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Review Article

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REVIEW

Insulating materials for realising carbon neutrality: Opportunities, remaining issues and challenges

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Abstract
The 2050 carbon-neutral vision spawns a novel energy structure revolution, and the construction of the future energy structure is based on equipment innovation. Insulating material, as the core of electrical power equipment and electrified transportation asset, faces unprecedented challenges and opportunities. The goal of carbon neutral and the urgent need for innovation in electric power equipment and electrification assets are first discussed. The engineering challenges constrained by the insulation system in future electric power equipment/devices and electrified transportation assets are investigated. Insulating materials, including intelligent insulating material, high thermal conductivity insulating material, high energy storage density insulating material, extreme environment resistant insulating material, and environmental-friendly insulating material, are categorised with their scientific issues, opportunities and challenges under the goal of carbon neutrality being discussed. In the context of carbon neutrality, not only improves the understanding of the insulation problems from a macro level, that is, electrical power equipment and electrified transportation asset, but also offers opportunities, remaining issues and challenges from the insulating material level. It is hoped that this paper envisions the challenges regarding design and reliability of insulations in electrical equipment and electric vehicles in the context of policies towards carbon neutrality rules. The authors also hope that this paper can be helpful in future development and research of novel insulating materials, which promote the realisation of the carbon-neutral vision.

1 | INTRODUCTION

The excessive consumption of fossil fuels has aroused a severe public concern about global warming [1], posing a serious threat to the world's economic development and even human survival [2]. To control the increase in global average temperature, in 2015, the Paris Agreement required countries and regions to reach a balance between sources and sinks of greenhouse gases in the second half of this century [3]. This balance implies an urgent need to achieve carbon neutrality [4], that is, the total annual CO_2 emissions from all anthropogenic sources being net-zero [5].

So far, 127 countries [6] and over 800 cities [7] have pledged to achieve or are considering the ambitious carbon neutrality target. Yet, different countries are heterogeneous and differentiated in terms of the electric sector composition, access to renewable resources, policies [8], and most importantly, stages of development. For example, China, as the largest coal consumer and carbon emitter in the world [9], has not yet reached the carbon-emission peak, while the European Union and the U.S. peaked in 2006 and 2007, respectively [10]. Owing to the temporally similar goals of realising carbon neutrality, China only has 30–40 years to accomplish what developed countries have achieved in 50–60 years, indicating considerable challenges [2].

While the challenges standing in the pathways to carbon neutrality vary on a country-by-country basis, electricity generally plays a central role in the future transition [11, 12]. Figure 1 presents the fuel and regional flows of CO_2 emissions in 2019 [13]. Given that the electric power generation and transportation sector are the two largest contributors to carbon emissions in most of the countries [14], both decarbonising the electric power generation and electrifying the transportation sector (including heavy-duty, air and marine traffic as well as light-to-medium-duty vehicles) are crucial for the neutrality goal [8, 15]. As a result, simultaneously integrating more renewables during the coal phase-out while investing more in future electric power systems and electric vehicles (EVs) are of vital importance.

Optimistic prospects regarding the development of future electric power system and electrified transportation have been foreseen globally. China, for example, has decided to increase the installed wind capacity to 1500–2600 GW while the solar capacity is expected to reach 2200–2800 GW by 2050, practically 10 times of those in 2020 [16, 17]. In addition, the EV sales in the U.S. are estimated to reach 60% market share around 2040 [18], from the current mere 2% of new sales [19].

The time frame for electricity-sector decarbonisation is short [20], whereas the challenges coming with the construction of future energy revolution are substantial. An open question remains regarding how to reliably and efficiently coordinate future electric power systems and massive electrified transportation as well as their insulation issues. There is also another issue. In the most developed countries, it is not easy to build overhead lines due to the opposition of local communities considering environmental impact and electromagnetic pollution so that in recent years the development of the power grid is done only thanks to compact electric power equipment, especially cables (underground or submarine).

Electric power equipment, which includes cables, transformers, capacitors, gas-insulated switches etc., is responsible for carrying, monitoring and regulating power flow and serves as an important part of the power grid. Being the core section of power equipment, the performance of insulating material directly determines the safety of power equipment. In the context of the construction of new power grid, the traditional power structure is transformed from thermal power to renewables, and the change in operation mode will bring severe challenges to insulating materials. For the newly developed countless EVs (i.e. electric aircrafts, electric ships, and EVs), the challenges faced by insulating materials are even more pronounced, which mainly refer to the changing of working

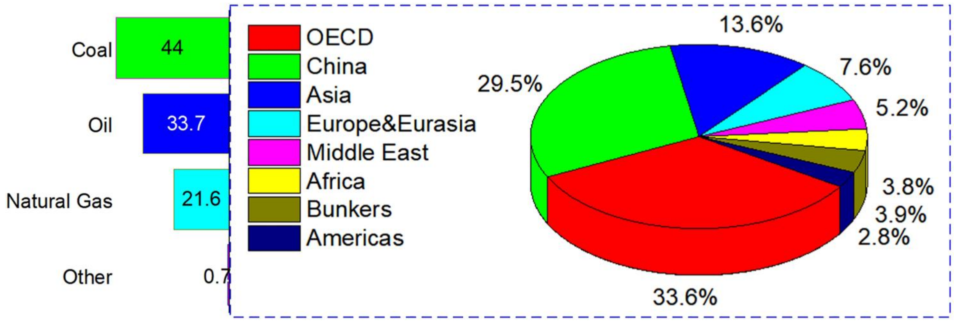

FIGURE 1 Fuel and regional flows of CO_2 emissions in 2019; Bunkers include international aviation and international marine bunkers; OECD members are excluded from Asia, Americas, and Europe and Eurasia [13]

conditions, voltage-mode transformation, as well as the requirements for insulating materials to be used under harsh conditions. For example, the surface charge accumulation of spacers should be solved for DC gas-insulated power transmission line (GIL), which greatly reduces the size of an offshore high voltage DC (HVDC) converter stations; space charge issues should be well-handled to realise the stable operation of DC electric power cables; the lower partial discharge (PD) inception of insulators under low pressure poses potential hazard to the safety of electric aircraft etc.

In addition, reducing the carbon dioxide emission is the key to achieve carbon neutrality. Standalone devices are used to work without need for power, which can be useful for monitoring purpose in isolated environments and offers basis for the future Internet of things (IoTs). Triboelectric nanogenerator (TENG), as a micro-energy harvesting devices, harvests tiny vibrational mechanical energy in the surrounding environment and converts it into electricity, which provides a new approach for future energy architecture and is helpful for reducing carbon dioxide emissions. Moreover, it has great potential in the replacement of massive batteries used to power sensors and wireless components used in future digital electric power system. However, the selection of insulating and more generally multifunctional materials to reach a desired output power should also be considered, especially for some extreme conditions.

Achieving the ambitious goal of carbon neutrality presents more challenges to the upgrading and innovation of insulating materials. This paper first talks about the engineering challenges constrained by insulators in future electric power system equipment and electrified transportation. The key in the development, selection and application of insulating materials in TENG is also discussed. Then, focussing on six key insulating materials, that is smart insulating material, high thermal conductivity insulating material, high energy storage density insulating material, extreme environment resistant insulating material, and environmental-friendly insulating material, their scientific issues, opportunities and challenges under the goal of carbon neutrality are discussed. It is hoped that this paper improves the understanding of the insulation problems of electrical equipment and EVs in the context of carbon neutrality. More importantly, we hope that this paper can be helpful in future development and research of novel insulating materials which promote the realisation of the carbon-neutral vision.

2 | EQUIPMENT INSULATION ISSUES IN FUTURE POWER GENERATION/TRANSMISSION AT LARGE

The leapfrogging of future electric power grid requires the innovation in electric power equipment. Insulation problems caused by increasing of electronic-based components (inverters), DC equipment, harsh working conditions (low pressure, high temperature) etc. have become more prominent. In this chapter, from a macro perspective, the insulation issues of electric power equipment for future power generation/transmission at large are discussed.

2.1 | Offshore wind power generation

Wind power generation does not produce any carbon emissions and is one of the most potential forms of power generation. The offshore wind power transmission calls for cross-sea large capacity electric power transmission [21]. For such kind of application, overhead power transmission lines and high voltage AC (HVAC) cable power transmission are not good choices because of the large construction cost and the large capacitive current, which severely reduce the power transmission efficiency [22]. The offshore wind power transmission based on HVAC and HVDC are currently considered for offshore transmission systems [23, 24]. However, due to the capacitive effect between the submarine cable and the ground, the HVAC has a large reactive current and cannot achieve long-distance power transmission. The longer the transmission distance of the HVAC power frequency AC mode, the smaller the active power that can be transmitted. In terms of construction cost, when the transmission distance exceeds the cut-off point (usually 50–100 km), the cost of HVDC direct current transmission is lower, as shown in Figure 2 [25]. Therefore, HVDC cable power transmission becomes the best choice for long-distance offshore wind power, which is also the main direction advocated by CIGRE [26].

2.1.1 | DC gas-insulated equipment

As an alternative plan to air-insulated substation, DC gas-insulated substation, which greatly reduces platform area to a maximum of 90%, has great potential to be used in the offshore platform [27, 28]. Spacers support the high voltage conductor in DC gas-insulated equipment [29], and their surfaces attract charges which modify electric field and greatly affect the safety of DC gas-insulated equipment.

The surface charge behaviours obey the field-dependent theory as shown in Figure 3 [30]. To be more specific, the surface roughness and micro morphology of conductors as well as the insulating gas property all affect the free charge in the gas side, and these charges play a significant role in determining the surface charge distribution. Homo-polar charges are usually due to both bulk charge transport and homo-polar charges from gas ionisation near HV conductor. Hetero-polar charges, on the contrary, are usually due to the gas ionisation over the grounded surface and polarisation near the homo-polar charge with high charge density, presenting as separated charge clusters [31]. At present, the charge origins have been sufficiently studied. However, concerns over surface charge accumulation, especially for insulation with complicated shapes and in real sized spacers inside GIL, are still lack of research. There is still very limited published research on real-sized spacers, and the industrialisation cases of DC gas-insulated equipment is few.

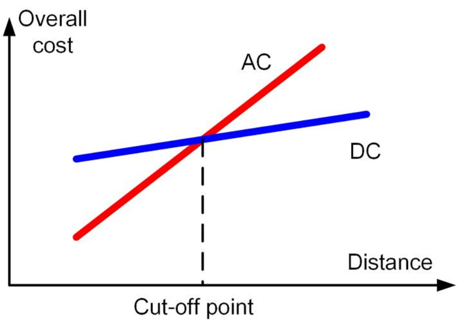

FIGURE 2 The relationship between the overall cost of the high voltage AC and high voltage DC power transmission systems as a function of transmission distance [25]

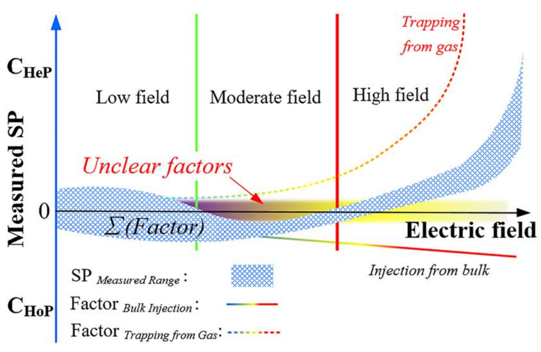

FIGURE 3 Field-dependent theory. SP, Surface potential; C_{HoP} stands for homo-polar charge; C_{HeP} stands for hetero-polar charge [30]

Apart from issues in terms of surface charge accumulation, SF_6, which has long been used as the main component of the gas insulation, has significant impact on the global warming. Investigating future environmental-friendly insulating gas has become a hot topic [32, 33]. Also, forward-looking research regarding recyclable solid insulating materials (i.e. polyethylene terephthalate [PET] and its composite materials) as a replacement of traditional epoxy composites for spacers has attracted more attention in recent years. More details regarding the above-mentioned environmental-friendly insulating materials are discussed in Section 5.5.

2.1.2 | DC cables

The development of polymeric cable insulating materials has gone through from natural rubber, polyvinyl chloride (PVC), synthetic rubber, polyethylene and crosslinked polyethylene (XLPE) [34–38]. At present, XLPE is the most popular HVDC cable insulating material, which is synthesised from the crosslinking of polyethylene. The long-term operation temperature of XLPE cables can reach 90℃ in HVAC cables [39, 40]. However, the crosslinking process transforms thermoplastic polyethylene into thermoset XLPE, which is very difficult to be recycled. Traditional methods to disposing of used XLPE are burning, pyrolysis or burying, which not only consume lots of energy but also pose negative impacts on the environment. The crosslinking and degassing process in the manufacturing of XLPE cables is very complicated and also results in a long production time and large energy consumption. The crosslinking by-products in XLPE, which could introduce microdefects and cause space charge accumulation [41–43], are always a threat to the safe operation of the HV cables. In view of the above shortcomings, XLPE cables can neither meet the requirement of large-capacity power transmission nor meet the requirement of sustainable development.

To mitigate the disadvantages of XLPE, thermoplastic materials, that is polypropylene (PP), are proposed to replace XLPE. The thermoplastic insulating materials are considered to possess excellent electrical insulation performance and also can be recycled [44–46]. The thermoplastic insulating materials do not require any crosslinking, thus completely avoiding the crosslinking process and consequently the adverse effect of the crosslinking by-products, energy consumption and CO_2 emission in the crosslinking process. The thermoplastic insulating materials are also anticipated to be recyclable, which not only have significant technique advantages but also possess considerable economic benefits. Results from a CIGRE working group have shown that compared to XLPE, the cost of thermoplastic material and overall cable system can be reduced by 14% and 17%, respectively, and the carbon emission during cable manufacturing can be reduced by 20% when using thermoplastic materials instead of XLPE [47]. In recent years, research on recyclable cable insulating materials has been extensively conducted [48, 49].

It is worth mentioning that the insulation issues of inverter motor which is used in wind power plant will be discussed extensively in Section 3.3.

2.2 | High voltage motors

As more and more renewables are integrated in the modern electric power systems, high voltage motors have been extensively implemented in various electric power generation sectors including the prevalent doubly fed induction generator (DFIG) in the wind power industry, hydro generators, among others. Electrical insulation systems are essential to the reliability of wind turbine generators and the DFIG equipped with a pulse width modulation (PWM) converter needs special measures of insulation design [50]. The inter-turn insolation of the HV motors may fail under PWM wave although the applied impulse voltage is much lower than the breakdown voltage of the dielectric, which has been explained by the accumulative insulation failure. Regarding the hydro generator, the inter-turn insulation failure may cause significant damage to the whole motor system including stator winding and core [51]. It was reported that more than 56% percent of failed power generators showed insulation damages [52].

2.2.1 | Ageing mechanisms

In large-capacity, high-voltage generators, premature failure of the winding insulation is initiated by corona discharge. Considering the special insulating structures in high-voltage generators, extremely uneven electric field is unavoidable especially for the areas with the small radius of curvature and short insulation distance such as the ventilation trenches of the iron core, the grooves out of the motor stator rods, and the inter-turns spacing of the wire rods etc. [53]. Corona discharge events may cause permanent damage on polymer dielectrics by ultraviolet luminescence, the collision of high-energy ion/electron and chemical reactions [54], which produces degradation by-products, accumulating on the surface of the defect voids and further intensifying PD activity [55]. As continuous corona discharge occurs, exposure to light, heat and electromagnetic energy cause local high temperature and accelerated ageing processes of the dielectrics including softening of insulating adhesives, carbonisation, oxidation etc. During the above-mentioned degradation process by the corona discharge, the surfaces of the insulation become rough and the discharge starts to be concentrated in the pits and develops into electrical tree damages inside the materials and finally lead to breakdown.

Besides the corona discharge events that drives the surface accumulative degradation of insulating materials, repeated pulses without initiating PDs can also cause insulation ageing and failure [56]. In this case, surface and space charge of the dielectrics play significant roles in the accumulative failure of the insulation. According to the space-charge-limited current (SCLC) theory, intense space charge injection may occur when the applied electric field exceeds a threshold. Under AC electric field, a large amount of charge injection/extraction causes recombination of trapped charges and releases energy to other electrons via radiation and forms high-energy charge carriers, which can attack and degrade polymer dielectrics [57–59]. Surface and space charge accumulation has been demonstrated as a cause of electrical ageing of dielectrics due to local field enhancement and electromechanical effect [60]. The Dissado–Mazzanti–Montanari (DMM) model was developed to explain the charge-induced long-term degradation of insulation [57, 61, 62].

2.2.2 | Corona resistant dielectrics

The rise of nanocomposite insulating materials has promoted the research in corona resistant dielectrics. In 1988, Johnson et al. found that the addition of a certain amount of nanoscale silica or alumina to the polymer can greatly improve the corona ageing resistance of the polymer matrix, and the nanocomposite insulating materials were widely used as the ground insulation of the mould-wound generators [63]. About 10 years later, DuPont together with ABB and Siemens developed corona-resistant polyimide films Kapton-CR and Kapton-FCR with the corona resistance 500 times higher than that of the original polyimide, which have been widely used in high-speed electric locomotives [64]. The Kapton-CR film has a sandwich structure, the middle layer is polyimide, and the upper and lower layers are 50–500 nm thick polyimide-based nanocomposite containing 10–30 wt% fumed alumina nanoparticles. Phelps Dodge Industries and GE have also published similar patents stating that the addition of 5–1000 nm of metallic or non-metallic oxide nanoparticles, such as TiO_2, SiO_2, Al_2O_3, ZrO_2, ZnO etc. to polymers such as polyimides, polyamides, nylons, epoxy resins etc., can greatly improve the ability to withstand the ageing of PWM pulse overvoltage.

It should be noted that the content of this section focusses on the insulation problems of HV motors, while the issues for inverter-fed motors for EVs are extensively discussed in Section 3.3.

2.3 | Power transformers

The invention of power transformers in the late 19th century enabled the long-distance bulk transmission of electrical power through HVAC power networks [65]. For future electric power system, there is no doubt that power transformers will continue playing an important role in the HVAC/HVDC hybrid power networks. Although dry-type transformers and gas-insulated transformers are generally utilised, the majority of power transformers used in the power networks are liquid immersed, where solid–liquid composite insulation systems are adopted.

To reduce the carbon footprint of power transformers, a complete life cycle assessment (LCA) including raw material selection, design, manufacture, transport, operation, maintenance, disposal and recycling is required. Based on over a century's experience, design and operation of power transformers have been highly optimised, with peak energy efficiency reaching over 99%. However, due to the sheer number of units and the long lifetime of power transformers, transformer losses including no-load loss and load loss become a major contributor to the carbon footprint of a transformer. For example, in Europe only, an estimated 4.5 million distribution transforms would produce 38 TWh of electrical losses and contribute 30 million tons of CO_2 emissions every year [66]. The European Commission set up an Eco-design Directive as early as in 2005 and issued the EU regulation 548 in 2014, a two-tier approach to further reduce transformer losses and hence improves the efficiency of the transformer [67]. One of the promising solutions is to adopt amorphous core materials, which can significantly reduce the transformer no-load losses (core loss). A case study showed that amorphous core-based distribution transformers can reduce the environmental impact by 40%–60% compared to the standard core-based transformers [68].

In terms of selecting insulating materials for new transformers, one recognised solution to reduce the environmental impact is replacing fossil-based mineral oils by alterative fluids, for example, ester liquids. Ester liquids including natural esters and synthetic esters are readily biodegradable and have high fire point, which contributes to improving the sustainability and fire safety of transformers. Ester liquids have been widely used in distribution transformers and increasingly used in transmission transformers up to 400 kV rating [69–71]. Ester liquid immersed transformers have also gained particular interests in safety critical applications, for example offshore platform, underground installation, data centre etc. Another approach to improve the usage of raw insulating materials is to adopt high temperature insulation systems in transformers, which can either increase the transformer thermal rating or prolong the transformer thermal lifetime. Options such as combining ester liquids with thermally upgraded paper or aramid paper are being widely investigated by the industry.

As for the large fleet of transformers in operation, oil regeneration including physical reconditioning and chemical reclamation is a well-developed technology to remove harmful chemicals such as printed circuit board (PCB), corrosive sulphur, general ageing by-products and hence improving the transformer health condition [72, 73]. However, it is noted that most of the present reclamation technologies involve high temperature burning-based reactivation process of the sorbent materials, which may produce harmful emission and may not be energy efficient. Research on developing alternative sorbent materials or the regeneration technique that have low environmental impact is encouraged.

Last but not the least, after transformer decommissioning, recycling of the transformer materials including copper, steel and oil will play a crucial role for reducing the transformer carbon footprint. Recycling has already been implemented to certain extent, but there is a clear potential for industry-wide coordination and optimisation of the process. This will ultimately extend the so called 'cradle-to-grave' analysis to 'cradle-to-cradle' analysis, a full circularity-based analysis to reduce the transformer carbon footprint and to improve the sustainability as a whole [66, 74].

2.4 | Power capacitors

In the new power system under the background of carbon neutrality, the massive access of alternative energy and EVs has accelerated the demand for distributed energy storage and flexible power transmission [75], among which power capacitors play an important role in many application fields [76]. Power capacitor is the core component of reactive power compensation devices and power electronic converter (PEC) valves to improve the power factor and achieve flexible transformation of the current type in the distribution system. AC filter capacitors reduce the harmonic components in current waveforms overlapped to the fundamental frequency, while the DC-link capacitors [77] are placed in the DC system to minimise ripple and voltage fluctuation, supporting the construction of a cleaner and more stable power network. According to the type of dielectric, power capacitors are classified [78] as aluminium electrolytic capacitors, oil immersed capacitors and dry film capacitors, in which metallised organic film capacitors have been rapidly developed resulting from their dominantly advantages, including low Equivalent Series Resistance (ESR), high stability of the capacitance value, withstanding of high peak voltage, maximum flexibility of adaption to the shape of the available space, environmentally friendly, and low risk of explosion. It is prepared by stretching a polymer film to a thickness of less than 10 microns followed by surface metal evaporation, slitting, winding, welding and final packaging [79]. At present, one of the commonly used dielectric films in commercial capacitors is biaxially oriented polypropylene (BOPP). It has many significant advantages [80] such as high self-healing properties and low and stable dissipation factor. However, the energy storage density of BOPP film is limited due to the low dielectric constant ($\varepsilon_r = 2.2$) of PP [81] and also cannot maintain stable work in high temperature environment [82]. The operating environment of the new power system possessing new features such as high voltage, large capacity and high proportion of power electronics has brought advanced challenges and requirements for capacitor insulation systems. The most highly anticipated improvement is the energy density of dielectric films [83]. Higher energy storage density will significantly reduce the volume and weight of the capacitor device given the same capacity. By incorporating inorganic ceramic particles into polymer matrix to form nanocomposites, the energy density has been significantly improved in many studies [84]. By increasing the capacity of the capacitors, more ohmic and dielectric losses will be generated, and it will significantly increase the operating temperature of the device that is destructive for the operational life of capacitors. In order to maintain the constant temperature of capacitor devices, the thermal conductivity of the dielectric material needs to be promoted [85], however, not at the expense of insulating performance. Some other properties [86, 87] such as PD resistance characteristics, self-healing ability, and the mechanism of electrode suppression of carrier injection also need further improvement to meet the challenges of future electric power grids.

2.5 | Power electronic devices and systems

Building a new type of power system with renewable energy as the main body is an important approach to achieve the goal of 'carbon neutrality'. The new power system evolution is characterised as the 'double high' including a high proportion of renewable energy and a high proportion of power electronic equipment. With the development of WBG semiconductors, the trend that the WBG devices replace their silicon-based counterpart is inevitable [88]. The WBG device has higher breakdown voltages, achieves faster switching speeds and lower switching losses, and withstands higher temperatures. To improve power conversion efficiency, increase power density and reduce system cost, power electronic devices tend to be designed towards high operating voltage, with their size getting smaller and smaller. However, the increased rated voltage and temperature directly lead to greater pressure for the packaging insulating material [89]. The insulation in power electronic devices and systems endures complex coupled electrical, thermal, and mechanical stresses, which urges the prominent need for research on failure mechanisms and condition evaluation [90].

The insulating materials used in power electronic devices and systems mainly include substrate and encapsulation insulating materials. The substrate materials provide both insulation and mechanical support for the power devices [91]. Additionally, the substrate must have good thermal conductivity to effectively dissipate the heat generated during the operation of the devices [92]. Table 1 lists four commonly used substrate materials and their performance comparison with SiC, the most popular WBG semiconductor material. Al_2O_3 ceramic tops the list over aspects of technical maturity and price. Besides, its coefficient of thermal expansion (CTE) is relatively large, and the dielectric constant is high. But compared with other ceramic materials, the thermal conductivity is very low, so it is not ideal for heat dissipation in high power density systems. BeO ceramic has the highest thermal conductivity, but the dust particles formed in the production are toxic and harmful to the human body and the environment. AlN ceramic is a relatively safe and promising material, its thermal conductivity is relatively high and its CTE is close to that of SiC. Si_3N_4 is another alternative material with a similar CTE to that of SiC, and it has the highest mechanical fracture toughness.

The role of the insulating material for encapsulation is to provide additional insulation and protect the power chips and metal interconnects from harsh environments such as moisture, chemicals, and dusts etc. [93]. Table 2 lists some commonly used insulating materials for encapsulation. Silicone gel is the most widely used encapsulating material, but it is generally used under 250℃. To improve the temperature resistance of silicone gels, researchers used inorganic fillers or modified silicone elastomers as encapsulating insulation, which can withstand temperatures above 250℃ over a certain time. Some polymers such as Polyimide (PI) and Parylene are also candidates as passivation agents on the chip surface to prevent the breakdown of the off-chip insulation under high voltage. In addition, thermosetting materials such as epoxy resins have sufficient mechanical strength and are also used as hard encapsulating materials, especially for the discrete small-sized power electronic devices with pin legs. However, hard

TABLE 2 Main properties of encapsulating materials

Material	Permittivity	Breakdown strength (kV/mm)	Temperature range (℃)
Silicone gel	2.79 (100 kHz)	16–20	−80–200
Polyimide	4.2 (1 MHz)	100–280	≤280
Parylene	2.65 (1 MHz)	275	≤260
Epoxy resin	3.3–4.0 (1 MHz)	35–40	−55–125

TABLE 1 Main properties of selected insulating materials for substrates

	Material	Al_2O_3	AlN	BeO	Si_3N_4	SiC
Mechanical properties	Tensile strength (MPa)	127.4	310	230	96	
	Flexural strength (MPa)	317	360	250	932	
	Elastic modulus (GPa)	310.3	310	345	314	500
	Density (kg/m^3)	3970	3260	3000	2400	
	Fracture toughness ($MPa \cdot m^{1/2}$)	3–5	2–3	1–2.5	4–7	
Thermal properties	Thermal conductivity (W/m · °C)	24	150–180	270	70	250
	CTE (ppm/℃)	6.0	4.6	7.0	3.0	3
Electrical properties	Resistivity (Ω cm)	$>10^{14}$	$>10^{14}$	$>10^{14}$	$>10^{10}$	0.02
	Dielectric loss ($\times 10^{-4}$)	3–10	3	3	2	
	Breakdown strength (kV/mm)	12	15	12	10	
Other	Relative cost	1×	4×	5×	2.5×	

Abbreviation: CTE, coefficient of thermal expansion.

encapsulating materials may exhibit cracks during thermal cycling, while soft encapsulants are thermally unstable at high temperatures. Therefore, the selection of encapsulating materials is often a trade-off between thermal stability and flexibility [94].

3 | INSULATION IN ELECTRIFIED TRANSPORTATION

Electrification is widely considered an attractive solution for reducing the oil dependency and environmental impact of transportation so as to achieve the goal of low carbon [95]. Insulating materials and systems for electrified transportation face new challenges due to voltage types and harsh environmental working conditions. This chapter discusses the emerging insulation concerns for the propulsion system of more-electric aircrafts and more-electric ships, as well as insulation issues for inverter motors used in road electrified transportation including electric cars, wind power generators, and high-speed railways.

3.1 | Insulating materials in more-electric aircrafts

The need to reduce the CO_2 emissions has become a growing challenge for the aviation industry. Approximately 98% of the world's aviation CO_2 is produced by aircraft with a gross take-off mass exceeding 25 metric tons. Figure 4 shows the fuel consumption sorted by aircraft class, from which it can be seen that the commercial single-aisle and twin-aisle airliners account for more than 90% of the fuel usage with regional jets consuming another 5% [96]. Electrifying these aircrafts would reduce CO_2 emissions one step further.

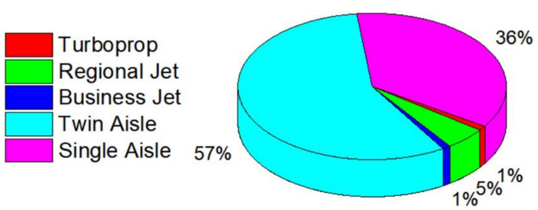

FIGURE 4 Fuel consumption by aircraft class [96]

However, the utilisation of a high throughput hybrid-electric propulsion systems of aircraft requires a power system rate up to tens of MW. From this standpoint, the corresponding voltage rate of the propulsion system shall be boosted to up to several kV. For example, an all-electric propulsion system for a twin-aisle aircraft requires at least 50 MW during takeoff. NASA estimates that the future hybrid-electric aircraft will need to operate at a maximum voltage of 20 kV, and its insulation must be able to withstand up to ~41 kV [97].

The increase in the voltage and power outputs has raised significant concerns in the degradation of insulating materials for power cables and electronic modules for the propulsion system, especially at elevated altitude conditions [97]. For example, according to the Paschen's curve, the gas breakdown voltage of two parallel plates with an air insulation distance of 3 mm at sea level is 10 kV. However, at an altitude of 10 km, arc will be triggered for the same electrode setup even for an air gap of ~10 mm. More importantly, the PD becomes a concern especially on insulator surface where air molecules are ionised. Cables as well as PCB in the propulsion system faces PD issues, and their insulation breakdown voltage should be even lower due to surface charge accumulation at DC voltage. In practice, according to IPC-2221B, the required insulation distance at 10 kV for components on a printed circuit board at sea level is 50 mm, which reached 15 times the Paschen's curve distance.

Table 3 summarises the requirements and emerging problems existed in the insulators for future more-electric aircraft [97]. The impact of heavy load resulting in an elevated operating temperature, as well as low gas pressure environment have placed stringent requirements on the insulator. To solve surface charging issues and developing insulating material and insulation system for the power cables, cable connectors as well as electronic components used in the propulsion system of more-electric aircraft are utterly important for developing the future hybrid-electric aircraft to reduce carbon emission. Further, the evolution to onboard higher transmitted power impacts cable technologies [98]: future cables will probably import more from high power transmission technologies, with introducing multi-layer extruded insulations instead of taped insulations, use of screened cables and moving to HVDC networks [99–102].

TABLE 3 Requirements and emerging problems for the insulation system for future hybrid-electric aircraft [97]

Key elements	Requirements	Emerging problems
Cable and accessory	Be able to operate at low pressures with high endurance to PDs; Alleviated effects on insulation in case of dv/dt; Be able to perform under high electric stress and temperature/temperature gradient subjected to the increased power density.	Over voltage at cable terminals; Tracking at insulator surface; Insulation ageing due to continuous PD.
Propulsion system	Sufficient clearance distance or capable insulation barriers for components in PCBs; PD resistance for IGBTs and PCBs especially at elevated temperatures; Be able to withstand PDs in encapsulating materials.	Surface PD, tracking or arc on the PCBs at low gas pressures; Electric treeing issues at low pressures and high temperature for the power encapsulating modules.

Abbreviations: PCB, printed circuit board; PD, partial discharge; IGBT, Insulated gate bipolar transistor.

These evolutions go with a reconsideration of presently used insulations mostly made of PI and polytetrafluoroethylene (PTFE) materials.

3.2 | Insulating materials in more-electric ships

Today, CO_2 emissions caused by maritime transportation account for roughly 3.3% of the global greenhouse gas emissions [103]. If left unregulated, this is expected to increase by more than three folds in the next 30 years [103]. A promising path towards addressing the environmental challenge is the electrification of ships. Higher energy efficiency, versatile ship design, and the possibility of incorporating clean energy sources provided by electrification lead to lower fuel consumption and carbon emission [104]. While the main reason for the maritime transportation electrification is to increase efficiency, reduce fuel consumption, and reduce carbon emission, various functional and strategic advantages are also enabled by going electric. These additional benefits include improved controllability, ship design flexibility [105], reduced noise and vibration [103], enhanced resilience [106], and versatile energy utilisation for high energy loads [106]. Unlike conventional ships that use mechanical shafts and gear boxes to transfer energy from a prime mover to propulsion systems, electrified ships utilise an onboard power system to distribute energy from power generators to propulsion systems and loads [103]. Most electrified ships today operate on AC shipboard power systems. However, advances in power semiconductors and PEC topologies render the use of DC distribution systems more favourable over AC [106]. Unlike AC that adds complexity due to reactive power and synchronisation among generators and loads, DC, being frequency agnostic, enables each generator to operate at the speed of their highest efficiency, which leads to overall efficiency improvement and lower energy consumption [104]. In addition, DC facilitates the use of alternative sources such as fuel cells, battery storage, flywheel, and eliminates the need for bulky low-frequency transformers. Developments in power electronics enabled significant advances in the electrification of ships. These have led to higher power density, higher efficiency, and multi-mission capabilities [104, 106]. Advanced PEC topologies and WBG power semiconductors including silicon carbide (SiC) and gallium nitride (GaN) devices, and power electronics building blocks (PEBBs) [107–109] enabled flexible and efficient shipboard medium-voltage direct current (MVDC) power distribution system that integrates multiple energy sources and loads.

Although it is clear that the electrification of ships offers both environmental and functional benefits, its fruition relies on the maturation of a range of new technologies and the proper handling of a set of emerging challenges. One of the most fundamental challenges that must be addressed is the challenge associated to dielectrics and electrical insulation. As existing and future ships are becoming more electrified, power generators, energy storage systems, and loads will be interconnected through PECs [110, 111], which leads to unique insulation challenges. However, electrical insulation coordination practices tailored to shipboard conditions are yet to be established. Much of dielectrics research reported has been application- or component-specific. To name a few, electric field analysis and high voltage experiments have been performed to investigate the impact of the spacing and the sharpness of power modules traces [112]. The triple point, where ceramic substrate, metallisation layer, and encapsulant join together, of direct bond copper (DBC) power substrates has been coated with high permittivity materials to increase partial discharge inception voltage (PDIV) [113, 114]. In some cases, additional copper metallisation layers have been placed into power substrates as equipotential rings for electric field grading [115]. When it comes to dielectrics research on power converters, PD characteristics in multilevel converters and fibre optic sensor array have been implemented to monitor PD [107–109]. In the case of dielectrics for cables and cable accessories, dielectric ageing behaviour under repetitive voltage stresses and polarity inversion on DC cables have been reported [116]. Also, medium-voltage cable accessories such as cable termination and cable joints were assumed to be utilised in PEC-driven systems, and the accelerated ageing caused by PD have been analysed and reported [117]. Studies on the insulator design of laminated busbars that provide low-inductance electrical connections and enable the operation of WBG power semiconductor at full-capacity have also been reported [118]. Correlations between defects including protrusions, cracks, and cavities that may exist in the bulk of an electrical insulator or in the conductor-dielectric interfaces and the likelihood of PD have been analysed [119]. In the case of high-frequency transformers, which provide galvanic isolation in MVDC PEC-driven systems, one of the factors that limits the size reduction potential enabled by the high-frequency switching of WBG power modules is electrical insulation [120, 121]. The effects of high-frequency voltage and high slew rate (dV/dt) have been reported [122, 123]. Finally, in the case of rotating machines, stator ground wall insulation that is subjected to high electrical stress compared to other parts has also been reported [124].

While a vast amount of knowledge has been generated through these component-level studies, further studies must be conducted to establish dielectrics and electrical insulation guidelines for all electric ships. A well-established guideline should be developed accounting for the fact that shipboard power systems are ungrounded [125–127] and that the atmospheric conditions are significantly different than terrestrial power systems due to the humidity and salinity of ship atmosphere [128].

3.3 | Insulating materials in inverter-fed motors

As more and more renewables are integrated in the modern electric power systems, invertor-fed motors equipped with frequency conversion, speed regulation systems have been extensively implemented in various electric power generation and transportation sectors including wind generators, high-speed railway as well as electric cars [129]. The global EVs fleet expanded significantly over the last decade, underpinned by supportive policies and technology advances. EVs are one of the solutions to meet global goals on climate change. Three main parts of EVs are motors, batteries and inverters in which electric motor fed by the inverter is the critical equipment of EVs. Industrial surveys and other studies have shown that 70% of rotating machine failures are due to insulation failure [130]. Hairpin winding motors are getting attraction to be used in EVs. Polyamide-imides (PAI) is the main insulating varnishing for these hairpin motor winding as shown in Figure 5a [131].

In automotive industry, a number of challenges are coming down the pipe specifically related to EVs. Typically, today we are using a lot of low voltage materials but there are requirements going forward towards a high voltage. The challenges are that the industry is pretty much divided between low voltage high current or high voltage low current and the penalties that exist with either option are potential compromises to performance and reliability. Currently, there are two schools of thought regarding EVs. One considers low voltage 48 V with high current up to 1200 A, and the other one, high voltage around 1250 V carrying around 50 A. The challenges that exist from that point of view are the safety measure and insulation. If we compare 15 years of safety measurements for combustion engine vehicles and EVs and if a car accident happens or insulation break during the 15 years operation, then a 1200 amps EV can cause severe damage to life as compare to combustion engine drive. So, what is tending to happen is moving towards high voltage low current as much because that

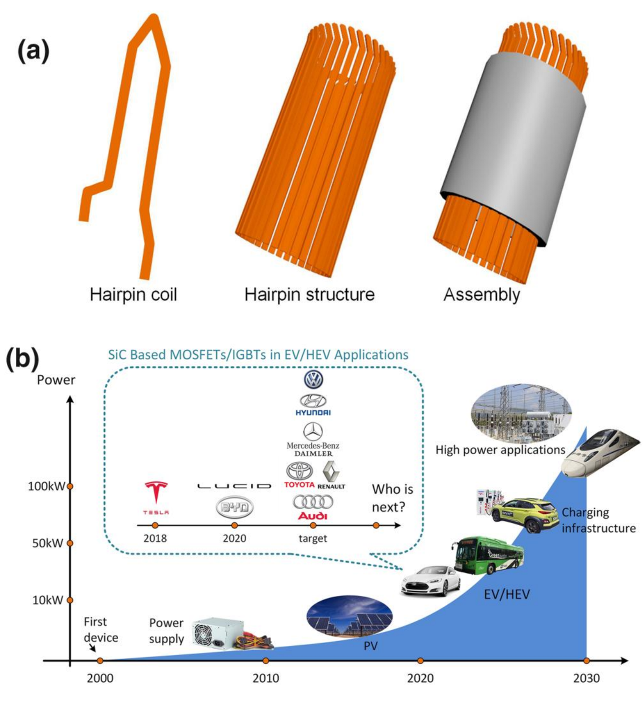

FIGURE 5 (a) Hairpin winding insulation for electric vehicle motors. (b) Power rating of wide bandgap power electronics inverters [131, 132]

reduces some of the burdens that otherwise have to be put in place when you are dealing with high current loads.

In recent years, SiC and Insulated gate bipolar transistor (IGBT)-based wide bandgap devices have increased the switching frequency and current carrying capacity of inverter-fed motors as shown in Figure 5b [132]. The fast-switching frequency and high dV/dt of inverters have raised the electrical stress at the motor terminals and deteriorate the motor insulating material [133]. The discharge phenomenon is much more complex under PWM waveforms. The PWM waveform parameters such as frequency, rise time, overvoltage, duty cycle, dead time and multilevel voltage waveforms have brought some challenges of non-uniform electric field distribution, partial/surface discharges and thermal transients, especially at the interface of motor slot and magnetic coil triple junction. The reliability of an electric motor depends upon its healthy insulation system. An extensive research work on bulk modification such as mixing nano/micro particles into the base polymer matrix and surface modification such as plasma treatment, fluorination, magnetron sputtering, and ion beam implantation has been adopted to enhance the electrical and thermal properties of these insulating materials. Pablo Gómez and M. Khalil Hussain from Western Michigan University, USA have developed some motor simulation models under PWM waveforms [134]. Jiandong Wu group of Shanghai Jiaotong University and the international group of Haoyang you from Ohio State University, USA have presented discharge phenomena under DC and PWM waveforms [135, 136]. T. Lebey from LAPLACE lab of Toulouse University, France has presented PD detection in motor winding under different pressure [137]. A group of scientists (Andrea Cavallini, Davide Fabiani, G. C. Montanari, Wang Peng and Shakeel Akram) from University of Bologna (Italy), Florida State University and Sichuan University have done most of the PD detection work at AC, DC, sinusoidal and PWM voltage waveforms under high frequency, short rise time and varying duty cycle [138–141].

4 | INSULATION IN TRIBOELECTRIC NANOGENERATOR (TENG)

By 2025, more than 30 billion objects would be linked to the IOTs, and a gigantically huge quantity of sensors will be used to monitor the status of all these objects [142]. Although the power required for each sensor is small, typically in the microwatt to watt range, the quantity of these sensors is sufficiently large. Considering the limited endurance and the huge resources for replacing backup power as well as the negative effect they bring to the environment, these huge numbers of batteries pose difficulties for achieving the goal of carbon neutrality. As an emerging energy harvesting method based on natural micro-energy harvesting, TENGs contribute to solve the problem of powering microelectronic devices as an approach for greatly reducing carbon emissions, which supports the carbon neutrality target.

TENG operates by a conjunction of triboelectrification and electrostatic induction through the contact-separation or relative sliding between two materials that have opposite tribopolarity [143, 144]. When two different materials get contacted, one surface is positively charged, and another is negatively charged. The voltage is induced between attached back electrodes when the two parts separate, and charge flows from external circuit, which provides periodical AC power.

Insulating material, as the key component for the friction layer, is the most important part of TENG. On the one hand, it generates electric charge when rubbed, which makes the potential between the two electrodes different, and on the other hand, it isolates the two electrodes to ensure that the electric charge flows in the external circuit, which is a necessary condition for the TENG.

The most frequently used insulating materials in TENG are polymers, especially those containing F element that are more likely to generate high-density surface charges [145, 146]. Therefore, PTFE and fluorinated ethylene propylene copolymer (FEP) are more favourable insulating materials for TENG [147]. With the advantages of biocompatibility, transfer-printable and transparency, polydimethylsiloxane (PDMS) is often used as some implantable or optical energy harvesters [148]. As for PI, its high temperature resistance also allows it to be used as a friction layer for TENG in some applications to maximise its output [149]. However, the manufacture of fluoride polymer is often accompanied by higher carbon emissions, while some biomass materials can minimise the carbon emissions of TENG. For example, very recently, some leaves with hydrophobicity in nature are found as very good insulating materials, which not only have good electric output but also reduce carbon emissions [150].

The selection of insulating materials for TENG is very wide, because essentially any two pieces of materials can have charge transfer when being rubbed, which constitutes the friction material pair of TENG. However, in practical applications, the selection of an excellent insulating material is not easy. For example, to maximise the output performance of TENG, material pairs with large differences in charge polarity should be selected as the friction layer, which requires a deep understanding of the charge transfer mechanism and triboelectric series. Since the insulating material in TENG undergoes continuous friction to replenish the charge, the wear problem is also very serious, and how to reduce the loss of the friction material is particularly important. On the other hand, the energy loss caused by friction force would reduce the output performance of TENG. Therefore, a material with high electrification ability and low friction coefficient is preferable. In addition, for some special environmental conditions such as high temperature, high humidity, human body etc., insulating materials are required to have better adaptability. Therefore, insulating materials with special properties such as high temperature resistance, super hydrophobicity, and biocompatibility have good application prospects. Apart from the above-mentioned concerns, with the continuous improvement of the output of TENG, the surface charges of insulating materials can accumulate to a very high level, more than 1 mC/m^2. How to keep these charges on the surface of the material for a long time without breaking down the material is also a serious challenge for the future development of insulating materials.

5 | FUNDAMENTAL ISSUES AND CHALLENGES FOR INSULATING MATERIALS

Based on the above-mentioned application background, many insulating materials have received increasing attention on the road to carbon neutrality. In this chapter, fundamental issues and challenges for these insulating materials, which include smart materials, insulating materials with high thermal conductivity, high energy storage density insulating materials, insulating materials for harsh condition, and environmentally friendly insulating materials, are discussed.

5.1 | Smart materials

Smart dielectrics, defined as functional composite materials, whose insulating properties can be automatically or semi-automatically adjusted with the electric field and the state parameters of the material itself, include adaptive field grading materials (adaptive dielectrics) and self-healing dielectrics capable of repairing electrical ageing damages. The development of ultra-high voltage transmission technology is closely related to the promotion of the goal of 'carbon neutrality', while the improvement of the voltage level of the equipment directly affects the operating life and safety of the insulating components. In addition, the extremely uneven distribution of the electric field through the insulating components in high-voltage equipment will bring great difficulties to the design and manufacture. High-voltage equipment based on traditional technology has encountered bottlenecks at higher voltage levels. The research and development of smart dielectric materials may play a significant role in reducing the design and manufacturing difficulty as well as lowering the cost and improving the operational reliability of high-voltage equipment.

Adaptive dielectrics utilise non-linearity of conductivity and dielectric permittivity to realise automatic control of the electric field in high voltage equipment. Although non-linear conducting properties have been widely reported, there are few studies on the field-dependent non-linear permittivity of field grading dielectrics. The theory of non-linear permittivity remains to be further improved, and related research is still in the process of finding materials with high levels of stable non-linear dielectric constants. At present, the research on the application of adaptive materials in high-voltage power equipment is mainly in the simulation design and laboratory model experiment stage. There is a lack of a direct experimental method for characterising the electric field distribution in internal insulation equipment such as cable accessories and capacitive wall-through bushings. In addition, to face a wider range of advanced power equipment, future adaptive materials will also be studied in terms of stability/compatibility as well as assembly characteristics.

The research objectives of future adaptive dielectrics include:

1) Find materials with a high level of stable non-linear dielectric permittivity.
2) Develop the field grading theory based on non-linear permittivity.
3) Evaluate the influence of the formula and sintering process of non-linear ceramic fillers on non-linear characteristics. Improve the technical methods for regulating the adaptive parameters of materials.
4) Establish a collaborative regulation method for multi-performance (electrical, thermal, mechanical etc.) of adaptive materials.
5) According to the insulating structure of the high voltage equipment (cable accessories, through-wall casings etc.), establish an electric field distribution measurement method to verify the effect of adaptive dielectrics on electric field distribution.

For the self-healing dielectrics, most self-healing materials can only cope with mechanical damages; studies on self-healing dielectrics against electrical damages have just started in recent years. The recently reported self-healing thermoplastic dielectrics based on defect-targeted magnetic heating mechanisms can be achieved by adding very low content of superparamagnetic nanoparticles while maintaining the excellent insulating properties of PP [151]. The current bottleneck of the magnetic healing approach is excitation of magnetic field on the insulation components in practical high voltage equipment. Microcapsule-based self-healing systems provide potential solutions for the development of self-healing cross-linked cable insulating materials [152]. For the traditional microcapsule-based and other extrinsic self-healing approaches, new material processing technologies compatible with cable extrusion processes are required. In addition, the introduction of fluids, curing catalysts and other healing agent components can decrease the electrical insulating properties of materials. Intrinsic self-healing has some advantages over extrinsic solutions. Intrinsic self-healing is the inherent ability of a material to recover from damages without the help of an external structure or reagent. These materials rely on chemical equilibria to dynamically reform their network on a molecular level by exchanging chemical bonds across the fracture plane. Their extension from mechanical applications [153] to the insulation field is on the way [154]. Besides, the electrical damages usually create large breakdown holes or occur inside the dielectrics (electrical tree), which makes it difficult to achieve damage surface closure, thus many self-healing methods for mechanical damage cannot be applied directly with serious damage. Therefore, healing processes operating at the earliest stage possible of the electrical damage, acting on pre-breakdown phenomena, are to be targeted.

Considering the multi-scale characteristics of electrical damages and the influence of electrical degradation by-products on the healing process and insulating properties of materials, removing degradation by-products, cleaning carbonised damage surfaces, and automatically closure of the tree channel and breakdown holes are also problems that need to be solved when designing self-healing dielectric materials [155].

Future research on self-healing dielectrics may be carried out around the following objectives:

1) Reveal the mechanisms of electrical damage initiation and propagation in dielectric materials and recognise the degradation by-products formed during the electrical ageing process of dielectrics.
2) Evaluate the feasibility of the existing self-healing approaches for different dielectric materials and application scenarios.
3) Manage absorption and removal of electrical degradation by-products. Researchers have developed the hybrid nanofillers by loading polyethyleneimide into mesoporous silica nanoparticles that can absorb the gaseous electrical degradation by-products (CO_2, H_2O etc.) generated in the electric tree channel thus effectively inhibiting the ageing rate of the electric tree and also improving the insulating properties of PP [156, 157]. These absorbing nanofillers can be used as auxiliary additives in self-healing systems.
4) Automatic closure of electrical damage surface. Researchers have introduced shape memory materials into self-healing systems, and shape memory assisted self-healing (SMASH) has attracted more attention [158–160]. The multi-responsiveness of SMASH materials (photon-responsive [161], electrical [162], magnetic [163] etc.) offers more opportunities for developing versatile self-healing dielectric systems.

5.2 | Insulating materials with high thermal conductivity

Overheating issue is one of the biggest bottlenecks in the electronic systems. The studies show that over 50% of the electronic failures are due to the overheating and the related issues [164]. Hence, the materials with both high electrical insulation strength and high thermal conductivity (HTC) play important roles in the electronic industries. The HTC insulating materials can be generally classified as III–V compounds, metallic oxides and carbon materials. Figure 6 shows the typically HTC insulating materials.

III–V compounds have numerous kinds of HTC insulating materials, among which, hexagonal boron nitride (h-BN) [165, 166] is the most representative. H-BN has a similar two-dimensional honeycomb structure with graphene, of which, the thermal conductivity was measured to be 360 W/(m·K) [2]. H-BN is a perfect heat-conductive additive, which can be in the shape of BN flakes [167, 168] or BN nanotube (BNNT) [169]. Hu et al. reported a silicone rubber (SiR)/BN flakes composite with an out-of-plane thermal conductivity of 7.62 W/(m·K) at 60 wt.% filler content. Aluminium nitride (AlN) is another highly heat-conductive insulating material, which has a

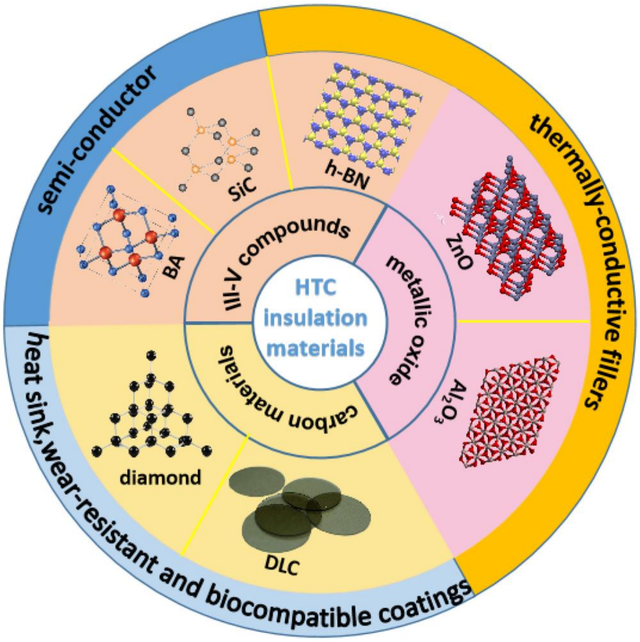

FIGURE 6 Typical high thermal conductivity insulating materials and their main applications

theoretical thermal conductivity of 320 W/(m·K) [170]. Due to its thermostability, non-toxic and low CTE, AlN is well implanted in electronic-packaging applications, including, heat dissipation film [170], insulation board [171], and thermal additive [172]. SiC [173], a third-generation semiconductor, has a high thermal conductivity of 490 W/(m·K), which is 3.3 times that of silicon. Meanwhile, it has higher breakdown field strength and similar carrier mobility compared to silicon. The SiC power devices have entered commercial production [174] and played crucial role in high-power electronic devices. Boron arsenide (BAs), a new material grown through a modified chemical vapour transport technique, was reported by three different research groups in 2018 [175–177]. The thermal conductivity of cubic BA is estimated over 1000 W/(m·K), much higher than that of SiC. It may be a promising thermal management material for high-power density electronic devices in the future.

Metallic oxides, such alumina (Al_2O_3) and zinc oxide (ZnO), have been widely used as reinforcing materials in thermal management. They have similar thermal conductivity of about 30 W/(m·K). Al_2O_3 is the most popular heat-conductive filler of commercial thermal interface materials (TIMs), due to its low CTE, high elastic modulus, relatively low dielectric constant, moderate thermal conductivity and low price [178, 179]. ZnO fillers usually mix with Al_2O_3 particles as supplement in TIMs due to the smaller particle size so that the thermal conductivity of the composites can be further improved [180]. It should be noted that some hydroxides, such as $Al(OH)_3$ and $Mg(OH)_2$, although have relatively low thermal conductivity of less than 1 W/(m·K), they are widely added in TIMs as heat-conductive fillers [181] due to the excellent flame retardancy.

Diamond, an allotrope of carbon, has the highest thermal conductivity (~2200 W/(m·K)) among numerous insulating materials. Due to the notable thermal and mechanical properties, diamond coatings are applied in many aspects, including, semiconductor, heat sink and wear-resistant coating [182]. Diamond-like carbon (DLC) is a hydrogenated amorphous carbon possessing the mixture of sp3 and sp2 carbon atoms. The thermal conductivity of DLC can reach the 50% of diamond [183], depending on the ratio of sp^3 carbon atoms. More importantly, DLC has extremely low frictional coefficient, so it is well applied as mechanical protective and biocompatible coatings [184].

Synthesis of high-quality bulk crystals of these HTC insulating materials is the greatest challenge, which has blocked the process of commercial application. For example, it is difficult to obtain over 2-inch single-crystal diamond films, because cracking is unavoidable when the size over 1 inch [185]. In addition, the influences of the defects, including doping, dislocation, and impurities, on the thermal properties remain to be studied.

5.3 | High energy storage density insulating materials

High energy storage density insulating materials are widely used in energy storage capacitors [186], which have significant advantages such as environment-friendly, high voltage resistance, long life, and ultrahigh power density. It meets the needs of the next generation of distributed energy utilisation and plays an increasingly significant role in power and electronic systems. Capacitors are also the irreplaceable energy storage devices in the fields of pulsed power, rail transit and electromagnetic energy weapons due to the extremely high charge and discharge rate [187]. The applications of high-performance capacitors in military and civil fields have attracted wide attention [188]. However, as an energy storage device, one significant disadvantage of capacitors is its low energy storage density. The energy density of the current commercial BOPP energy storage capacitor is less than 2 J/cm^3 [189], which is much lower than the counterparts, such as batteries and supercapacitors. Dielectric materials with higher energy storage density are highly expected to support the development of high energy storage capacitor devices. For linear dielectrics, the maximum energy storage density U can be obtained [190] as

$$U = \frac{1}{2}\varepsilon_0\varepsilon_r E_b^2 \quad (1)$$

where ε_0 is the vacuum permittivity, ε_r and E_b are the relative permittivity and breakdown field strength of the dielectric material, respectively. Non-polar polymer materials represented by PP have high breakdown field strength but relatively low relative permittivity. In contrast, polyvinylidene difluoride (PVDF, high polarity) possesses higher relative permittivity, but the breakdown field strength is strongly weakened [191]. Therefore, how to break through the inherent contradiction between the relative permittivity and the breakdown field strength of polymer materials and achieve simultaneous improvement is essential to obtain the high energy storage capacitor dielectric materials.

The nanocomposites, by incorporating inorganic ceramic particles in the polymer matrix, can simultaneously take advantage of the strong polarisation of the ceramic particles and the high electrical strength of the organic matrix [192]. It has become an efficient way to obtain high energy storage density materials and the dielectric performance can be effectively tailored by modifying the morphology, filling concentration and spatial distribution of inorganic particles [193, 194]. However, the micro-mechanism of the interface between particles and polymer, which is demonstrated to be important for dielectric properties of nanocomposites, is still not definitely investigated [195]. In addition to experimental methods, as the rapid improvement of computing performance, simulation has become another effective way to study the dielectric properties of composite materials. From micro to macro scale, many advanced simulation methods including first principles, molecular dynamics, Monte Carlo simulation, phase field simulation and finite element analysis have been significantly developed [196–199]. These methods can not only predict dielectric parameters but also reveal the microscopic mechanisms behind dielectric phenomena at different scales. The data-driven strategies [200] are accelerating the design and development of high energy density materials.

5.4 | Insulating materials at harsh condition

Nuclear is a zero-emission clean energy source, and nuclear power has been used for over 60 years all around the world. In China, the first nuclear power plant (NPP) was established in 1991 in Zhejiang Province, after which the development of nuclear power was encouraged to be even faster. Till the end of 2021, 53 nuclear power units had been in commercial service with a total installed capacity of 5.46 × 10^7 kW. Due to the operating of nuclear power, approximately 1.25 × 10^8 t standard coal consumption could be saved in 2021, which equally led to the reduction of CO_2 emission of 3.28 × 10^8 t [201]. Nuclear power plays an important role in the process of carbon neutrality.

However, one essential issue that must be taken into consideration for the development of nuclear power is safety. Once a nuclear leakage accident occurs, a catastrophic tragedy to human beings may be formed. Polymeric insulating materials have been used in a wide range of applications in NPP, and these materials have to be exposed to harsh conditions during their lifetime in service, for example high energy radiation such as gamma-ray and high temperature. The degradation of the material is accelerated under such conditions, and consequently the electrical and mechanical properties are to be degraded to introduce insulation failure that threatens the safe operation of NPP [202]. Accordingly, from the viewpoint of safety, it is of great significance to investigate the degradation behaviour of polymers at harsh conditions.

Investigations on radiation-induced ageing of polymers started in the 1950s; silicone, polyethylene (PE), PVC were selected as test objects to estimate the gamma-ray radiation-induced variations in dielectric properties [203]. Dielectric constant, dielectric loss, resistivity and breakdown strength were employed as the parameters for discussion. In the following 30 years, research on radiation effect has been performed with respect to a much wider range of polymers, and radiation-induced conductivity, tensile strength, elongation at break, elastic modulus, and melting point were also put onto the list for discussion, which further improved our understanding of the radiation effect on polymer dielectrics. It has been summarised that the threshold of material change begins to occur at 1 kGy, significant damage at 100 kGy and severe damage at 1000 kGy [204]. The reason for the material change is attributed to the radiation induced chemical reactions such as crosslinking, chain scission and oxidation [205]. With the progress of measuring technique of solid dielectrics, studies are enabled to examine some other aspects such as carrier trap, space charge behaviour, surface tracking and water treeing phenomena within/on polymer materials. Thermal stimulated discharge current (TSDC) has been employed to extract trap distribution of polyetheretherketone (PEEK) [206], PP [207], polyethylene napthalate (PEN) [208], and polyethersulfone (PES) [209]; the re-arrangement of ruptured chain and the appearance of polar groups are responsible for the variation in carrier trap feature. Space charge accumulation in gamma-ray irradiated low density polyethylene (LDPE) has been observed through the pulsed electro-acoustic (PEA) method due to the radiation induced oxidation in LDPE [210]. Increase of imaginary part of permittivity close to 100 kHz correlated to polymer oxidation and antioxidant effects have been seen in dielectric spectroscopy measurements in XLPE, EPR and ethylene-vinyl-acetate (EVA) insulation [211–213]. Surface dielectric property of polybutylene naphthalate (PBN) is improved by gamma-ray radiation induced crosslinking reaction [214]. Water treeing as a special ageing behaviour in XLPE exposed to gamma-ray radiation has been investigated; the water tree becomes shorter with the radiation dose because of the radiation induced variations in crosslinking degree, crystallinity and the size of spherulite [215].

Several studies mentioned above mainly focus on the radiation effect. However, in a real NPP, the ageing of polymer usually occurs under radiation and high temperature conditions, especially for insulation of cables installed inside the nuclear power containment [216]. It is thereby necessary to investigate the ageing behaviour under multi-stress of radiation and heat [217]. XLPE, SiR and ethylene propylene rubber (EPR) are the most popular materials used as cable insulation in NPP. For XLPE, after ageing under heat and gamma-ray radiation, oxidation occurs heterogeneously in material depth, which is owing to the oxygen diffusion [216]. An antioxidant plays effective role against oxidation only when its content exceeds a critical value around 0.04 wt% [218]. For EPR, the degradation of polymer main chains is induced by oxidation as well. The antioxidant plays an important role to prolong the lifetime of EPR in the aspect of thermal oxidation with a critical content of 0.5 phr [219]. For SiR, the degradation mechanism is completely different from those of XLPE and EPR, which are prepared by crosslinking that involves thermal and radiation-induced oxidation [220]. The combined ageing with radiation and heat has been also conducted for flame-retardant crosslinked polyolefin (FR-XLPO). Gamma-ray radiation with a proper dose can mitigate the degradation induced by thermal stress conducted simultaneously or subsequently through crosslinking reaction [221]. To sum up, polymers with different molecular structures go through different degradation processes under the combined thermal-radiation stresses. More efforts should be taken to reveal the ageing mechanism of polymers under multi-stress conditions.

In order to improve the material property to withstand the harsh condition, attempts have been made by tailoring the material structure or re-selecting the polymer matrix. Micro-sized hexagonal boron nitride (M-h-BN) and nano-sized silica (n-SiO_2) have been concurrently added into LDPE to achieve a synergistic modification of thermal and electrical properties. Enhancement of 30% in thermal conductivity and reduction of 23% in carrier mobility are obtained meanwhile with M-h-BN of 20 wt% and n-SiO_2 of 3 wt% [222, 223]. After gamma-ray radiation of 1000 kGy, the composite exhibits a carrier mobility 60% lower than that of the virgin LDPE, indicating a pronounced electrical property stability against the radiation. High melting point polymer, PET, has been examined as well with respect to gamma-ray radiation induced ageing. The presence of aromatic groups in its molecular chain contributes to the great endurance to radiation induced degradation [224]. Aromatic groups contained epoxy resin is modified via nano-alumina (n-Al_2O_3) to withstand the gamma-ray radiation and chemical corrosion that would emerge with the occurrence of loss of coolant accident (LOCA) [225], which reveals the effectiveness of inorganic nano-particle and aromatic group against the harsh condition in NPP.

In the future, the development of nuclear power featured with high temperature gas cooled reactor will require much higher safety operation of electrical systems. Hence, the polymer insulating materials are going to face new challenges at harsh conditions. The following issues are suggested to be considered in related research fields:

1) Effects of the harsh conditions, for example radiation, heat, flame, high pressure steam, and chemical erosion, on electrical and mechanical properties of polymers should be further clarified. The dependence of the polymer properties upon the microstructures in response to the harsh conditions should be clearly established.
2) Modification of the properties of polymer materials should be performed by tailoring the physical or the chemical structures, where the inclusion of micro- or nano-sized inorganic filler and/or organic compound containing aromatic rings may be suggested. Polymer materials with intrinsically higher melting temperature or better radiation resistance should be developed.
3) Effective methods for estimating the ageing degree of polymer material exposed to the harsh conditions should be further investigated, and modelling of polymer ageing at the mentioned conditions should be performed, which would better exhibit the mechanism of the degradation manner and provides valuable information for insulation design at the harsh conditions.

5.5 | Environmental-friendly insulating materials

In this section, issues for both solid and gaseous environmental-friendly insulating materials are discussed.

1) Solid environmental-friendly insulating material

Solid environmental-friendly insulating materials, represented by PP, have shown great advantages and potential as insulating materials for HVDC cables, including environmental friendliness, high temperature resistance and high breakdown strength [226–228]. Environmental-friendly insulating materials are expected to play a more important role in future cable insulation systems and gradually replace XLPE [229, 230]. Remarkable achievements have been made in environmental-friendly DC cable insulating materials. Various strategies and material systems are proposed to improve the electrical properties of environmental-friendly DC cable insulating materials, including control of crystalline morphology and aggregate structure [231], multi-olefin monomer copolymerisation [232], blending with other thermoplastic polymers [233, 234], nanocomposite [235, 236], and grafting chemical group [237]. The introduction of these methods has greatly improved the performance of PP-based environmental-friendly DC cable insulating materials and even surpassed the performance of the currently widely used XLPE. In order to further promote the development of environmental-friendly DC cable insulating materials, the following problems need to be addressed.

The mechanical and electrical properties of the material should be synergistically optimised to meet the overall requirements of cable production, transportation, laying and operation. The environmental-friendly insulating material should have mechanical flexibility, high temperature integrity, excellent insulating properties and low cost. Particular attention should be paid to the mechanical and electrical properties of materials at extreme temperatures to overcome the challenges of low temperature brittleness and stark decrease in electrical properties at high temperatures [238].

Space charge behaviour, including charge origin, transport, accumulation and dissipation processes of the material should be comprehensively studied. Space charge plays a very important role in affecting the performance of insulating materials under DC electric field, especially at high temperatures. To ensure the safe operation of HVDC cable systems, the problem of space charge accumulation must be solved. Moving from XLPE to thermoplastic insulation represents in principle as an advantage over crosslinked insulation as cross-linking by-products are avoided. Considerable progress was achieved in

XLPE grades by reducing the amount of by-products and investigating processes that could be virtually by-products free [239]. This has represented large research efforts to master XLPE properties. It is necessary to clarify the space charge behaviour and mechanism in environmental-friendly DC cable insulating materials from a microscopic level, which is helpful to design the insulating materials and structures in a targeted manner to reduce the impact of space charge [240]. Designing interfaces is another way to explore to mitigate space charge effects, both from volumic insulation and surface flashover perspective [241, 242]. Naturally, power cables are not the only domain where the question of recyclability and environmental-friendly poses. Epoxies are widely used in insulations, with concerns of non-recyclability as well as the shadow on health issues with bisphenol A contents. For spacers in GIS, it appears that the thermoplastic PET is a promising alternative material to epoxy. Indeed, long term tests on real-size insulators made of commercially available PET under 400 kV AC and for more than 2 years were successful. Verification for HVDC application are to be explored [243]. High temperature thermoplastics [244] as Polyamides, Polyetherimides and Polyketones (PEEK, and PEK), and, as stated previously, fluorinated thermoplastics [101] may represent alternatives to epoxies and polyimide thermosets in a number of applications.

Nanocomposite and chemical grafting are two promising methods to develop environmental-friendly insulating materials with tuning their properties. But the method of nanocomposite encounters many problems, such as the uniformity and dispersion of the nanoparticles. This is notably the case in cable application where mass production is required [245]. Compared with nanocomposites, chemical grafting modification can avoid the problem of nanoparticle agglomeration and maintain good insulating properties. However, the molecular mechanism on regulating electrical properties of chemical grafting still needs further direct experimental study. An in-depth understanding of the relationship between the grafted chemical structures and the electrical performance of the insulating materials can provide a guidance for the rational material structural design, which can be further facilitated by computational simulation, machine learning and big data techniques [246, 247]. Chemical–physical modelling techniques notably are more and more used as a guide to anticipate polarisability, energy levels etc.; modifications are imparted by material modification, and this constitutes real opportunities for designing materials with targeted dielectric properties.

However, once these fundamental properties are mastered, the overall performances of such environmental-friendly insulating materials under their use stresses that can be extreme conditions regarding field and temperature and need to be evaluated in detail. For example, research on the long-term operating characteristics of environmental-friendly DC cable insulating materials is still lacking, and further detailed evaluation is required.

In addition, the environmental friendliness of the proposed materials should be demonstrated. Thermoplastic insulating materials are generally considered as environmental-friendly materials, for example the recycled materials may be used as construction materials with the addition of inorganic fillers [248]. However, with going to more and more fillers containing materials, the control and the treatment of theses recyclable materials may become complicated. The packaging sector is probably the most advanced domain where the impact of material recycling is considered [249]. Indeed, packaging applications are stricter in terms of material quality and mechanical properties than urban furniture or construction sector for example. Nevertheless, the increasing amount of nanoparticles in the materials and the cumulative effects of these substances may lead to variation in the final properties. For insulating materials as other, direct experimental characterisation of their environmental impact is still lacking. It is necessary to further propose the recycling scheme and demonstration of the used environmental-friendly insulating materials.

2) Gaseous environmental-friendly insulating material

Sulphur hexafluoride (SF_6)-based gas insulated equipment (GIE) with the advantages of small footprint, high reliability, and long dimensional cycle has been widely used in high voltage transmission and distribution systems. However, SF_6 is listed as the most greenhouse gas with the Global Warming Potential (GWP) value of 23,500 and atmospheric lifetime more than 3200 years, and the annual consumption of SF_6 in GIE is more than 7000 tons in China, equivalent to 120 million tons of CO_2 [250]. Currently, extensive studies have been dedicated to evaluating the comprehensive performance of environmental-friendly gas insulating medium including Fluorinated nitrile (C_4F_7N), Fluorinated ketone (PFKs, $C_5F_{10}O$, $C_6F_{12}O$) and Hydrofluoroolefins (HFOs, HFO-1234ze(E), and HFO-1336mzz(E)) [251–254]. The main progresses and problems have been summarised as follows.

The basic insulation performance of environmental-friendly gas has been explored, and CO_2, N_2 or air utilised as the buffer gas is required to meet the minimum operating temperature of GIE [255, 256]. Moreover, environmental-friendly gas insulating medium is more sensitive to the electric field inhomogeneity than that of SF_6 and its relative dielectric strength at high pressure (above than 0.4 MPa) is inferior [257]. The design or optimisation principle of the GIE insulation structure that meets the gas insulation characteristics needs to be further clarified. For arc-quenching scenarios, some problems such as insulation performance decline, solid by-products precipitation exists, which is closely related to the complex molecular structure and poor recovery characteristics of environmental-friendly gas insulating medium [258, 259]. The optimisation of the circuit breaker structure, adjustment of gas composition and auxiliary arc extinguishing means should be addressed.

The decomposition of environmental-friendly gas under discharge or thermal conditions generate gaseous by-products including fluorocarbons (CF_4, C_2F_6, C_3F_8, and C_3F_6), oxocarbons (CO, and COF_2), cyanide (CF_3CN, C_2F_5CN, and C_2N_2) and HF, HCN [260–263]. The content of by-products demonstrates the linear increase trend with the failure duration and severity. That is to say, the main insulating gas is continuously consumed and the 'cumulative effect' of the decomposition by-products is an issue. Considering the insulating property is closely related to the main insulating gas content and some by-products are highly toxic or corrosive, the accumulation of large content of decomposition by-products will aggravate the failure severity and threaten the service life of GIE. Therefore, further explorations on the generation regulation strategy and on-line monitoring method of the decomposition product should be conducted. For the gaseous by-products, the addition of regulatory gas such as O_2 and the development of selective component adsorbent should be considered. Moreover, the modification method of solid metal and non-metal materials to inhibit the solid by-product precipitation and interfacial corrosion should be investigated. Further, there exists a correlation between decomposition characteristics and the attributes of electrical or thermal faults. By extracting the characteristic decomposition by-products that can reflect the fault type and severity, the condition evaluation method of environmental-friendly GIE based on the decomposition component analysis (DCA) can be proposed.

The comprehensive performance of environmental-friendly gas insulating medium needs further verification from actual application. Several manufacturers including General Electric (GE), ABB, Xuji Group, et al. have launched the C_4F_7N, $C_5F_{10}O$, $C_6F_{12}O$ based 145 kV GIS, 420 kV GIL, 245 kV CT and 10 kV Ring Main Unit and put them into operation in some EU countries, South Korea and China [264, 265]. Long term live assessment of the insulation, stability, and material compatibility performance of the environmental-friendly gas insulating medium is necessary. In addition, the biosafety of C_4F_7N, $C_5F_{10}O$ and their main decomposition by-products also needs to be further clarified before large scale application [266]. The safety protection measures and operation and maintenance procedures should also be formulated.

6 | CONCLUSION

This paper provides a summary review on insulating materials in the framework of the ambitious carbon neutrality targets which many countries want to achieve in the forthcoming years. This paper discusses the engineering challenges constrained by insulators in future electric power equipment/devices and EVs. Insulating materials, including intelligent insulating material, high thermal conductivity insulating material, high energy storage density insulating material, extreme environment resistant insulating material, and environmental-friendly insulating material, are categorised and their scientific issues, opportunities and challenges under the goal of carbon neutrality are discussed.

However, it has to be admitted that the content of this paper is certainly not exhaustive. Due to space limitations and the limited ability of the author's team, there are still many important aspects, such as high-field breakdown of insulating materials, corona-resistant and tracking-resistant insulating materials etc., which have not been covered. In addition, the overview of the advanced power equipment for future electric power grid is not complete.

Nevertheless, we do hope that the discussion and suggestions can provide reference for researchers in the field of insulating materials, EVs, as well as high voltage engineering, especially in selecting research topics and breakthrough points so as to contribute to the shared goal of carbon neutrality in the near future.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest that could be perceived as prejudicing the impartiality of the research reported.

DATA AVAILABILITY STATEMENT

The data used to support the findings of this study is included within this paper.

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