

Recyclable Polypropylene-based Insulation Materials for HVDC Cables: Progress and Perspective

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Abstract—Polypropylene (PP)-based recyclable materials have attracted tremendous interest for HVDC cable insulation applications due to their superior electrical properties, *e.g.*, high thermal stability and superior recyclability. Compared with crosslinked polyethylene (XLPE), PP-based materials exhibit the advantages of not only higher working temperatures but also facile and efficient cable manufacturing with reduced costs, which are highly desirable in HVDC cable manufacturing. Considering their promising advantages, PP-based materials have received significant attention from both academia and industry in the field of HVDC cable insulation. In order to adopt PP as a cable insulation material, the mechanical flexibility of PP should be improved. However, regulations of the mechanical properties inevitably influences the electrical properties of PP. So extensive research has been conducted on the regulation of the mechanical and electrical properties of PP. This review summarizes the research progress on recyclable PP-based materials for HVDC cable insulation applications. Particular attention is placed on the electrical property regulations and material structure-property relationships. The challenges that remain to be addressed and the opportunities for future studies on PP-based recyclable HVDC cable insulation materials are also presented.

Index Terms—polypropylene, HVDC cable, dielectric materials, recyclable insulation materials.

I. INTRODUCTION

WITH the development of UHV power transmission, more and more HV power transmission lines are under construction or will be constructed [1]–[4], for example, about 800,000 km of UHV transmission lines will be constructed in China [5]–[7]. Most of these transmission lines are overhead transmission lines. However, there are several limitations of overhead transmission lines, such as the adverse impact on the landscape and natural environment along the transmission

lines, the compression of human living space because of the complicated electromagnetic environment, as well as the vulnerability to natural disasters (*e.g.*, lightning and icing). Considering the aforementioned limitations, some changes should be made in power transmission methods. Underground cable power transmission is an excellent option to solve these problems. The flexible HVDC power transmission is also the main direction advocated by CIGRE [8]. Moreover, offshore wind power sending and marine resource utilization also call for cross-sea large capacity electric power transmission [9]. For such applications, HVAC cable power transmission is not a good choice because of the large capacitive current which severely reduces the power transmission efficiency [10]. So HVDC cable power transmission is almost the only choice. Also, in some large cities, there is not enough space to build new overhead transmission lines. Indeed, some large cities in China have proposed a plan to build underground pipe galleries to accommodate electric power transmission, gas transmission, information transmission and underground railways to save the scarce land resources [11]–[13]. Considering these urgent demands, large capacity HVDC power cables should be developed.

The development of polymeric cable insulation materials has gone through several stages, including natural rubber, polyvinyl chloride, synthetic rubber (butyl rubber and ethylene propylene rubber), polyethylene, and crosslinked polyethylene (XLPE) [14]–[21]. Since the 1950s, low density polyethylene (LDPE) has been used in MV and HV cables because of its excellent insulation properties, simple processing technology and good flexibility. However, the poor thermal properties restrict the further application of LDPE in high voltage large capacity power cables. In order to improve the poor mechanical properties of LDPE at high temperatures, LDPE is crosslinked. After crosslinking, the thermoplastic LDPE is transformed into thermoset XLPE, and the heat resistance of XLPE is greatly improved. The long-term operation temperatures of XLPE cables can reach 90 °C in HVAC cables [22]. Although XLPE has been used as cable insulation material for about 60 years [23], there are some issues with XLPE. First, XLPE is a thermoset material which is difficult to be recycled. Traditional methods of disposing used XLPE are burning, pyrolysis or burying underground, which not only consume a lot of energy, but also have a negative impact on the environment. Second, the crosslinking and degassing

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process in XLPE manufacturing is very complicated and requires precise control. These processes also result in long production time, which is estimated to be 5–10 times longer than that of thermoplastic materials. Third, the crosslinking by-products are always a problem in XLPE insulation, which can easily introduce microdefects and cause serious space charge accumulation under the DC electric field [24]–[26]. In view of the above shortcomings, XLPE can neither meet the requirements of large-capacity HVDC cables, nor can it meet the requirements of sustainable development. Therefore, it is urgent to develop high-performance recyclable thermoplastic HVDC cable insulation materials to replace XLPE and meet the requirements of large-capacity HVDC cable power transmission.

Recyclable HVDC cables use thermoplastic materials as the insulation materials, which do not require any crosslinking process, thereby completely avoiding the adverse influence of crosslinking by-products [27]–[29]. The thermoplastic insulations can also be recycled after service, which not only has significant technical advantages, but also has considerable economic benefits. A CIGRE working group has compared the cost of thermoplastic insulation and XLPE [30]. The results show that the cost of thermoplastic material can be reduced by 14% compared with XLPE. The overall cost of the cable system can be reduced by 17% and the carbon emission during cable manufacturing can be reduced by 20% when using thermoplastic materials instead of XLPE.

The development of recyclable cable insulation materials has become the forefront of power cable academia and industry studies [31]–[34]. At present, the main technical routes to develop recyclable cable insulation materials include the blending of LDPE and high-density polyethylene (HDPE), and the blending of polyethylene (PE) and polypropylene (PP). Among the proposed methods, PP-based materials are promising because of their high operation temperature (*i.e.*, above 110 °C) and excellent insulation performance compared with XLPE. Prysmian developed a PP-based thermoplastic HVDC cable prototype in 2015 [35]. In recent years, research on recyclable cable insulation materials have been extensively conducted worldwide [36]. In particular, researchers in China have obtained relatively mature PP-based recyclable cable insulation material, and the trial production of PP-insulated HV cable has been carried out.

II. RECYCLABLE POLYPROPYLENE INSULATION MATERIAL

PP is obtained by the polymerization of propylene monomer. The polymerization process and the chemical structure of PP are shown in Fig. 1.

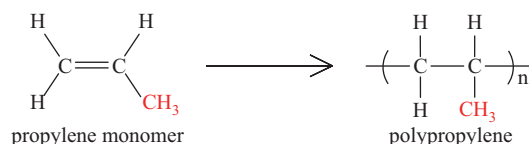


Fig. 1. Polymerization process and chemical structure of polypropylene.

Due to the presence of the methyl group in propylene (the red area in Fig. 1), PP can be classified into three different configurations according to the position of the methyl groups, isotactic polypropylene (iPP), syndiotactic polypropylene (sPP) and atactic polypropylene (aPP). The structures of the three different PPs are shown in Fig. 2, the methyl groups in iPP are located on the same side of the main polymer chain, while the methyl groups in sPP are alternatively located on both sides of the main polymer chain. In addition, the methyl groups are randomly located on both sides of the main polymer chain in aPP.

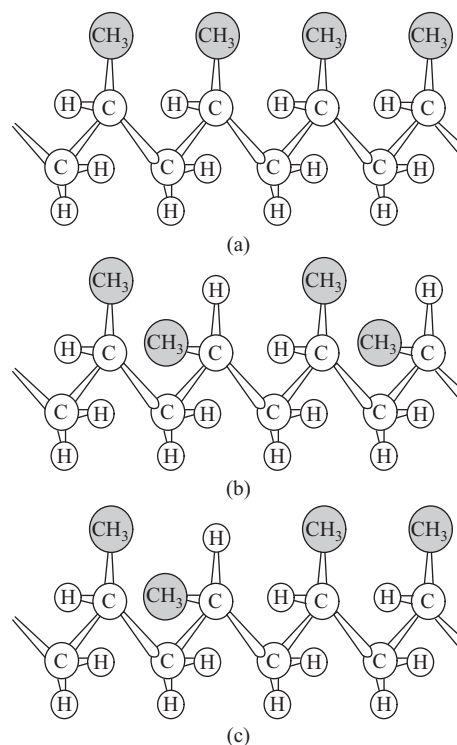


Fig. 2. Polypropylene with different structures, (a) isotactic polypropylene, (b) syndiotactic polypropylene and (c) atactic polypropylene.

Due to their different structures, the properties of iPP, sPP and aPP are quite different. iPP is a semi-crystalline polymer with a melting temperature of about 165 °C and the long-term operation temperature is about 130 °C. iPP possesses excellent heat resistance and insulation properties, but poor mechanical flexibility at low temperatures. More than 90% of PP in industrial production and applications are iPP. sPP is also a semi-crystalline polymer with a melting temperature of about 135 °C. Because of the relatively good mechanical flexibility and excellent insulation performance, sPP has been regarded as a potential recyclable cable insulation material and has been previously studied. K. Yoshino *et al.* have developed a model cable based on sPP, which exhibits higher performance than XLPE insulation [37], [38]. However, since the methyl groups in sPP are alternatively located on both sides of the main polymer chain, the polymerization of sPP is difficult and expensive, which limits the application of sPP. In addition, the lower melting temperature of sPP (*i.e.*, 135 °C) restricts the operation temperature of sPP-insulated cables, so it is

difficult to raise the capacity of sPP-based cables. As for amorphous aPP, it has not been extensively used because of its low molecular weight, poor mechanical properties and low temperature resistance.

Considering the insulation performance, operation temperature and material cost, iPP is an ideal recyclable DC cable insulation material. Table I lists the performance of commonly used polymeric cable insulation materials. Compared with other materials, PP exhibits stronger temperature resistance, higher volume resistivity and breakdown strength, indicating that PP is more suitable for large-capacity HVDC cable insulation applications.

TABLE I
ELECTRICAL PERFORMANCE OF COMMONLY USED
INSULATION MATERIALS

Material	ϵ_r	ρ ($\Omega\cdot\text{m}$)	E_b (kV/mm)	T_m ($^{\circ}\text{C}$)
XLPE [36]	2.3	0.9×10^{15}	300	105 (softening)
LDPE [39]	2.2	3.1×10^{15}	345	108
HDPE [39]	2.3	2.2×10^{15}	450	132
LDPE/HDPE [40]	2.2	3.2×10^{15}	450	115
PP [29]	2.3	2.2×10^{16}	400–600	165

ϵ_r : dielectric constant, ρ : DC volume resistivity, E_b : DC breakdown strength, T_m : melting temperature.

However, the mechanical flexibility, especially the brittleness at low temperature of PP should be improved to meet the demand of cable applications. To solve this problem, many methods have been adopted to regulate the mechanical properties of PP, such as blending with thermoplastic elastomers (i.e., ethylene propylene diene monomer (EPDM) and polystyrene-ethylene-butylene-styrene (SEBS)) and copolymerization with other olefins [41]–[44]. Although these methods can readily regulate the mechanical properties of PP, the electrical properties are often deteriorated. To realize the comprehensive performance regulation of PP, it is more important to further enhance the electrical properties of PP, especially at high temperatures. For DC cable insulation applications, space charge accumulation is a big threat. Particularly, with the increased working temperature of PP, space charge accumulation and insulation performance deterioration under high temperatures are the main problems for the electrical property regulation of PP-based recyclable insulation materials.

III. METHODS FOR ELECTRICAL PROPERTY REGULATION OF POLYPROPYLENE

Recently, PP has become a hot topic in the research of recyclable HVDC cable insulation materials for its remarkable performance. A series of methods for regulating the electrical property of PP-based insulation materials have been developed, including: (1) nucleating agent modification, (2) copolymerizing modification (3) blending modification, (4) chemical grafting modification, and (5) nanocomposite modification. These regulation methods are reviewed in detail in the following sub-sections.

The aggregation structure of PP plays an important role in charge carrier transport, thus affecting the electrical properties of PP. PP is a semi-crystalline polymer, in which the crystallization phase and amorphous phase coexist. The electrical

breakdown strength of the amorphous phase is significantly lower than that of the crystallization phase. Therefore, the electrical breakdown process of PP often occurs along the spherulite boundaries. Therefore, larger spherulite with distinct boundaries would result in decreased electrical breakdown strength [45]. So, reducing the spherulite size and increasing the spherulite number can effectively improve the electrical properties of PP. Based on controlling the aggregation structure of PP, a series of electrical property regulation strategies have been developed, including the addition of nucleating agents, copolymerization with other olefin monomers, and blending with other thermoplastic polyolefins.

A. Nucleating Agent Modification of Polypropylene

Isotactic PP can crystallize into monoclinic- (α), trigonal- (β) and orthorhombic- (γ) phases. The three types of crystal phases have significant differences in melting point, density, and electrical properties. Under normal processing conditions, PP is in α -phase. However, the α -phase can be artificially converted into β -phase by adding nucleating agents. Zha *et al.* added β nucleating agent *N,N'*-dicyclohexylterephthalamide (DCTH) to convert iPP from α -phase to β -phase, and studied the effect of the crystal phase on the space charge and trap level distribution in iPP (Fig. 3). The results show that the β -phase can introduce deep charge traps and then reduce the charge carrier mobility and inhibit the space charge accumulation, which finally improve the electrical performance of iPP [46]. Zhou *et al.* also obtained similar results using aryl amide derivative (TMB-5) as the β nucleating agent, which achieves improved DC breakdown strength and space charge suppression under 100 kV/mm in iPP at room temperature [47], [48]. However, it should be noted that the nucleating agents are mostly small organic molecules or organic salts, which may migrate or even directly ionize under high temperatures and high electric fields [49], resulting in space charge accumulation and the deterioration of electrical properties under high temperatures and high electric fields. Also, it is necessary to further evaluate the long-term stability of the induced β -phase under thermal cycling.

B. Copolymerizing Modification of Polypropylene

By copolymerizing with other olefin monomers, the electrical properties of PP copolymers can be regulated. Common monomers include ethylene, butane and styrene. Copolymerized PP can be classified into homopolymer, block copolymer and random copolymer according to the type and spatial arrangement of the monomers [50]. The molecular structures of different copolymers are shown in Fig. 4a. Meng *et al.* studied the influence of copolymerization structure on the electrical properties of PP copolymers by comparing four different PP copolymers [51]. The results show that compared with block copolymer, the random copolymer exhibits higher mechanical flexibility and more excellent electrical properties, which is more appropriate for HVDC cable insulation (Fig. 4b-e). Xu *et al.* compared the microstructure and electrical properties of homopolymer and block copolymer of PP [52], and found that the ethylene-rich segments in block copolymer may act as nucleation agents, which accelerate the crystallization process

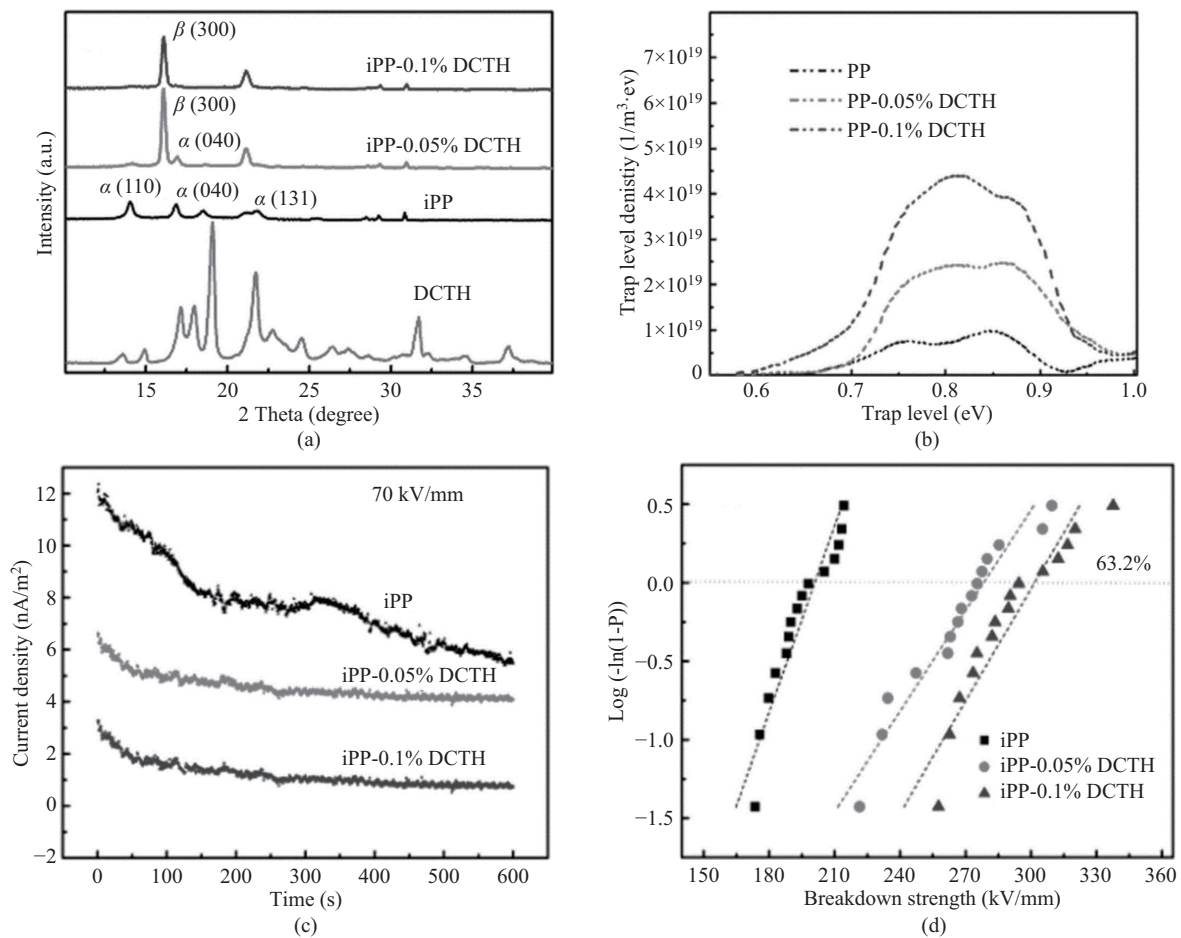


Fig. 3. Structure and electrical properties of the polypropylene with induced β phase. (a) X-ray diffraction patterns, (b) trap level distributions, (c) electrical conduction current, (d) DC breakdown strength of iPP with 0.05 wt. % and 0.1 wt. % DCTH. Reproduced with permission [46]. Copyright 2015, AIP Publishing LLC.

and reduce the spherulite size of PP, thereby improving the electrical properties. I. L. Hosier *et al.* studied the effect of ethylene monomer content on the breakdown strength of ethylene-propylene copolymers [53] and found that high ethylene content reduces the breakdown strength of the copolymers at high temperature. Although the electrical properties of PP can be improved by copolymerization, it is complicated to precisely control the composition and chemical structure of the copolymers. So present research on PP copolymers is primarily based on the commercial available materials, which restricts further investigations.

C. Blending Modification of Polypropylene

Compared with copolymerization, blending is a more practical approach in industrial applications to regulate the properties of polymers. The aggregation structure of PP can also be regulated by blending with other thermoplastic polyolefins. Dang *et al.* prepared PP/elastomer blends with different elastomers [54], and then studied the effect of elastomer type on the aggregation structure and electrical properties of PP. The results show that the good compatibility between PP and propylene-based elastomer can reduce the spherulite size and increase the spherulite density of PP, which leads to higher DC volume resistivity in PP/propylene-based elastomer blends

compared with PP/ethylene-based elastomer blends (Fig. 5). However, the low operation temperature of the thermoplastic elastomer results in degraded electrical performance of the blends compared with pristine PP at high temperatures. Gao *et al.* also compared the effect of elastomer types on the electrical properties of PP, and obtained a similar conclusion [55]. The compatibility between the thermoplastic polyolefin and PP has a significant effect on the properties of the blends. The synergetic performance improvement can only be obtained when the interfaces of the two phases are fully combined. Therefore, the selection of the thermoplastic polyolefin is particularly important in the blending modification of PP [56]. Moreover, some other methods, such as adding compatibilizer and copolymerization, have also shown the potential to improve the compatibility between elastomer and PP matrix [57], [58]. Compared with PP, the thermoplastic elastomers usually show lower melting temperatures. The different thermal properties of PP and elastomer would cause phase separation under long-term high-temperature operations, which forms PP-rich or elastomer-rich regions inside the PP-based blends. Such phase separation may deteriorate the long-term electrical performance of PP-based blends. Therefore, phase separation during long-term high-temperature operations is still a key issue for PP-based blends in DC cable insulation applications

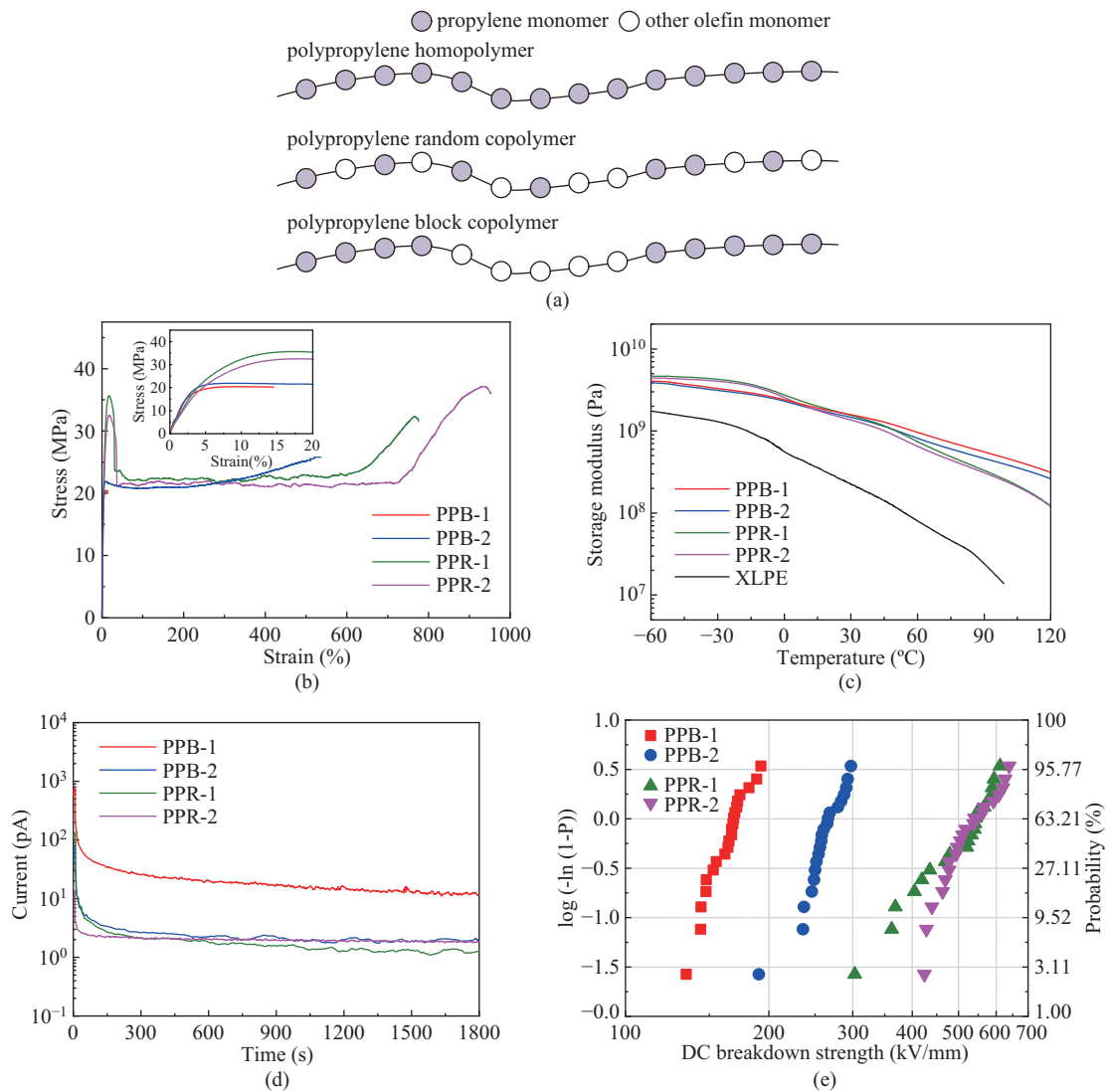


Fig. 4. Structures and electric properties of PP-based copolymers. (a) Structures of different polypropylene copolymers, (b) tensile stress-strain curves, (c) storage modulus from DMA tests, (d) DC leakage current and (e) DC breakdown strength of different polypropylene copolymers. Reproduced with permission [51]. Copyright 2019, IEEE.

and needs to be further investigated.

D. Chemical Grafting Modification of Polypropylene

Grafting polar functional groups onto the non-polar polymer chains is an effective way to regulate the charge transport characteristics and enhance the electrical properties of polymers. Chemical grafting modification has been used in XLPE insulation. It has been shown that grafting maleic anhydride can inhibit space charge accumulation by 50%–70% in XLPE. Other polar groups, *e.g.*, carbonyl, nitro, cyano, aromatic rings and saturated aliphatic acids, can also increase the volume resistivity of PE and other α -polyolefins [59]–[63]. The physical mechanism of the enhanced electrical performance in grafted polymers is that the grafted polar groups could introduce deep traps. The deep traps may capture the charge carriers and reduce the charge carrier mobility, leading to increased volume resistivity and DC breakdown strength, as well as suppressed space charge accumulation.

Our research group has grafted maleic anhydride (mah),

which contains a carbonyl group, onto the molecule chain of PP by melt grafting [64]. With the grafting of mah, the breakdown strength of PP increases by 13.5%, and the space charge injection threshold electric field increases from 33.5 kV/mm of pristine PP to 46.7 kV/mm of PP-g-mah. The results of thermally stimulation current test show that the grafted mah group introduces a large amount of deep traps with the trap level of 0.7–1.0 eV in PP, and the trap level density increases by 4.4 times. The introduced deep traps are responsible for the inhibition of homocharge injection and accumulation. To further reveal the origins of deep traps in grafted polymers, our research group simulated the electronic band structure and 3D electric potential distribution in PP and styrene grafted polypropylene (PP-g-St) [65]. It shows that the different electronic band structure of the grafted polymer compared with the pristine polymer is responsible for the introduction of deep traps in grafted polymers (Fig. 6). By carefully choosing the grafted groups to optimize the band structure of the grafted polymers, the electrical properties

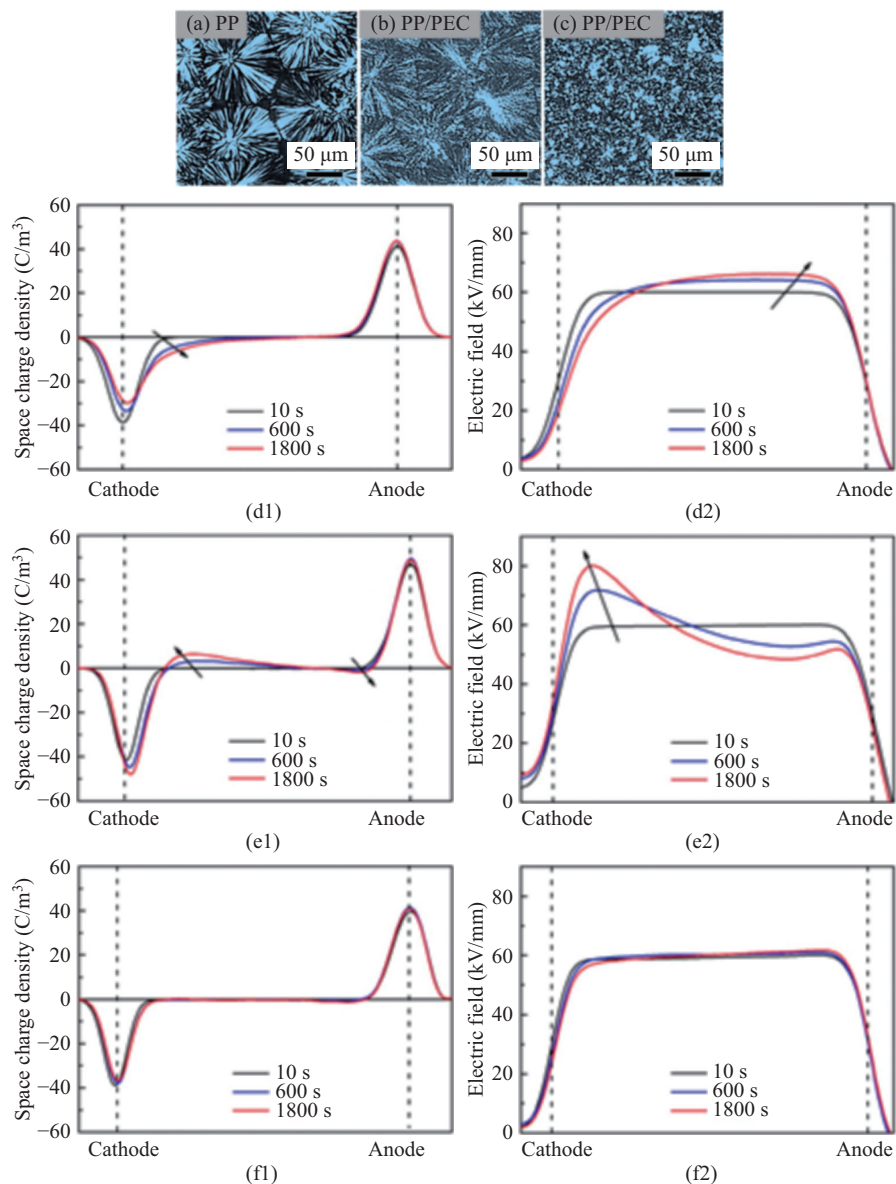


Fig. 5. Structure and space charge behaviors of polypropylene blends with different elastomers. (a-c) POM images of the morphologies, (d-f) space charge and electric field distribution of PP, PP/PEC, PP/EOC, respectively. Reproduced with permission [54]. Copyright 2016, Society of Chemical Industry.

can be further regulated, which provides a pathway for the structure design of high performance HVDC cable insulation materials.

Zha *et al.* studied the effect of different maleic anhydride grafting contents on the microstructure and electrical properties of PP [66]. It was found that grafting maleic anhydride changes the aggregation structure of PP and reduces the spherulite size. The polar groups inhibit the space charge accumulation by changing the trap level distribution. When the grafted maleic anhydride content is 2%, the temperature dependence of the volume resistivity of PP can be significantly weakened.

Although chemical grafting of polar groups can improve the electrical properties of PP, the grafting process is usually carried out by melt extrusion and initiated by initiators (generally peroxide, *e.g.*, dicumyl peroxide). Using such a grafting process may result in the degradation of PP. Also, the residual

initiators and the by-products of the grafting reaction are detrimental to the electrical properties of PP. For the reasons aforementioned, the chemical grafting process needs further optimization.

E. Nanocomposite Modification of Polypropylene

As the new generation insulation materials, nanocomposite dielectrics show great advantages in improving the electrical properties of polymer insulation, such as breakdown strength, volume resistivity, corona aging lifetime, space charge accumulation, partial discharge, dielectric loss and electrical treeing [67]–[72]. Nanocomposite dielectrics have been extensively studied in various insulation materials, such as LDPE, XLPE, epoxy resin, silicone rubber, and ethylene-propylene rubber [73]–[77]. With the development of recyclable PP insulation, extensive studies have been carried out on the PP-based nanocomposites to improve the electrical properties [78]–[83].

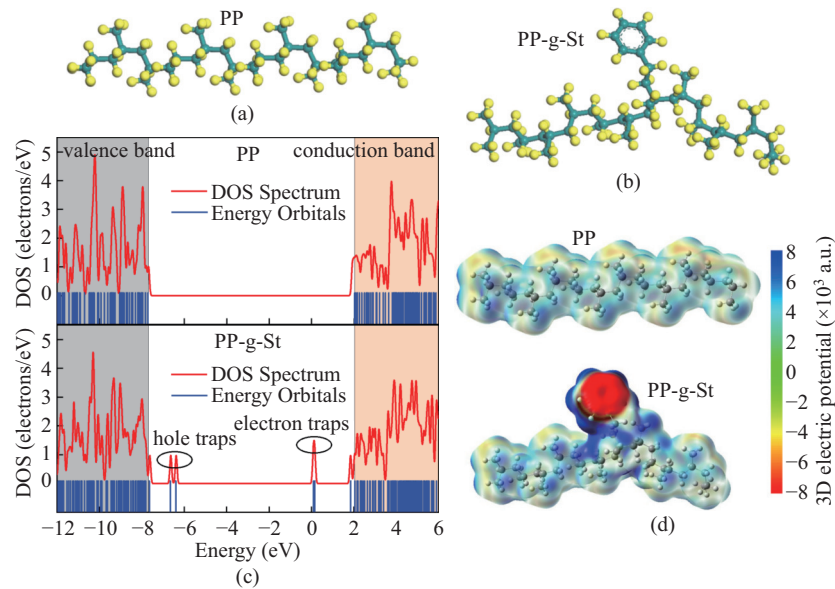


Fig. 6. Structure and properties of polypropylene and styrene grafted polypropylene. (a) Structure of polypropylene, (b) structure of styrene grafted polypropylene, (c) DFT simulated band structure of PP and PP-g-St, (d) 3D electric potential distribution in PP and PP-g-St. Reproduced with permission [65]. Copyright 2020, IOP Publishing Ltd.

Our group modified iPP with the surface-modified MgO nanoparticles [84]–[87] and found that MgO nanoparticles can introduce deep traps at the interfaces between the nanoparticles and the PP matrix to suppress the homocharge injection from the electrodes. The nanocomposites with 3 phr MgO nanoparticles exhibit the highest DC breakdown strength, which is 29% higher than that of pure PP, indicating that the nanocomposite can significantly improve the electrical properties of PP, which expands the application of polymer nanocomposites in recyclable PP insulation (Fig. 7).

The dielectric properties of nanocomposites are closely related to the dispersion and distribution of the nanoparticles. Generally, inorganic nanoparticles and organic polymers are incompatible, so it is necessary to surface-modify the nanoparticles to improve the compatibility and promote the dispersion of the nanoparticles. Jiao *et al.* used different silane coupling agents to modify SiO₂ nanoparticles and prepared PP/SiO₂ nanocomposites [88]. The results show that the interfacial structures formed by different silane coupling agents have important influence on the nanoparticle dispersion and electrical properties of the nanocomposites. That is, promoting the dispersion of the nanoparticles can significantly suppress the space charge accumulation. The SiO₂ nanoparticles surface-modified with polydimethylsiloxane result in the minimal space charge accumulation. L. S. Schadler *et al.* studied the electrical properties of PP nanocomposites with SiO₂ nanoparticles modified with anthracene and polymethacrylate [89]. The effect of grafting density and molecular chain lengths of polymethacrylates on the nanoparticle dispersion were discussed, which shows that the polymethacrylate-modified SiO₂ nanoparticles have better dispersion in PP, and the corresponding DC breakdown strength of the PP/SiO₂ nanocomposite can be increased by 33%. He *et al.* investigated the effect of nanoparticles surface modification with different alkyl silane agents on the electrical properties of PP/MgO nanocompos-

ites [90]. The results indicate that different alkyl groups in the silane agent may influence the trap level distribution of the nanocomposites, thereby affecting the electrical properties. The PP nanocomposite with octyltrimethoxysilane surface-modified MgO nanoparticles shows the highest trap level density and the best comprehensive electrical properties.

In order to combine the advantages of introducing polar groups and nanoparticles, Zhou *et al.* introduced polypropylene-graft-maleic anhydride (PP-g-mah) surface modified MgO nanoparticles into PP [91]. The PP-g-mah not only benefits from the uniform dispersion of nanoparticles by the similar physical and chemical properties of PP-g-mah and PP, but also increases deep charge trapping sites by the polar groups in PP-g-mah. The resultant PP-based nanocomposites show superior electrical insulation properties with enhanced resistivity and breakdown strength. To directly reveal the effects of interfacial regulation, the Kelvin probe force microscopy (KPFM) technology with nanoscale spatial resolution is developed to probe the interfacial trap distribution using a nano-isothermal surface potential decay (nano-ISPD) method. It is shown that with the surface modification of PP-g-mah, more deep traps are introduced at the interfacial region (Fig. 8).

In addition to conventional nanoparticles, *e.g.*, MgO, SiO₂, TiO₂, Al₂O₃, and ZnO, some new types of nanoparticles with specific functions, such as fullerene and nano-scaled mesoporous nanoparticles, are introduced into PP nanocomposites. Dang *et al.* utilized the high electron affinity of fullerene to introduce deep traps in PP [92], which achieves effective space charge suppression at very low doping content (*i.e.*, 0.1 wt%) and increases the volume resistivity of PP by nearly an order of magnitude. Yang *et al.* carried out a structural design to load polyethyleneimine (PEI), which can capture the degradation products during electric tree growth, into nano-sized mesoporous SiO₂ [93]. With the introduction of such

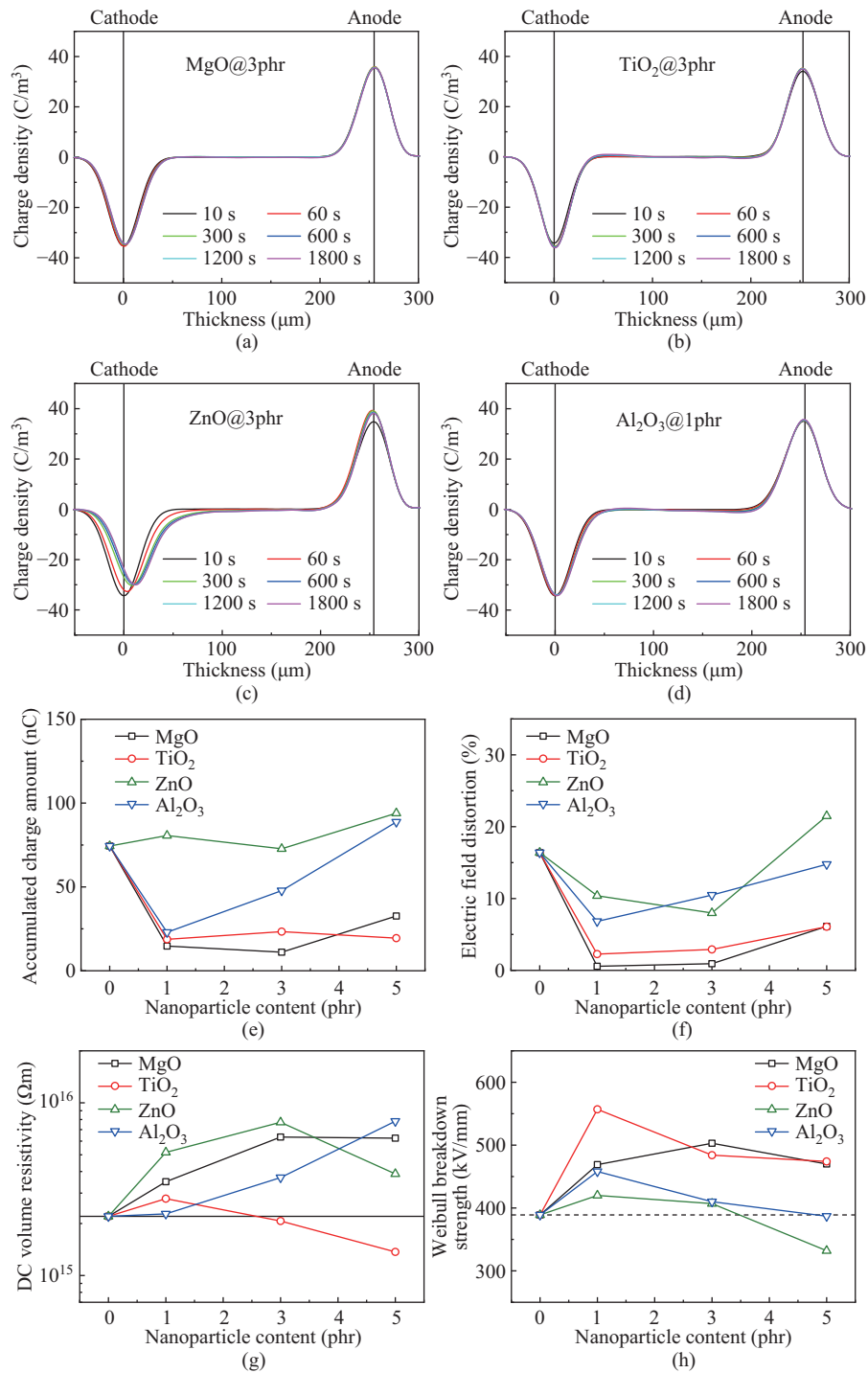


Fig. 7. Electrical properties of polypropylene nanocomposites with different nanoparticles. (a)-(d) space charge distribution under 60 kV/mm DC electric field, (e) accumulated space charge amount, (f) electric field distortion factor, (g) DC volume resistivity under 60 kV/mm, (h) Weibull characteristic breakdown strength. Reproduced with permission [87]. Copyright 2017, IEEE.

nanoparticles, the DC breakdown strength of PP increases by 16 %, and the electric tree growth is delayed by 5 times as the result of capturing the electrical degradation products by the loaded PEI.

It is worth noting that deep traps play an important role in regulating the electrical properties of PP, but the origins of deep traps introduced by different regulation methods are still under investigation. It has been shown that deep traps can

be introduced at the crystal/amorphous interface by changing the crystallization of PP [86]. Moreover, deep traps can be introduced by the functional group grafting and the interface of the nanoparticles [65]. The characteristics of these deep traps should be further studied to reveal the mechanism of the electrical performance enhancement. In addition, the aforementioned modification methods by nanocomposite to improve the electrical properties of PP are still in the initial stage. The

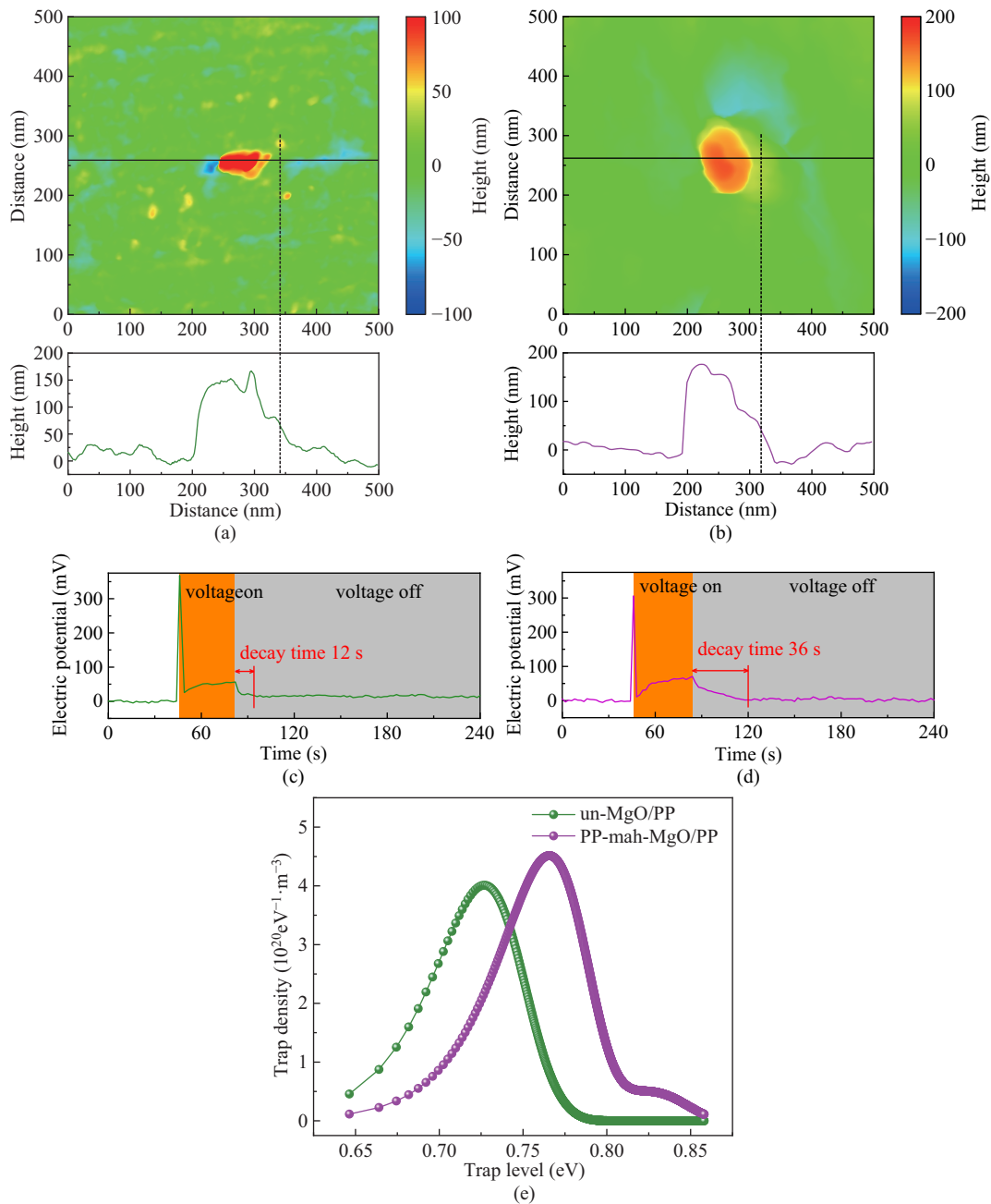


Fig. 8. Local charge trap level distribution in PP-based nanocomposites characterized by Kelvin probe force microscopy. (a-b) topography of the un-MgO/PP and PP-mah-MgO nanocomposite samples, (c-d) time dependent surface potential decay at the nanoparticle/polymer interfaces in un-MgO/PP and PP-mah-MgO nanocomposites, (e) local charge trap level distribution at the interfacial region obtained from the surface potential decay curves. Reproduced with permission [91]. Copyright 2020, Elsevier B.V.

characterization of the electrical properties is primarily carried out at room temperature. Since one of the advantages of recyclable PP insulation is the improved operation temperature, the effects of high temperature and long-term stability of the PP-based nanocomposites should be further studied. Moreover, scale-up and robust production of the nanocomposites should be further developed to produce large-scale and high-quality nanocomposites.

IV. CONCLUSION AND PERSPECTIVE

Recyclable polymeric materials, *e.g.*, polypropylene, have shown significant advantages in HVDC cable insulation ap-

plications, including environmental-friendliness, high temperature resistance and strong dielectric properties, and are extensively expected to serve as the next generation HVDC cable insulation material to replace XLPE. Exciting achievements have been made in the field of recyclable HVDC cable insulation material in recent years. To promote the research and application of recyclable HVDC cable insulation material, there are still some key points that need to be noted and further studied.

1) High-performance insulation materials with co-optimized mechanical and electrical properties, which fulfill the requirements in cable manufacturing, installation and operation,

should be developed. The developed material must satisfy various demands, including mechanical flexibility, high temperature integrity, low temperature resistivity and excellent electrical properties. Especially for higher DC voltage level and higher operation temperature, the enhanced electrical properties and insulating performance need to be further investigated.

2) Related studies so far have shown that the nanocomposite regulation of the electrical properties is strongly related to the microscopic interface characteristics between the nanoparticles and polymer matrix. Some models of the interfacial interaction between nanoparticles and polymer matrix have been established to explain the enhanced electrical properties in the nanocomposites [94], [95], whereas the validity of these models should be comprehensively and carefully verified. Therefore, the in-depth mechanisms, especially at nanoscale, still need to be further studied. Based on the insightful understanding of the interfaces, the electrical property regulation methods can be established to guide the material structure design.

3) Space charge characteristics, including charge origination, transport, accumulation and dissipation, play an essential role in determining the electrical performance of the insulation material operating under DC electric field, particularly at high temperatures. The space charge accumulation can directly affect the conductivity, breakdown strength and long-term reliability of the DC insulation materials. Space charge behavior is also a vital issue in HVDC cable operation, which is connected to the lifetime and reliability of the cable. So, the space charge behavior in recyclable PP-based HVDC cable insulation materials should be systematically investigated, especially at high temperatures.

4) In addition to nanocomposite, other modification methods should be researched and developed. Compared with nanocomposite, chemical grafting modification avoids the problems associated with nanoparticle aggregation, which has shown promising potential in the development of high performance recyclable HVDC cable insulation materials. In-depth fundamental research should be carried out on the relationship between the chemical structure and physical properties, which can provide guidance on the rational material structure design. Computational simulation, as well as machine learning and big data technology, can further assist the material structure design process.

5) The overall performance of PP-based HVDC cable insulation material under extreme operation conditions needs to be investigated. The performance and lifetime of PP-based insulation materials should be comprehensively evaluated to assist with the design and manufacturing of PP-based HVDC cables.

With the increasing demand on HVDC cable power transmission, the extruded polymeric HVDC cables should play a more important role in the future power grid. Considering the advantages of PP-based materials, the recyclable PP-based insulation materials should have promising potential in future HVDC cable insulation materials.

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