High Voltage

Review Article

Progress in and prospects for electrical insulating materials

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Abstract: The performance of electrical equipment and devices is determined to a great extent by the properties of their insulating materials. In power systems and electrical devices, insulating materials have to work in extreme circumstances that can include high temperature differences, intense radiation, and strong electric fields. Such conditions demand high-quality insulating materials with superior electrical, thermal, and mechanical properties as well as resilience to other environmental stresses. This study focuses on advances in insulating materials since the early 20th century and reviews the many developments in their properties and applications, including electric breakdown strength, thermal conductivity, temperature resistance, corona resistance, and specific energy storage in dielectrics. Some research progress on other properties is also covered, such as non-linearity and radiation resistance. Investigations into the properties of insulating materials can greatly assist in understanding their interface effects and composite structures, which in turn is helpful for discovering methods to improve the performance of electrical devices. Future directions for research are proposed to guide new investigations and support the development of even better insulating materials.

1 Introduction

At the beginning of the 20th century, insulating materials were mainly natural substances, such as mica, rubber, marble, and pitch. These dielectrics were produced directly from plants or from minerals or other inorganics, and they had poor electrical properties. Their low resistivity and breakdown strength limited the insulation capabilities and power levels of electrical equipment as well as the use of dielectrics in specialised electrical instruments. These natural materials are regarded as the first generation of insulating materials.

In the 1950s, with global economic growth, there was a steadily increasing demand for electric power [1], leading to rapid expansion in the areas of electrical equipment and insulating materials. With the development of polymers, the insulator market was flooded with synthetic materials such as resins, insulating varnishes, impregnated insulating fibres, and composites [2]. These materials had better electrical properties than first-generation insulating materials and were easy to produce, so they became widely used in all kinds of equipment, even in extreme conditions. Polymeric materials hence became the second generation of insulating materials.

The key advantage of polymers was that their properties could be adjusted by changing their chemical composition and molecular structure. As progress was made in characterising these properties, designing molecular structures, and studying polymers' specialised properties, research on polymer insulating materials was highly successful. Manufacturers could produce insulators in various categories, meet high quality standards, and tailor the materials to meet a wide range of specifications.

However, as the level of power systems continued to grow, polymers encountered a bottleneck. Problems of space charge accumulation, aging, pollution flashover, and polarity reversal in combined fields became important thresholds [2]. Moreover, the rise of renewable energy sources such as solar, wind, tidal, and nuclear made it imperative to explore the properties of materials that could be used in these specialised environments. The demand for better insulating materials called for a third generation: nanodielectrics, which appeared in the 21st century. Nanodielectrics are prepared by adding certain nanoscale fillers into a polymer matrix to yield better electrical, thermal, and mechanical properties. When the particle pretreatment methods and the content or category of fillers are adjusted, nanodielectrics tend to have greater breakdown strength as well as better high-temperature resistance and space charge suppression.

Progress and developments in insulating materials began in the early 20th century, spurring the creation of new electrical equipment. Novel applications of this equipment in turn demanded the development of better materials. For example, the use of trichlorodiphenyl in insulating oil increased the specific energy storage of capacitors. However, trichlorodiphenyl is harmful to humans and hence needed to be replaced by another dielectric liquid [1]. Replacement of SiC non-linear resistors with ZnO varistors greatly increased the non-linear coefficient and surge energy absorption capacity of insulators.

Clearly, the performance of electrical equipment and devices is greatly determined by the properties of their insulating materials. This paper presents evidence to show that more attention needs to be focused upon improving the properties of insulating materials and expanding the range of their applications.

2 New challenges for insulating materials

Electrical insulating materials are the foundation of electrical equipment. Hence, progress with insulating materials determines the future development of power systems. A new generation of power systems requires novel developments in insulating materials within several fields:

(a) The development of solar, wind, tidal, and nuclear energy necessitates improvements in capacitors to enable high specific energy storage.
(b) The construction of ultra-high and extra-high voltage power lines demands that insulating materials work under conditions of combined voltages, space charge accumulation, and polarity reversal.
(c) The widespread demand to produce sustainable and environmentally friendly technology means using renewable materials and decreasing negative environmental effects during the production, manufacturing, application, and recycling of insulating materials.

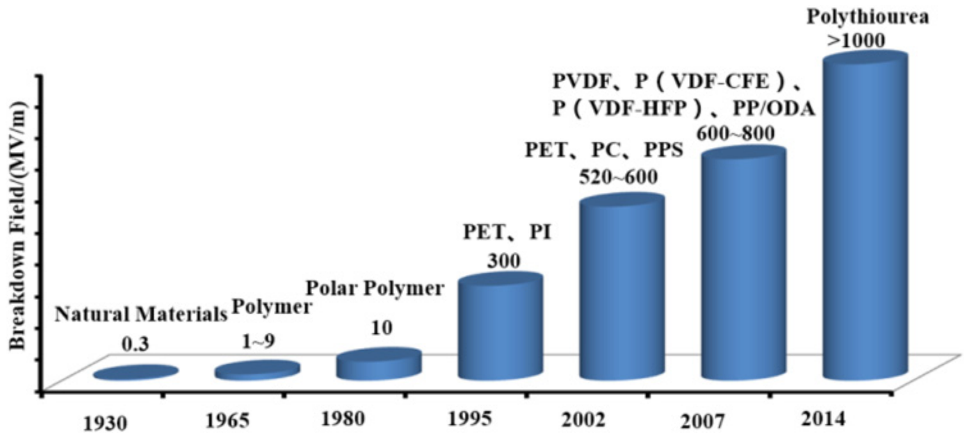

Fig. 1 *Progress in the electric strength of insulating materials [9–22]*

(d) The development of high-speed rails and electric vehicles brings the challenges of improving motor capacity and braking.
(e) Advancements in spacecraft call for the development of insulating materials with greater radiation resistance and better space charge dispersion.

Electrical equipment experiences numerous common and recurring problems, including space charge accumulation in DC current cables, surface corona with high-speed motors, surface flashover in gas-insulated switchgears, corona with insulators in severe environments, wide temperature variations between day and night at high altitudes, and strong radiation and heat accumulation during photovoltaic power generation in spacecraft [1]. Such problems have increased the urgency to develop insulating materials with better electric strength, capacity, conductivity, temperature resistance and so forth.

3 Progress with insulating materials

The past two decades have witnessed many valuable discoveries and improvements in insulating materials. Their electrical, thermal, and mechanical properties have been enhanced, expanding their application and offering the possibility of further improvements for electrical equipment.

3.1 Materials with high electric strength

Dielectrics always break down at submicron or nanoscale weak points, such as the interface between the electrode and the dielectric, or the various interface regions within the dielectric. It is possible to influence the breakdown strength of dielectrics by adjusting the shape of the electrodes, the smoothness of the samples, and the combination of filler and matrix [3–8].

Natural dielectric materials have many internal defects that limit their breakdown strength. As the power levels of systems have increased, polymers have emerged as the materials best able to meet the corresponding demands. However, the breakdown strength of polymers can be affected by many factors. First, polar polymers have higher electric strength, as the polar groups can trap electrons. Second, increasing the molecular weight decreases the degree of crystallinity and the size of spherulites, making it more difficult to build a discharge channel inside the polymer and thereby leading to higher breakdown strength. In addition, research has shown that after a polymer is crosslinked, its melting point increases, thus increasing its breakdown strength (especially in high-temperature regions). Overall, the development of polar polymers has increased the electric strength of insulating materials.

It is possible to reduce the defects in a polymer film by adjusting the catalyst content of the polymer's precursor. Thus, the breakdown strength of a polymer can be enhanced using different production methods. For example, the breakdown strength of polyester (PET) films has increased by more than 200 MV/m since the late 20th century. Fig. 1 illustrates some of the developments in creating insulating materials with high electric strength. Differences in precursors and reaction conditions lead to various structures and defects in polymers, and correspondingly different electrical and mechanical properties. In this way, film polymers with higher breakdown strength can be obtained. For example, high-quality films with uniform thickness and few surface defects can be obtained using polythiourea; a polythiourea film only 1 μm thick can withstand a field of over 1000 MV/m [21].

Factors such as interface properties, particle dispersion, and filler–matrix compatibility influence the breakdown strength of composites. Fig. 2 shows the Weibull distribution in breakdown strength of crosslinked polyethylene (XLPE) composites. XLPE with nanosized SiO_2 particles has a higher breakdown strength than either pure polymers or composites with microsized particles. Consequently, the breakdown properties of composites can be improved by modifying filler surfaces, adjusting the interfaces between particles and matrices, and improving production methods.

3.2 Insulating materials with high thermal conductivity

High operating temperature and heat accumulation cause the temperature of equipment to increase, which is the main factor in the degradation of insulation systems. Temperature fluctuations inside electrical equipment can lead to looseness between devices, reducing their lifespan. It is therefore important to improve the thermal properties of insulating materials [1].

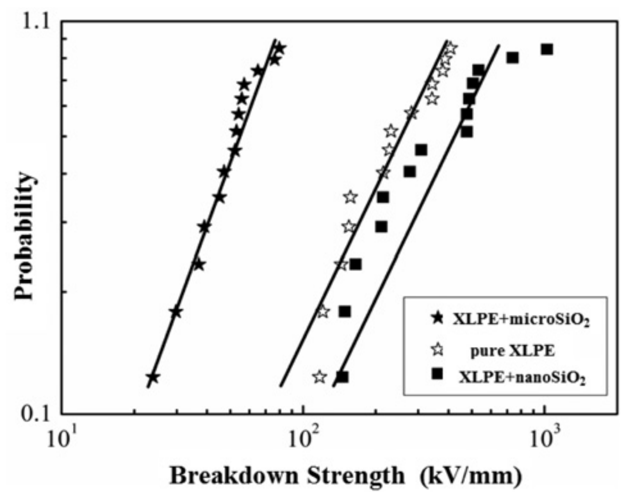

Fig. 2 *DC breakdown strength of XLPE composites filled with microsized and nanosized SiO_2 [23]*

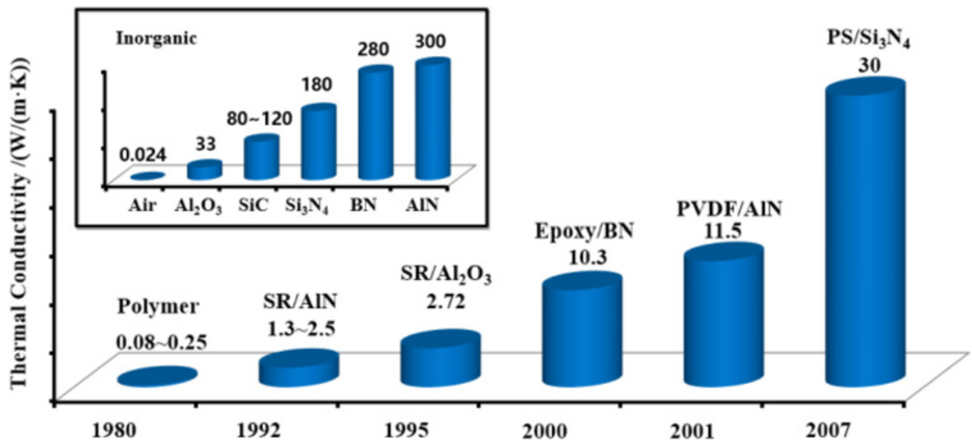

Fig. 3 *Progress in developing insulating materials with high thermal conductivity [24, 31–44]*

Metals conduct heat through free electrons, whereas polymers conduct it through the vibration of atoms, groups, and chains – that is, through phonons. Structural features such as the number of free electrons, lattice characteristics, dipolar polarisation, the number of polar groups, molecular weight, the degree of crosslinking, and the degree of orientation influence the thermal conductivity of insulating materials. For example, polystyrene can conduct heat through its large number of electron conjugation bonds, increasing its thermal conductivity to 30 W/m·K.

The main method for improving the thermal conductivity of insulating materials is to add nanoparticles into polymers. Changing the type, content, and surface modification method of the nanoparticles can enhance a nanocomposite's thermal conductivity. Surface modification is used to improve the dispersion and combination of particles within a matrix [21]. Although metallic fillers have high thermal conductivity, thermal conductivity paths can only form when the filler content is high enough [24, 25]. This requires a certain density of metal powder [26–29]. Hence, filling polymers with metal powders is not an effective way to improve their thermal properties. The main advances in developing insulating materials that have high thermal conductivity in applications have involved: (i) developing new kinds of thermally conductive fillers, such as aeolotropism fillers; (ii) creating methods of dispersing nanoscale fillers into conductive macromolecule polymers; (iii) identifying methods of doping fillers with different grain sizes; and (iv) optimising the moulding technology used to create insulating materials [30].

Fig. 3 illustrates the research progress made in developing insulating materials with high thermal conductivity. Various kinds of fillers – such as Al_2O_3 [28], SiC [29], Si_2N_4 [26, 27], BN [31–33, 45], BNNT [34, 35] and AIN [36, 46] – have been modified and added into different matrices, including epoxy [32, 36–38], polyamide [39–43] and silicone rubber [46]. Through careful selection of the size and content of fillers, the thermal conductivity of the polymer can be improved by several hundredfold.

3.3 Insulating materials with high heat resistance

At high temperatures, insulating materials' state, density, impurity mobility, charge transport, and degree of thermal degradation change significantly. Impurities within these materials lead to severe chemical reactions, such as oxidation, ozonisation, and hydrolysis, causing degradation. The heat resistance of insulating materials is therefore an important factor in applications.

Polymers with high heat resistance are generally composed of aromatic rings or heteroaromatic rings on backbones containing strong chemical bonds and have the molecular structure of rigid chains. By adjusting the molecular structure of polymers, we can improve the thermal properties of insulating materials. Fig. 4 illustrates such developments in insulating materials with high heat resistance. In the 1950s, organosilicon and organofluorine polymers were the primary heat-resistant insulating materials of choice. From the 1960s, aromatic and heterocyclic polymers with high temperature resistance were developed and put into broad use. In the 1970s, researchers turned to studying the processing characteristics and applications of insulating materials – for example, to increase their heat resistance by reducing the active impurities and introducing heat-resistant fibres. Subsequently, the types of heat-resistant materials grew, and their properties were comprehensively studied. The rigidity of polymer molecules can be increased by crystallisation, crosslinking and doping using fillers with rigid molecular structures, thus enhancing the polymer's heat resistance. For example, polyimide incorporated with silicone can withstand temperatures up to 330°C.

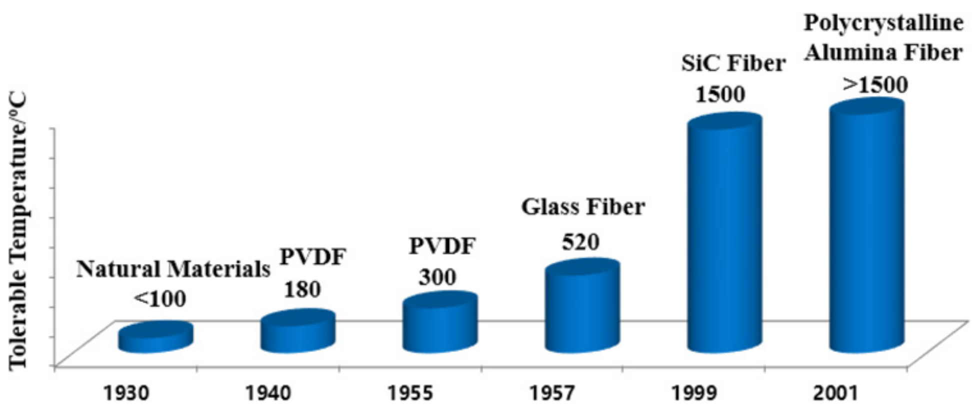

Fig. 4 *Progress in insulating materials with high heat resistance [22, 24, 29–33, 36, 44, 47]*

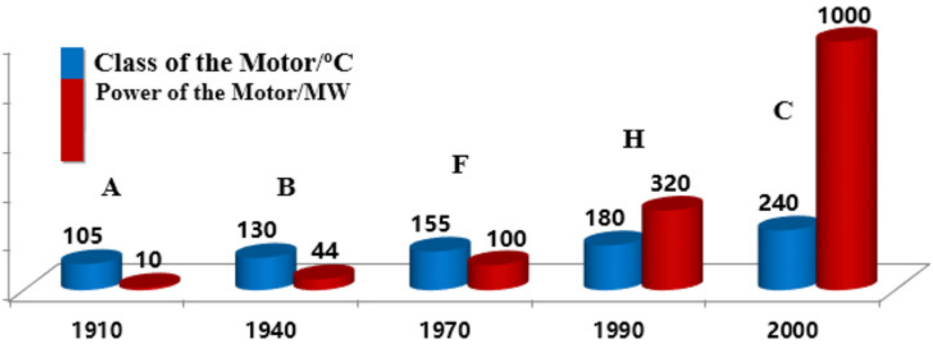

Fig. 5 *Development of motors in terms of temperature and power [1, 22, 24, 30, 31, 35, 36]*

Along with the increasing heat resistance of insulating materials, the upper temperature limit of motors increased from class A (105°C) to class C (240°C). At the same time, motor power increased tenfold from 10 to 1000 MW, as shown in Fig. 5.

3.4 Insulating materials with low-temperature resistance

When materials are used at very low temperatures, the random thermal motions of the atoms and molecules in the materials slow down, the dipoles freeze, the structure of the materials tends to become ordered, and the density and mobility of carriers decrease. As a result, the properties of insulating materials change significantly. For instance, the thermal conductivity and thermal expansion coefficient decrease, the tensile and compression properties increase, the dielectric loss increases, and the AC breakdown strength increases. These trends are presented in Table 1.

Fig. 6 shows the growing AC breakdown strength of insulating materials at very low temperatures. With the development of materials that work better at low temperatures, the electrical, thermal and mechanical properties of insulating materials have improved since the 1980s. For example, the breakdown strength at 77 K increased nine times, from 30 MV/m with cable paper to 280 MV/m with high-density polyethylene.

To improve the mechanical properties of insulating materials at low temperatures, it is important to increase their elongation percentage and tensile strength [22]. For films, water absorption is a severe problem, since films absorb water from the air, which then degrades the polymer's electrical properties. Hence, it is vital to improve the production of films with low water absorption so as to reduce the impact of water. Current trends in the development of insulating materials working at low temperatures are: optimising the insulating system to reduce the distortion field; expanding the application of insulating materials that have good properties at very low temperatures; and researching materials with low dielectric loss and low capacitivity, such as superconductors and superconducting magnets.

3.5 Insulating materials with high corona resistance and tracking resistance

Insulating materials working outdoors or in moist and polluted environments become corroded [55–59] and form tracks that conduct carriers, accompanied by the formation of organic semi-conductive materials, carbonisation, and graphitisation at the materials' surface. Spark discharge is the main aging factor causing these surface conducting tracks. The value of the spark discharge current is between partial discharge and arc discharge. The most useful method for reducing the surface tracks in insulating materials is to weaken the oxidation function and suppress the formation of a dry zone. There are several ways to increase these materials' tracking resistance: (i) reduce the number of benzene rings and groups, as they are easily oxidised; (ii) use polymers that have a low pyrolysis rate and produce few conjugated systems; (iii) develop new materials with low surface energies; (iv) prevent the formation of a continuous water film at the material's surface; and (v) suppress the current at the material's surface. In addition, doping polymers with nanofillers using crystallised water can reduce tracking because the nanofillers are oxidised during spark discharge and thereby clear the free carbon at the material's surface. The crystallised water inside the composites is then released, flushing the carbon granules over the material and suppressing surface tracking.

Table 1 Trends in polymer properties at low temperatures [22, 48]

Mechanical property	Trend	Electrical property	Trend
Tensile strength	Increase	Relative permittivity	Decrease
Elastic modulus		Dielectric loss	
Elongation percentage	Decrease	Volume resistivity	Increase
Impact strength	Increase or decrease	AC breakdown strength	

The corona resistance of insulating materials affects the lifetime of electrical devices, especially in PWM inerter-fed motors [60–63]. The main methods for improving a material's corona resistance are optimising the insulation system of the device, reducing the electric field distortion at the electrode, suppressing the accumulation of space charge, developing flaky materials, coating the surface of the device with semiconducting materials, and adding inorganic nanoparticles to the polymer [4, 64–74]. Research has shown that around the nanoparticles doped into polymers are many orderly arranged spherulite structures [8, 75, 76]. Such structures can prevent the development of electric erosion and thereby help polymers resist corona and partial discharge. For example, a small amount of TiO_2 nanoparticles can improve the corona resistance of epoxy resin (see Fig. 7). TiO_2 nanoparticles can improve the distribution of electric fields in polymers and make the inner field tend to be uniform. With high conductivity, TiO_2 nanoparticles can form a shielding layer to capture and conduct charges, thereby increasing the composite's corona resistance. The depth of electrical erosion was the mean value of five depths obtained with a probe (diameter of 1 mm).

Thinner and more uniform films are required to make devices smaller and more efficient. However, finding methods to produce thin, uniform films with high corona resistance is a challenge [77, 78]. Kapton®FCR films can resist corona discharge for over 10,000 h in the same test environment. However, methods for producing corona-resistant films still need improvement.

Fig. 8 illustrates the developments that have been achieved in creating insulating materials with high corona resistance. In 50 years, the corona discharge resistance time and applied electric field of insulating materials have greatly increased. In the 1960s, molecules and other groups in natural materials such as mica sheets were chaotic and could withstand a field of 10 MV/m for less than 10 h. Inorganic oxides have better thermostability, inoxidisability and ultraviolet shielding. Incorporating these with nanoscale inorganic oxides diffuses the space charges, optimises the charge distribution inside the polymer, and increases the

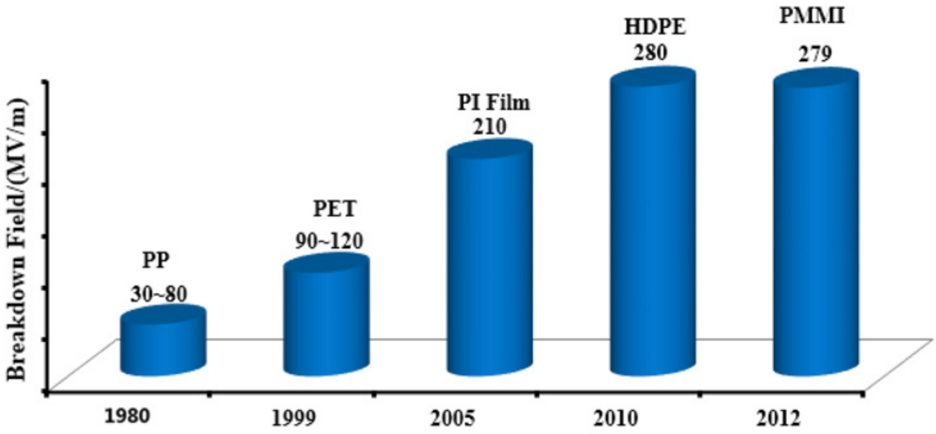

Fig. 6 *Progress in AC breakdown strength of insulating materials at liquid nitrogen temperature (77 K) [48–54]*

insulating material's corona resistance. Elastic mica tape with nanofillers has a more ordered structure than plain mica and can resist corona discharge for over 200,000 h.

Sometimes, insulating materials have to work in very strong fields and therefore discharge more easily. Improving the dispersion of nanofillers and importing films with fewer defects can extend the films' lifetime by 22 times (to 5500 min) during corona discharge in a higher field of 40 MV/m [79].

3.6 Insulating materials with high energy storage density

Clean energy sources such as solar, wind, and tide, as well as hybrid electric vehicles, require the development of smart, highly efficient power grids. We also urgently need to increase the current for large loads in military and electrical systems. Hence, we require devices with greater energy storage density, and this in turn necessitates further developments in insulating materials [83–86].

Increasing the permittivity and electric strength of linear insulating materials can boost their energy density to 0.7 J/cm^3. In contrast, the permittivity of non-linear materials can be increased to an optimal value to prevent untimely intensities of polarisation saturates in low fields. The main method of heightening the energy storage density of non-linear materials is to enhance their electric strength. Exploring antiferroelectric materials with higher energy storage density is another important way.

Natural materials first used as insulators, such as plants and minerals, have low energy density. With the development of polymers, the energy density of capacitors increased tenfold. Subsequently, doping metal nanoparticles into polymers and obtaining nanocomposite films of micron thickness increased the energy density over a hundredfold. To increase the energy density of materials, we need them to have high electric fields and permittivity. However, it is difficult to increase these two dielectric properties simultaneously. Polymers with high electric fields and low energy loss tend to have low permittivity (2–4) [83]. One solution is to combine them with ceramic. By controlling various filler factors, including size, dispersion, content, and interfaces, we can influence the dielectric properties of ceramic-polymers [87–91]. However, finding a way to achieve low dielectric loss while raising the dielectric constant is still a challenge in high-voltage and high-energy-density capacitor applications.

Fig. 9 shows the energy-density values of various insulating materials in experiments and applications. In the 1960s, the technique for producing mica paper was poor, and once the paper film came into contact with the aluminium foil and the heat accumulated as the current passed through, the film melted, which decreased the energy storage of devices. Compared with other dielectrics, much more attention has been focused upon polymers due to their super-high breakdown field, low fabrication temperature, and flexibility. For example, PVDF-based materials were found to have larger permittivity because of their ferroelectric nature [14, 92–96]. With developments in insulating materials and production technologies, the energy density of polymers has reached 25,000 J/L with high breakdown strength. The breakdown strength of imported films made of P(VDF-CTFE) or P(VDF-HFP) has reached 800 MV/m – 100 times that of earlier materials [18].

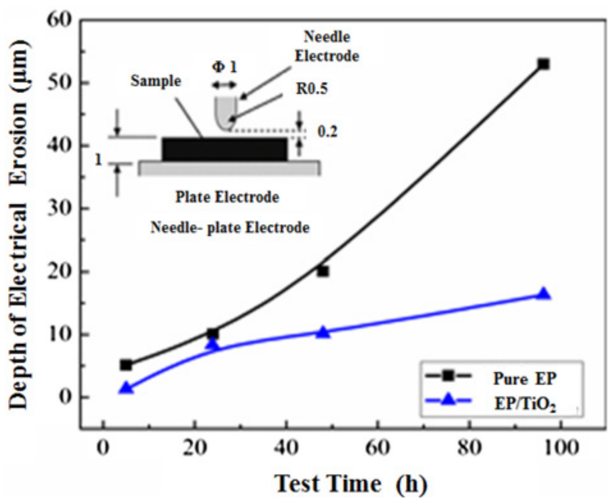

Fig. 7 *Corona-resistant properties of epoxy and epoxy/TiO_2 nanocomposites at 60 Hz, 6 kV AC voltage*

4 Other progress in insulating materials

Other properties of insulating materials have also been studied and improved during the past century, in response to the evolution of power systems and electric devices; these properties include non-linearity, radiation resistance and corrosion resistance.

The electric field inside non-linear materials can initially be homogenised to suppress space charge accumulation. This improves their electrical properties. Research into non-linear materials has controlled for grain growth by changing the additives, reducing grain size through adjusting the formula, optimising the method for improving the grains' microstructural homogeneity, and modifying the nanoparticles' surfaces. In the future, non-linear insulating materials should have a high electric potential gradient as well as high energy absorption capability. In addition, voltage-dependent resistors with a chip multilayer structure and low sintering temperature are materials worth researching.

Radiation resistance impacts a material's lifetime. In certain circumstances, insulating materials face electromagnetic radiation from naturally radioactive materials, particle accelerators, electric

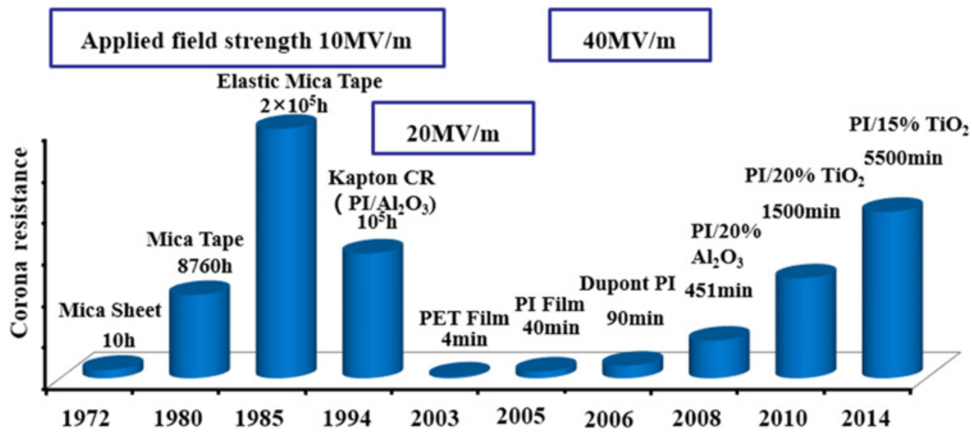

Fig. 8 *Progress in insulating materials with high corona resistance [79–82]*

devices, lightning, the Earth's surface, and outer space. The electromagnetic radiation acting on insulating materials stimulates active particles, causing chemical aging. We can increase the radiation resistance of insulating materials by adjusting their microstructure via doping them with fillers (such as carbon black, Pb, Ba and B) [22], according to the amount and type of radiation and the environment in which it is experienced. Some research has been done to develop radiation-resistant materials with high purity and low chlorinity.

Eco-friendly, clean and sustainable insulating materials have been rapidly developed in recent years. In the future, insulating materials must be economical as well as easy to recycle and reuse. On the one hand, any potential harm to humans must be taken into account when insulating materials are used for a long time; to reduce such harms, new kinds of clean materials should be utilised – for example, heat-resistant fibres can replace asbestos, mixed gas and halothane can replace SF_6 and so on. On the other hand, the application of recyclable materials should be expanded by getting rid of unrecyclable materials – for example, replacing thermosetting plastics with biodegradable thermoplastics such as poly(*p*-phenylene oxide), replacing mineral oil with plant oils that have a higher fire point, replacing synthetic resins with reproducible inorganic powders and so on. Moreover, we must take into account the noise, pollution, and waste generated during the production, use, and recycling of insulating materials. Flame-proof and self-extinguishing materials such as phenol-biphenylene/epoxy nanocomposites have been studied to replace materials with a low fire point.

5 Summary and forecast

Insulating materials have been developed for over a century. From natural materials to organic polymers, from pure polymers to nanocomposites, there have been continuous developments, and the electrical, thermal and mechanical properties of insulating materials have been greatly improved [1]. For example, breakdown strength has increased 3000 times, from 0.3 to 1000 MV/m; thermal conductivity has increased 30 times, from less than 1 W/m·K of pure polymer to 30 W/m·K of nanocomposite; the highest temperature that materials can withstand has been increased 15 times, from 100°C for natural materials to 1500°C for crosslinked and crystalline materials; the electric strength of materials at very low temperatures has been increased ninefold, from 30 MV/m for paper to 280 MV/m for imported films; the corona resistance has been increased from 10 to 40 MV/m; the corona resistance lifetime has been increased 5000 times by doping nanofillers into PI films; the experimental energy density has been increased 2500 times; and the energy density in applications has been increased 520 times.

There are still many challenges and problems – for example, the structure of the interfaces between nanoparticles and matrices is not clear, the adjustment of interfaces is still underdeveloped, nanoparticle dispersion is inadequate, and the repeatability of relevant experiments is low. Thus, we urgently need to simultaneously develop individual materials and improve the comprehensive performance of composites.

The findings of this study suggest new directions for insulating materials: matching the characteristics of materials with the specific application demands for devices; improving the technology of interface adjustment and manufacturing; exploring surface modification methods and dispersion techniques for nanoparticles; creating multilayer designs for materials and devices to be used in smaller, thinner, cleaner products; studying the connections between microscopic, mesoscopic and macroscopic structures; and applying analogue simulations and computer calculations to analyse the formation of interfaces [1, 3, 4, 6–8].

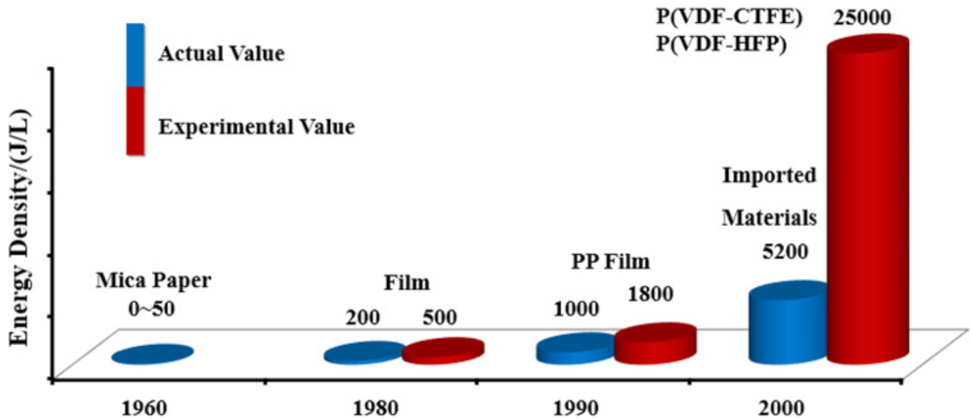

Fig. 9 *Progress in insulating materials with high energy storage density, experimentally and in applications*

Overall, insulating materials are poised to experience a phase of rapid development due to global concerns over environmental protection and continued developments in their electrical, thermal and mechanical properties.

6 Acknowledgments

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