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Overview of Development of Insulating Materials

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1.1 Introduction

Electrical insulating materials are the foundational components that enable safe and efficient operation of power and electronic systems. By preventing unwanted current flow, insulators allow voltages to be maintained and electric fields to be controlled within equipment. Over the past two centuries, the field of insulating materials has evolved dramatically, encompassing solids such as ceramics, polymers, resins, and paper, liquids such as mineral oils, and gases such as sulfur hexafluoride (SF_6), air [1–6]. Historically, the selection of insulating materials has been a key driver for advancements in electrical technology. In the earliest days of telegraphy and power distribution (mid-19th century), engineers relied on natural materials like cotton, silk, paper, rubber, and oils – materials that were readily available but often exhibited limited durability [6–8]. As electrical systems evolved toward higher voltages and more demanding operating environments, the limitations of these early insulating materials became increasingly apparent: Many materials age, degrade, crack, or absorb moisture over time, thereby weakening their performance [9, 10]. This has driven continuous innovation in insulating materials, closely intertwined with the progress in electrical engineering itself. Each generation of insulating materials has addressed the shortcomings of its predecessors and opened up new possibilities for system design, such as enabling higher voltage transmission or reducing equipment size. Therefore, the development of insulating materials can be viewed as a synergistic evolution alongside electrical infrastructure.

In modern times, polymer materials such as plastics and resins, liquid materials such as mineral oil, synthetic esters, and silicone oil, and gas materials such as SF_6 have dominated many electrical devices due to their excellent fundamental properties, including dielectric performance, mechanical properties, chemical stability, and thermal properties [8, 11–15]. However, some new challenges that were

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not fully considered in earlier times have also emerged: environmental sustainability and lifecycle management of these materials have become key concerns [16–18]. In the context of urgent climate agreements, insulating materials now face new challenges and requirements: they must not only exhibit outstanding electrical performance but also remain low-carbon, eco-friendly, and sustainable throughout their entire lifecycle. Many traditional insulating materials, including cross-linked thermosetting polymers, insulating oils, and certain gas or vacuum-based insulating materials, pose recycling or environmental risks when discarded or leaked [19–21]. Additionally, as reliability requirements increase, materials capable of self-monitoring/warning of health status, self-adapting, or even self-healing damage are gaining favor [22–28]. These considerations are driving a new round of research into next-generation insulating materials, which must not only be excellent in performance but also possess low-carbon, eco-friendly, sustainable, and smart characteristics. This chapter will review this evolutionary journey, from the earliest natural insulating materials to the latest low-carbon, eco-friendly, and smart solutions. We will see how early engineers relied on natural materials such as paper, rubber, glass, and oil, and how the emergence of synthetic polymers, specialty fluids, and smart insulating materials revolutionized electrical insulation technology.

1.2 Historical Evolution of Insulating Materials

1.2.1 Early Insulating Materials in Electrical Engineering

The development of electrical insulating materials is closely related to the development of electrical technology. Early researchers and engineers experimented with any material that could separate and protect conductors. Up to about 1925, naturally occurring products such as bitumen, natural rubber, mica sheets, cotton cloth, silk, paper, wood, and ceramics were the standard insulators, as shown in Table 1.1 [32, 33]. For example, cotton and silk tape were used to wrap early wire conductors. Mica, a mineral that can be split into thin flexible sheets, was prized for its high dielectric strength and heat resistance in applications like commutators and early capacitors. Oil-impregnated paper became a widely used insulating material in insulated cables and transformer windings due to its ability to improve dielectric properties by removing air [8]. Even today, some high-voltage direct current (HVDC) cables still use impregnated paper or paper-polypropylene laminates as insulators, although they are gradually being replaced by polymer materials [34, 35]. These materials are widely used because they are simple to obtain and show high electrical impedance, but their performance under electrical and thermal stress is very limited.

One notable natural insulator was natural rubber, derived from latex, which was one of the first insulating materials used for electrical wires in the early 19th century [32]. Unvulcanized rubber was tried as early as 1810, followed by vulcanized rubber after Charles Goodyear’s patent in 1844. Natural rubber softens easily at high temperatures and becomes brittle at low temperatures. The vulcanization process greatly improved the stability of rubber by adding cross-links (sulfur bonds)

Table 1.1 Early commonly used insulating materials, their properties, and applications.

Era (approx.)	Common insulating materials	Key properties and applications
1800s (early)	**Glass, porcelain:** for telegraph and early power line insulators; very durable, high dielectric constant (~6), but brittle and heavy [6]. **Cloth (cotton, silk):** wrapped on wires and coils; flexible but moisture-sensitive [9]. **Shellac, wax, and asphalt:** coatings on windings and cables; provided dielectric protection but could soften with heat [29]. **Mica:** used in motors, commutators, and early capacitors; excellent dielectric strength and temperature endurance [3].	1850s telegraph lines using glass insulator knobs on poles. First undersea cable (1851) with gutta-percha insulation.
1900~1920	**Natural rubber:** used for wire insulation and cable jackets by late 1800s; soft and prone to cracking unless vulcanized [30, 31]. **Oil-impregnated paper:** introduced for high-voltage cables/transformers; dielectric strength: up to ~10–20 kV/mm when thoroughly impregnated [8].	1910s transformers with oil-paper insulation; rubber-insulated wiring in early homes (rubber often wrapped in cloth for protection).

between polymer chains, marking one of the first instances of chemically modified insulating material to enhance performance [30, 36]. Additionally, another crucial early insulator was gutta-percha, a resinous polymer obtained from the Palaquium gutta tree. In 1843–1847, it was introduced as a wire insulating material, especially for submarine telegraph cables. Gutta-percha is a thermoplastic material that can be heated and coated onto the surface of wires, then hardened to form a tough and waterproof coating. It remained the primary insulating material for submarine cables for decades, including the first transatlantic cable in 1858. However, gutta-percha was expensive, available only from Southeast Asia. And it tended to degrade (becoming brittle) when exposed to oxygen and sunlight over time.

In overhead power lines and early electrical equipment, ceramics and glass were essential insulating materials [37]. By the 1850s, glass insulators were used on telegraph lines, and porcelain, a type of ceramic made from clay, was later found to offer superior mechanical strength and better resistance to weathering. Porcelain insulators were widely used to support high-voltage lines and busbars due to their excellent dielectric strength and weather resistance [6, 38]. These inorganic insulators are essentially permanent, as they do not undergo significant aging, but they are heavy and prone to brittleness. Thus, although polymers have replaced many insulation tasks, ceramics remain crucial in areas with high requirements for stability in extreme environments, such as substation pillar insulators and bushings.

By 1920, the electrical industry had identified a range of natural insulating materials and combinations such as paper oil systems and rubber fabric composites. However, the pursuit of improved performance including higher breakdown voltage, greater temperature resistance, and reduced aging was constrained by the inherent limitations of these natural materials. This laid the foundation for the introduction of synthetic materials.

1.2.2 Advent of Synthetic Insulating Materials

From the late 1920s to the 1940s, the field of insulating materials underwent a revolution with the emergence of synthetic polymers and resins. The first major breakthrough was the synthesis of phenolic resin by Bakelite in 1907, also known as Bakelite plastic [39]. It is regarded as the first completely synthetic plastic in human history, marking the beginning of the polymer era. In the electrical industry, Bakelite and similar phenolic resins were rapidly adopted as insulating varnishes and molded parts in the 1910s to 1930s. For example, phenolic laminates made by impregnating paper or fabric with phenolic resin and curing it were used as switchboard panels, tube sockets, and connector bases. These applications fully utilized the excellent electrical insulation properties and heat resistance of Bakelite, which could withstand temperatures of approximately 150 °C. By the 1930s, phenolic resin had largely replaced shellac and asphalt for impregnating high-voltage coils, providing more durable insulation material for transformer and motor windings [40].

Following phenolic resin, other synthetic thermosetting resins were subsequently developed. The first were urea-formaldehyde resin and melamine-formaldehyde resin (1930s), which offered superior electrical properties and lighter colors. Urea-formaldehyde resin is commonly used for appliance insulation and socket components, while melamine-formaldehyde resin, with its better moisture resistance, is used for high frequency insulation. Furthermore, in the field of rubber and elastomers, researchers have synthesized synthetic rubber that can replace natural rubber. Neoprene (polychloroprene) was invented in 1931 as one of the first oil-resistant rubber insulators. Styrene-butadiene rubber (SBR) became widely available in the late 1930s as a rubber substitute for wires and cables [32]. These synthetic elastomers could operate at higher temperatures and had improved aging characteristics compared to natural gum rubber.

More importantly, in the late 1930s, thermoplastic polymers began to be used in insulating materials. Polyvinyl chloride (PVC) was first synthesized in 1872, but it was in 1926 that Waldo Semon at B.F. Goodrich discovered how to plasticize PVC to make it flexible and processable. By 1935–1936, PVC insulating materials were being tried on wires and cables. However, early PVC had higher dielectric constant and dissipation factor, which limited it to low voltages despite improvements in compounding. Moreover, polyethylene (PE) was accidentally discovered in 1933 by researchers at Imperial Chemical Industries (Fawcett and Gibson) through the high pressure polymerization of ethylene. During World War II, PE was used in coaxial cables for radar and radio because of its excellent high frequency insulation properties, including a low dielectric constant of around 2.3 and low dielectric loss. By the late 1940s, commercial PE insulating materials began to be used in radar cables and some power cables. Early PE had a relatively low melting point (approximately 105 °C), but it exhibited extremely strong chemical inertness and high dielectric strength (approximately 25–30 kV/mm). In addition, polytetrafluoroethylene (PTFE), famously known as Teflon, was discovered accidentally by Roy Plunkett at DuPont in 1938. PTFE has unmatched chemical inertness, extremely low dielectric constant (approximately 2.0) and loss, and can withstand temperatures up to 260 °C, making it the gold standard insulating material for high-temperature and high frequency applications. Its discovery was another key moment in polymer science [41, 42].

During the early stages of synthetic material development, liquid and gas insulating materials also made innovative progress (Table 1.2). In the 1930s, polychlorinated biphenyls (PCBs) were introduced as synthetic insulating oils, with higher flash points and dielectric constants than mineral oils, and were adopted as insulating media for transformers and capacitors. PCB-based insulating oil enabled capacitors to achieve smaller sizes and higher efficiency, as its higher dielectric constant allowed for greater capacitance per unit volume. However, PCBs were later found to be highly toxic and environmentally persistent, leading to their gradual phase-out in the 1970s. Another milestone was the synthesis of SF_6 as in the early 20th century (first prepared in 1900) and its recognition as an exceptional dielectric gas. By the late 1940s, SF_6 was being studied for use in high-voltage equipment, though its widespread adoption had not yet taken hold.

This era established the pivotal role of polymers in electrical engineering. The successive development of numerous synthetic insulating materials significantly expanded the design possibilities for electrical equipment. Improvements in dielectric strength and thermal stability enabled equipment to achieve miniaturization and higher reliability. For example, in motors and transformers, enameled wires made of copper and coated with synthetic resin paint such as polyvinyl alcohol or polyester replaced traditional insulation materials like silk and shellac. This substitution allowed the operating temperature limit to increase from 90 °C to 130 °C or higher. Overall, by 1950, the foundation for modern insulation systems had been established: combinations of natural and synthetic materials were widely adopted, but there was an increasing trend toward fully synthetic solutions designed for specific requirements.

1.2.3 High-Performance Insulating Materials and Composite Systems

From the 1950s to the 1970s, polymer science experienced significant development and maturation, leading to the emergence of high-performance insulating materials capable of withstanding harsh environments such as high mechanical stress and elevated temperatures. During this period, many insulation systems that are still in use today were developed, and polymers invented in the 1930s and 1940s were gradually improved.

Table 1.2 Summarizes the new synthetic insulating materials introduced up to the middle of this century.

Material	Type	Key properties and impact
Phenolic resin	Thermoset solid	Heat resistant (150 °C) and good electrical properties; enabled molded electrical parts (switches, coil forms). First synthetic polymer insulator, replacing natural shellac and wood in many components.
PVC	Thermoplastic solid	1910s transformers with oil-paper insulation; rubber-insulated wiring in early homes (rubber often wrapped in cloth for protection).
PE	Thermoplastic solid	Excellent dielectric constant (~2.3) and high volume resistivity; dielectric strength ~25 kV/mm. Used in radar/coax cables and gradually in power cables. Low melting point limited early use to moderate temperatures.
Rubbers (neoprene, SBR)	Solid	Suitable for insulated cables in industrial environments and wire insulation in high-temperature environments (e.g., motors, vehicles, etc.), without rapid degradation.
Askarel (PCB oil)	Liquid dielectric	High dielectric constant fluid for transformers/capacitors, nonflammable.
SF_6 gas	Gas dielectric	Dielectric strength about 2–3 times that of air; chemically inert, arc-quenching; laboratory synthesis began in 1900, and practical applications began in the 1940s.
Epoxy resin	Thermoset solid	A versatile resin (reaction of epichlorohydrin and bisphenol-A, pioneered by Castan and Greenlee in the late 1930s). Commercial epoxy resins became available ~1947–1950. By 1945, epoxies were used for encapsulating electronic components and soon for laminating (e.g., in printed circuit boards and insulating structural parts). Epoxies offered excellent adhesion, mechanical strength, and electrical insulation, though they are rigid and require additives for toughness.

While polyethylene had been used in cables, its relatively low melting point meant that high operating temperatures could deform it. In the 1950s, cross-linked polyethylene (XLPE) technology, which involves forming a thermosetting network structure in polyethylene using peroxides or radiation, was introduced. XLPE combines the excellent dielectric properties of PE with significantly improved thermal stability, which allows continuous operating temperatures up to ~90–110 °C [43]. By the late 1960s and 1970s, XLPE cable insulating materials began to replace traditional oil-paper cables in medium-voltage power grids. The 1970s

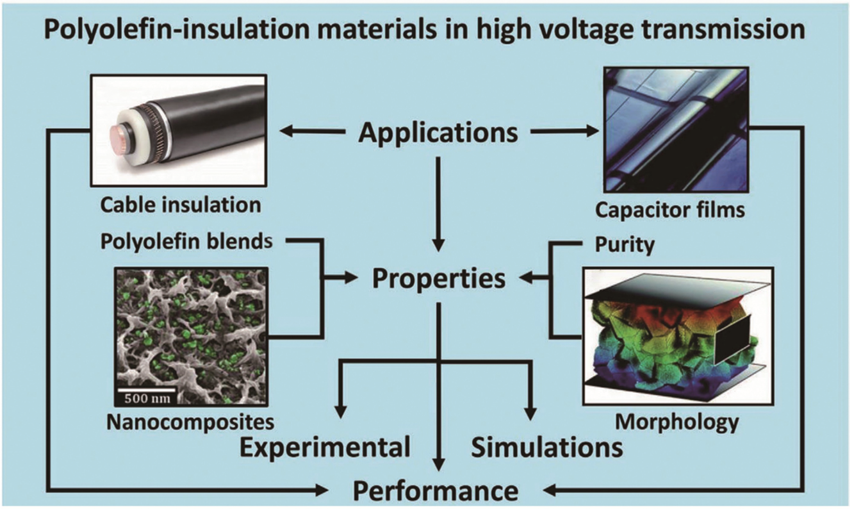


Figure 1.1 Polyolefin insulating materials for high-voltage insulation. *Source:* [12] / John Wiley & Sons / CC BY-NC 4.0.

marked the application of XLPE in medium-voltage cables, supporting voltage up to approximately 33 kV. Subsequently, improved formulations of XLPE, enhanced with additives to inhibit the formation of water trees, began to be used at high-voltage transmission levels ranging from 66 to 230 kV. Today, XLPE has become the standard insulation material for power cables due to its excellent balanced performance. In addition to PE, other types of polyolefins also play an important role in the field of electrical insulation (Figure 1.1). Isotactic polypropylene (IPP) was discovered in 1954. Although its electrical properties are similar to those of PE, with a dielectric constant of approximately 2.2 and extremely low dielectric loss, it exhibits a higher melting point of around 165 °C and can withstand higher operating temperatures [44]. In the 1960s, polypropylene (PP) began to replace paper and polystyrene as the dielectric film for capacitors, as it could be made into very thin PP films and could self-clear after dielectric breakdown [45, 46]. PP film capacitors became a common choice for AC applications in the 1970s and 1980s. However, due to the low-temperature brittleness and processing difficulties of PP, it was not initially as widely used in cables as PE. In recent years, PP has once again gained attention as an eco-friendly cable insulating material due to its ease of recycling.

Beyond PTFE, other melt-processable fluoropolymers were developed in the 1950s–1960s, such as fluorinated ethylene propylene (FEP) and ethylene tetrafluoroethylene (ETFE). These materials offer superior dielectric and thermal properties while providing more convenient processing methods such as extrusion molding. By the 1970s, FEP and ETFE were used for wire insulation in aerospace and computer cabling, where thin-walled, high-performance insulation materials were required. Notably, since its introduction around 1945, epoxy resin has rapidly become the basis for electrical insulating materials due to its high cohesive strength, strong adhesion, good flexibility, excellent thermosetting properties, and

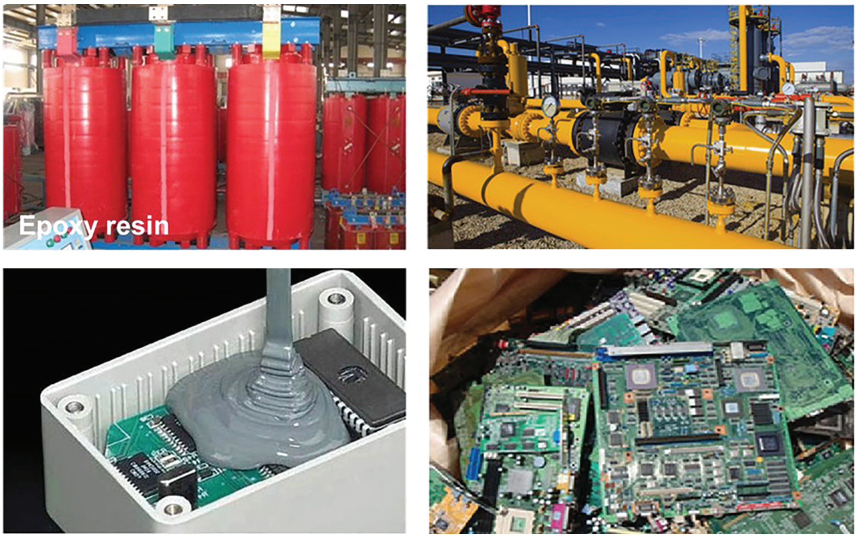


Figure 1.2 Photos of epoxy resins used in transformer insulating bushings, curing agent, encapsulant, and electronic materials. *Source:* [48] / with permission of Elsevier.

stable chemical corrosion resistance. In the 1950s and 1960s, epoxy resin was combined with fillers such as aluminum oxide and silicon dioxide to produce molded high-voltage insulation components, including bushings, instrument transformer housings, and coil spacers for rotating machines (Figure 1.2) [47, 48]. Epoxy resin materials enabled the mass production of small insulating components with superior dielectric strength and mechanical stability. Moreover, by the 1960s, epoxy glass laminates such as G10 and FR-4 boards had become the standard material for printed circuit boards and many structural insulation boards, replacing the phenolic paper laminates previously used for high-performance applications.

A major milestone in the field of high-voltage transmission was the development of composite polymer insulators in the 1960s and 1970s. Traditional ceramic insulators were heavy and prone to brittle failure. To improve performance, researchers introduced composite insulators with a glass fiber core as the base material, covered with a polymer, typically made of silicone rubber or ethylene propylene diene monomer rubber. Silicone rubber, which debuted in the 1940s, is a high-temperature elastomer with excellent hydrophobic properties, making it suitable for external insulation applications to prevent moisture ingress and reduce leakage current [36, 49, 50]. Early applications in the 1960s were not very successful, but by the 1970s, especially the 1980s, polymer composite insulators gradually gained trust. They are lighter in weight, more resistant to damage, and can achieve excellent antipollution performance due to their hydrophobic surface, which continuously repels water. Currently, composite insulators are widely used in transmission lines and substations as an alternative to ceramic insulators (Figure 1.3).

In addition, to meet temperature requirements above 180 °C, a class of heterocyclic and aromatic polymers has emerged. Notably, DuPont commercialized polyimide

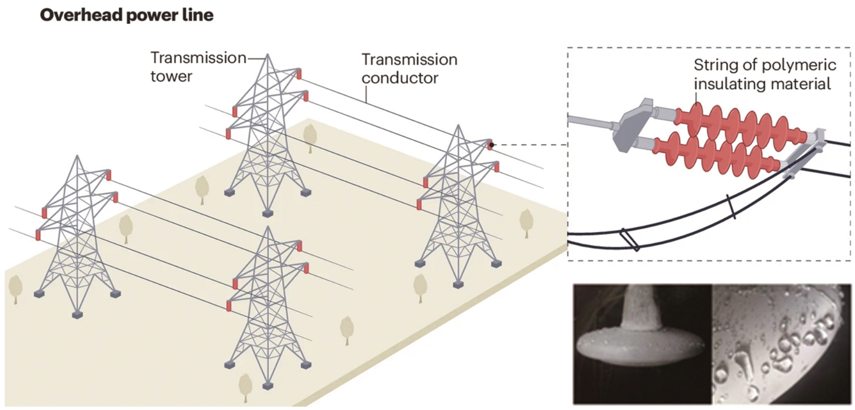


Figure 1.3 External insulation applications of silicone rubber insulating materials. *Source:* [1] / Springer Nature.

under the trademark name “Kapton®” in 1961. Kapton® polyimide film can operate within a temperature range from absolute zero (4 K) to 400 °C and only carbonizes at extremely high temperatures [13]. The industrialization of polyimide (PI) resin has a history of half a century. Its excellent high-temperature resistance, chemical stability, and mechanical properties make it an important material in high-tech fields as an engineering plastic and composite material [51]. Furthermore, its dielectric strength of 150–200 kV/mm (in film form) makes it suitable for extreme applications, such as insulation material for spacecraft wiring, high-temperature flexible circuits, and insulation material for special motors and generators, as shown in Figure 1.4. Similarly, other high-performance polymers such as polybenzimidazole (PBI), polysulfone, polyetheretherketone (PEEK, introduced in 1978), and polyphenylene sulfide (PPS) were developed in the 1960s and 1970s, providing engineers with material options to meet extreme thermal and electrical requirements.

During the 1960s, the insulation systems of large rotating machines (generators, motors, etc.) achieved technological advancements through the adoption of F-class (155 °C) and H-class (180 °C) materials. This was achieved by using mica paper tapes bonded with epoxy or PI resins on coils, and fiber glass cloth, in place of older asphaltic mica or shellac-based systems. This improvement enabled generators and motors to operate more reliably at higher temperatures for longer periods, thereby increasing their power density. For example, in the 1970s, the use of epoxy mica vacuum pressure impregnation (VPI) processing for generator stator windings eliminated air gaps and achieved significantly optimized dielectric properties.

Meanwhile, gas and liquid insulation technologies have also made significant progress. By the 1960s, SF_6 gas insulation technology had been fully adopted in the power industry. Power companies deployed SF_6 circuit breakers and gas-insulated substations (GIS), which could handle extremely high voltages (up to 500 kV) in a compact layout, something air insulation technology could not achieve. Furthermore, in terms of liquids, safer silicone oil and synthetic ester liquids replaced PCB oil as transformer coolants during the 1970s and 1980s. At the same

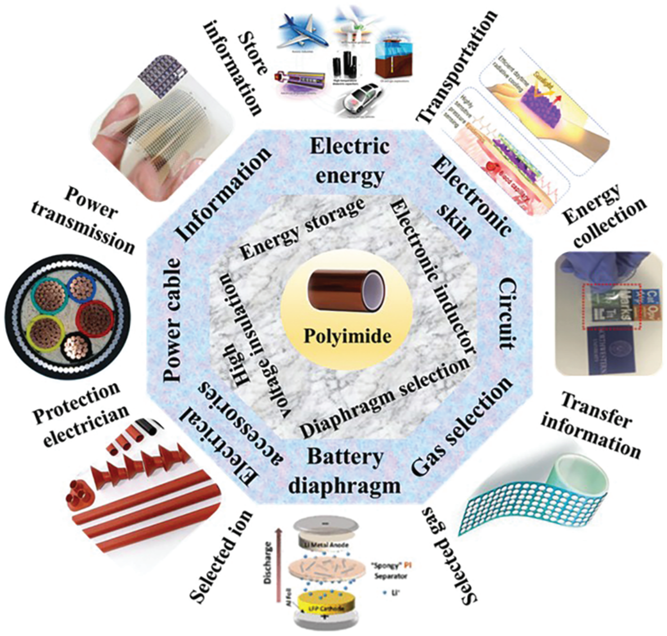


Figure 1.4 Summary diagram of PI applications. *Source:* [13] / John Wiley & Sons.

time, advancements in refining processes, including the removal of impurities and improved oxidation stability, enhanced the quality of transformer oil and contributed to extending the service life of transformers.

In summary, by the late 20th century, the electrical industry had at its disposal a rich toolkit of insulators: Solids: cross-linked polyolefins (XLPE), ethylene propylene rubber (EPR), and PVC for low voltage, epoxies and polyesters for structural parts, polyimides and silicones for high-temperature, and engineering thermoplastics for specialized uses. Liquids: mineral oil with inhibitor additives, silicone oil for high-temperature transformers, and new ester-based oils (starting to appear as fire-safe or biodegradable options). Gaseous insulating materials include air and SF_6, with SF_6 being the dominant choice in enclosed high-voltage equipment because of its reliability and space-saving benefits, despite its higher cost. The historic progress made during this period also laid a solid foundation for cutting-edge research in the present day.

1.3 Modern Insulating Materials and Current Challenges

Current electrical infrastructure relies heavily on polymer insulating materials, from underground cables and substation equipment to home wiring and electronic products, as shown in Figure 1.5. The widespread use of polymer materials is due to their excellent electrical insulation properties, mechanical flexibility, lightweight

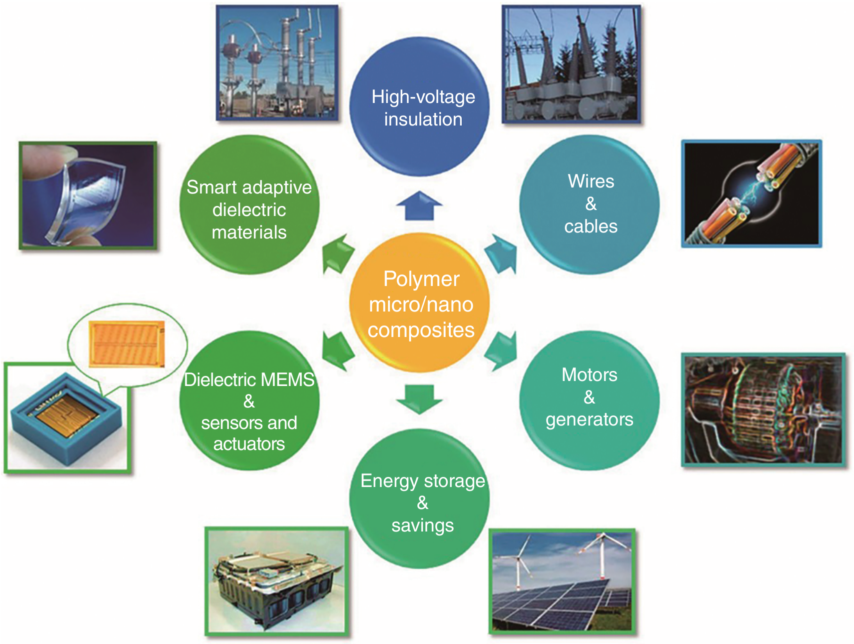


Figure 1.5 Application scenarios for modern polymer insulating materials. *Source:* [52] / MDPI / CC BY 4.0.

characteristics, and scalable manufacturing processes. The most common insulating materials currently used in practical applications include epoxy resin, XLPE, PP, EPR, PVC, PI, cellulose insulating paper, etc. In addition, liquid insulating materials such as mineral oil and natural ester fluids, as well as gas insulating materials such as SF_6, C-FN/CO_2 mixtures, and dry air, also play an important role in modern insulation systems. Despite the great success of modern insulating materials, several challenges and concerns have become apparent.

1.3.1 Aging and Degradation

The first issue is aging and performance degradation during operation. All organic insulating materials undergo aging under electrical, thermal, and environmental stress. Taking XLPE cables as an example, electrical aging can lead to the formation of electrical tree structures initiated by partial discharge, which involves microscopic sparks occurring within material defects. These microscopic damage pathways can penetrate the insulation layer and ultimately lead to insulation breakdown. Although XLPE cables have high long-term reliability, water trees may still form if trace moisture and impurities combine under electrical stress over decades [53, 54]. In inverter-powered motors or HVDC systems, due to the presence of high frequency components in the voltage waveform, polymer insulating materials may suffer from electrical erosion or space charge accumulation. This electrical aging leads to long-term degradation of material performance, occurring well before complete failure. Therefore, monitoring and diagnosing the health of insulating materials is critical for power system maintenance. Traditional condition monitoring relies on methods such as measuring loss factor, partial discharge, or chemical analysis of transformer oil. However, for solid polymer insulating materials, changes in the material may be subtle such as minor chemical changes or microporosity, making it difficult to detect aging phenomena on-site before a failure occurs [24]. This has driven the development of smart self-diagnostic materials, which will be discussed in detail in subsequent chapters. These materials can emit optical warning signals when significant internal degradation takes place. Additionally, thermal aging results from prolonged exposure to thermal-oxidative conditions, leading to brittleness or loss of elasticity. Environmental aging is caused by factors such as ultraviolet radiation, rain, ice accumulation, or radiation in space applications, and remains an ongoing challenge for insulating materials. Therefore, researchers must continue to refine material formulations, such as by adding antioxidants, stabilizers, or nano-filler reinforcements, to slow down the aging process.

1.3.2 Recycling and Disposal

As mentioned earlier, many polymer insulating materials such as XLPE, PI, EPR, and epoxy resin are thermosetting materials and cannot be recycled through melting and reprocessing. Landfilling such materials is unsustainable, while incineration, though it can recover some energy, may release carbon dioxide and potentially toxic compounds, posing significant risks to the environment and human health [9, 21]. Given the growing regulatory and economic pressures, there is an urgent need to explore recycling methods for such thermosetting materials or to design and synthesize new recyclable insulating materials. Even thermoplastic insulating materials like PVC and PE face recycling challenges when they are part of composite materials containing metals and other polymers. Additionally, separation and cleaning pose significant challenges [55, 56]. Currently, innovative solutions, including chemical recycling (depolymerization), converting polymers into fuels or other chemicals, or reprocessable cross-linked polymers (glass-state polymers), are under active research. Life cycle assessments indicate that recycling plastics can save significant amounts of energy and reduce carbon emissions. For example, recycling one ton of plastic instead of incinerating it can avoid approximately three tons of carbon dioxide emissions. The cable industry has already initiated recycling efforts, such as mechanically processing XLPE into secondary products or using crushed XLPE as a filler. However, current commercial technologies have not yet achieved fully closed-loop recycling.

1.3.3 Environmental Impact

Some traditional insulating media have significant environmental drawbacks. Certain solid insulating materials, such as PI and XLPE, are difficult to self-heal or recycle due to their permanently cross-linked networks. For instance, with a large number of early XLPE cables reaching the end of their service life and waste generated during the manufacturing process, the issue of disposal has become increasingly severe. For example, it is estimated that in Germany alone, approximately 150 000 tons of waste cable, much of which contains XLPE and PVC, are generated annually [57]. Typically, these wastes are landfilled or incinerated, causing significant environmental pollution. Mineral oils used in transformers and certain electrical equipment may leak and cause soil contamination, and their raw materials are derived from petroleum. This has prompted the transformer industry to adopt plant-derived natural esters as more biodegradable and sustainable alternatives, which is a significant advancement in the field of liquid insulation technology. Another example is SF_6, a gas widely used as an insulating and arc-quenching medium in high-voltage switchgear. However, SF_6 has an extremely high global warming potential ($GWP \approx 25\,200$ times that of carbon dioxide) and an atmospheric lifetime of up to 3200 years, and losses during handling make it a significant source of greenhouse gas emissions. Additionally, it is worth noting that while pure SF_6 is nontoxic, its byproducts such as SF_4, SOF_2, and S_2F_{10} are toxic and corrosive [58]. Therefore, for climate and safety reasons, alternatives such as new fluorocarbon or fluoroketone gases, or vacuum and solid insulation designs are being sought to replace SF_6 [59]. Although SF_6 gas insulation exceeds the scope of “insulation materials” in the solid/polymer sense, this example highlights the widespread industrial pursuit of eco-friendly insulation solutions to reduce its environmental footprint.

1.3.4 Performance Limits

Modern electrical equipment requirements, such as higher operating temperatures, higher electric fields (e.g., compact dry-type DC capacitors, transformers, power electronic devices, etc.), and radiation-rich environments (space and nuclear power), are pushing many insulating materials to their performance limits. Solid insulating materials such as PI, XLPE, PP, and epoxy resin now not only need to withstand thermal stresses exceeding 130 °C but must also cope with increasingly intense electric fields, radiation, and mechanical loads. This has driven the development of high-temperature polymer alloys, functionally graded materials, and nanocomposites, which incorporate fillers that suppress electric fields or create traps to enhance partial discharge resistance and inhibit space charge accumulation. Liquid insulating materials, including mineral oil, natural esters, and silicone liquids, must address limitations related to oxidative stability, moisture resistance, and thermal degradation, particularly in high-voltage and high-temperature applications. Gas insulating materials, particularly SF_6, are under pressure due to environmental concerns, leading to alternatives like C_4-FN/CO_2/N_2 mixtures, which must balance dielectric performance with environmental safety.

In summary, while the insulating materials in use today represent a pinnacle of decades of development and generally perform remarkably well, they face a dual mandate of improving basic characteristics such as high breakdown strength, low loss, and long-term reliability, while enhancing sustainability and functionality like self-healing, recyclability, and reduced environmental impact.

1.4 Toward Green and Smart Insulating Materials

To address the challenges above, the past few years have seen rapid advancements in the science of insulating materials. As shown in Figure 1.6, researchers are rethinking material design at the molecular level to support goals such as recyclability, degradability, and environmental friendliness in line with the circular economy. Meanwhile, efforts are focused on extending service life through self-healing, enabling smart functions like self-warning and adaptability, and improving overall performance such as weather resistance. This section will introduce some of these cutting-edge directions, which will be explored in detail in subsequent sections.

1.4.1 Eco-Friendly and Sustainable Insulating Materials

In recent years, a strong emphasis has been placed on low-carbon, eco-friendly, and sustainable insulating materials, which is highlighted by the Paris Agreement and global climate initiatives. The goals are to reduce the environmental impact of

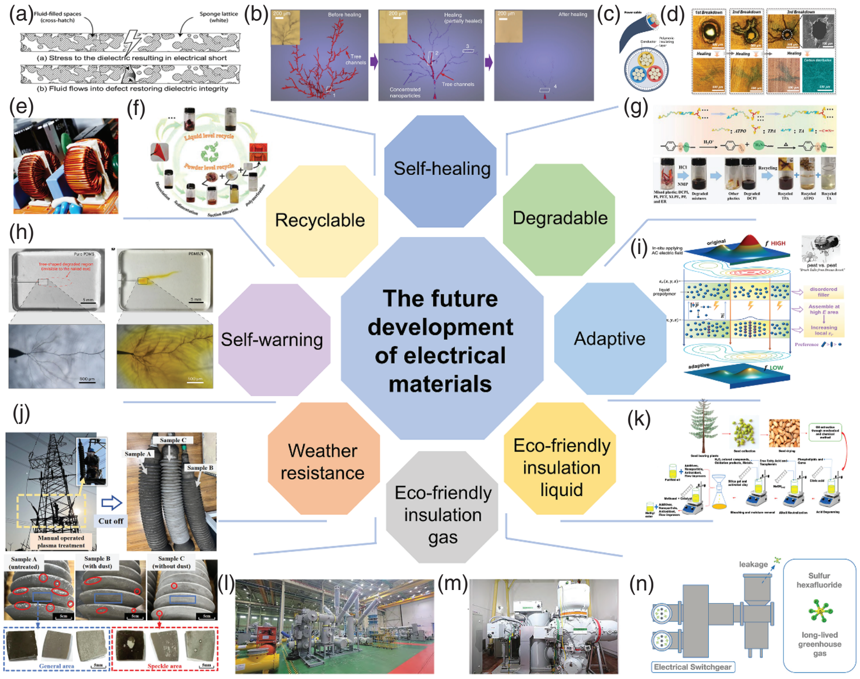


Figure 1.6 The future development of electrical materials. (a–d) Self-healing (e, f) recyclable. (g) degradable, (h) self-warding. (i) adaptive and (j) weather resistance electrical materials. (k) Eco-friendly insulation liquid. (l–n) Eco-friendly insulation gas. *Source:* (b) [23] / Springer Nature, (d) [60] / American Chemical Society, (e) [61] / American Chemical Society, (f) [61] / John Wiley & Sons, (g) Baoquan Wan et al., 2023 / With permission of John Wiley & Sons, (i) [62] / John Wiley & Sons / CC BY 4.0, (j) [49] / with permission of Elsevier, (k and M) [63] /American Chemical Society and (n) [63] / with permission of American Chemical Society.

insulating materials across their life cycle, from production to end-of-life disposal, and to ensure safety and reliability under green energy transitions.

Traditional thermoset materials such as XLPE, epoxy, and PI are not recyclable because, once cured, they cannot be remelted. This results in significant disposal challenges, with waste materials like scrap cables and electronic components contributing to the growing problem of electronic waste [11, 13, 64]. To address this issue, one revolutionary idea is to construct a polymer network that functions like a thermosetting material during operation, offering high mechanical strength and good heat resistance, but can be reverted to a dynamic state when recycling is required. Such polymers are typically referred to as covalent adaptive networks (CANs) or glass-like polymers (vitrimers) [65–67]. This concept was first proposed in the field of general polymer science in the 2010s and has begun to be gradually applied to the field of insulating materials. For example, Wan et al. [26] designed a dynamic covalent adaptable polyimide (DCPI) composite dielectric film to address the limitations of non-recyclability and limited durability of traditional PI materials after curing. By introducing dynamic imide bonds into the PI molecular structure, the DCPI maintains high insulation and mechanical strength while gaining degradability and reprocessability. As shown in Figure 1.7, the film can be completely degraded in acidic solutions at room temperature, allowing the recovery of high purity monomers for resynthesizing PI films. Notably, the regenerated films exhibit almost no loss in mechanical and electrical performance, offering a viable solution for the closed-loop recycling of PI materials.

There is a renewed interest in using renewable and degradable materials for insulation, especially where ultra-long life is not required. For instance, natural ester fluids derived from vegetable oils such as soybean or rapeseed have emerged as alternatives to mineral oil in transformer applications. These fluids are biodegradable and have higher fire points, which enhances fire safety. By 2020, natural ester insulated transformers had been widely used in distribution and even transmission

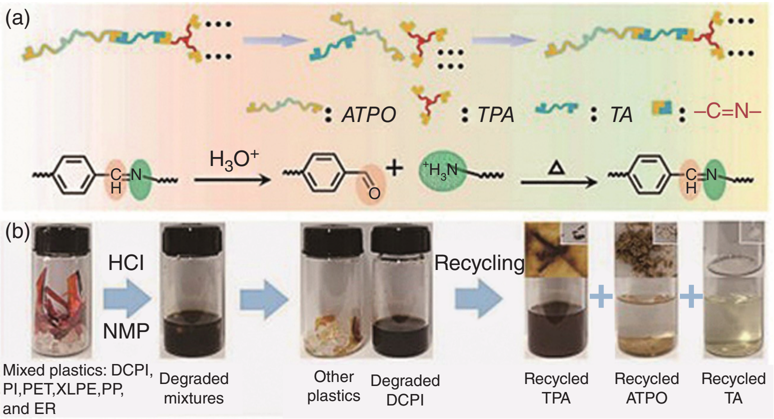


Figure 1.7 (a) Schematic and recycle process of DCPI films. (b) Depolymerization and recycling process of DCPI from the mixed plastic wastes containing PI, PET, XLPE, PP, and ER. *Source:* [26] / John Wiley & Sons.

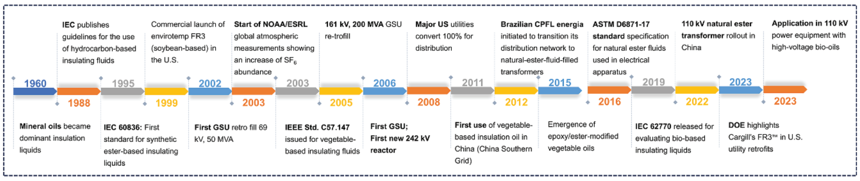


Figure 1.8 The historical development of insulating oil (from mineral oil to eco-friendly and sustainable insulating oil). *Source:* Adapted from [4, 69].

levels, effectively reducing the risk of soil contamination in the event of leakage. In addition, we have summarized the historical development of insulating liquids, from the initial use of mineral insulating oil (1960) to the first synthetic ester-based insulating liquid (1995), and finally to the current use of eco-friendly bio-based insulating oil in electrical equipment (Figure 1.8). This provides a brief overview of how insulating liquids have evolved from initial performance requirements to current environmentally driven green insulation. In the field of solid insulation, researchers are exploring the use of cellulose nanofibers, starch-based polymers, and other biopolymers as dielectric materials. Wang et al. [69] prepared a biodegradable bio-based fluorinated epoxy resin by introducing Schiff base bonds and fluorine substituents into traditional epoxy resins, achieving excellent mechanical and dielectric properties, as well as the ability to be recycled and acid-catalyzed degradation (Figure 1.9).

As the global energy transition towards low-carbon and sustainable development, power systems face the major challenge of reducing greenhouse gas emissions. In the field of gas insulation, traditional SF_6 gas is widely used in high-voltage electrical equipment, such as gas-insulated switchgear and gas-insulated transformers, due to its high insulation strength and excellent arc-quenching performance [18, 70, 71]. However, SF_6 has a GWP as high as 25 200 and an atmospheric lifetime of up to 3200 years, making it one of the primary restricted industrial greenhouse gases. Therefore, the synthesis and search for high-performance, high-safety, and long-term reliable eco-friendly insulating gases to replace SF_6 has become a focus of attention in academia and industry. As shown in Figure 1.10, we have summarized the entire historical development process from the first synthesis of gas media in 1889 to the current use of numerous eco-friendly gases and new eco-friendly insulating gas equipment. This marks that the world has entered an important historical stage of transformation and development in gas insulation.

1.4.2 Smart Insulating Materials

As a new type of material, smart materials have broken through the concept of traditional insulating materials and can better adapt to various complex service conditions. These materials often exhibit some remarkable properties, such as self-warning of health status, self-adaptive changes to the working environment, and self-healing after mechanical and electrical damages.

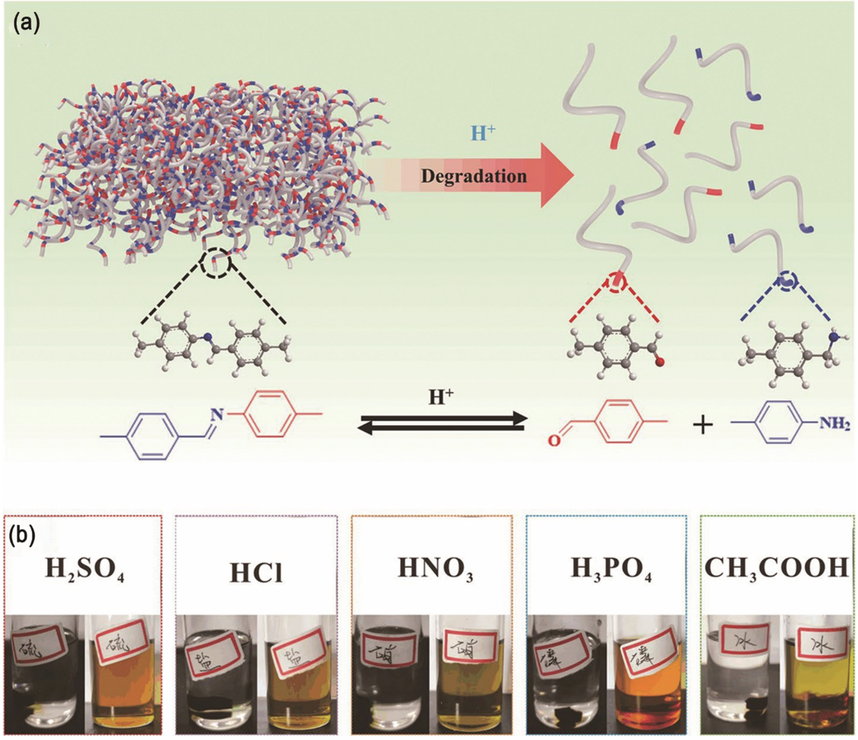
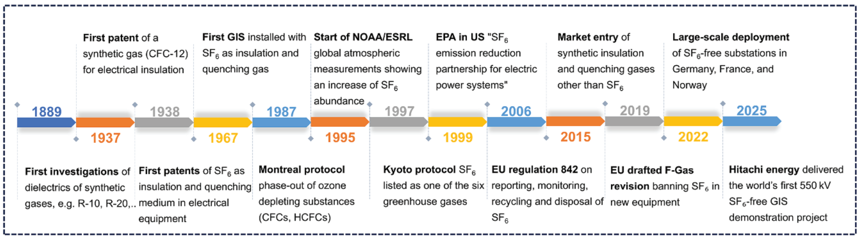


Figure 1.9 (a) Degradation mechanism of Schiff base epoxy thermosets. (b) Digital photographs of SA-BTB-EP/DDM and degradation rates at different acids. *Source:* [69] / Springer Nature.



First investigations of dielectrics of synthetic gases, e.g. R-10, R-20,..

First patent of a synthetic gas (CFC-12) for electrical insulation

First patents of SF$_6$ as insulation and quenching medium in electrical equipment

First GIS installed with SF$_6$ as insulation and quenching gas

Montreal protocol phase-out of ozone depleting substances (CFCs, HCFCs)

Start of NOAA/ESRL global atmospheric measurements showing an increase of SF$_6$ abundance

Kyoto protocol SF$_6$ listed as one of the six greenhouse gases

EPA in US "SF$_6$ emission reduction partnership for electric power systems"

EU regulation 842 on reporting, monitoring, recycling and disposal of SF$_6$

Market entry of synthetic insulation and quenching gases other than SF$_6$

EU drafted F-Gas revision banning SF$_6$ in new equipment

Large-scale deployment of SF$_6$-free substations in Germany, France, and Norway

Hitachi energy delivered the world's first 550 kV SF$_6$-free GIS demonstration project

Figure 1.10 Timeline and key events in the development of insulating gases (from SF$_6$ to eco-friendly gases).

Electrical aging is a major challenge faced by most electrical equipment, and the emergence of smart insulating materials has provided an innovative solution to this issue. Electrical aging often does not significantly change bulk electrical measurements until near failure. A visual or optical early warning could allow operators to replace or repair insulating materials well before failure, improving system reliability and safety. A typical example is the polydimethylsiloxane-based

(PDMS-based) polymer composite material developed by Huang et al., which changes color when subjected to electrical aging [24]. In this material, a specially designed chromophore (color indicator) is incorporated in trace amounts into the insulating material. During normal operation, the material remains stable, and the indicator remains in an inert state, so the insulating material retains its original color (white or colorless). However, if the polymer experiences significant electrical degradation, characterized by the generation of high-energy electrons and chemical byproducts such as free radicals, particularly oxygen free radicals formed through polymer chain scission, the indicator molecules undergo a chemical transformation and shift to a colored state (Figure 1.11a). This results in visually detectable color changes in the aged areas, providing a clear warning signal before complete failure occurs. The advantage of this method lies in its autonomy, as it does not require the embedding of power sources or electronic components in the material. The chemical reaction of the material itself provides the signals, which are continuous and distributed.

During operation, insulating materials inevitably face unfavorable conditions such as voltage fluctuations, sudden temperature changes, and contamination.

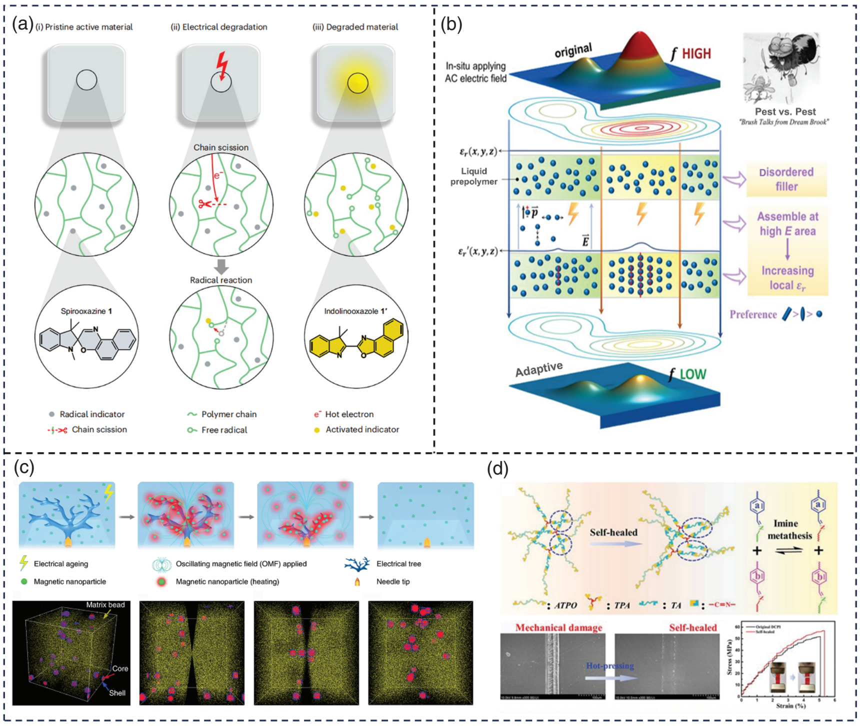


Figure 1.11 (a) Electrical-degradation-indicating materials and visualization of the free radical induced chromogenic mechanism. *Source:* [24] / Springer Nature. (b) 3D schematic of the permittivity-electric field evolution. *Source:* [62] / John Wiley & Sons / CC BY 4.0. (c) Schematic diagram of electric trees self-healing induced by functionalized nanoparticles. *Source:* [23] / Springer Nature. (d) Schematic and mechanism of self-healable process of DCPI films based on dynamic imine bonds. *Source:* [26] / John Wiley & Sons.

Therefore, the development of new self-adaptive insulating materials is particularly important. These materials can sense and respond to local electric field variations, actively adjusting their permittivity or conductivity to alleviate electric field distortions and enhance breakdown strength and system stability [25]. Moreover, self-adaptive insulating materials enable demand-driven design, which reduces redundancy and improves system compactness and cost-effectiveness. They also signify the transition from the “passive insulation” mechanism of traditional insulating materials to the “active regulation” mechanism of adaptive insulating materials, offering unique advantages under complex electrical, thermal, and mechanical stresses. As shown in Figure 1.11b, Shen et al. [62] proposed a fabrication method for adaptive dielectric gradient insulators based on in situ alternating current field-induced self-assembly. High aspect ratio fillers are driven to align in chain-like structures in high field regions, enabling spatial tuning of permittivity and mitigating field concentration. This method integrates electric field responsiveness, self-organized structuring, and structure freezing, demonstrating smart environmental adaptability and providing a new pathway for adaptive high-voltage insulation design.

Additionally, electrical insulation failures are usually caused by microscale defects that grow under the influence of electrical, thermal, and mechanical stresses. If these minor damages could heal themselves, the service life, and reliability of insulating materials could be greatly enhanced. This has inspired researchers to study self-healing insulating materials, i.e. materials that can repair electrical or mechanical damages (or at least mitigate its effects) without human intervention. Research on self-healing insulating materials currently focuses on two types: extrinsic and intrinsic strategies. Among these, the main method for extrinsic self-healing strategies is the microcapsule method, embedding tiny capsules of a liquid healing agent in the solid insulation. When electrical trees or cracks grow, they rupture nearby capsules, releasing monomers or liquid to fill the gaps and polymerize, thereby repairing the insulating material. As shown in Figure 1.11c, by introducing superparamagnetic γ-Fe_2O_3 nanoparticles into the electrical cable insulating material PP, effective self-healing of electrical damage has been achieved [23]. Driven by conformational entropy, surface-modified γ-Fe_2O_3 nanoparticles can detect defects caused by electrical treeing. Simultaneously, these nanoparticles autonomously migrate to the electrical treeing channel region and utilize an oscillating magnetic field to perform localized heating of the damaged area. This process increases the local temperature of the defect region by 24 K compared to the surrounding polymer matrix, thereby triggering targeted melting and damage repair of the PP. Another strategy is to enable intrinsic self-healing

capabilities in polymer networks after damage through molecular structure design. This can be achieved by incorporating dynamic covalent bonds such as disulfide bonds, Diels-Alder bonds, borate ester bonds, imide bonds, and hydrazone bonds, as well as reversible noncovalent interactions including van der Waals forces, hydrogen bonds, ionic bonds, host-guest interactions, and coordination bonds into the network structure [67, 72–77]. The reversible rearrangement of dynamic bonds is used to achieve self-healing after damage, which effectively extends the service life of the material. As shown in Figure 1.11d, the dynamic reversible properties of imine bonds were used to achieve self-healing of PI after mechanical damage, and the self-healed PI basically maintained its original performance level [26].

1.5 Summary and Outlook

The development of insulating materials has been driven by the evolving demands of electrical technology. In recent years, the urgent need for low-carbon, eco-friendly, and sustainable development has also become a significant driving force behind this progress. In this chapter, we first review the evolution of insulating materials, tracing the development from simple materials such as glass, rubber, oil, and paper to advanced engineering materials including cross-linked polymers, SF_6 gas, and composite materials. These advancements have enabled us to rely primarily on polymer dielectric materials for electrical infrastructure, which can maintain excellent properties for decades even under harsh conditions. However, modern materials not only require outstanding performance but must also strike a balance between high-performance, smart, low-carbon, eco-friendly, and sustainable characteristics. This also represents the new set of challenges facing insulating materials today: addressing environmental concerns such as how to recycle, degrade, or safely dispose of large volumes of polymer insulating materials; ensuring long-term reliability by managing performance degradation during service life and enabling self-healing or self-warning after damage; and achieving performance breakthroughs, including the ability to withstand higher operating temperatures and electric fields.

In summary, with the comprehensive implementation of the green transformation of electrical insulation, the research and development of insulation materials is entering a new era centered on low-carbon, eco-friendly, sustainable, and smart. Meanwhile, the research and development of insulation materials has gradually become a multidisciplinary challenge involving electrical engineering, materials science, and chemistry. The accumulated experience over the past century regarding the relationship between material structure and performance is being utilized by researchers to design new types of electrical insulating materials that combine excellent basic properties with multifunctionality. Subsequent chapters will delve deeper into insulating materials from the perspectives of different insulation types and the latest research findings.

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