Minireview Article

Nonlinear Conductivity Theory Research of Composite Materials

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ABSTRACT

Composite materials with nonlinear electrical conductivity are widely used to solve the problem of electric field concentration in cable accessories. This paper systematically summarizes the conductive mechanisms and influencing factors of inorganic conductive particles and polymer matrices, analyzes several mechanisms and applicable conditions in various conductivity theories, and illustrates the effects of filler type, content, and temperature on the conductive performance of composite materials. Then, the research progress of the nonlinear electrical conductivity theory of composite materials is reviewed, and the application of single filler doping multi-dimensional filler co-doping and in the field of nonlinear electrical conductive materials is introduced. The purpose of this paper is to enable researchers to fully understand the current state of research on nonlinear electrical conductivity. We believe that this work is of great significance for the study of nonlinear electrical conductivity.

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Keywords: polymer materials; nonlinear conductivity; conductive mechanism.

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1. INTRODUCTION

High-voltage direct current (HVDC) transmission technology has been widely applied in the field of power transmission due to its advantages of low current loss, large transmission capacity, high operational stability, low environmental impact, and economic efficiency. However, cable accessories, as an indispensable part of power systems, often become the most frequent failure points. According to the statistical data on faults in power cable systems, cable accessories account for up to 70% of all faults [1]. This is mainly because in DC systems, the insulating polymer is easily charged and accumulates charges when subjected to external conditions, leading to uneven electric field distribution within the insulation. Under the combined effects of space charge accumulation, local discharge, overheating, and electrode branching, the insulating performance of the polymer gradually deteriorates, ultimately leading to irreversible breakdown [2-3].

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In order to deal with this problem, as early as the 1960s, some scholars proposed the use of materials with certain conductance characteristics to optimize electric field distribution. However, the problem of concentration of electric field intensity in cable attachment media has not been studied in depth [4]. Subsequently, through the unremitting exploration of scholars at home and abroad, it is found that the use of nonlinear conductivity composite materials can maintain good insulation properties under normal working conditions. When the accumulation of space charge causes the internal field strength to reach a specific threshold, these materials can accelerate the release of space charge through the transient high conductivity mechanism, thus ensuring that the electric field strength inside the cable attachment medium is always maintained within a relatively safe range [5-8].

In this paper, the conductive mechanism and influencing factors of inorganic conductive particles and polymer matrix are summarized, and the research progress of nonlinear conductive materials is reviewed in order to provide a new reference scheme for HVDC transmission.

2. CONDUCTION MECHANISM OF NONLINEAR CONDUCTIVE COMPOSITES

At present, regarding the electrical conduction mechanism of electric field-controlled composite materials, the generally accepted theories include percolation theory, effective medium theory, quantum tunneling effect theory, field emission theory, etc. [9] These theories explain the electrical conduction mechanism of composite materials under different conditions from different perspectives.

2.1 PERCOLATION THEORY

In 1957, J. M. Hamersley proposed the percolation theory for the first time in the study of liquid flow phenomena in disordered porous media. Later, it was found that percolation is also common in polymer composites, and conductive composites can be prepared by adding a certain amount of conductive particles to the insulating polymer matrix, which is precisely because there is a "percolation phenomenon" between the concentration of conductive particles in the polymer matrix and the conductivity of the composite material, that is, with the increase of the concentration of filler in the composite material, The conductivity of the composite material is not proportional to the increase, but when the concentration of the filler particles inside the composite material increases to a threshold, the conductivity will increase abruptly, and the change range is very obvious. After this threshold, the conductivity of the composite slowly increases as the packing concentration continues to increase [10-12]. Specifically described as: when the concentration of the conductive filler is low, the average distance between the filler inside the polymer matrix is large, at this time, the conductivity of the composite material is mainly considered to be the conductivity of the polymer matrix. When the concentration of conductive fillers in the polymer continues to increase, the average distance between the fillers inside the polymer will gradually decrease, until the concentration of the filler exceeds a certain critical value, a conductive network can be formed between the filled particles, thus making the conductivity of the composite material suddenly increase, this phenomenon is called percolation phenomenon. The critical value is called the percolation threshold [13-14]. Percolation theory is a macroscopic explanation of the conductivity phenomenon of composite materials, which does not involve the essence, but can be widely used because it is easy to understand.

2.2 EFFECTIVE MEDIUM THEORY

The effective medium theory can not only be used to study and explain the influence of the relationship between matrix and filler on the conductivity of composite materials, but also to explain the influence of the basic microscopic morphology and distribution of filler on the overall conductivity, and is also helpful in predicting the conductivity of some composite materials [15]. Effective medium theory can be divided into two parts: classical effective medium theory and universal equation theory of effective medium. Among them, the classical effective theory can be understood as the average theory in essence, ignoring the small-range effect around the seepage threshold, so it is not easy to hold within the overall doping amount range, which is divided into two theories of homogenization and non-homogenization, respectively proposed by Bruggeman and Maxwell-Gamett [16-17]. In order to make up for the shortcomings of the effective medium theory, Mclachlan proposed the universal equation of effective medium [18]. However, since the premise of this theory is to require the doping to be 100% full of the entire polymer, the practical application of this theory is greatly limited [19].

2.3 QUANTUM TUNNELING THEORY

The theory was first introduced in 1963, and is a universal tunnel conduction equation derived by Simmons based on quantum theory. It can be described as follows: under different applied voltages, the insulation between electrodes will have a potential barrier that changes in size and width, and this microscopic difference will lead to a change in the current density of the composite material on the macro level. In view of other studies and theoretical analysis, the theory explains the conductive mechanism of composite materials as follows: when the spacing of conductive fillers in the composite materials is very small, electrons will transition between them to form a conductive network, so that the composite materials conduct electricity. However, when there are fewer fillers, the spacing between fillers is too large to form current conduction, and the tunneling current of the composite material will decrease exponentially, that is, the tunneling phenomenon only occurs between conductive fillers very close to each other [20-22].

2.4 FIELD ELECTRON EMISSION THEORY

This theory is similar to the quantum tunneling effect theory, which both believe that the tunneling effect is the main behavior of the composite material's electricity conduction, but the field electron emission theory has a different explanation for the cause of electron tunneling. According to Beek's research, the tunneling effect should be a special case of field emission inside conductive particles. When the distance between conductive particles is less than 10nm, the field intensity between the particles will be very large, and the conductive particles will interact with each other and transfer charges, resulting in the generation of current [23-26].

To sum up, both quantum tunneling theory and field electron emission theory believe that the conductivity of composite materials is related to the size of the applied field strength. In addition, other theories believe that the conductivity of composite materials is related to the concentration of fillers and other factors. In most cases, the conductive mechanism of polymer composites may be a combination of multiple theories, so it is relatively complex.

3 FACTORS AFFECTING NONLINEAR CONDUCTIVITY CHARACTERISTICS

In practical applications, the selection of polymer matrix is generally fixed, so the type, content and temperature of filler are the key factors affecting the nonlinear conductivity of composite materials.

3.1 INFLUENCE OF FILLER TYPE AND CONTENT

The conductivity characteristics of different fillers are different. Take the most common modified fillers as an example, ZnO is an anisotropic ceramic material, and its nonlinear conductivity characteristics are due to the typical disordered grain boundary structure inside. The nonlinear conductivity characteristics of SiC are derived from the particle surface oxidation, field emission and local heating theories, etc. Both of these fillers can effectively improve the nonlinear coefficient of composite materials [27]. However, carbon allotrope has a high conductivity, which can significantly improve the conductivity of composite materials when used as modified fillers, but the improvement of nonlinear coefficient is very limited, and the high content of fill will lead to the loss of nonlinear conductivity of composite materials [28]. Therefore, it can be concluded that the size, shape and grain boundary of the same kind of filler will also affect the nonlinear conductivity characteristics of the composite material, and we can improve the performance of the nonlinear conductivity composite material by adjusting the above parameters.

3.2 INFLUENCE OF TEMPERATURE

The insulating materials in HVDC cables are mostly polymer insulating materials, which have low heat conduction. The conductivity of insulating materials is greatly affected by temperature. When there is a temperature gradient in DC equipment, the conductivity of insulating materials will change significantly, resulting in local charge accumulation and changes in electric field distribution, which will adversely affect the service life of insulating materials. Therefore, it is necessary to study thermal insulation materials.

Huang et al. concluded by finite element simulation that the temperature gradient would cause the accumulation of heterpolar charge on the low temperature side of the anode of XLPE, resulting in the distortion of the electric field, and with the increase of the temperature gradient, the distortion rate of the electric field would also increase[29].

Teng et al. doped PDA-coated (BT60) particles into epoxy resin matrix to prepare composite materials. It is found that this method can significantly optimize the resistivity and temperature characteristics, thus optimizing the distribution of the DC electric field and maintaining the DC breakdown strength. The DC breakdown strength of the epoxy composite containing 20 wt% BT60-PDA is 99.5% of that of pure epoxy at 90 ℃, while the simulated maximum electric field is 49% lower compared to the latter [30].

Zhang et al. uniformly dispersed boron nitride nanosheets (BNNSs) in the SEBS phase to construct a thermal conductivity network structure based on the dual seepage process. The results show that the introduction of BNNSs significantly improves the direct current (DC) breakdown strength and space charge suppression. In addition, the thermal conductivity of the composite has increased threefold[31].

The conductivity characteristics of nonlinear conductivity composites are determined by the properties of the material and the external environment, so the influence of temperature on the nonlinear threshold field strength and nonlinear coefficient should be fully considered when designing the nonlinear conductivity modification scheme.

4 MEASURES TO IMPROVE THE PROPERTIES OF NONLINEAR CONDUCTIVE COMPOSITES

The addition of modified fillers is essentially to introduce impurities into the polymer matrix, and the content of the filler is too low to ensure the stability of the nonlinear conductivity of the composite. If the content is too high, the interface gap of the composite dielectric will lead to accelerated aging and deterioration, and the breakdown strength will be greatly reduced. In addition, due to the involvement of high-voltage insulation equipment, electronics industry, and spacecraft components and other application fields, the nonlinear threshold field strength and non-linear coefficient need to adapt to different application scenarios, and the thermal conductivity and mechanical strength of the composite material after the introduction of the filler can be adapted to the working environment for a long time should also be taken into account.

In recent years, scholars at home and abroad have devoted themselves to the study of improving the comprehensive properties of nonlinear conductive materials and the control of non-linear parameters. He et al. summarized the methods to improve the properties of nonlinear conductance composites and regulate the parameters of nonlinear conductance composites, which mainly include: artificial control of filler distribution, the use of nano-filler with high aspect ratio, the influence of polymer matrix, the influence of internal grain boundaries of the filler and the shape of the filler on the nonlinear parameters, and the addition of a second filler [32]. It is more feasible to control the parameters of the filler itself and introduce a second filler, which is mainly used to improve the performance of nonlinear conductive materials.

4.1 PARAMETER CONTROL OF A SINGLE FILLER

The dimension, size, shape and grain boundary of the filler are the key factors that affect the nonlinear conductivity of the composite. By adjusting the parameters of the filler itself, the seepage threshold of the filler can be effectively reduced and the nonlinear conductivity of the composite can be adjusted.

In order to study the uneven distribution of electric fields in insulated devices and components, Hu et al. used nano-sic, nano-ZnO and micro-ZnO particles as fillers. Five kinds of nonlinear conductive composites based on epoxy resin (EP) were prepared (nano-SiC/EP, nano-ZnO/EP, micro-ZnO/EP, nano-SiC/ZnO/EP and nano-micro-SiC/ZnO/EP). The mass fraction of inorganic fillers was 1, 3 and 5 wt%, respectively. The DC voltage characteristic results of the composite are shown in FIG. 1 and FIG.2. The conductivity and nonlinear coefficient of the composite increase with the increase of inorganic filler content. Under the same conditions, the conductivity and nonlinearity of SiC/EP are higher than nano-ZnO/EP and micro-ZnO/EP. The addition of zinc oxide and sic at the same time has a significant effect on the nonlinear coefficient of the composite. When the ratio of micro-ZnO to nano-SiC is 2:3, the nonlinear coefficient of the composite reaches a maximum of 3.506, which is significantly higher than that of other samples. Compared with nano-SiC/EP, micro-ZnO/EP and nano-ZnO/EP composites with inorganic filler content of 5 wt%, the nonlinear coefficients of the composites are increased by 0.82, 2.48 and 5.01 times, respectively [33].

In order to solve the problem of mechanical degradation of nonlinear conductivity composite materials with high filling content, Gaska and other scholars prepared graphite nanosheets/low density polyethylene (LDPE) composite materials. When the mass fraction of the filler was only 5%, The composite material has shown obvious nonlinear conductivity characteristics, and its mechanical properties are almost at the same level as before modification [34]. Nie et al. use silver-coated zinc oxide whiskers to modify silicone rubber materials, so that the pressure sensitive field strength of silicone rubber composite materials can be significantly reduced and the nonlinear coefficient can be increased at a lower filler content [35].

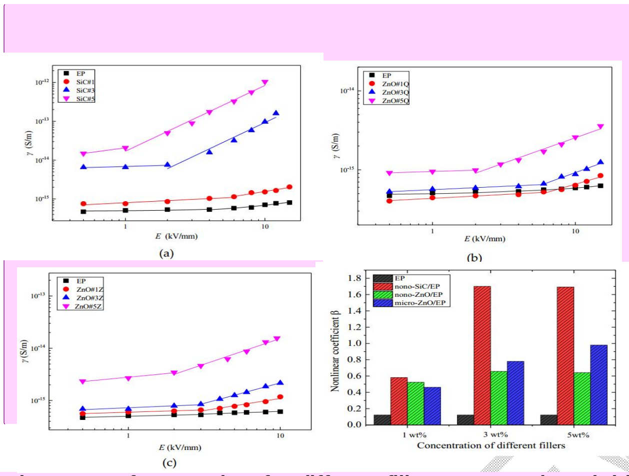

Figure 1. γ-E characteristic curves of composites for different filler concentrations (with respect to epoxy resin (EP)): (a) nano-SiC/EP composites; (b) nano-ZnO/EP composites; (c) micro-ZnO/EP composites. And nonlinear coefficients of three composite materials at different mass fractions[33].

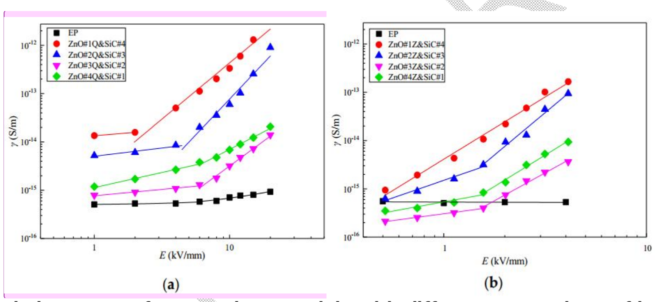

Figure 2. γ-E characteristic curves of composite materials with different proportions of inorganic fillers: (a) nano-ZnO/SiC/EP nanocomposites; (b) micro-ZnO/SiC/EP micro/nanocomposites [33].

4.2 ADOPTING TERNARY COMPOUND SYSTEM

Although the properties of the composites can be significantly improved by adjusting the parameters of the filler itself, the modification effect of a single filler on the polymer matrix is limited. In order to further improve the comprehensive energy of the composite material, a second filler is needed [36]. The scheme of using ternary composite system to improve the nonlinear conductivity of composite materials was proposed by Hidehito Matsuzaki and other scholars from Toshiba Corporation of Japan. By adding small diameter conductive or semi-conductive particles into ZnO/polymer system, more conductive channels can be formed under the condition of lower filler content. A nonlinear conductivity composite with excellent properties was obtained [37-38]. With the development of the material field, there are more options for second fillers.

Yang et al. used silicone rubber-based materials with nonlinear conductivity to improve the field distortion and charge accumulation problems in cable accessories by adding graphene (G; 0.25 phr) and alumina particles (Al_2O_3; 5-40 phr) to SR to obtain good nonlinear electrical conductivity and relatively high breakdown strength, as well as thermal conductivity. The results are shown in FIG. 3 and FIG. 4. At 23℃, the nonlinear conductivity α of $0.25G/40Al_2O_3/SR$ composite reaches 6.68, and the breakdown field strength E_0 is 49.1kV/mm, which is comparable to that of pure SR. In addition, at 23℃, the thermal conductivity of $0.25G/30Al_2O_3/SR$ is $0.28Wm^{-1}\ K^{-1}$, which is 114.98% higher than that of pure SR, indicating the application prospect of $G/Al_2O_3/SR$ composites as cable components [39].

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Iranian scholars Mashkouri et al used two-dimensional laminated exfoliated graphite (EG) to make the original ZnO/ high density polyethylene (HDPE). The pressure sensitive field strength of the HDPE composite system can be adjusted in the range of 40~400 kV/m, so as to meet different application requirements. With the increase of EG filler content, the nonlinear threshold field strength of the composite decreases from 400 kV/m to 40 kV/m[40].

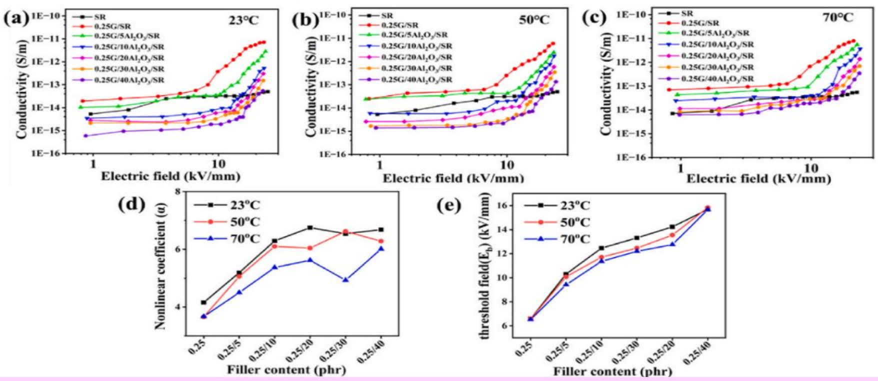

Figure 3.DC conductivity at (a) 23 ℃, (b) 50 ℃, (c) 70 ℃, and (d) The nonlinear coefficient α and (e) the threshold field strength E_b of the G/Al_2O_3/SR composites[39].

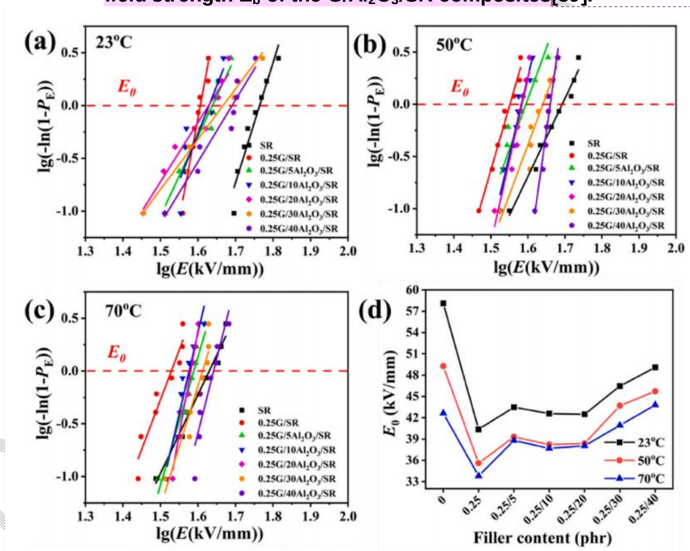

Figure 4.The Weibull distribution of breakdown strength at (a) 23 ℃ (b) 50 ℃ (c) 70 ℃, and (d) the characteristic breakdown field E_0 of the SR composites [39].

5. CONCLUSION

1) The nonlinear conductivity of polymer materials is affected by a variety of charge transport mechanisms, but there is no unified theory to explain the nonlinear conductivity mechanism of different composite systems under different external environments.

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2) The content of the filler in the composite has a certain influence on the formation of the nonlinear conductive seepage path, the injection of charge carriers and the interface effect between the filler and the matrix, thus affecting the difficulty of the formation of the nonlinear conductivity characteristics of the composite.

3) The preparation of nonlinear conductive polymer materials shows the trend of filler development from binary composite to ternary composite, and the new nonlinear conductive composite materials provide more reference schemes for the material selection of cable accessories.

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Manuscript Number: **Ms_JMSRR_127176**

Dear Chief Editor,

I hope this message finds you well. I have completed my review of the manuscript titled "**Nonlinear Conductivity Theory Research of Composite Materials** submitted to **Journal of Materials Science Research and Reviews**. I appreciate the opportunity to contribute to the peer-review process for this submission.

In my assessment, I have carefully evaluated the manuscript and provided detailed comments and suggestions aimed at improving its quality and contribution to the field as highlighted in the reviewed work.

The work is rich in content and has followed the journals format. However, there are few areas that need little adjustments, which I have outlined in my review. These include Abstract, Introduction, and results presentation, which I believe need to be addressed as the manuscript can be considered for publication.

I believe the work has the potential to make a meaningful contribution to the literature.

I trust that my feedback will be helpful to the authors and assist them in enhancing the quality and clarity of their work. Please let me know if there are any additional steps you would like me to take or if you require further clarification on any of the points raised in my review.

Thank you once again for the opportunity to review this manuscript. I look forward to hearing from you regarding the next steps in the review process if need be.
Best regards,

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