEI has been a research focus in polymer science. Its molecular structure endows it with excellent dimensional stability, UV resistance, low water absorption and outstanding electrical performance, making it widely used in electronic components, connectors and switchgear [1]. In addition, PEI has strong resistance to chemical corrosion and can withstand the erosion of various solvents and chemical substances, enhancing its application potential in harsh environments.

However, PEI materials still have some drawbacks during their development: 1) Although PEI has a relatively high glass transition temperature (T_g), degradation may still occur at extreme high temperatures (beyond its thermal stability limit), affecting its dielectric performance. When the requirements for high-temperature applications are rather strict, the thermal stability of PEI may not meet the demands of specific fields. 2) In dielectric applications, although PEI has good dielectric properties and low loss characteristics, its dielectric constant (ε_r) is relatively low, which limits its application in some high-performance dielectric materials. Improving the dielectric properties of PEI and enhancing its electric field tolerance remain an important direction that researchers focus on. Therefore, the modification and optimization of polyetherimide materials will be a long-term and

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arduous task in the field of polyetherimide research. The following provides an overview of PEI. On this basis, the research progress in polyetherimide dielectric materials in recent years is expounded. Finally, the existing problems of polyetherimide dielectric materials are summarized.

1. Overview of Polyetherimide

PEI is a high-performance thermoplastic engineering plastic and holds an important position in the field of materials science. In 1964, General Electric Company (GE) of the United States first carried out research and development on PEI. The molecular structure of PEI is connected by ether bonds (-O-) and imide groups (-C=O-N-), and this unique structure endows PEI with outstanding performance. Due to its special molecular structure, PEI exhibits excellent thermal stability and has a T_g as high as 217°C. This characteristic enables PEI to maintain excellent mechanical properties and dimensional stability in high-temperature environments, and it is widely used in fields such as aerospace, electronics and electrical appliances, and automobiles, where the heat resistance of materials is strictly required. However, in some strong oxidizing acids or extreme environments with high temperature and high humidity, the performance of PEI may deteriorate.

At present, the main material for commercial film capacitors is biaxially oriented polypropylene (BOPP), with an ε_r of approximately 2.2 and an energy storage density of only 1 to 3 J/cm^3. This relatively low energy storage density makes BOPP capacitors unable to meet the growing demands for miniaturization and lightweighting [2]. In addition, the operating temperature of BOPP capacitors is generally below 120°C, while the maximum operating temperature of BOPP cannot exceed 105°C, which limits its application in high-temperature environments. Therefore, the development of polymer dielectric materials with high energy storage density and high-temperature resistance has become an urgent task [3].

To further expand the performance and application fields of PEI, researchers modified it by adding inorganic materials, conductive materials, and preparing all-organic composite materials, with the aim of enhancing its high-temperature energy storage performance. These studies not only enhance the competitiveness of PEI in the field of dielectrics, but also open up new prospects for it in a wider range of application fields.

2. Research Progress of Polyetherimide in the Dielectric Field

In the vigorous development process of modern materials science, polymer matrix composites have become the focus of research due to their unique performance advantages and broad application prospects [4]. With the increasingly strict requirements for material performance in various industries, such as the pursuit of materials with high electrical conductivity and excellent dielectric properties in the electronics field, the desire for materials with high energy density in the energy storage field, and the urgent need for stable dielectric materials in high-temperature environment application scenarios, the research on polymer matrix composites has been continuously deepened and expanded [5]. Among them, the three major categories of conductive fillers/polymer composites, inorganic nano-fillers/polymer composites, and all-organic polymer composites each demonstrate unique performance characteristics and research directions, attracting the attention and exploration of numerous researchers. In recent years, a series of remarkable research progress has been achieved.

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2.1. Conductive filler/polyetherimide

In the field of materials science, common conductive fillers include silver, nickel, carbon nanotubes (CNT), and graphene (GnP), etc. Unlike high dielectric inorganic fillers that need to be added in large quantities to enhance the ε_r of the composite material, according to the percolation theory, a small amount of conductive fillers can achieve a similar effect. When the content of conductive fillers approaches the percolation threshold, the ε_r of the composite material will increase significantly. In polyetherimide composites, the addition of conductive fillers can significantly improve the electrical conductivity. In addition, conductive fillers also improve the dielectric properties of the composite materials. A high ε_r of the filler can increase the ε_r of the composite material, and by surface modification and optimizing the interface interaction between the filler and the matrix, it helps to reduce dielectric loss.

Yang Luxi et al. [6] mixed multi-walled carbon nanotubes (Ox-MWCNTs) of different mass fractions with N-methylpyrrolidone (NMP) solvent and ultrasonically applied them. They added PEI particles and stirred for dissolution. After the film was cast and laid, it was vacuum-dried at different temperatures to prepare a series of Ox-MWCNTs/PEI composite films. When the mass fraction of Ox-MWCNTs is $\geq 1\%$, the film shows nonlinear electrical conductivity characteristics, and the percolation threshold is approximately 3%. At this time, the nonlinear coefficient of the 3% Ox-MWCNTs/PEI sample reaches 6.2. The dielectric constant of the 3% Ox-MWCNTs/PEI composite film increased from 6.04 to 14.56 at 1000Hz, and the dielectric loss only increased from 0.02 to 0.14.

Mohsin Ali Marwat et al. [7] prepared GO@ZO packing by coupling zinc oxide (GO) with graphene oxide (ZO), and prepared polyvinylidene fluoride - hexafluoropropylene containing different mass percentages of GO@ZO packing by solution casting. BP blends were used as the top layer and linear PEI as the bottom layer, resulting in different bilayer composites (xBP-L, where x represents the mass percentage of GO@ZO nanofillers). Studies show that the 2BP-L composite film exhibits excellent performance. At an electric field of 527 MV/m, the discharge energy density reached 12.63 J/cm^3 with only 2 wt% GO@ZO filler, which was approximately 426.3% higher than that of pure PEI material. Furthermore, the xBP-L nanocomposites prepared by adopting this new type of bilayer heterostructure design have been significantly enhanced in both dielectric properties and breakdown strength. The ferroelectric layer at the top provides a relatively high dielectric constant, while the PEI layer at the bottom ensures a relatively high breakdown strength [8].

2.2. Inorganic nanomaterials/polyetherimide

Since the concept of nano-dielectrics was proposed, nanoparticles have been widely embedded in polymers to enhance the performance of nanocomposites in terms of electrical, mechanical, physical and thermal properties, etc. [9] Nanodielectrics are expected to become a new generation of high-voltage insulating materials, and this potential has attracted widespread attention, especially in suppressing partial discharge, reducing space charge, increasing breakdown strength (Eb), and enhancing the storage characteristics of capacitors. Nanoparticles play a key role [10]. Over the years, many attempts have been made and various nano-fillers such as Al_2O_3, HfO_2, $BaTiO_3$, TiO_2, MnO_2, and

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MgO have been used. However, the selection criteria for nano-fillers remain unclear to this day.

In the field of polymer-based nanocomposites, the energy density is often enhanced by incorporating nano-fillers with high ε_r . However, with the enhancement of the applied electric field, the energy loss of such nanocomposites increases and the charge-discharge efficiency decreases, resulting in a decrease in Eb and limiting their application. Recent studies have shown that adding wide bandgap nitride and oxide (such as BNNS, Al_2O_3) to polymer matrices can effectively reduce conductive loss and significantly increase Eb[11]. However, due to its relatively low ε_r , the improvement of energy density of nanocomposites is still limited. It can be known from this that the band gap is closely related to ε_r and jointly determines the final energy density of nanocomposites.

Lulu Ren et al. [12] prepared a series of composites by adding SiO_2, ZrO_2 and TiO_2 nanoparticles to PEI at different volume fractions ranging from 1 vol% to 9 vol% respectively. Studies show that TiO_2 nanocomposites exhibit large dielectric loss and low charge and discharge efficiency due to their narrow band gap and high electrical conductivity. SiO_2 nanocomposites have a wide band gap and high charge-discharge efficiency, but the low ε_r limits the improvement of energy density. The ZrO_2 nanocomposites, on the other hand, possess both a wide band gap and a moderate ε_r, demonstrating excellent comprehensive performance.

He Li et al. [13] prepared PEI ternary polymer nanocomposites containing barium titanate nanoparticles (BTNPs) and BNNSs. The research finds that the composite system of 1.27 vol% BTNPs and 6.05 vol% BNNSs has the best performance. At 150 °C and 1000 Hz, the ε_r of this composite material reaches 3.74, the dielectric loss is 0.00715, and the Eb reaches 547 MV/m. The charge and discharge efficiency of this ternary composite material is high, and the efficiency is close to 90% under an electric field of 300 MV/m. After 50,000 cycles of charge and discharge at room temperature and 150 °C, the discharge energy density shows almost no attenuation.

Na Zhang et al. [14] proposed a novel laminated MgO/ polyetherimide (MgO/PEI) composite nanomaterial, in which the insulating layer was placed near the electrode to prevent charge injection, and the spatial distribution of MgO nanoparticles was optimized to construct deeper internal trap energy levels. This structure integrates the optimization of the electrode/dielectric interface and the bulk phase of the polymer dielectric, thereby significantly regulating charge migration, suppressing conduction loss and improving the overall energy storage performance at high temperatures. The optimized composites with 5.0 wt% MgO added to the polarization layer and 1.0 wt% MgO added to the insulation layer achieved ultra-high Ue at both 150 °C and 200 °C.

2.3. All-organic polymer composite materials

In the research on enhancing the dielectric strength of polymers, using inorganic nanoparticles of different shapes and structures as fillers is a common approach. Organic composite dielectric materials have advantages in processing, dielectric, mechanical properties and environmental friendliness, making all-organic composite dielectric materials a current research hotspot in the field of dielectrics [15].

Bo Peng et al. [16] prepared CD/PEI all-organic composite films by introducing cyclodextrin (CD) into PEI through solution blending based on the molecular isolation strategy. Studies show that with the increase of CD content, the introduction of CD significantly suppresses the conduction loss of PEI

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under high temperature and high electric field, increases Eb, and reduces the leakage current density. CD isolates the PEI molecular chain through volume effect, increases the chain spacing, interrupts the conjugation of aromatic rings, and hinders the formation of electron channels, thereby reducing conduction loss [17]. For example, 1.5 wt% CD/PEI composite films perform exceptionally well under an electric field of 150°C and 500 kV/mm. Their energy storage density reaches 2.68 times that of pure PEI, and they have good cycling stability, with charge and discharge cycles exceeding 100,000 times.

Bin Zhang et al. [18] blended 1,4,5, 8-naphthalenetetraformic anhydride (NTCDA) with PEI to prepare all-organic dielectric polymer composites. Research shows that the relationship between current density and electric field verifies that under high temperature and high electric field, the charge conduction within the material follows the jump conduction model. Adding 0.5 vol% of NTCDA can reduce the charge jump distance, form more charge traps, and thereby enhance Eb at high temperatures. Among them, the discharge energy density of 0.5 NTCDA/PEI composite material reaches 5.1 J/cm^3 at 150°C, and the discharge efficiency is 90%. At 200°C, the discharge energy density is 3.2 J/cm^3 and the discharge efficiency is 85%.

Song Ding et al. [19] prepared thin films by mixing PEI and polyarylether urea (PEEU) in NMP in different proportions. The introduction of PEEU reduced the leakage current density under high temperature and high field, increased the trap depth and enhanced the charge capture ability. The PEI/15% PEEU film has the highest Eb, which is 736 MV/m at room temperature and 672 MV/m at 150°C. The discharge energy density of this film at 150°C and 550 MV/m reaches 4.84 J/cm ^{3}, and the efficiency exceeds 90%.

3. Conclusion and Outlook

As a high-performance dielectric material, PEI has demonstrated extensive application potential in multiple fields such as aerospace, electronic devices, and automobiles, thanks to its outstanding thermal stability, mechanical strength, and chemical stability. Nevertheless, the stability of PEI at high temperatures and its relatively low ε_r limit its performance in some specific applications. In order to improve the dielectric performance of PEI, researchers have modified and optimized it through various means, such as adding conductive fillers, inorganic nano-fillers and developing all-organic composite materials to PEI. These efforts have not only effectively enhanced the dielectric properties of PEI materials, but also provided new directions for their applications in high-energy-density capacitors, electronic components and high-temperature environments.

At present, the research on polyetherimide dielectric materials is still in the process of continuous development. Although certain progress has been made, there are still some problems that need to be solved urgently. Firstly, the balance between the thermal stability and dielectric properties of PEI remains a challenge. How to enhance its operating temperature and electric field tolerance while ensuring the material's performance remains the focus of future research. Secondly, the selection and optimization of fillers remain the key to enhancing the performance of composite materials. How to rationally design the types, sizes, and distributions of fillers to achieve the best dielectric and mechanical properties still requires further exploration.

Based on this, in order to obtain dielectric materials with high temperature resistance and high energy storage density that can be practically applied, the future research directions of PEI composites

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should focus on the following aspects:

1) Matrix selection: Previous studies have basically chosen commercial PEI systems as the matrix for composite materials. In recent years, new types of PEIs developed by the reaction of different diamines and dianhydride monomers have also achieved good energy storage properties. Therefore, using these PEIs as the matrix may lead to the acquisition of composite materials with even better performance.

2) Selection of fillers: The addition of high dielectric materials such as alumina (Al_2O_3), barium titanate ($BaTiO_3$), perovskite structural materials, etc., which have a relatively high ε_r, can enhance the electrical energy storage density of the composite materials. Studying how to select the appropriate type, form (nanometer, micrometer), dispersibility, etc. of fillers can effectively increase the energy storage density of materials.

3) Structural optimization of composite materials: It is expected to obtain composite materials with excellent energy storage properties by reducing film defects, introducing electronic traps or preparing layered structures.

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