

Development of epoxy resin with superior breakdown strength: A Review

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ABSTRACT

Epoxy resin (EP) has been widely utilized in electrical equipment and electronic devices due to its fascinating electric, thermal, and mechanical properties. However, the complex insulation structures of modern power devices in high-voltage direct current systems pose several challenges for EP-based dielectrics. The most significant among these challenges is the need for EP to stably operate under greater electric fields, requiring superior breakdown strength. This paper summarizes the key factors influencing the breakdown strength of EP and reviews reported methods for enhancing this property. Recognizing the limitations of existing approaches, we propose that the emerging technology of molecule design offers a potentially optimal solution for developing EP with enhanced breakdown strength. Furthermore, we anticipate the future development direction of EP with satisfactory insulation properties. We believe that enhancing the breakdown theory of solid dielectrics, exploring new research and development methodologies, and creating environmentally friendly EP with high performance are primary focus areas. We hope that this paper will offer guidance and support for the future development of EP with superior breakdown strength, proving valuable in advancing EP-based dielectrics.

KEYWORDS

Epoxy resin, breakdown strength, electrical insulation.

To reduce carbon dioxide emissions in the power system, clean energies, such as wind, solar, and hydropower, must be efficiently converted into electricity and transported over significant distances. High-voltage direct current (HVDC) transmission technology has emerged as a crucial solution to ensure stable energy transmission, improve transmission capacity, and reduce transmission line losses. HVDC technology is a crucial energy strategy of the 21st century, optimizing energy allocation, improving grid reliability, promoting clean energy utilization, and driving low-carbon economic development^[1-5]. The progression of HVDC systems depends heavily on the reliable functioning of power equipment. However, during prolonged operations, these components may encounter malfunctions. Among these, insulation faults have been identified as a prevalent cause of failures in power equipment, with electric breakdown being a severe and consequential malfunction^[6]. Electric breakdown incidents in high-voltage equipment can significantly damage insulation systems, leading to equipment failure or paralysis, posing serious risks to the stability of the power supply^[6]. Therefore, enhancing resistance to electric breakdown in power equipment is crucial for maintaining a stable power supply in the HVDC system. The insulation properties of power equipment are primarily influenced by the characteristics of the insulation dielectrics. Epoxy resin (EP) is a widely accepted insulation dielectric in power systems due to its competitive electrical, thermal, and mechanical properties^[7-10]. However, the complex insulation structures of modern power devices in HVDC systems pose several severe challenges for EP-based dielectrics. Currently, EP faces the critical challenge of inadequate breakdown strength, which hinders the development of advanced EP-based power equipment operating at higher voltages. Additionally, a growing demand exists for EP-based dielectrics with superior breakdown strength in electronic devices and electric vehicles^[6,11].

Therefore, exploring EP and EP-based dielectrics with enhanced breakdown strength is essential for developing advanced EP-based power equipment and electronic devices.

1 The theory of solid breakdown

Theoretical research on electric breakdown in solid dielectrics began in the early 20th century. Despite numerous efforts by several scholars, the complex physics processes involved in electric breakdown remain unclear^[12-20]. The breakdown phenomena in solid dielectrics are diverse and can be categorized into several types, including electric breakdown, free volume breakdown, thermal breakdown, electromechanical breakdown, space charge effect, and aging^[12-21]. Figure 1 illustrates the relationship between the electric field and time associated with different breakdown mechanisms. The breakdown voltage ranges from about 10^5 to 10^{10} V/m, with breakdown times varying from 10^{-9} to 10^3 s. Breakdown incidents lasting more than 10^3 s are considered aging and will not be discussed further in this paper.

Breakdown in solid dielectrics can be classified into two categories based on the underlying mechanism: intrinsic breakdown (electric breakdown and free volume breakdown) and extrinsic breakdown (thermal breakdown, electromechanical breakdown, and space charge effect)^[6]. While intrinsic breakdown is primarily determined by the intrinsic properties of the material and typically occurs at electric field strengths ranging from 10^9 to 10^{10} V/m, extrinsic breakdown is influenced by external factors, often resulting in a lower breakdown strength than intrinsic breakdown.

1.1 Intrinsic breakdown

1.1.1 Electric breakdown

Electric breakdown theories, such as Zener breakdown^[16] and

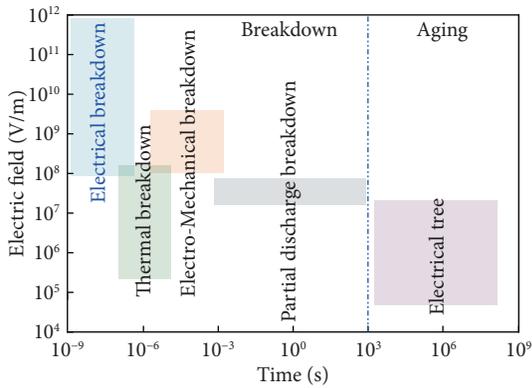


Figure 1 Relationship between electric field and time related to breakdown mechanisms^[6,18].

avalanche breakdown^[17], are commonly used in semiconductors. Following Zener breakdown theory, electrons in the valence band can tunnel into the conduction band under high electric fields, initiating current flow and breakdown. However, the avalanche breakdown theory suggests that free carriers within the solid are accelerated by the applied electric field and collide with neighboring atoms, resulting in collision ionization and the generation of new carriers. These newly generated carriers undergo the same acceleration and collision ionization process, causing an electron avalanche effect that ultimately leads to breakdown.

1.1.2 Free volume breakdown

In polymers, the volume can be divided into two components: occupied and unoccupied (also known as free volume). Artbauer^[17] proposed the free volume breakdown theory, which suggests that carriers in solid polymers are accelerated within the free volume and rapidly accumulate energy. Building upon the principles of Zener and avalanche breakdown theories, this theory explains the breakdown behavior of polymers below their glass transition temperature. Specifically, an increase in temperature results in a greater amount of free volume in the polymer, consequently reducing the breakdown strength.

1.2 Extrinsic breakdown

1.2.1 Thermal breakdown

When subjected to an electric field, dielectrics generate heat and experience loss while dissipating heat to the surrounding environment. The thermal accumulation and dissipation can influence the breakdown strength of dielectrics^[18]. When the heat generation rate exceeds the heat dissipation rate, the heat accumulation will increase the temperature of dielectrics and resulting increase in temperature leads to greater conductivity, amplifying conductive losses and further heat accumulation, causing the temperature to rise once more. Ultimately, these conditions may lead to the degradation and carbonization of the dielectric, culminating in breakdown due to the combined effects of electricity and heat, a phenomenon known as thermal breakdown.

1.2.2 Electromechanical breakdown

Researchers have discovered that in thermoplastic polymers with a low elastic modulus, breakdown above the glass transition temperature is closely linked to their elastic modulus^[19,20]. Applying an electric field exerts a compressive force on dielectrics with a low elastic modulus, causing a decrease in thickness. This reduction results in a higher electric field strength. The intensified electric field, in turn, further reduces the material's thickness. Ultimately,

breakdown occurs due to the combined effects of the electric field and mechanical force acting on the dielectric material.

1.2.3 Space charge effect

Matsui et al.^[21] discovered that the accumulation of space charge in polymer dielectrics can significantly distort the electric field. When the peak of this distorted field surpasses the discharge threshold, dielectric breakdown occurs rapidly.

Notably, the breakdown theories mentioned above are often interconnected in solid dielectrics. Depending on the specific type of polymer and environmental conditions, the primary breakdown phenomenon and mechanism can vary. Instances occur where multiple breakdown phenomena coexist within the dielectric material. In reality, the breakdown of solid dielectrics typically arises from a combination of multiple influencing factors. Hence, when studying the breakdown of polymers, it is essential to flexibly apply various theories to comprehensively understand the underlying mechanisms.

2 Method for improving the breakdown strength of EP

As a common polymer dielectric, the breakdown strength of EP is influenced by extrinsic factors, such as fillers^[22–30], temperature^[27–29], impurities^[31–33], and curing processes^[8,34,35], as well as intrinsic factors, such as chemical structures^[11, 36–38]. Recently, scholars have made significant progress in studying the electric breakdown of EP. Current mainstream technologies for enhancing the breakdown strength of EP include incorporating nano and micro fillers into the EP matrix^[22–30, 39–45], reducing impurity levels^[31–33], optimizing curing processes^[34,35], and modifying the molecular structures of EP or hardeners^[36–38].

2.1 Adding fillers

Since nanodielectrics emerged in 1993, their remarkable dielectric properties have attracted considerable attention worldwide^[46]. Consequently, various nanofillers, such as nano Al_2O_3 , SiO_2 , TiO_2 , MgO , and multiwalled carbon nanotubes (MWCNTs), have been incorporated into polymers to produce nanocomposites with superior breakdown strength. As shown in Figure 2, there were fewer than 150 publications on the impact of nanohybrids on the electric breakdown of EP before 2011. However, over the past decade, research in this field has significantly expanded, indicating scholars' focus on creating EP/nanocomposites with satisfactory insulation properties.

Li et al.^[27,28] conducted experiments adding nano TiO_2 to the EP matrix and measuring the DC breakdown strength at different temperatures. The study revealed that the nanofiller content was

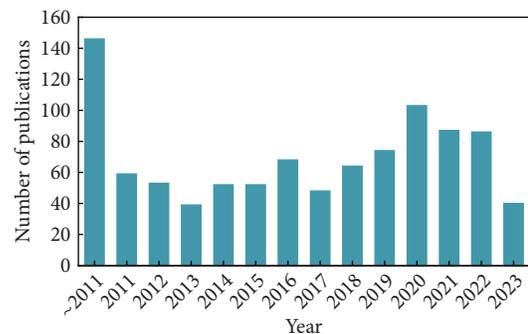


Figure 2 Publications on nanofillers and the electric breakdown of EP in recent years.

crucial in influencing the electric breakdown of the composites. As depicted in Figure 3(a), the breakdown strength of EP composites initially increased and decreased with an increase in the nanofiller content. It was observed that EP with 1 wt% nano TiO₂ fillers exhibited the highest breakdown strength of 122.31 kV, showing an improvement of 11.7% compared to pure EP. Additionally, breakdown experiments were conducted at various temperatures, as shown in Figure 3(b). The results indicated a gradual decrease in the breakdown strength of EP composites with increasing temperature. In addition to TiO₂, Li et al.^[39] incorporated MWCNTs into EP and found that 0.05 wt% MWCNTs resulted in satisfactory DC breakdown strength, as shown in Figure 3(c). Furthermore, he reported that samples with thinner thickness exhibited higher breakdown strength, as illustrated in Figure 3(d).

In addition to nano TiO₂ and MWCNTs, nano Al₂O₃ has been widely utilized to enhance the breakdown strength of EP. According to Preetha and Thomas^[40] and Mohanty and Srivastava^[41], incorporating Al₂O₃ into EP composites effectively increased the breakdown strength, with the optimal filler content being 5 wt%. Chen et al.^[43] delved deeper into the impact of different nano Al₂O₃ types on the DC breakdown strength of EP composites. By adding 2 wt% γ and α Al₂O₃ into the EP matrix and conducting DC electric breakdown experiments, they observed that the breakdown strength of pure EP was 71.3 kV/mm. For the γ and α Al₂O₃ composites, the breakdown strengths were 55.6 kV/mm and 76.1 kV/mm, respectively. Chen's study highlighted that not all the nano Al₂O₃ materials could enhance the breakdown strength significantly, as seen with γ Al₂O₃. Therefore, selecting suitable fillers is crucial for improving the overall insulation performance of EP composites.

Researchers have identified the interaction between the polymer matrix and nanofillers as crucial in influencing electrical insulation properties^[42,47,48]. They found that modifying the interaction between fillers and polymer molecules can significantly enhance the breakdown strength of nanocomposites^[47,48]. Consequently, many scholars have proposed various surface modification technologies for nanofillers to improve interactions with host molecules^[45,49,50]. Lü et al.^[50] utilized plasma fluorination to graft chemical bonds with F elements onto the surface of nano Al₂O₃, thereby enhancing the interaction between fillers and the EP matrix. In Figure 4(a), the X-ray photoelectron spectroscopy (XPS) analysis indicated a reduction in O1s in Al₂O₃ after plasma treatment, with a new peak representing F elements appearing in the fluorinated fillers. Moreover, compared to untreated nanofillers, fillers subjected to plasma treatment significantly enhanced the DC breakdown strength of EP. In Figure 4(b), the maximum DC breakdown strength for EP with plasma-treated nano Al₂O₃ fillers was approximately 42 kV, compared to only around 38 kV for the sample with untreated fillers. Furthermore, for nanocomposites with higher filler content, the untreated nanofillers worsened the breakdown strength of EP composites, whereas the composites with treated fillers maintained a satisfactory breakdown strength.

Although surface modification enhanced the interaction between fillers and the polymer, it also complicated the sample preparation process, a factor not favored by scholars and applications. To achieve satisfactory nanocomposites without the need for surface treatment of nanofillers, studies have added nano polyhedral oligomeric silsesquioxane (POSS) into polymers, creating composites with improved breakdown strength^[25,26,48,51]. As

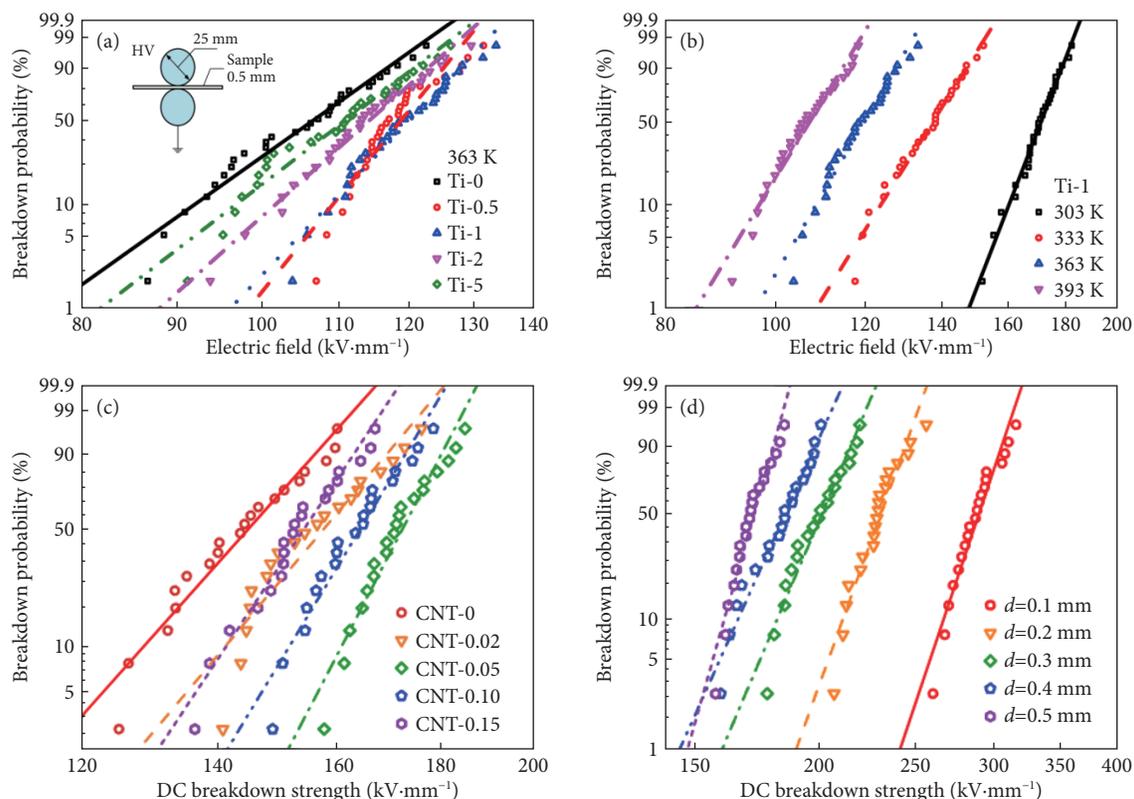


Figure 3 Weibull distributions of the DC breakdown strength of (a) EP/TiO₂ nanocomposites and (b) EP composites with 1 wt% nano TiO₂ at different temperatures (reprinted with permission from Ref [27], © 2022, AIP Publishing). Weibull distributions of the DC breakdown strength of (c) EP/MWCNTs nanocomposites and (d) EP composites with 0.05 wt% MWCNTs under different thicknesses (reprinted with permission from Ref [39], © 2023, The Authors).

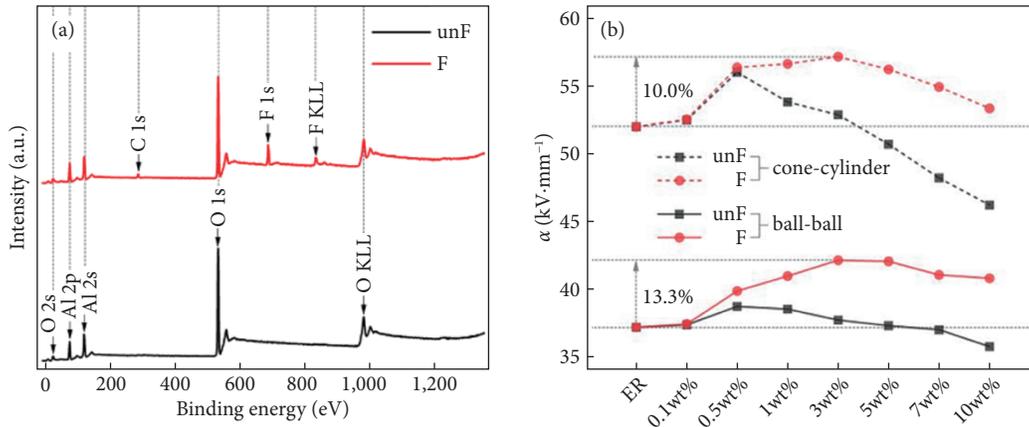


Figure 4 (a) The XPS spectra of nano Al_2O_3 fillers before and after plasma fluoridation. (b) The DC breakdown strength of EP composites. Reprinted with permission from Ref [50], © 2021, Society of Plastics Engineers.

POSS fillers have different chemical groups on the surface, they may have varying influences on the interfacial interaction and breakdown strength. For instance, Huang et al.^[25] incorporated octa(aminophenyl)silsesquioxane (a type of POSS) into EP to form EP composites. However, this research demonstrated that this nanofiller was ineffective in enhancing the breakdown strength of EP. In Figure 5, two types of POSS fillers (OG-POSS and ECH-POSS) are depicted. Aslam et al.^[26] found that both OG-POSS and ECH-POSS contributed to a greater DC breakdown strength. They reported that for neat EP, the DC breakdown strength was 201.65 kV/mm, while it increased to 231.32 and 236.07 kV/mm for the OG-POSS and ECH-POSS nanocomposites, respectively. Similarly, Li et al.^[51] discovered that adding 2 wt% OG-POSS effectively improved the AC breakdown strength of EP from 78 to 90 kV/mm.

Almost all reports concerning nano dielectrics with enhanced breakdown strength emphasize the importance of nanofiller interface effects in determining electrical performance^[47–53]. Moderate nanofiller levels have been observed to introduce deeper traps within polymers, thereby inhibiting charge transport in solid dielectrics and augmenting breakdown strength^[52]. However, an excess of nanofillers leads to a decline in the breakdown performance of epoxy composite materials. The surplus nanofillers precipitate overlapping interface regions among nanoparticles, thereby enlarging the volume of these interface regions and attenuating the inhibitory role of the interface effects on carrier migration^[52,53]. The carriers can accelerate in the enlarged interface regions, accumulate energy, and impact the excitation, collision, and ionization of polymer molecules. Consequently, this leads to a

diminution in breakdown performance.

Alongside nanohybrids, adding microfillers to the polymer matrix can enhance electrical insulation properties. A recent study by Yang et al.^[30] revealed that incorporating a large amount of microdiamonds can increase the breakdown strength of EP as shown in Figures 6. After adding 60 wt% microdiamonds to the EP, the AC breakdown strength of composites was improved by 42.6% compared to adding Al_2O_3 with same mass fraction as illustrated in Figure 6(e). Furthermore, Figure 6(f) also demonstrates that EP composites with 60 wt% microdiamonds exhibit excellent insulation properties, even at high temperatures. For the excellent insulation properties, Yang et al.^[30] interpreted that the microdiamonds repressed the charge accumulation at the polymer/filler interfaces, optimized the electric field, and suppressed the field distortion.

Despite the satisfactory electrical insulation properties of EP composites containing nanofillers or microfillers, their practical utilization presents challenges. Adding fillers increases the viscosity and density of EP^[54], and the uneven distribution of nanofillers or microfillers within the composites compromises their electrical and mechanical properties^[55]. Additionally, although nanofillers may scatter high-energy electrons, some electrons can still collide with polymer molecules. Consequently, a polymer matrix with inferior breakdown strength becomes a weakness in the electrical insulation of the composites, making them susceptible to discharges and even electrical breakdown.

2.2 Reduce impurity content

During the production of polymers, inevitable impurities exist,

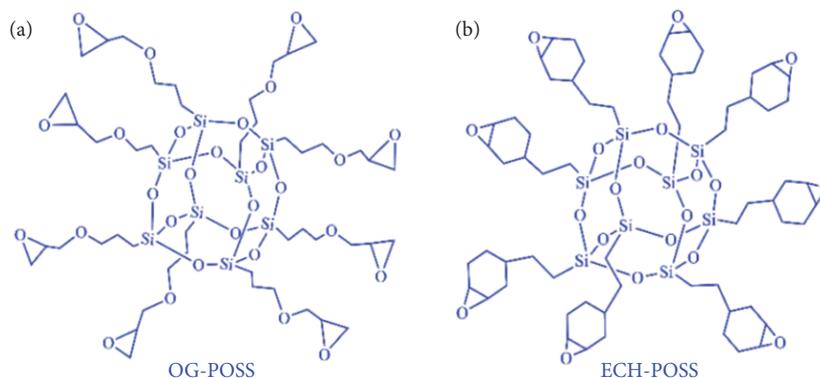


Figure 5 The structure of POSS. (a) OG-POSS and (b) ECH-POSS. Reprinted with permission from Ref [26], © 2021, The Authors.

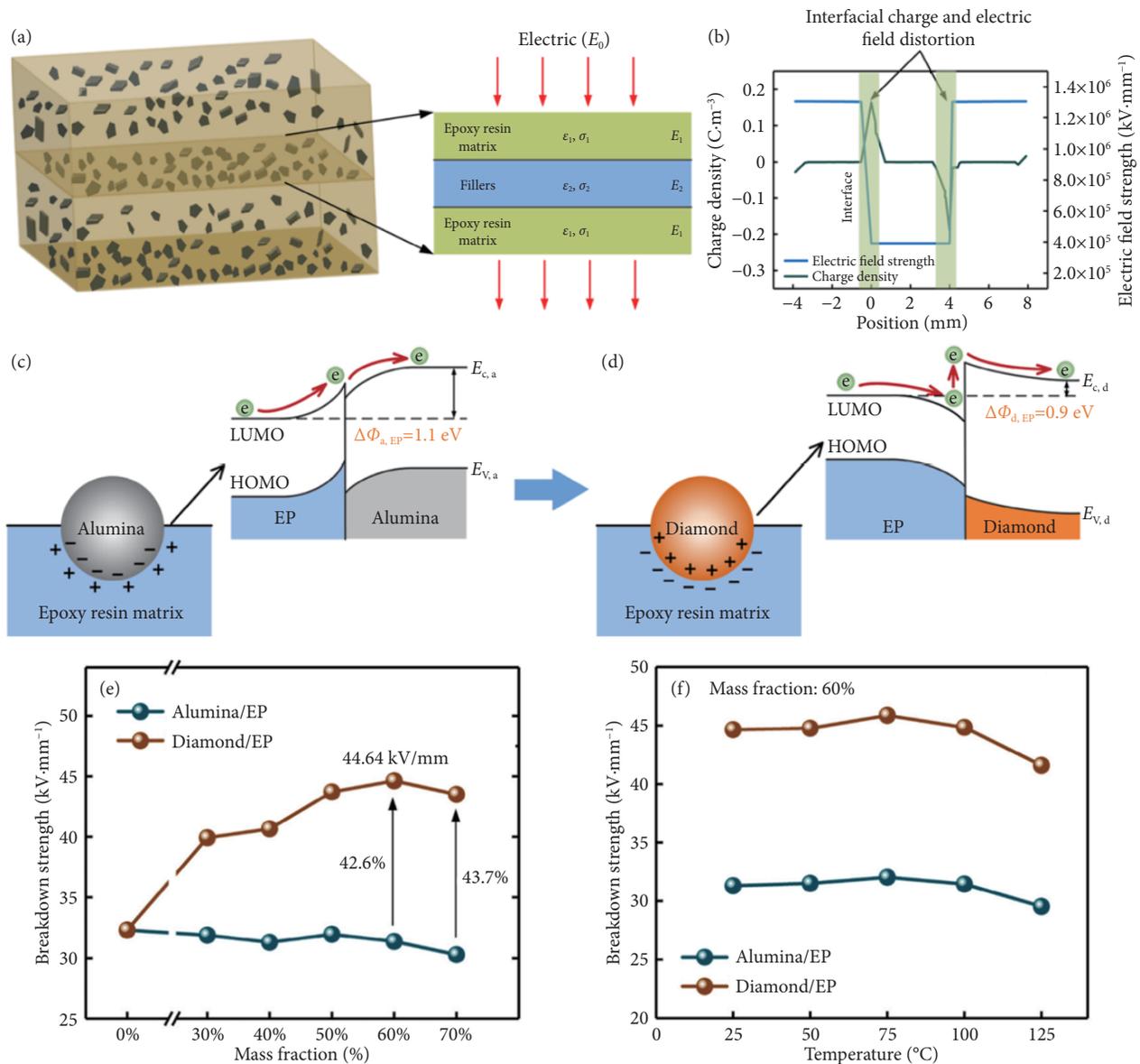


Figure 6 Design principles for diamond/epoxy composites. (a) Three-dimensional structure of epoxy composites. (b) Electric field distortion at the epoxy matrix–inorganic filler interface. (c) Energy band structure of the interface between the diamond and epoxy matrix. (d) Charge transport at the diamond–epoxy matrix interface. (e) Breakdown strength of EP composites. (f) Breakdown strength of EP composites at different temperatures. Reprinted with permission from Ref [30], © 2021, Elsevier Ltd.

such as metal particles, free impurity ions, and small molecule compounds^[31–33,56], and their influence on electric breakdown cannot be ignored. EP, produced from epichlorohydrin and bisphenol A in an alkaline environment (NaOH or KOH), inherently contains slight chloride, sodium, or potassium ions. Generally, these impurities can form insulation defects that weaken the properties of electric insulation. Scholars have conducted numerous experiments to study the influence of anions and cations on EP breakdown strength, yielding valuable conclusions^[31–33].

Du et al.^[31] introduced additional epichlorohydrin into EP to study the effect of chlorine-containing impurities on breakdown strength, as seen in Figures 7(a) and 7(b). Their findings indicated that increased epichlorohydrin and chlorine content led to decreased breakdown strength. Similarly, Liu et al.^[32] and Wang et al.^[33] discovered that sodium ions are detrimental to maintaining excellent breakdown strength in EP. As shown in Figures 7(c) and 7(d), the preparation of EP with varying sodium ion contents and

subsequent AC breakdown experiments revealed that introducing sodium ions weakened the breakdown strength of EP composites. Consequently, impurities can compromise insulation properties, emphasizing the necessity of reducing impurity content to maintain the satisfactory breakdown strength of EP.

2.3 Optimizing the curing process

EP is a thermosetting polymer that undergoes a curing process, during which it forms a three-dimensional crosslinked network with a hardener aided by an accelerator. This curing process can significantly impact the crosslinked structure of EP and consequently affect its macroproperties. Li et al.^[34] investigated the influence of accelerator content on the breakdown strength of EP. They varied the accelerator dosage at 0.1%, 0.2%, 0.5%, and 1% of the epoxy matrix, labeling the samples as DMD-1 to DMD-4. Figure 8 presents the electric breakdown results, analyzed using Weibull distribution. Notably, the DMD-3 sample with 0.5%

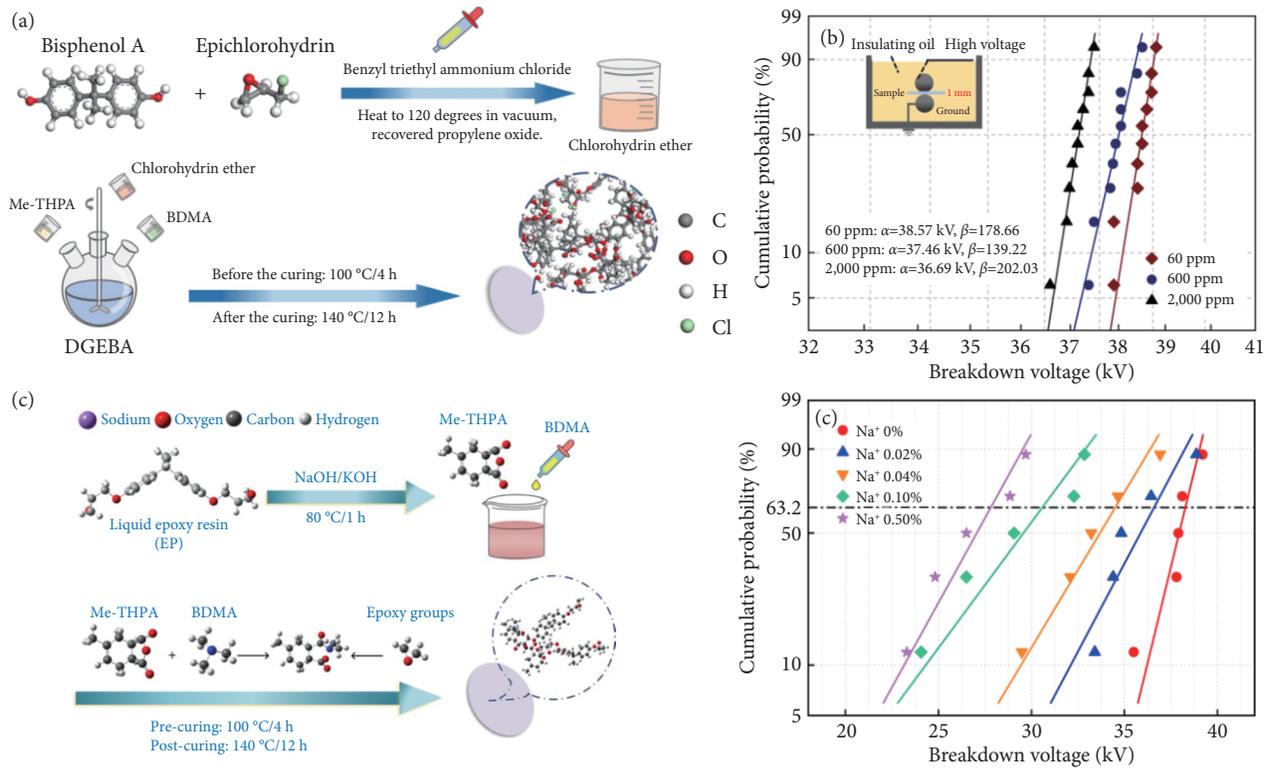


Figure 7 The influence of chloride and sodium ion on the AC breakdown strength of EP. (a) The preparation process and (b) breakdown results by Weibull distribution of EP with different chloride contents (reprinted with permission from Ref [31], © 2023, The Minerals, Metals & Materials Society). (c) The preparation process and (d) breakdown results by Weibull distribution of EP with different sodium ion contents (reprinted with permission from Ref [32], © 2022, The Minerals, Metals & Materials Society).

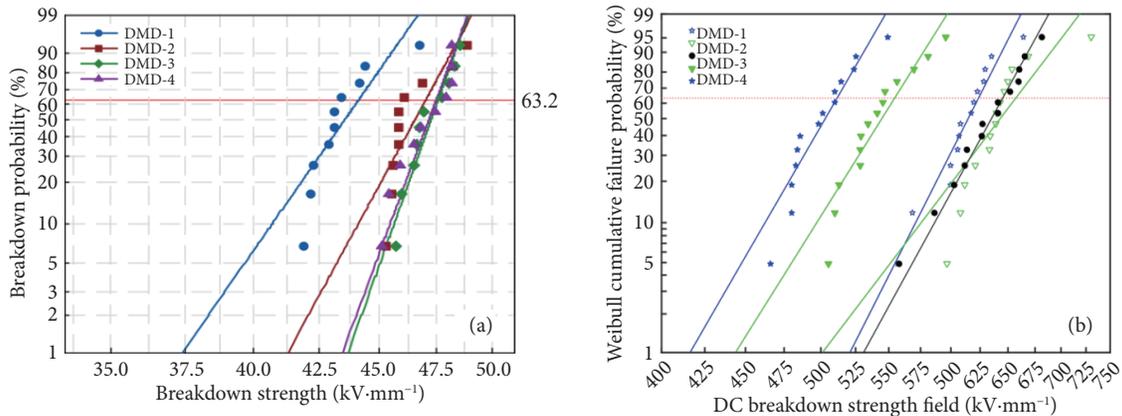


Figure 8 (a) The influence of the accelerator on the electric breakdown strength of EP (reprinted with permission from Ref [34], © 2023, IEEE). (b) The influence of hardener dosage on the breakdown strength of EP (reprinted with permission from Ref [57], © 2017, The Authors).

accelerator exhibited the highest breakdown strength of 47.57 kV/mm. Furthermore, this sample demonstrated the most stable breakdown data among all samples. Hence, the breakdown strength of EP can be optimized by carefully adjusting the accelerator content.

Similarly, studies have found that the dosage of the hardener can influence EP properties. Alhabill et al.^[57] altered the resin-to-hardener ratio to produce EP with different network structures. They observed that when the ratio was equal to 0.8, the EP displayed the lowest DC conductivity, the highest breakdown strength, and a higher glass transition temperature. Similar findings were reported by Wang et al.^[58], who observed that an inappropriate stoichiometric ratio between EP and hardener compromised the

dielectric properties of EP composites. Guo et al.^[59] prepared a mixed hardener comprising methyl hexahydrophthalic anhydride (MeHHPA) and hexahydrophthalic anhydride (HHPA) and studied the impact of the mixed ratio on breakdown strength. Figure 9, obtained from Ref. [59], represents the AC breakdown strength of the samples through Weibull distribution. Notably, when the hardener comprised 80% MHHPA and 20% HHPA, the sample exhibited a superior breakdown strength of 102.2 kV/mm. Thus, strict control over EP, hardener, and accelerator dosage is crucial for achieving high-performance epoxy materials.

In addition, the curing temperature and time can influence the electric properties of EP. As seen in Figure 10, Li et al.^[834] found that curing parameters, including procuring and postcuring time,

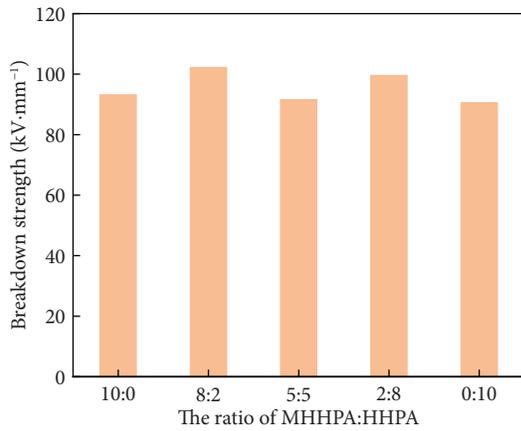


Figure 9 The influence of mixed hardeners on the electric breakdown strength of EP^[59].

could also influence the breakdown strength of EP. By tailoring the curing temperature, they comprehensively evaluated the DC conductivity, dielectric loss, and AC breakdown strength and selected the optimal curing regime for electric insulation.

2.4 Tailoring molecular structure

As thermosetting polymers, the microproperties of EP are intricately linked to the structures of the hardener and epoxy molecules. Researchers have made significant efforts to tailor

molecular structures to enhance the breakdown strength of EP.

2.4.1 Hardener molecular structure

Zhao et al.^[38] studied the influence of methyl and carbon-carbon double bonds in the anhydride molecule on the dielectric properties of EP composites. Figure 11(a) illustrates their selection of five different hardeners: methyl tetrahydrophthalic anhydride (MeTHPA), tetrahydrophthalic anhydride (THPA), HHPA, MeHHPA, and methyl nadic anhydride (MNA). EP samples were prepared using these hardeners, and breakdown experiments were conducted. The AC breakdown strengths of the EP samples, analyzed using Weibull distribution, are presented in Figure 11(b). Evidently, EP cured with MNA exhibited the lowest breakdown strength, while samples cured with MeTHPA demonstrated the highest insulation properties. Consequently, the researchers concluded that MeTHPA was the most suitable for EP in insulation systems among the various hardeners tested.

In addition to traditional hardeners, scholars have explored new hardeners to enhance EP breakdown strength. Luo et al.^[60] developed new hardeners named 1EZ and 2E4MZ, as depicted in Figures 12(a) and 12(b). They blended these hardeners with epoxy and fabricated samples to create high-performance EP polymers. The DC breakdown strengths of the samples crosslinked by the two hardeners were denoted as EP-1EZ and EP-2E4MZ, as shown in Figures 12(c) and 12(d). In Figure 12(c), the breakdown strength of EP-1EZ decreased with an increased content of the hardener. The sample cured with 1 phr 1EZ exhibited the highest

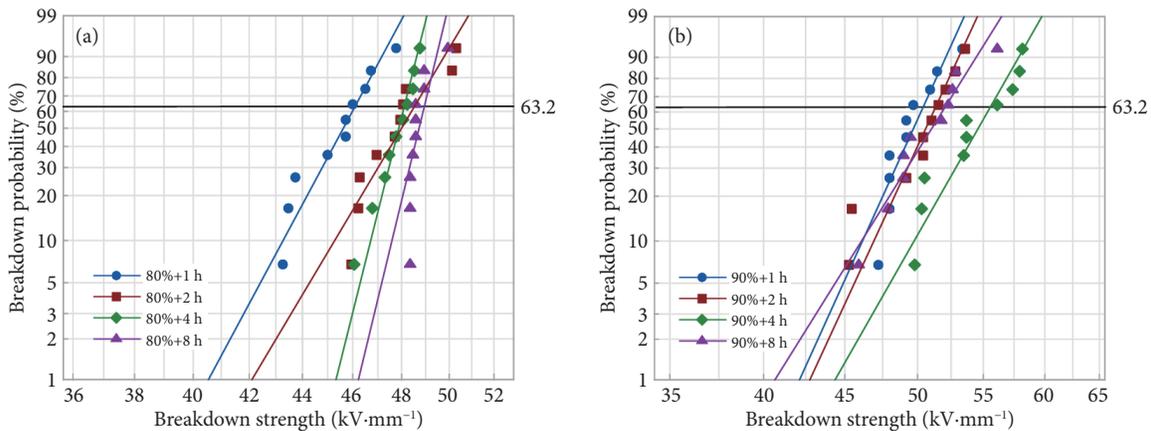


Figure 10 Effects of postcuring time on AC breakdown characteristics of samples with (a) 80% primary curing degree and (b) 90% primary curing degree less than 25°C. Reprinted with permission from Ref [34], © 2023, IEEE.

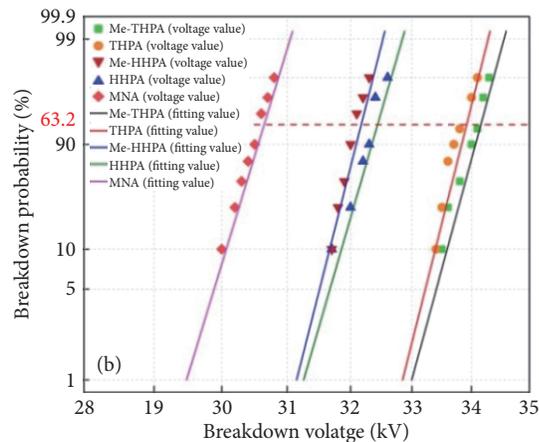
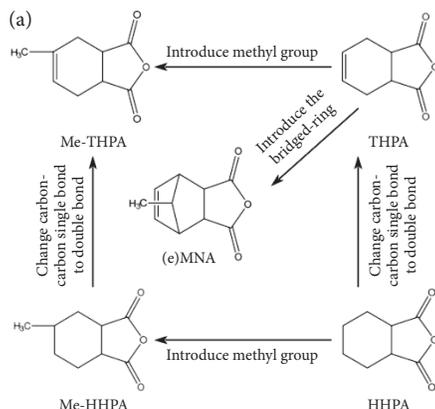


Figure 11 (a) The molecular structure of hardener and (b) breakdown strength of EP. Reprinted with permission from Ref [38], © 2021, IEEE.

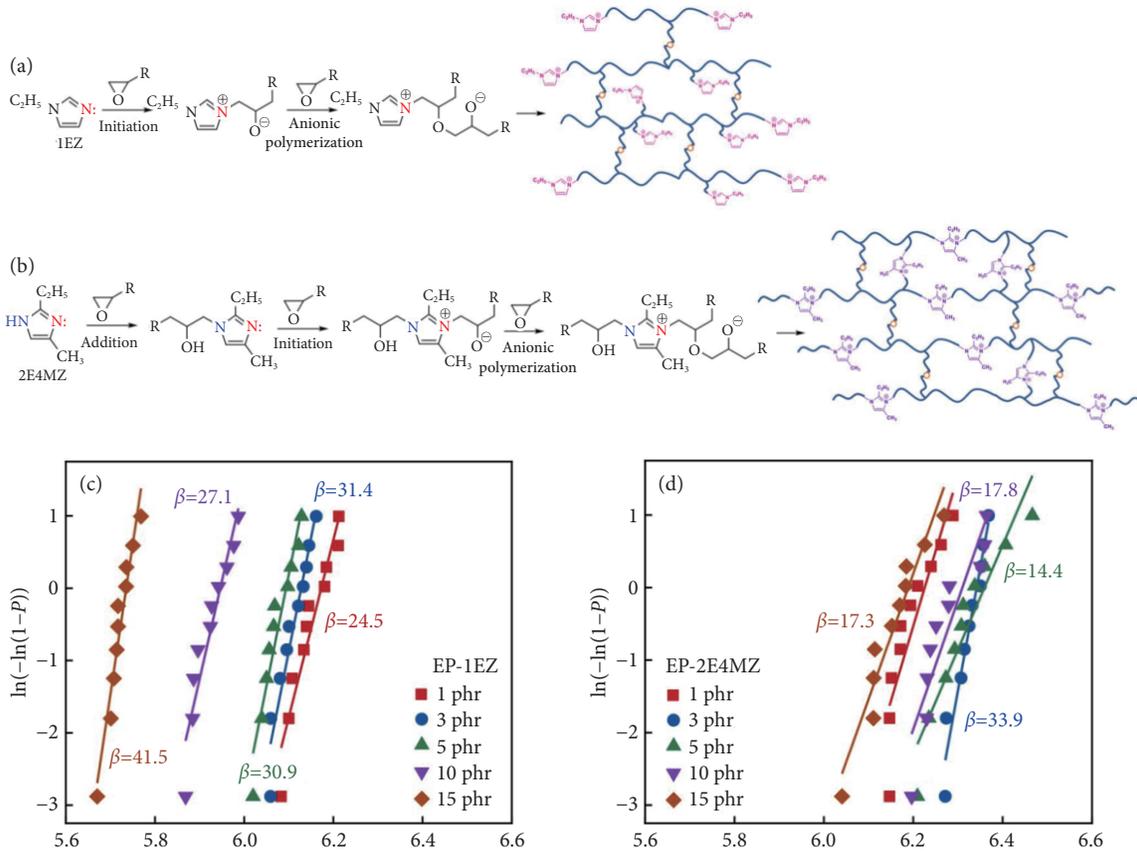


Figure 12 Reaction mechanism of EP/imidazole system. (a) Imidazole with pyridine-type nitrogen (1EZ) and (b) imidazole with both pyridine- and pyrrole-type nitrogen (2E4MZ). The breakdown strength by Weibull distribution of (c) EP-1EZ and (d) EP-2E4MZ. Reprinted with permission from Ref [60], © 2021, The Authors.

breakdown strength. Figure 12(d) presents the breakdown strength of the EP-2E4MZ sample analyzed through Weibull distribution. It was observed that the trend of breakdown strength was not a simple function of hardener dosage. When the dosage of 2E4MZ was below 5 phr, the breakdown strength increased with the hardener dosage. However, when the dosage exceeded 5 phr, the breakdown strength gradually decreased. Following these experimental results, they enhanced the breakdown strength and energy density of EP films. Mao et al.^[61] developed a fluorine hybrid hardener, as shown in Figure 13(a). The breakdown strength from Mao's research is displayed in Figure 13(b) and 13(c). The sample cured with S_{3FAN-C} outperformed the others at a thickness of 20 μm . The breakdown strength of the S_{3FAN-C} sample

at 0.1 mm was also competitive. Furthermore, Zhang et al.^[62] prepared fluorinated hardeners and produced EP with exceptional breakdown strength and heat resistance.

2.4.2 Epoxy molecular structure

A common method for tailoring epoxy molecular structure involves grafting other segments onto the epoxy group. This group exhibits strong reactivity and can easily undergo ring-opening reactions and react with other molecules under specific conditions. Gong et al.^[11] conducted a study in which they grafted hydroxy-terminated polydimethylsiloxane (HTPDMS) onto epoxy molecules and prepared samples with varying HTPDMS fractions to improve insulation properties in wet environments.

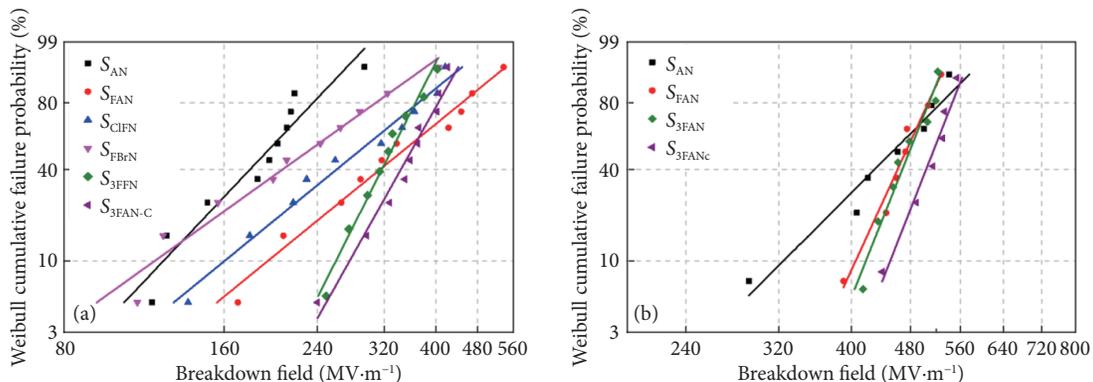


Figure 13 The breakdown strength by Weibull distribution of samples with thicknesses of (a) 0.1 mm and (b) 20 μm . Reprinted with permission from Ref [61], © 2022 Elsevier B.V. All rights reserved.

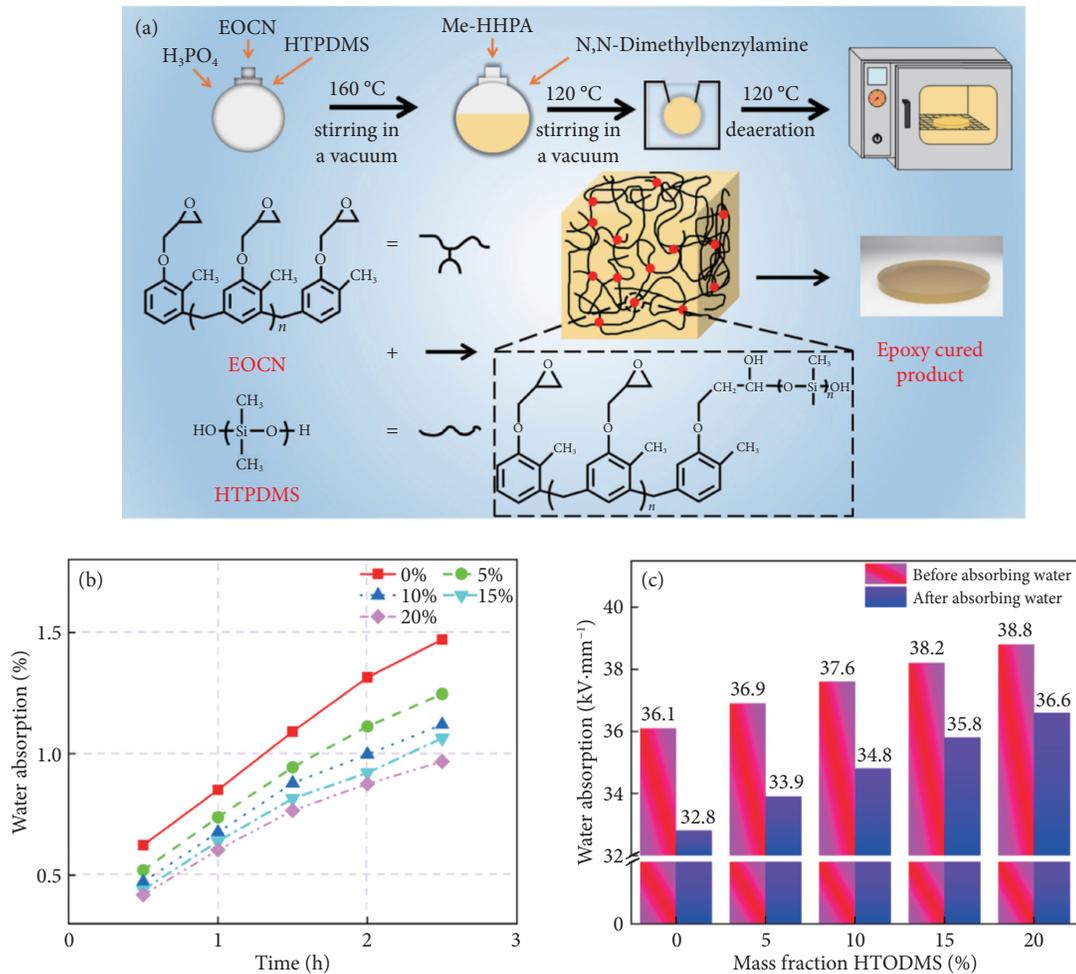


Figure 14 (a) The preparation of HTPDMS-modified EP. (b) The water absorption and (c) AC breakdown strength of EP modified by HTPDMS. Reprinted with permission from Ref [11], © 2021 Wiley Periodicals LLC.

Figure 14 shows the experimental results. After grafting HTPDMS, moisture resistance significantly improved. The water absorption decreased notably with higher HTPDMS fractions, as depicted in Figure 14(b). Furthermore, as illustrated in Figure 14(c), increased HTPDMS fractions led to superior breakdown strength in both dry and humid environments. They interpreted that the increased breakdown strength of HTPDMS-modified EP in humid environments resulted from the reduced moisture-induced electron migration influence. For the modified EP with 20% HTPDMS, the AC breakdown strength after absorbing

water was 36.6 kV/mm (increased by 11.6% compared to EP).

By tailoring hardener molecular structures, researchers have successfully developed fluorine hybrid EP with enhanced breakdown strength^[61,62]. Similarly, constructing high-performance fluorine hybrid EP by customizing epoxy molecular structures holds great potential. In a study by Yang^[63], EP with increased trap levels was created by grafting bisphenol AF (BPAF) onto epoxy molecules, resulting in a 22.29% improvement in breakdown strength. Furthermore, CF_3 groups in the molecules facilitated the formation of deep traps, effectively inhibiting charge transport

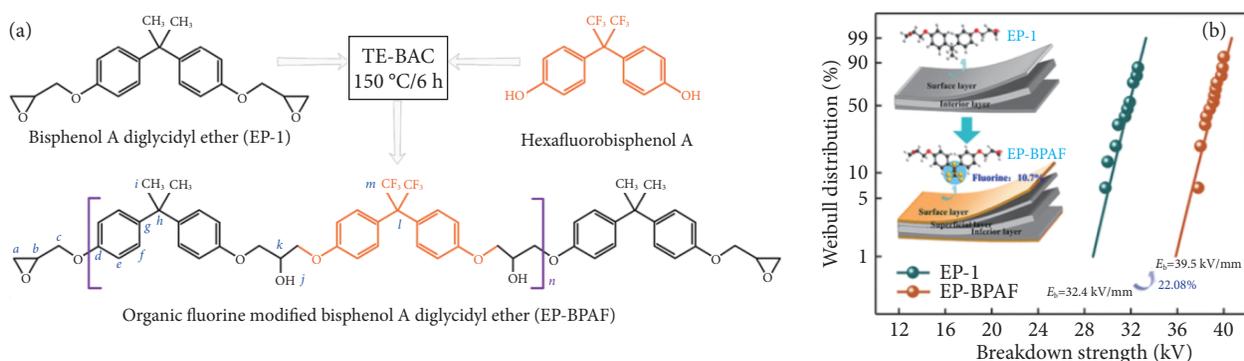


Figure 15 The preparation process and breakdown strength of fluorine hybrid EP. (a) The preparation process of fluorinated EP. (b) The AC breakdown strength of fluorinated EP. Reprinted with permission from Ref [63], © 2021, American Chemical Society.

within the dielectrics. As depicted in Figure 15, after grafting BFAF onto epoxy molecules, the AC breakdown strength of the fluorinated EP increased from 32.4 to 39.5 kV/mm. This enhancement demonstrates the effectiveness of tailoring epoxy molecular structures to achieve superior performance in fluorine hybrid EP.

Li et al.^[64] also reported similar findings in their research. They utilized surface fluorination at various temperatures to create fluorine hybrid EP with different F fractions. It was observed that higher fluorination temperatures led to greater grafting of CF_n groups into the EP matrix. Using density functional theory simulations, they discovered that CF_3 groups played a dominant role in increasing electron trap levels within the fluorinated EP, significantly enhancing breakdown strength. In Figure 16, after surface fluorination at 80 °C for 1 hour, the DC breakdown strength of the EP increased by 27.64%. In addition to the studies above, Sun et al.^[37] explored the construction of high-performance fluorinated

EP by grafting CF groups onto epoxy molecules as seen in Figure 17(a). This resulted in fluorinated EP with superior DC breakdown strength and degradable and self-healing properties. As shown in Figure 17(b), the DC breakdown strength of the EP was improved from 61.68 to 67.45 MV/m. Sun explained that the enhanced breakdown strength in fluorinated EP stemmed from the increased bond energy of the C–F groups, resulting in a higher trap level in the fluorinated EP.

3 Problems existing in the study on improving the breakdown strength of EP

3.1 The theoretical work on electric breakdown of dielectrics still needs improvement

Despite the significant contributions of numerous scientists in

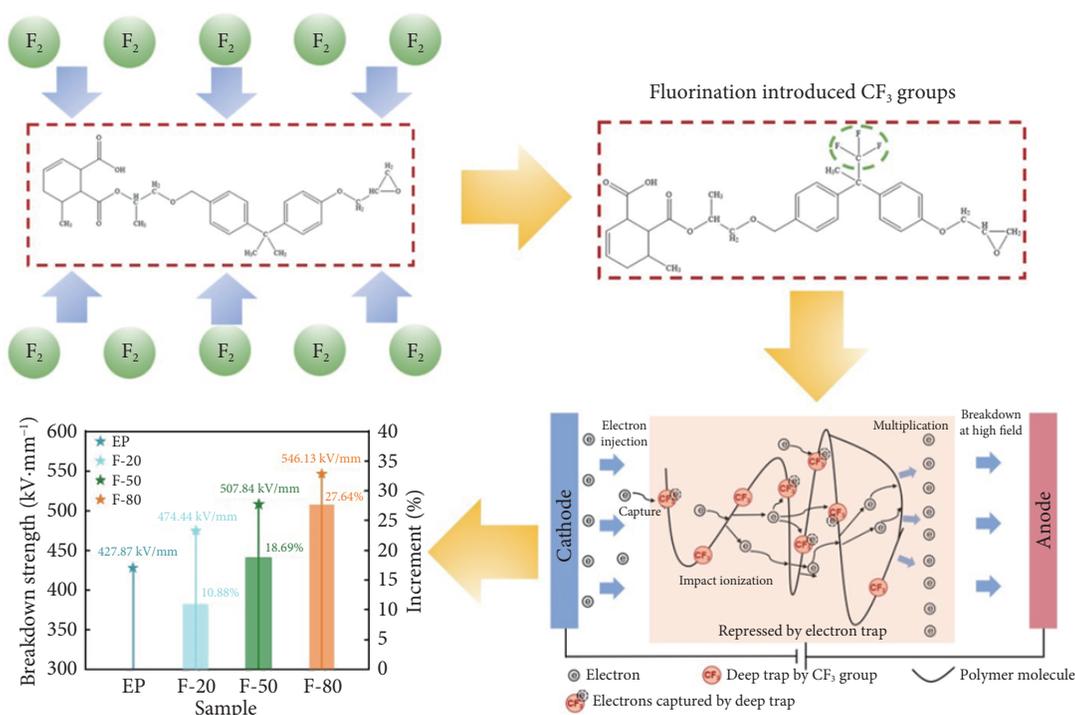


Figure 16 The mechanism of surface fluorination on improving the breakdown strength of EP. Reprinted with permission from Ref [64], © 2023, American Chemical Society.

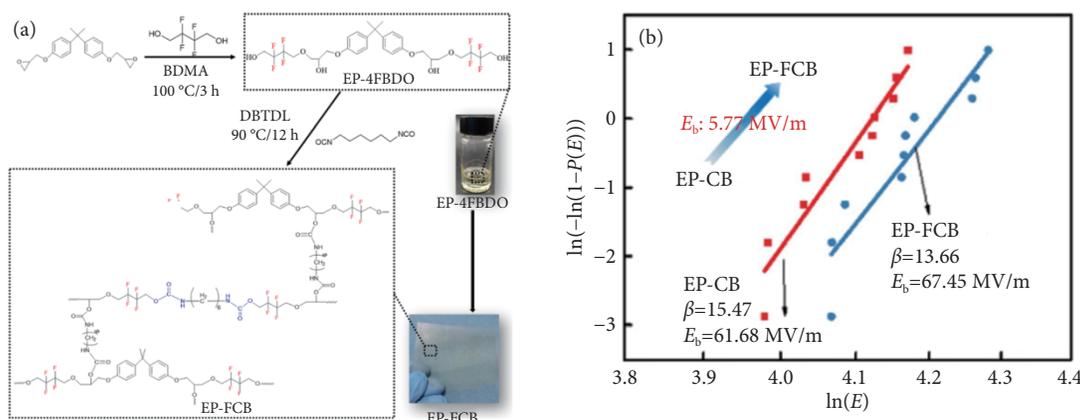


Figure 17 The synthesis process and breakdown strength of fluorinated EP. (a) The synthesis process of fluorinated EP. (b) The breakdown strength of fluorinated EP. Reprinted with permission from Ref [37], © 2023, The Royal Society of Chemistry.

exploring the theoretical mechanisms of electric breakdown^[12–21], a considerable gap remains in unveiling the mysteries of breakdown mechanisms. The charge dynamic processes of insulation materials are complex and influenced by numerous factors, posing challenges in proposing a theoretical model that can comprehensively encompass all electric breakdown phenomena. Additionally, the underlying relationship between long-term aging and short-term breakdown remains unclear. The impacts of temperature, sample thickness, and waveform of the source on the micro process of electric breakdown still require further elucidation.

3.2 Advanced technology for exploring high-performance EP lags behind

Currently, the predominant method of enhancing the breakdown strength of EP is through nanohybrid techniques^[22–28,39–45,49–54]. However, as mentioned previously, nanohybrid approaches have not significantly improved the unsatisfactory breakdown strength of epoxy matrices. The hosting polymer with unpleased insulation strength remains a weakness in nanocomposites, making them susceptible to discharge and breakdown^[61–64]. Thus, the key to unlocking high-performance EP lies in developing an EP matrix with superior breakdown strength. Molecule design technology is an effective method for optimizing high-performance polymers, demonstrating success in creating dielectrics with exceptional breakdown strength for energy storage capacitors^[65–68]. Nevertheless, the journey to developing EP with enhanced breakdown strength through molecule design is just commencing. Furthermore, rapid advancements in artificial intelligence (AI) and big data technology have introduced new perspectives on material development^[69,70]. In the future, leveraging AI-assisted molecular design based on existing databases could enable the customization of high-performance materials, followed by the synthesis of innovative dielectrics.

3.3 The development of environmentally friendly EP has shortcomings

With growing awareness of environmental issues, the demand for developing environmentally friendly electrical materials in power generation is increasing. The evolution of environmentally friendly EP includes advancements in eco-friendly, self-healing, and degradable EP. While some studies have synthesized environmentally friendly EPs using itaconic acid, their properties, particularly their electrical insulation performance, still lag behind traditional EPs^[71]. The objective of developing self-healing materials is to prolong their service life and minimize the impact of insulation damage on power supply^[37,72–73]. However, enhancing the dielectric strength of EP to bolster their resistance to electrical damage can reduce insulation damage occurrence, thereby limiting insulation faults. Consequently, the immediate demand for self-healing EP is not pressing. Progress in degradable EP technology contributes to EP recycling and reduces carbon emissions during epoxy production. In recent years, scholars have focused on the mechanical and thermal properties of degradable EPs^[74–77]. Recently, some scholars reported degradable EPs with high-performance electric insulation^[78–80]. However, the evolution of degradable EPs with satisfactory insulation properties for application remains a critical area for further development.

4 Conclusions and prospect

Based on the review of studies aimed at enhancing the breakdown strength of EP, significant progress has been made, providing

valuable insights and a solid foundation for future research. In our assessment, the key areas for the development direction of high breakdown strength EP are as follows:

(1) Leveraging existing polymer breakdown theories, experiences, and molecular design technology to precisely design, evolve, and manufacture EP molecules and products with superior breakdown strength.

(2) Optimizing the preparation processes for EP by exploring new synthetic technologies and enhancing existing methods to minimize industrial impurities during EP synthesis and production. This approach will improve the breakdown strength of EP.

(3) Emphasizing environmental considerations and the imperative for sustainable development by focusing on developing environmentally friendly EP. Concurrently, targeted efforts will be directed toward enhancing the electrical insulation strength of these new products to meet the requirements of the electrical insulation sector. Ultimately, this will produce high-performance, environmentally friendly EP for advanced electrical equipment and electronic devices.

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Additional information

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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