IEEE Access
Multidisciplinary ⋮ Rapid Review ⋮ Open Access Journal

Received January 15, 2019, accepted March 7, 2019, date of publication March 21, 2019, date of current version April 26, 2019.

Digital Object Identifier 10.1109/ACCESS.2019.2906662

Review of Nonlinear Conductivity Theory Research of Modified Composite Materials

CHENYANG LIU[1,2], YING ZHENG[3], BO ZHANG[1], XIAOQUAN ZHENG[1], SIQUAN HU[2], AND KUN HAN[2]

[1]State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, Xi'an 710049, China
[2]State Grid Xuji Group Corporation, Xuchang 461000, China
[3]Faculty of Humanities and Foreign Languages, Xi'an University of Technology, Xi'an 710049, China

Corresponding author: Chenyang Liu (liucheyan@126.com)

This work was supported by the National Natural Science Foundation of China under Grant 51377126.

ABSTRACT Nonlinear conductivity dielectric is widely used today to solve high energy discharge problems in many fields (such as spacecraft charging and motor insulation). This paper summarized the research about the nonlinear conductivity theory of composite materials. In particular, the key issues about nonlinear conductivity studies of composite materials are proposed. The nonlinear conductivity mechanisms of two typical filler particles (SiC and ZnO) are introduced. In addition, two kinds of nonlinear conductivity theory, including the traditional dielectric conduction theory and the polymer conduction theory, are reviewed. Several mechanisms in each of the conduction theories are analyzed, and also, the advantages and disadvantages of these theories are presented in detail. Thus, the purpose of this paper is to let the researchers know the nonlinear conductivity research status fully. We believe this paper is of great significance for the nonlinear dielectric conductivity researchers.

INDEX TERMS Nonlinear conductivity, composite materials, dielectric conduction theory, polymer conduction theory.

I. INTRODUCTION

Nowadays, polymers are widely used in spacecraft dielectrics because of its excellent insulation performance. But high energy electrons by irradiation in space plasma environment can penetrate the spacecraft surface and finally accumulate inside the dielectrics that can make dielectrics deep charged which can lead to discharge pulses to occur, this may seriously affect the normal operation of spacecraft, for example [1], the electrons accumulation leads to the destruction of solar panels. Therefore, in order to solve this problem, researchers have created a kind of special material whose conductivity can change (increase) as the electric fields increase. People call this material property as nonlinear conductivity. This kind of material usually is also called ''smart'' insulation material [2]–[4].

Many studies have shown that, using ''smart'' insulation materials with their nonlinear conductivity properties on spacecraft can reduce, or eliminate, the discharge pulse frequency, thus achieving an anti-static purpose [5]. As we all know, the conductivity of nonlinear materials can present an exponential increase with increase in electric field under a high electric field condition. When the high energy electrons accumulate in the material to a certain degree, the internal potential in the dielectrics will increase. However, due to the nonlinear conductivity property of the ''smart'' materials, material conductivity can rise instantly, and make the accumulated charges fast released; this can reduce the risk of spacecraft charging greatly [6]. However, due to some reasons such as secrecy requirements of space race between countries in the world, very few literatures about nonlinear conductivity materials of spacecraft can be found, so this article summarized the previous studies which are about nonlinear conductivity materials, and put forward the improving ways, the purpose is to give a positive reference for subsequent researchers in space field.

The associate editor coordinating the review of this manuscript and approving it for publication was Bora Onat.

II. SEVERAL KEY ISSUS INVOLVED IN

A. CHARGE TRANSPORT WAY

At present, the common way to prepare the modified composite materials is adding nano or micro-filler to based

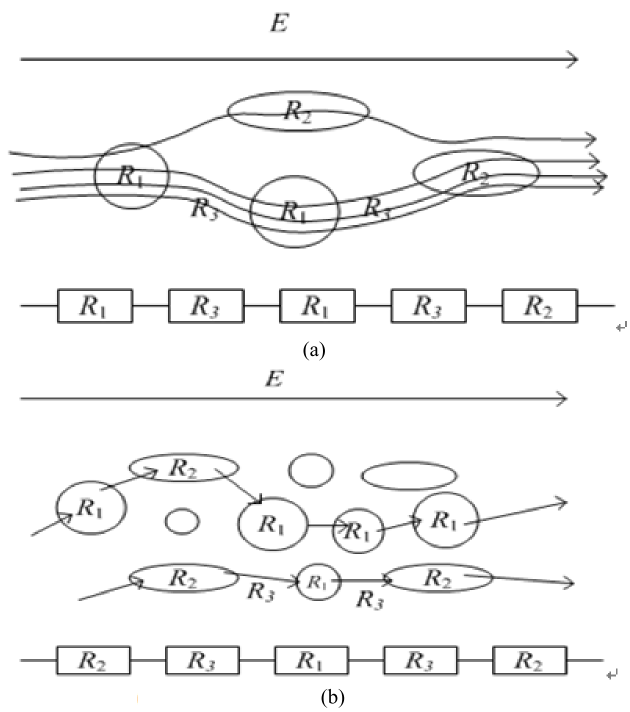


FIGURE 1. Charge transport ways. (a) Ohm conductivity path *E*. (b) Nonlinear conductivity path.

polymer resin [7], [8]. But the modified composite materials have complicated physico-chemical structures [9], when an electric field is applied, it would like to become much complicated to study how the internal carriers (charges) transmit in material inside. In a general way, in order to explore the charge transport mechanism of nonlinear conductivity materials, assume that there are two conductive paths existing—the ohm conductivity path and nonlinear conductivity path [10], as shown in Fig. 1. R_1 represents the resistance of modifier filler, R_2 represents the equivalent resistance of other reinforced material, R_3 represents the part resistance of based polymer resin, and $R_1 < R_2 < R_3$. Fig. 1 just shows one kind of possible path which the assumed current flows in, of which circle represents the modifier filler particle, the other blank area represents the based resin. Fig.1a shows the ohm conductivity path, in the ohm conductivity area, the flowing direction of current keeps the same with the electric-field vector, the current flowing path will always follow the principle of minimum resistance. Fig.1b shows the nonlinear conductivity path with a high electric-field, the current flowing path doesn't keep the same with the electric-field vector, the conduction path of carriers is chaotic, the conductivity mechanism will follow the interface principle of conduction particles. In fact, the real models of charge transport way are far more complicated than above [10], [11], so it is important for us to study the charge transport mechanism of nonlinear conductivity materials.

B. CONDUCTIVITY THEORY

Until now, about the research of conductive composite polymers, there are many conduction theory results which have been considered. One is the classic conductivity theory, another is the derived conduction theory. The classic conductive theories are as follows [44]: (1) hopping conduction theory (2) Schottky effect (3) Poole-Frenkel effect (4) tunneling effect (5) space charge-limited current effect. The derived conduction theories of polymer are [12]: (1) percolation theory (2) effective medium theory (3) tunneling-effect conductivity theory (4) Field emission theory. Of course, there are also other conductive mechanisms existed, but due to the intrinsic structure complexity of dielectrics, it has not yet built a unified theory which can explain all the nonlinear conductivity phenomenon until now. So aiming at the nonlinear mechanism explanation at present, researchers just can use one or several kinds of conductivity mechanisms together to interpret the nonlinear conductivity behavior of composite materials, thus, it has an important significance to develop the conductivity theory of nonlinear composite materials.

III. NONLINEAR CONDUCTIVITY RESEARCHES OF COMPOSITE MATERIALS

A. NONLINEAR CONDUCTIVITY THEORIES OF COMPOSITE MATERIALS

The modified composite materials usually are composed of based polymer resin and inorganic semi-conducting filler, or another reinforced material is added to enhance their mechanical properties. Most of the result showed that the modified composite materials presenting a nonlinear behavior has relations with the type, mass fraction and particle size of inorganic semi-conducting filler and so on. So the next part will introduce the conductivity theory of inorganic semi-conducting filler and composite materials in the first place.

1) TYPICAL CONDUCTIVITY MECHANISM OF INORGANIC SEMI-CONDUCTING FILLER

a: SiC INORGANIC SEMI-CONDUCTING FILLER

The corona phenomenon at the end of stator winding is a difficult problem for researchers all the time. In the early days, researchers had tried to add SiC filler to modify the insulating paint, the purpose is to solve this corona problem [13], [14]. The results showed that the modified SiC/paint can reduce the corona level to a certain degree. Since SiC was found to have a nonlinear conductivity behavior, later researchers also used SiC to create the nonlinear resistor and so on [15]. However, a large number of experimental studies still have not unified the nonlinear conductivity mechanisms of SiC filler until now. Furthermore, the nonlinear properties of SiC filler show a connection with the particles contacting each other. There are several conduction theories of SiC nonlinear behavior as follows:

② *Particle surface oxidation*

SiC itself has a very small resistivity, about 1-10$\Omega \cdot$ cm. A thin SiO_2 layer which is colloidal exists around the SiC particle surface usually, it has a high resistivity ($10^6 10^8 \Omega\cdot$ cm). When a high electric-field is applied, a big voltage-drop will develop in this thin layer, so generating a high electric-field. This time the conductive carriers can pass through this thin layer and form a tunnel current [16]. When the applied electric-field increases, the number of carriers will grow rapidly and the nonlinear conductivity behavior will show up [17].

② *Electronic emission*

Because SiC particles have a highly irregular shape, there are some prominent angle contact areas when the particles are accumulated. An air gap with certain distance could exist between two adjacent particles. When an electric-field applied in this gap exceeds their critical electric-field value, electronic emission will occur automatically at the edges or sharp corners of the SiC particles. When the applied electric-field is low, an internal current flows in the area between two particles contacting each other, and then the conductivity theory still obeys Ohm's law. If the applied electric-field grows continually, more and more air gaps begin to ''participate'' in conduction, this is equivalent to enlarging the effective contact area, lowering the SiC resistivity, and making the nonlinear conductivity phenomenon happen [18], [19].

② *Local heating theory*

This theory assumes that, due to the contact area distribution difference between two adjacent SiC particles, when the applied electric-field increases, the resistivity of SiC itself will decrease because of thermal resistance effect. This is the reason why SiC has a nonlinear conductivity behavior [19].

Of course, besides the above three theories there exists other conductive mechanisms for explaining SiC nonlinear behavior. Some researchers conclude that, because SiC itself has a complicated structure, a single theory cannot be used to explain its nonlinear phenomenon well, two or more theories should be combined together to interpret its nonlinear conductivity characteristics.

b: ZnO INORGANIC SEMI-CONDUCTING FILLER

In 1968, researchers just began to explore the study for ZnO piezo-resistor. ZnO as an excellent ceramic material which has an extensive application in power equipment, such as ZnO arrester, nonlinear switch, circuit break and so on [20]–[22]. G.D. Mahan *et al.* [23] proposed the two-step transportation model for ZnO nonlinear conductivity behavior, the researcher assumed that the thickness of boundary layer between two ZnO particles is not the same completely, if the thickness of boundary layer between two-phase particles is very thin (<3nm), due to the existed defects inside, a large number of electronic states can accumulate in interface layers, this will make the negative charge areas to build.

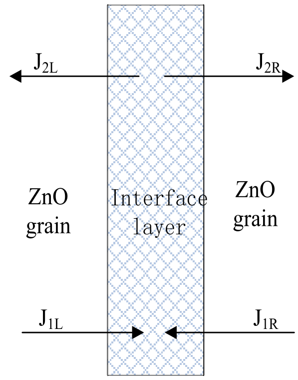


FIGURE 2. **Charge transport model [23].**

Therefore, the other side of grain in order to keep the electrostatic equilibrium will show a positive charge characteristic.

Fig. 2 shows the charge transport path in ZnO. When the electric-field is applied, electrons will inject into interface layer between two adjacent ZnO grains, and then pass into another ZnO grain, so four currents can be built near the interface layer, as shown in Fig. 2. J_{1L} represents the left-side injected current; J_{1R} represents the right-side injected current; J_{2L} represents the left-side extracted current; J_{2R} represents the right-side extracted current.

When the current state tends to be balanced, equation will be written as following [23]:

$$(J_{2R} + J_{2L}) / (J_{1R} + J_{IL}) = 1 \tag{1}$$

With the low electric-field, thermal excitation is the main factor for charge transmitting, and then the current density equation of thermal excitation is [23]:

$$J = J_0 \cdot \exp(-V_B/kT) \tag{2}$$

V_B-potential barrier, eV; k-boltzman constant, 1.38 10^{-23}J/K; T- temperature, K. Both sides of potential barrier on interface layer will change, charges may increase in the side of which potential barrier decreases due to tunnel effect, thermal current density will turn to [23]:

$$J = J_{2R} - J_{1R} = J_0 \cdot e^{(-\beta V_B)\left\{S^{\wedge(V_R)} - e^{-\beta(V_R - V_B)}\right\}} \tag{3}$$

S-the reformed interface charge, C; $^\wedge V_R$-tunnel effect factor. This model could explain some nonlinear conductivity phenomenon of ZnO under low electric-field condition.

When the applied electric-field increases, on the one hand, distortion near the barrier areas will occur, as shown in Fig. 3. A lot of holes at valence band in one side could be generated near the ZnO interface layer, on the other hand, many negative charges are generated correspondingly. A large number of holes can make the potential barrier reduced further, the tunnel effect becomes more obviously. This also proves that tunnel effect is the main reason which leads to the current increase in composite material [29]. As we all know, current is the result of electrons transport, we define ''hole'' the purpose is just to study its characteristics easily, ''holes''

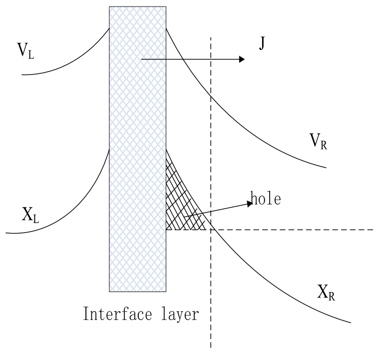


FIGURE 3. Hole assistant tunnel model [29].

increasing makes tunnel effect more obviously, more and more electrons could hop and penetrate the potential barrier, so current increases. Thus, this model by researchers also called it hole-assistant tunnel model, it can explain why ZnO presents nonlinear conductivity characteristics under high electric-fields preliminarily.

c: OTHER INORGANIC SEMI-CONDUCTING FILLERS

Of course, besides the above introduced two kinds of inorganic semi-conducting filler (SiC and ZnO), which all have an excellent nonlinear behavior, there are other semi-conducting fillers like WO_3 [24], Al_2O_3 [25] and SiO_2 [26] etc., they all show a certain nonlinear behavior. So as for these fillers which have different crystal structure, the nonlinear conduction mechanism is different too, this suggests that different crystal structure can have different charge transport mechanisms. Now the nonlinear conductivity theory about composite materials are still scattered, even as for one semi-conducting filler, researchers all have given different explanation theories for the nonlinear conductivity mechanism, it is more difficult to establish an accurate theory model. Therefore, as for the composite materials, what is mixed with nonlinear inorganic semi-conducting filler, we need to combine the conduction mechanism of semi-conducting filler with the conduction mechanism of polymer resin together to explain their nonlinear conductivity behavior. The mechanism exploration of the semi-conducting filler is still an important work for researchers.

2) NONLINEAR CONDUCTIVITY THEORY OF MODIFIED COMPOSITE MATERIALS

The conduction theories of polymer are: traditional dielectric conduction theory and polymer conduction theory.

a: TRADITIONAL DIELECTRIC CONDUCTION THEORY

The traditional dielectric conduction theories are: hopping conduction theory, Schottky effect theory, Poole-Freankel effect theory, tunnel effect theory and space charge limited current (SCLC) theory and so on. The details of these theories is as follows:

① *Hopping conduction theory:*

Most of polymers are composed of non-crystal and crystal areas together. In the non-crystal area, electrons must overcome the potential barrier between one crystal grain and another crystal grain to transport, when low electric field is applied, electrons will gain low energy(eV) to hop, but this time they will be influenced by the thermal action mainly, thermal action will make electrons-hopping to happen so that the hopping conductivity γ is built [27], the current density can be shown as:

$$J = \frac{qn_0 v\delta}{3} \cdot \exp\left(-\frac{\mu_0}{kT}\right) \cdot \sin\left(\frac{q\delta E}{2kT}\right) \tag{4}$$

q-charge, C; n_0-carrier concentration,1/m^3; ν-vibrational frequency, Hz; δ-carrier hopping distance, m; μ_0-potential barrier, eV; k-Boltzmann constant, 1.38×10^{-23}J/K; T-temperature, K; E-electric field, V/m.

Eq.4 shows that carrier hopping conduction process is closely related to temperature. It suggests that the current density increases with the temperature; this may explain why few polymers can have a nonlinear conductivity behavior under low electric field.

② *Schottky effect:*

Due to some factors influence, when the electric field applies to the composite material sample, the potential barrier of interface between cathode and sample surface will decrease, this leads to an increase of thermal electron emission current flowing in cathode. The current flowing past the material inside will increase too, this phenomenon researchers call it Schottky effect [28]. Equation is:

$$J = AT^2 \cdot \exp\left(\frac{\beta_s\sqrt{L} - \phi_D}{kT}\right) \tag{5}$$

A-Richardson Dushman constant; β_s -Schottky coefficient, $\beta_s = \sqrt{\frac{e^3}{4\pi\varepsilon\varepsilon_0}}$; ε, ε_0-relative dielectric constant, vacuum permittivity; Φ_D -work function, eV; T-temperature, K.

③ *Tunnel effect:*

Tunnel effect is also called as potential barrier penetration; let us see the charge transport mechanism. When a high electric field is applied to composite material sample, that will make the interface barrier inclined, next leads to that one part of carriers overcome the potential barrier and current density increases instantaneously [29]. The equation about current density changing with electric field is:

$$J = AE^2 \cdot \exp(-B/E) \tag{6}$$

A,B-constants related to work function, E-electric field, V/m.

④ *Poole-Frenkel effect:*

Under a high electric-field environment, the "donor level" on positive charges could overcome the potential barrier to "capture" the thermal emission electrons, this will lead to the increase of electronic current, this phenomenon is "Poole-Frenkel effect" [30]. Its current density is calculated as:

$$J = B \cdot E \cdot \exp\left[\left(\beta_F \cdot \sqrt{E} - \phi\right)/\mathrm{kT}\right] \tag{7}$$

B-constant; β_F -Poole-Frenkel factor, $B_F = \sqrt{e^3/\pi\varepsilon\varepsilon_0}$.

⑤ *Space charge limited current (SCLC):*

Also, the carriers could be captured by localized states in the transmission process, and accumulate in materials to build the space-charge areas, thus the number of carriers which transmit from cathode to anode will be limited. Therefore, the current density will be limited by the carrier mobility [31]. At this time, the current density can be written as:

$$J = \frac{9}{8} \cdot \frac{\varepsilon\varepsilon_0 \mu U^2}{d^3} \tag{8}$$

μ-carrier mobility, $m^2/V \cdot s$. when the applied voltage grows continually, the injected carriers will be captured by traps more and more, this process cannot quit until the whole traps are filled with the carriers. Therefore, the subsequent injection carriers will not be captured so that they can shift free inside materials. Once the applied voltage reaches the critical value U_L, the space charge limited current (SCLC) would be generated, this leads to the current density inside materials increasing quickly.

b: POLYMER CONDUCTION MECHANISM

Aiming at the conductive polymer, researchers obtained some approximative conductivity theories previously [32]–[34]. The agreed opinion aiming at the question why composite materials have a nonlinear conductivity behavior is that: because materials inside have some free charges (carriers), charges (carriers) with different shifting speed could transport and build a current under a high electric-field environment [35], [36]. This is the main reason to make the nonlinear conductivity phenomenon happen. In other words, to produce a nonlinear conductivity behavior for composite materials, two rules must be satisfied: (1) charges (carriers) need to be presented, and (2) a conductive pathway must be built by electric field.

① *Percolation theory:*

At present, there are two typical percolation theories which are used widely: one is statistical percolation theory, another is interface thermodynamic theory. Let's see two theories as follows.

In 1973, Kirkpatrick S proposed the statistical percolation theory. This theory considers the composite material sample to be a whole system, the whole system looks like an array which consists of many 2-dimensional or 3-dimensional points (keys). Consider the filler particles to be filled with these points or keys, once the filler particles concentration value exceeds its percolation threshold, the possession probability of points or keys exceeds a certain value, the conductive particles will be united and like to be seen as an unchained chain, it appears to show a long-term correlation behavior. Its mathematical equation is given by eq.9 [37].

$$\sigma = \sigma_p \cdot (\varphi - \varphi_c)^x \tag{9}$$

σ-conductivity of composite material, S/m; σ_p-conductivity of filler, S/m; φ-filler concentration, wt%; φ_c-percolation threshold, wt%; x-index, corresponding to the system dimension. This theory is appropriate for the situation that the amplitude of filler concentration value just exceeding the percolation threshold is very small. For 3-dimension system, $x = 1.9$. Of course, there is also another theory existed, -double percolation theory, it suggests that the whole system with a low filler concentration could have a good conductive effect.

The interface thermodynamic theory which is most representative could be the theory proposed by Miyasaka. This theory explains that, when the filler particles add to the polymer matrix, there are n number of conductive particles (volume V_0, surface S_0), so the surplus interface energy g in unit volume can be calculated as [38]:

$$\Delta g = KnPS_0 \tag{10}$$

K-the unit surplus interface energy; P- a constant which is related to dispersion degree of filler particles in polymer matrix, P=1 (the particles are dispersed very well), whereas P<1. If dispersion degree of filler particles in the polymer matrix is well initially but changes finally to an aggregation state, the final surplus interface energy is denoted as $\Delta g*$. Thermodynamics theory is needed to explain the mechanism of change in the polymer matrix. Thus, the critical volume fraction of filler particles can be deduced as [38]:

$$\varphi_c = 1 + \left[\left(\gamma_f^{1/2} - \gamma_p^{1/2}\right)\left(1 - e^{-et/\eta}\right) + K_0 e^{et/\eta}\right] \cdot \frac{S_0}{V_0} \cdot \frac{1}{\Delta g^*} \tag{11}$$

γ_f-the surface tension on conductive particles; γ_p-the surface tension on polymer matrix; t-mixing-molding time of composite materials; η-melt viscosity of polymer macromolecule. This theory considers that, the interface between polymer matrix and conductive particles is an important factor to affect the conductive channels building. Because the surface properties of conductive particles will change during the preparation process, the surplus interface energy of whole system will be generated. If the surplus interface energy increases more and more, the conductive channels between particles will be built, this is the reason why conductivity of composite material increases nonlinearly.

The above introduced two percolation theories can be used to explain the conductivity changing phenomenon of some conductive composite materials in situations, but it just considers from statistical view and simply thinks about that the building of conductive channel network depends on the filler concentration, they ignore the interface interaction between polymer matrix and conductive particles. The interface thermodynamic model focuses on the interface interaction between polymer matrix and conductive particles. However, because the shape of filler particle is usually not a perfect sphere, this makes the measuring of the surface tension very hard. In fact, it is hard to find the real conduction mechanism of composite materials. Thus, there are many limitations for the two percolation theories to be used widely.

② *Effective media theory:*

There are two effective-media theories which are typical, one is Bruggeman theory(uniform effective-media theory and non-uniform effective-media theory); another is Maxwell-Wagner theory [39].

Let us see the uniform effective-media theory first, assume that filler particles can disperse in polymer matrix uniformly, and the conductivity σ_m of composite material can be calculated as:

$$\frac{\varphi_h(\sigma_h-\sigma_m)}{\sigma_h+A\sigma_m}+\frac{(1-\varphi_h)(\sigma_l-\sigma_m)}{\sigma_l+A\sigma_m}=0 \quad (12)$$

σ_h-conductivity of filler particles, S/m; σ_l-conductivity of polymer matrix, S/m; φ_h-mass fraction of filler particles, wt%; A-demagnetizing factor, $A=(1-L)/L$, $L = 1/3$ means that the shape of filler particles is an ideal sphere.

Now introduce the non-uniform effective media Bruggeman theory, this theory assumes the filler particles are coated by polymer matrix absolutely, the conductivity of composite material will show as following:

$$\frac{(\sigma_m-\sigma_h)^3}{\sigma_m}=(1-\sigma_h)^3\cdot\frac{(\sigma_l-\sigma_h)^3}{\sigma_l} \quad (13)$$

The two Bruggeman theories are considered under an ideal situation. In fact, filler particles cannot be coated by polymer matrix totally and uniformly, the particle shape also cannot be a perfect sphere. So Bruggeman theories are just an approximate theory, it doesn't consider the condition what is about the thermodynamics and kinetics influence on the conductivity variation of composite materials, therefore, it cannot explain the building mechanism of conductive channel network fundamentally, so the effective media theory still has a big application limitation until now.

Based on the effective media theory, McLachlan *et al.* [40] proposed the GEM (General Effective Media) theory, the mathematic equation is:

$$\frac{\varphi_h\left(\sigma_h^{1/t}-\sigma_m^{1/t}\right)}{\sigma_h^{1/t}+A\sigma_m^{1/t}}+\frac{(1-\varphi_h)\left(\sigma_l^{1/t}-\sigma_m^{1/t}\right)}{\sigma_l^{1/t}+A\sigma_m^{1/t}}=0 \quad (14)$$

t-index, $t= m_l m_h/(m_l+m_h)$, m_l, m_h-component parameter of polymer and filler respectively. Although GEM theory can reflect the current variation in composite materials accurately, the structural parameters derived from Eq.14 cannot satisfy with that of conductive particles, so this theory application is limited.

Maxwell-Wagner theory is also called as infinite dilution theory. Assume that filler particles can disperse in polymer matrix uniformly, the conductivity of polymer matrix can be thought as 0 S/m approximately, so the conductivity of composite materials can be calculated as [40]:

$$\sigma_m=\sigma_h\left(1-\frac{3}{2}f\right) \quad (15)$$

σ_h-conductivity of filler particles, S/m; f-mass fraction of filler particles, wt%. Suppose that the shape of filler particle is elliptical, Eq.15 can be written as:

$$\sigma_m=\sigma_h\cdot\left(1-\frac{f}{1-L_f}\right) \quad (16)$$

L_f-demagnetizing factor.

Suppose that the shape of filler particle is spherical, the Maxwell-Wagner equation will be written as [40]:

$$\frac{\sigma_m-\sigma_h}{\sigma_m+2\sigma_h}=f\cdot\frac{\sigma_l-\sigma_h}{\sigma_l+2\sigma_h} \quad (17)$$

Eq.17 is suitable for explaining the nonlinear conductivity phenomenon of composite materials in case of low filler mass fraction (< 10 wt%), however, it cannot be used to explain the nonlinear behavior of composite materials with high filler mass fraction (> 20 wt%) [41].

③ *Tunnel conduction theory:*

Tunnel conduction theory proposed is based on the quantum mechanics theory. It can be divided into three types: Ezquerra conduction theory, low-temperature thermal hopping conduction theory and Simmons conduction theory.

Ezquerra *et al.* [42] calculated the conductivity σ_{dc} of composite materials between two particles, the equation is:

$$\sigma_{dc}=\sigma_f\cdot\exp(-2\,x_t \mathrm{d}) \quad (18)$$

σ_f-conductivity of filler particles, S/m; d-tunnel distance between two particles, m; $x_t=\sqrt{2\,mv(\mathrm{T})/h^2}$, m is the quality of charge, $\nu(\mathrm{T})$ is the potential barrier between two particles, h-Planck constant, $6.63\times10^{-34}J\cdot$s. the Eq.18 suggests that the conductivity of composite material changes exponentially, it is corresponding to the conductivity increasing of filler particles. When the mass fraction of filler increases, the distance between two particles will decline, this will lead to the conductivity of composite materials increasing.

The theory of low-temperature thermal hopping conduction is explained as follows: the value of mass fraction of filler reaches to the percolation threshold, this time the conduction process of carrier will not depend on the contacting area between two particles, and it only depends on the electrons hopping process between two particles. Therefore, the current density which flows through the composite material inside under low temperature environment can be calculated as [43]:

$$J(\varepsilon)=J_0\cdot\exp\left[-\frac{\pi\chi\omega}{2}\left(\frac{|\mathrm{E}|}{E_0}-1\right)^2\right] \quad (19)$$

J_0-Equivalent conductivity of gap between two particles, S/m; ω-Gap width, m; E-electric field between two particles, V/m; E_0-constant, $4\mathrm{V}_0/\mathrm{e}\omega$; $\chi=\sqrt{2\,mV_0/h^2}$, m-electron mass, 9.1×10^{-31}kg.

Eq.19 shows that the current density decreases with the decrease of gap width. When the gap width between two particles is very large, the current density flowing through composite materials will be small. In other words, the large gap width makes the tunnel effect between two particles happen hard.

In 1963, Simmons studied the conduction characteristics of insulating film between two metal electrodes and deduced the tunnel conduction equation as [44].

$$J=J_0\cdot\left[\overline{\varphi}\cdot\mathrm{e}^{-A\varphi^{-1/2}}-(\overline{\varphi}+\mathrm{eV})\cdot\mathrm{e}^{-A(\overline{\varphi}+\mathrm{eV})^{1/2}}\right] \quad (20)$$

And $J_0=e/2\pi h(\beta\Delta S)^2$, $A=(4\pi\beta\Delta\mathrm{S}/h)(2\,m)^{1/2}$.

φ-the average potential barrier, eV; m-electron mass, 9.1×10^{-31}kg e-electron charge 1.6×10^{-19}C V-applied voltage, V; β-correction factor; ΔS- tunnel distance between two particles, m; h-Planck constant, 6.63×10^{-34}J·s.

When the applied electric field is low, eV≈0, the average potential barrier $\overline{\varphi} >>$ eV . Because of $A\varphi^{-1/2} \gg 1$, Eq.20 will be simplified as [44]:

$$J = J_L \overline{\varphi} V \cdot e^{-A\varphi^{-1/2}} \tag{21}$$

And $J_L = \left[(2\,m)^{\frac{1}{2}}/\Delta S\right] \cdot (e/h)^2$. If ΔS=S, $\overline{\varphi} = \varphi_0$. Then, we can get the tunnel conduction equation under low electric field [44]:

$$J = \frac{3(2\,m\varphi)^{1/2}}{2\,S} \cdot \left(\frac{e}{h}\right)^2 \cdot V \cdot e^{\left[\frac{-4\pi S}{h}(2m\varphi_0)^{1/2}\right]} \tag{22}$$

If the applied electric-field is medium, $V < \varphi_0/e$. In this situation, ΔS=S, $\overline{\varphi} = \varphi - eV/2$, and then deduce a new equation, as shown in Eq.23 [44].

$$J = \frac{e}{2\pi h S^2} \cdot \left\{\left(\varphi_0 - \frac{eV}{2}\right) \cdot \exp\left[-\frac{4\pi S}{h}(2m)^{1/2}\left(\varphi_0 - \frac{eV}{2}\right)^{1/2}\right] - \left(\varphi_0 + \frac{eV}{2}\right) \cdot \exp\left[-\frac{4\pi S}{h}(2m)^{1/2}\left(\varphi_0 + \frac{eV}{2}\right)^{1/2}\right]\right\} \tag{23}$$

Similarly, when the applied electric field is high, V> φ/e, $\Delta S = S\varphi_0/eV$, $\overline{\varphi} = \varphi_0/2$. The tunnel conduction equation will become as following [44]:

$$J = \frac{2 \cdot 2e^3 E^2}{8\pi h\varphi_0} \left\{\exp\left[-\frac{8\pi}{2.96heE}(2m)^{1/2}\varphi_0^{3/2}\right] - \left(1 + \frac{2eV}{\varphi_0}\right) \cdot \exp\left[-\frac{8\pi}{2.96heE}(2m)^{1/2}\varphi_0^{3/2}\left(1 + \frac{2eV}{\varphi_0}\right)^{1/2}\right]\right\} \tag{24}$$

Simmons tunnel conduction theory studied the nonlinear conductivity behavior of composite materials with three different electric-field conditions (low, medium and high), but many research demonstrate that the conduction mechanism of composite materials cannot be always consistent with the above three electric-field conditions, this theory also couldn't explain the nonlinear conductivity performance of composite materials well.

④ *Field emission theory:*

Van proposed the field emission theory in 1962. This theory supposes that, when the gap width between two adjacent particles is <10nm, this will lead to the emission field producing under high electric field, thus the current density will increase, it can be calculated as [45]:

$$J = AE^n \cdot e^{-\frac{B}{E}} \tag{25}$$

J-current density, A/m^2; A-tunnel frequency; E-electric field, V/m; n-index, 1∼3; B-constant. Eq.25 shows that the current density increases exponentially with the increasing of electric field, it doesn't need to consider the temperature influence, so this theory is widely used to interpret the nonlinear conductivity behavior of composite materials.

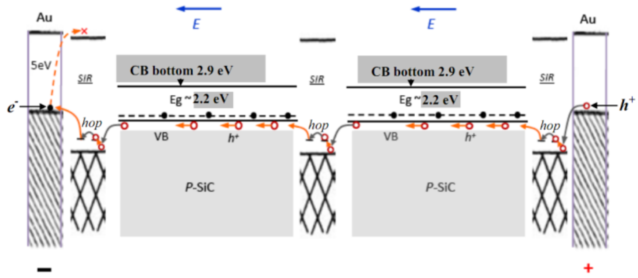


FIGURE 4. **The speculated hole injection and conduction model [46].**

The above stated conductivity theories can explain the nonlinear conductivity behavior of one part of modified composite materials under different electric-field conditions, however, researchers still cannot use one kind of theory to explain the nonlinear conductivity behavior of all modified composite materials, so the nonlinear conductivity theories are limited in many fields; many theories ignore the nonlinear conductivity characteristics of filler particles, and cannot combine the filler particle characteristics with the polymer well, so it needs to improve the nonlinear conductivity theory continuously.

Wang *et al.* [46] studied the nonlinear conductivity mechanism of Nano p-SiC/Silicone rubber composite, he proposed a nonlinear conductivity mechanism which is shown in Fig.4. The model can be explained as follows: First, hole injection by electrode is more likely to occur because the Schottky barrier height should be much lower compared to that of electron injection. For instance, if we assume the silicone rubber band gap is 6∼8eV, then the hole injection barrier height is around 1-3 eV, whereas the barrier height for electron injection is about 5 eV. Second, even if there are injected electrons, they will likely recombine with holes in p-SiC, therefore not contributing much to the steady-state current. The overall charge process may occur as follows: holes (h+) are Schottky injected from the anode, which travel through the neighboring thin rubbery layer via hopping across localize states near the silicone rubber valence band (VB) tail, followed by bulk conduction in the SiC VB. After that, it is postulated that holes are further internally emitted from p-SiC VB to the VB of the neighboring silicone rubber layer, followed by hopping transfer through that silicone rubber layer and so on, until eventually recombining with electrons at the cathode.

Liu *et al.* [47] proposed an interface conduction model that can be solved to explain the nonlinear conductivity behavior of composite materials. Liu suggested that the conductivity behaviors of nonlinear composite materials are different at low and high electric field conditions. This consideration colligates the semi-conducting interface conduction model and typical polymer interface conduction model well [48], [49]. This is because filler particles adding to a polymer matrix will generate many interfaces between the particles and the polymer matrix; a potential barrier at the contact interface exists also. When the electric field is low, the conduction carriers drift to the interface (Fig. 5a). The low-energy carriers

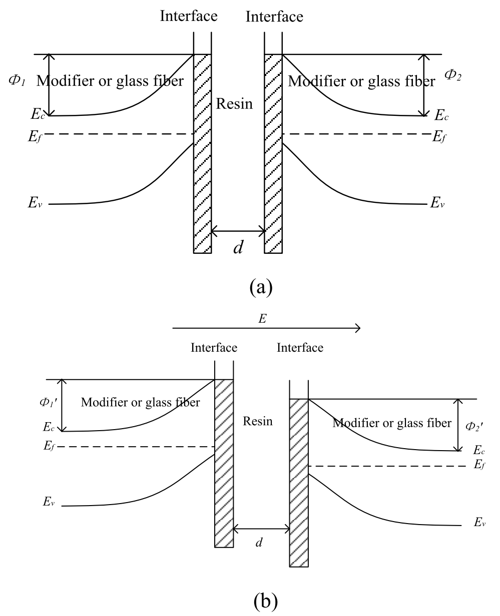


FIGURE 5. The potential barrier at the interface of Liu *et al.* [47]. (a) Without applied electric-field. (b) With applied electric-field.

are detained by the potential barrier at the interface. Some, but very few, of the higher-energy carriers, with thermal motion, may pass through the barrier and continue to travel onwards. When a strong electric field is applied, the barrier profile of the interface is tilted (Fig. 5b). One side of the barrier becomes higher than the other. With thermal motion, more carriers (electrons and holes) can tunnel through a tilted barrier than an un-tilted barrier. As a result, the current conduction increases with the applied electric field. This model can explain many nonlinear conductivity phenomenon of composite materials under high electric field condition well.

B. NONLINEAR CONDUCTIVITY RESEARCHES STATUS AT HOME AND ABROAD

Bai *et al.* [50] used one kind of nonlinear filler to modify the polyimide (PI), and studied how the filler mass fraction affects the conductivity characteristics of polyimide composite material. This research results showed that, when the mass fraction of filler is less than 5 wt%, the conductivity of polyimide composite material decreases slightly, the author thought that maybe this phenomenon is caused by the interface effect, but the researcher didn't give a deep discussion. but for filler mass fraction more than 5 wt% the conductivity of polyimide composite material would increase greatly. And the nonlinear conductivity of modified polyimide composite material could change observably, because more electrons can pass through the polymer layer due to the ''heat electrons hopping effect'', this makes the electronic conductivity of composite material increase. The proposed nonlinear conductivity mechanism is difference from the traditional strong electric-field theory totally, the filler particles adding to the polyimide will lead to a strong electric-field near the particles, when a high voltage is applied, filler particles conduct ''in series'' and lead to a nonlinear current which occurs at the polyimide layer. Also, the reason of dielectric constant increasing was studied. This work mainly used the hopping conduction theory to interpret the nonlinear conductivity phenomenon.

Steven Boggs ever found that conductivity of composite materials has an exponential increasing relationship with the electric-field, E [51]:

$$\gamma(E) = A \cdot \exp(B \cdot E) \tag{26}$$

$\gamma(E)$-conductivity, S/m; A- conductivity of material with 0 kV/mm, S/m; B- nonlinear coefficient. Aiming at this kind of nonlinear dielectrics whose permittivity and conductivity can change with the electric field, the researcher deduced the mathematic model as [51]:

$$J(E,T) = \gamma(E,T)E + E\frac{d\varepsilon(E,T)}{dt} + \varepsilon(E,T)\frac{dE}{dt} \tag{27}$$

Eq.27 suggests that the conductivity of material can increase with the increase of electric-field, the current density has a complicated relationship with the temperature, this equation only suggests that the AC current density changes with the frequency (50Hz$\sim$300kHz), however, the author didn't have a research for the DC conductivity changing, so there is still a large application limitation for this model.

Wei *et al.* [52] in his simulation studies evaluated the response behavior with different voltage excitations for the nonlinear composite material. The researcher simulated the polarization and depolarization process of linear and nonlinear dielectrics, the results indicated that, the way which measures the surface potential decay of self-discharge process of nonlinear composite material, to reflect the nonlinear conductivity degree for composite material, is feasible. Also the researcher had a depolarization and AC characteristics test for the SiC/PE composite materials, the test results and simulation results showed a good consistency finally. However, this research doesn't propose a real model for the nonlinear conductivity mechanism of modified composite.

Zhang [53] prepared the nonlinear LDPE composite materials with SiC filler, the author found that the nonlinear barrier layer can prevent the electrical tree from developing obviously, the conductivity and nonlinear coefficient of LDPE composite materials can increase with the increasing of nano-SiC contents, and COMSOL software was used to simulate the electric-field distribution in SiC/LDPE cable, and simulation results indicated that the added nonlinear barrier layer in cable insulation sides could uniform the electric field distribution at the defects on the cable insulation surface effectively and restrain the electrical tree formation. This work studied the nonlinear conductivity behavior of SiC/LDPE, and believed that the SiC adding makes the distance between two particles small, this makes a strong tunneling effect happen to generate a nonlinear conductivity changing, the classic conductivity theory was used to interpret the nonlinear conductivity changing. However, this research didn't consider the nonlinear characteristics of SiC filler.

Hu and Xie [54] used the simulation ways to explore the effect mechanism about nonlinear composite materials how to improve the electric-field distribution degree. Results indicated that the ratio $E_{\text{Vmax}}/E_{\text{aver}}$(the maximum electric-field on vertical direction/the average electric-field) of nonlinear composite materials compared with that of linear composite materials is declined obviously, the ratio is from 9.06 decreasing to 1.03, and this improves the electric-field distribution degree largely. Fig. 6 shows their simulated electric-field distribution and power-loss results of the nonlinear composite materials. This work suggests the nonlinear conductivity material has a good electric-field distribution property, but it cannot interpret the nonlinear mechanism.

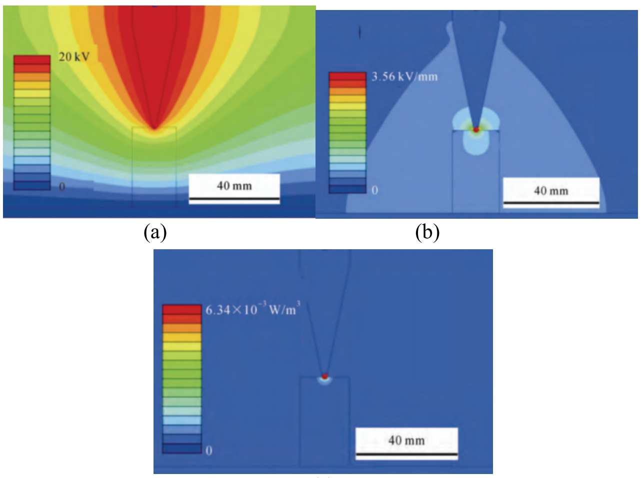


FIGURE 6. Hu and Xie's [54] simulation results of nonlinear materials. (a) Potential results. (b) Electric-field results. (c) Power-loss results.

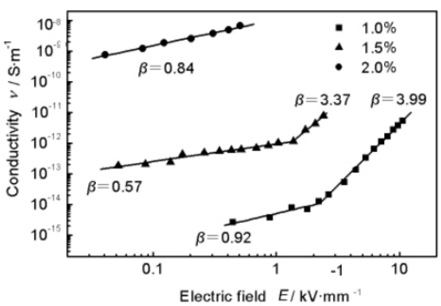


FIGURE 7. γ-E curve [55] of nonlinear materials.

Han *et al.* [55] aimed at the problem about how magnetic field affects the DC conductivity characteristics of CNT/LDPE composite materials, his research showed that, the magnetic field can lead to the crystallinity increasing of LDPE, volume resistivity increase too; Fig. 7 shows the nonlinear conductivity coefficient of Han's research [15]. The Fig.7 indicates that, when the CNT mass fraction increases from 1 wt% to 2.5 wt%, the CNT/LDPE composite materials showed an excellent nonlinear-conductivity behavior. As the CNT mass fraction increases, the electric-field value corresponding to the conductivity coefficient changes suddenly. The electric field E at the point of sudden change is the nonlinear conductivity threshold. This work suggests that the filler mass fraction has a main effect on the nonlinear conductivity threshold, the percolation theory could be used to explain the nonlinear conductivity changing of composite material, but it just gives an experimental research, the nonlinear conductivity mechanism is still not observed.

Wu *et al.* [5] added one kind of inorganic semi-conducting filler to PI and PTFE matrix and prepared a new nonlinear composite material, the researcher had studied the relationship between volume conductivity and filler mass fraction, temperature, electric-field, etc., results showed that filler mass fraction has a large influence on the nonlinear conductivity threshold value of nonlinear composite material. This work discussed the intrinsic conductivity and ionic conductivity of materials, it believes the nonlinear conductivity of modified PI can change largely under irradiation environment, the author explored that the nonlinear conductivity changing of composite materials can be attributed to the tuneling effect. But this work cannot give a theoretical analysis why the nonlinear conductivity of modified PI and PTFE is obvious than that of pure PI and PTFE.

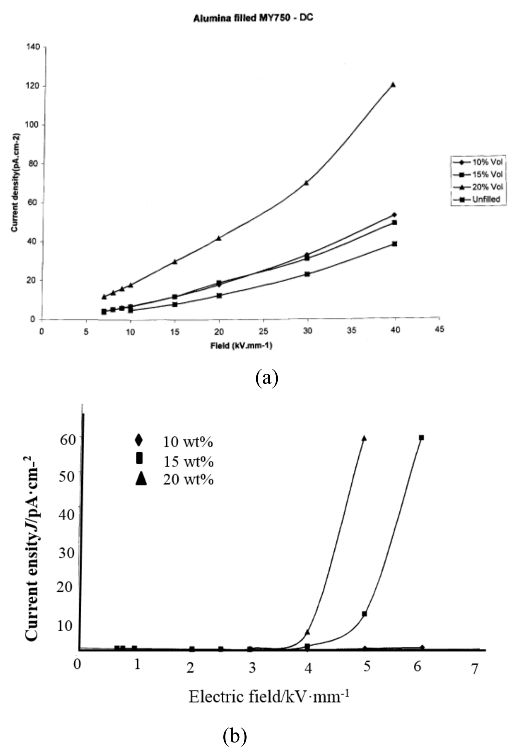


FIGURE 8. *J-E* curve of EP composite materials [56]. (a) Al_2O_3/EP composite materials. (b) ZnO/EP composite materials.

Donnelly and Varlow [56] had compared the nonlinear conductivity properties of Al_2O_3/EP with that of ZnO/EP composite materials (10 wt%, 15 wt% and 20 wt%) respectively under DC electric-field, as shown in Fig. 8. Results suggest that the Al_2O_3/EP and ZnO/EP composite materials all showed a nonlinear conductivity feature, when the Al_2O_3 mass fraction is 20 wt% and the applied electric-field grows to 30 kV/mm, the conductivity of Al_2O_3/EP composite material begins to increase nonlinearly and the value to be 7×10^{-15}S/m. However, the ZnO/EP composite material could present an obvious nonlinear-conductivity behavior just at 4 $\sim$ 5 kV/mm, test results also proves that the nonlinear conductivity performance of ZnO modifying EP are better than Al_2O_3. The author explained the nonlinear conductivity of composite materials by using the classical conductivity theory. This work gives an interesting explore for the nonlinear conductivity changing with different semi-conductor filler, but this research lacks the mechanism discussion about filler variety influence on the nonlinear conductivity changing of modified EP.

Lin *et al.* [57] studied the nonlinear conductivity performance of graphite/EP composite materials. They found that the*I-V* curves of EP composite materials with different graphite mass fraction are different, when the electric-field reaches a certain value, composite materials can present a reversible nonlinear-conductivity behavior. They proposed a nonlinear conductivity theory which combines the Non-linear Random Resistor Network (NLRRN) model with another theory named Dynamic Random Resistor Network (DRRN) model. Researchers called it Non-linear Dynamic Random Resistor Network (NDRRN) model. This model could explain why graphite/EP composite materials show a nonlinear conductivity behavior. But this theory model is used to explain the nonlinear conductivity for the conductive polymer, it cannot explain the nonlinear conductivity phenomenon of all insulating materials, and it ignores the interface effect, that makes this theory applied in a narrow field.

In 2006, Imai *et al.* [58] studied the electrical properties of EP composite materials modified with non-layered silicate and micro silicon dioxide, and observed the volume resistivity changing property with temperature, as shown in Fig. 9(a). The results indicate that, the filler mass faction with 1.5 wt% modified the EP composite materials whose volume resistivity is lower than that of pure EP, when the temperature increases, the resistivity of EP composite materials presents a nonlinear decreasing trend. On the other hand, the author studied the permittivity ε_r changing property with temperature, as shown in Fig.9(b). Test results showed that the permittivity has a small variation with the temperature increasing. This work studied how the interface area will influence on the conductivity changing of composite materials, also the researcher thought that the classic conductivity theory would interpret the nonlinear phenomenon, however, the detailed equation of nonlinear conductivity cannot be given.

Donnelly and Varlow [3] added ZnO filler to the epoxy and prepared the ZnO/EP composite materials, the researcher observed the AC conductivity characteristics of modified EP with different ZnO filler mass fraction, as shown in Fig. 10. The results showed that the nonlinear effect of 20 wt% modified ZnO/EP composite materials was remarkable. When the

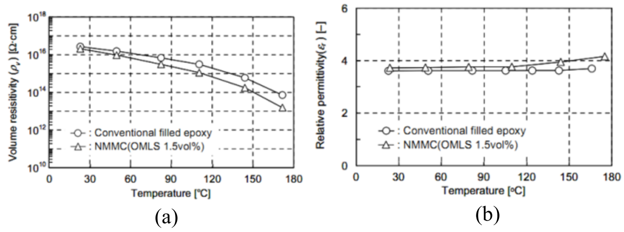


FIGURE 9. Nonlinear characteristics of EP composite materials of Imai *et al.* [58]. (a) Al_2O_3/EP composite materials. (b) Permittivity changing.

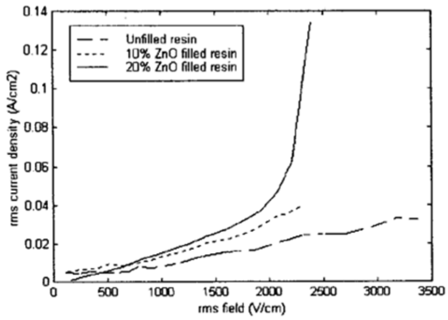


FIGURE 10. Donnelly and Varlow [3] nonlinear conductivity of modified ZnO/EP.

electric-field reached 2000 V/cm, the curve of nonlinear conductivity was expected to have a sharp slope, but the pure EP behaved linearly. These results also show that adding ZnO makes the modified ZnO/EP composite materials to have a nonlinear conductivity behavior. However, the author didn't give a discussion for the nonlinear behavior.

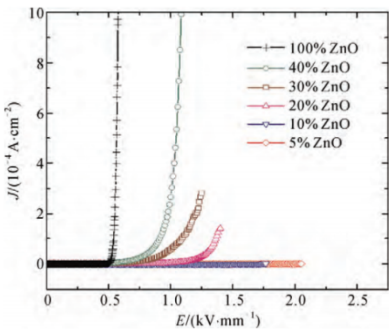


FIGURE 11. Nonlinear conductivity research of Xie *et al.* [59].

Xie *et al.* [59] used ZnO varistor-filler with different mass fraction to modify the silicone rubber (SiR), and studied their nonlinear conductivity characteristics, the results are shown in Fig. 11. The results showed that, when the ZnO varistor-filler mass fraction is >10 wt%, the ZnO/SiR composite material just can appear to have a slight nonlinear-conductivity trend, when the ZnO varisotr-filler mass fraction are 30 wt% and 40 wt%, the nonlinear coefficient of ZnO/SiR composite material can reach 16.9 and 17.4 respectively. Therefore, the nonlinear behavior of ZnO/SiR composite material becomes better with the increasing of filler mass fraction, the researcher suggested that because the mass fraction of filler exceeds the percolation value (percolation theory), the composite material with high mass fraction has an obvious nonlinear phenomenon. But this research did not consider the filler mass fraction increasing what has an influence on the breakdown-strength decreasing of ZnO/SiR composite material, it still has a large application limitation in fact.

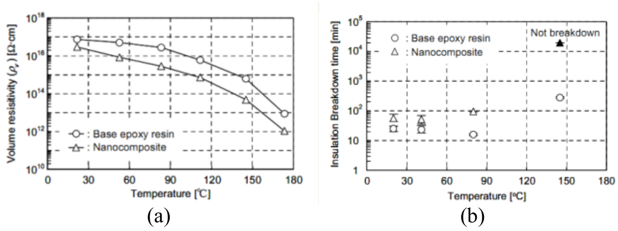


FIGURE 12. **Imai *et al.* [60] conductivity changing of nano-layered silicate/EP composite materials. (a) Volume resistivity. (b) Breakdown time.**

Imai *et al.* [60] has investigated with the nano-layered silicate/EP composite materials, and studied its volume resistivity changing behavior as the temperature increases. The results indicated that, the resistivity of silicate/EP composite materials showed a nonlinear decreasing trend with the temperature increasing, and the resistivity of nano-layered silicate/EP composite materials was lower than that of pure EP under the same temperature environment, as shown in Fig. 12. Kim *et al.* [21] also studied the temperature influence on the breakdown time of composite materials, and found that the breakdown time of nano-layered silicate/EP composite materials was much longer than that of pure EP under 10 kV at 1 kHZ. When the temperature was 145°, the breakdown time of nano-layered silicate/EP composite materials was expected to be 2×10^4 min, whereas it was just 280 min for pure epoxy. This work suggests that the temperature can influence on the conductivity changing of modified EP, but it lacks the mathematical derivation for the conductivity mechanism.

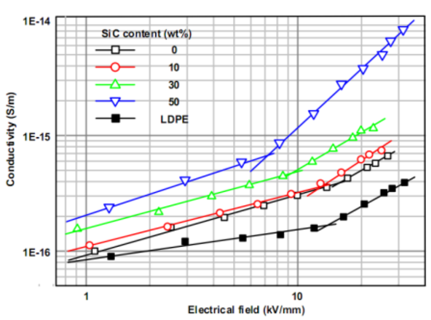


FIGURE 13. **Nonlinear conductivity research of Li *et al.* [61].**

Li *et al.* [61] had a nonlinear conductivity research for the EPDM/SiC composite materials(Fig.13), the researcher found that the nonlinear conductivity threshold of modified SiC/EPDM composite decreases and nonlinear behavior increases with the SiC filler mass fraction, filler mass fraction increase leads to the average distance between SiC particles decreasing, and makes the DC conductivity increase of EPDM/SiC. Also, the space charge behavior with the LDPE was studied, the SiC/EPDM composite sample will have a better interface accumulation. The author proves that the space charge-limited current effect (SCLC) is the main reason to make the nonlinear conductivity phenomenon happen.

Liang *et al.* [62] studied the nonlinear conductivity behavior of SiC/EP spacers, results showed that SiC/EP spacers have an obvious nonlinear conductivity phenomenon, the author observed that the nonlinear conductivity increasing can contribute a lot to the increase of SPD rate.

Although many researchers have studied many nonlinear conductivity task for the modified composite materials, such as the relationship between nonlinear conductivity and the flashover of modified composite material, the nonlinear conductivity behavior with the space charge, the nonlinear conductivity with the SPD(Surface Partial Discharge) and so on [63]–[65], the nonlinear conductivity mechanism is hard to be unified. The listed researches cannot give a unified theory to explain the nonlinear conductivity behavior, in the next article we will introduce the nonlinear conductivity theory.

The discussions above summarized the state of nonlinear conductivity theory of composite materials. There are many application limitations in some situations, because the modified composite materials may have complicated structures. The carriers transport mechanisms in such materials are often very hard to observe. The nonlinear theories need to be developed further in the future. It is important for us to continue to pursue deeper research into the nonlinear conductivity mechanisms.

IV. CONCLUSION

The purpose of this work is to summarize the nonlinear conductivity theories status of composite materials at home and abroad. Two important contributions of this research work are as follows:

(1) Two key problems about nonlinear conductance in composite material are described. They are the charge transport pathway and the conductivity theory. When the charges are transmitting within a composite material, they would pass through two regions (Ohm region and nonlinear Ohm region). Due to the complexity of composite material structure itself, until now, studies of charge transport behavior and properties were difficult.

(2) The advances in nonlinear conductivity experiments of new composite materials at home and abroad are summarized. Furthermore, the nonlinear conductivity mechanisms of filler particles are analyzed. Many researchers suggest that the nonlinear conductivity behavior of composite materials is corresponding to the filler category. SiC and ZnO filler all have a very famous nonlinear feature. At last, the two kinds of nonlinear conductivity theory (traditional dielectric conduction theory and polymer conduction theory) are introduced, and the limitations of every theory are analyzed also.

ACKNOWLEDGMENT

Special thanks for Professor ShuT.Lai working in MIT, who helped to revise this paper. The author would like to thank them all.

REFERENCES

[1] T. Kawasaki, S. Hosoda, J. Kim, K. Toyoda, and M. Cho, "Charge neutralization via arcing on a large solar array in the GEO plasma environment," *IEEE Trans. Plasma Sci.*, vol. 34, no. 5, pp. 1979–1985, Oct. 2006.

[2] J. Robertson and B. R. Varlow, "The use of non linear permittivity fillers for the purposes of stress grading within cables," in *Proc. 7th Int. Conf. Properties Appl. Dielectric Mater.*, Jun. 2003, pp. 1210–1213.

[3] K. Donnelly and B. R. Varlow, "AC conductivity effects of non-linear fillers in electrical insulation," in *Proc. Annu. Rep. Conf. Elect. Insul. Dielectric Phenomena*, Oct. 2000, pp. 132–135.

[4] B. Z. Han, W. M. Guo, and Z. H. Li, "Research on the non-linear conductivity characteristics of NANO-Sic silicone rubber composites," *J. Funct. Mater.*, vol. 39, no. 9, pp. 1490–1493, 2008.

[5] J. Wu, Y. L. Kang, Z. J. Zhang, and X. Q. Zheng, "Study on the deep dielectric charging protection technology of two typical polymers on spacecraft," *Vacuum Cryogenics*, vol. 18, no. 1, pp. 26–132, 2012.

[6] M. P. Cals, J. P. Marque, and C. Alquie, "Direct observation of space charge evolution in e-irradiated Kapton films," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 27, no. 4, pp. 763–767, Aug. 1991.

[7] K. Terashima, H. Sukuki, M. Hara, and K. Watanabe, "Practical application of ±250-kV DC-XLPE cable for Hokkaido-honshu HVDC link," *IEEE Trans. Power Del.*, vol. 13, no. 1, pp. 7–16, Jun. 1998.

[8] T. Tanaka *et al.*, "Dielectric properties of XLPE/Sio2 nanocomposites based on CIGRE WG D1.24 cooperative test results," *IEEE Trans. Dielectr. Electr. Insul*, vol. 18, no. 5, pp. 1482–1517, Oct. 2011.

[9] J. K. Nelson, *Dielectric Polymer Nanocomposites*. New York, NY, USA: Springer, 2010.

[10] P. Peng, "Study on conductivity modification technology of PI composites," M.S. thesis, School Elect. Eng., Xi'an Jiaotong Univ., Xi'an, China, 2015.

[11] C. Liu, "Nonlinear conductivity characteristics of modified epoxy composite materials in simulated space electron-injection environment," Ph.D. dissertation, School Elect. Eng., Xi'an Jiaotong Univ., Xi'an, China, 2017.

[12] W. M. Guo, "Research on nonlinear conductive characterstics and mechanisms of polyethylene composites filled with inorganic filler," Ph.D. dissertation, College Elect. Electron. Eng., Harbin Univ. Sci. Technol., Harbin, China, 2010.

[13] J. Budnick, "Reticulant doped semiconductive epoxies and plastics for high voltage applications," in *Proc. IEEE Conf. Nucl. Sci. Symp. Med. Imaging*, Nov. 1991, pp. 1004–1008.

[14] R. Malamud, I. Cheremisov, G. Shumovskaya, and T. Stepanova, "The principles of obtaining stable semiconducting compositions and reliable anti-corona designs for high voltage windings," in *Proc. IEEE Conf. Electr. Insul.*, May 2009, pp. 91–95.

[15] J. A. Martinez and D. W. Durbak, "Parameter determination for modeling systems transients-part V: Surge arresters," *IEEE Trans. Power Del.*, vol. 20, no. 3, pp. 2073–2078, Jul. 2005.

[16] H. Zhang, S. Wu, and H. Xie, "Study on the conduction mechanism of SiC corona suppression coating," *Trans. China Electrotechnical Soc.*, vol. 9, no. 3, pp. 60–63, Aug. 1989.

[17] X. M. He, "The conduction mechanism of SiC nonlinear resistor (in Chinese)," *Electroceram. Arrester*, vol. 1, no. 6, pp. 47–49, 1992.

[18] T. Terashige, N. Ishimoto, and K. Okano, "The effect of sintering atmosphere on electrical characteristics of fe_2TiO_5 pellet ceramics sintered at 1200ÂřC for NTC thermistor," *J. Phys., Conf. Series*, vol. 118, no. 9, pp. 401–406, 1998.

[19] L. Guo *et al.*, "Study progress of silicon carbide non-linear property (in Chinese)," *Insulating Mater.*, vol. 2, no. 3, pp. 60–64, 2005.

[20] S. Jayaraman and C. H. Lee, "Observation of two-photon conductivity in GaAs with nanosecond and picosecond light pulses," *Appl. Phys. Lett.*, vol. 20, no. 10, pp. 392–395, Oct. 1972.

[21] I. Kim, T. Funabashi, H. Sasaki, T. Hagiwara, and M. Kobayashi, "Study of ZnO arrester model for steep front wave," *IEEE Trans. Power Del.*, vol. 11, no. 2, pp. 834–841, Apr. 1996.

[22] R. Wu, Y. Wang, Z. Yan, Z. He, and L. Wang, "Design and testing of a novel inductive pulsed power supply consisting of HTS pulse power transformer and ZNO-based nonlinear resistor," *IEEE Trans. Plasma Sci.*, vol. 41, no. 7, pp. 1781–1786, Jul. 2013.

[23] G. D. Mahan, L. M. Levinson, and H. R. Philipp, "Theory of conduction in ZnO varistor," *J. Appl. Phys.*, vol. 50, no. 4, pp. 2799–2812, Apr. 1979.

[24] A. Zakria and Y. Wang, "Studies on fabrication conditions and doping of new low voltage WO 3-based varistor (in Chinese)," *J. Funct. Mater.*, vol. 30, no. 3, pp. 299–301, 1999.

[25] S. Park, K. Cho, J. Jung, and S. Kim, "Annealing effect of Al_2O_3 tunnel barriers in HfO_2-Based ReRAM devices on nonlinear resistive switching characteristics," *J. Nanosci. Nanotechnol.*, vol. 15, no. 10, pp. 7569–7572, Oct. 2015.

[26] S. Vijayalakshmi, H. Grebel, Z. Iqbal, and C. W. White, "Artificial dielectrics: Nonlinear properties of Si nanoclusters formed by ion implantation in SiO_2 glassy matrix," *J. Appl. Phys.*, vol. 84, no. 12, pp. 6502–6506, Sep. 1998.

[27] N. F. Mott, "Conduction in non-crystalline materials III. Localized states in a pseudogap and near extremities of conduction and valence bands," *Phil. Mag.*, vol. 19, no. 160, pp. 835–852, Sep. 1969.

[28] A. L. Smith and R. Breitwieser, "Richardson-dushman equation monograph," *J. Appl. Phys.*, vol. 41, no. 1, pp. 436–437, Nov. 1970.

[29] W. Jin, *Dielectric Physics*. (in Chinese), Beijing, China: China Machine Press, 1995.

[30] P. Notingher, A. Toureille, J. Santana, L. Martinotto, and M. Albertini, "Study of space charge accumulation in polyolefins submitted to ac stress," *IEEE Trans. Dielectrics Electr. Insul.*, vol. 8, no. 6, pp. 972–984, Dec. 2001.

[31] G. G. Raju, R. Shaikh, and S. Ul Haq, "Electrical conduction processes in polyimide films - I," *IEEE Trans. Dielectr. Elect. Insul.*, vol. 15, no. 3, pp. 663–670, Jun. 2008.

[32] J. S. Huh, H. R. Hwang, K. M. Lee, H. G. Byun, and J. O. Lim, "Sensing mechanism of conducting polymer sensor for volatile organic compounds," in *Proc. IEEE Conf. Mater. Sci.*, Feb. 2003, pp. 331–336.

[33] S. A. Moiz *et al.*, "Polaron hopping mechanism of conducting polymer," in *Proc. Int. Conf. Emerg. Technol.*, Oct. 2012, pp. 1–6.

[34] K. Doblhofer and C. Zhong, "The mechanism of electrochemical charge - transfer reactions on conducting polymer films," *Synth. Metals*, vol. 43, no. 91, pp. 2865–2870, Jun. 1991.

[35] J. Tahalyani, K. K. Rahangdale, and K. Balasubramanian, "Measuring semantic similarity 554 between words using web search engines," *Rsc Adv.*, vol. 6, no. 74, pp. 69733–69742, 2016.

[36] Y. Qiao and L. Shen, "Conductive mechanism of organic conducting polymer materials," *J. Shanxi Radio Tv Univ.*, vol. 1, no. 2, pp. 104–105, Apr. 2005.

[37] S. Kirkpatrick, "Percolation and conduction," *Rev. Modern Phys.*, vol. 45, no. 4, pp. 574–588, Oct. 1973.

[38] K. Miyasaka, "Mechanism of electrical conduction in electrically-conductive filler-polymer composites," *Int. Polymer Sci. Technol.*, vol. 6, no. 13, pp. 41–48, 1986.

[39] R. Landauer, "Electrical conductivity in inhomogeneous media," *AIP Conf. Proc.*, vol. 40, no. 1, pp. 2–45, 1978.

[40] D. S. McLachlan, M. Blaszkiewicz, and R. E. Newnham, "Electrical resistivity of composites," *J. Amer. Ceram. Soc.*, vol. 73, no. 8, pp. 2187–2203, Aug. 1990.

[41] A. Celikyilmaz, D. Hakkani-Tur, P. Pasupat, and R. Sarikaya, "Conduction in heterogeneous systems," *Adv. Electrochemistry Electrochem. Eng.*, vol. 2, no. 2, pp. 17–48, May 1962.

[42] T. A. Ezquerra, M. Kulescza, C. S. Cruz, and F. J. Baltá-Calleja, "Charge transport in polyethylene-graphite composite materials," *Adv. Mater.*, vol. 2, no. 12, pp. 597–600, Dec. 2010.

[43] B. Xi and G. Chen, "The mechanism of electrical conduction in polyethylene/carbon black composite," in *Proc. 6th Int. Conf. Properties Appl. Dielectric Mater.*, Jun. 2000, pp. 1015–1018.

[44] J. G. Simmons, "Electric tunnel effect between dissimilar electrodes separated by a thin insulating film," *J. Appl. Phys.*, vol. 34, no. 9, pp. 2581–2590, Sep. 1963. doi: 10.1063/1.1729774.

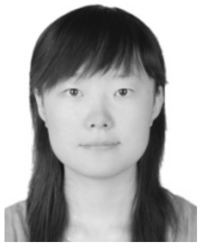
[45] L. K. H. V. Beek and B. I. C. F. van Pul, "Internal field emission in carbon black-loaded natural rubber vulcanizates," *J. Appl. Polymer Sci.*, vol. 6, no. 24, pp. 651–655, Nov. 1962.

[46] X. Wang *et al.*, "Mechanisms leading to nonlinear electrical response of a nano p-SiC/silicone rubber composite," *IEEE Trans. Dielectrics Elect. Insul.*, vol. 17, no. 6, pp. 1687–1696, Feb. 2010.

[47] C. Liu, X. Zheng, and P. Peng, "The nonlinear conductivity experiment and mechanism analysis of modified polyimide (PI) composite materials with inorganic filler," *IEEE Trans. Plasm. Sci*, vol. 43, no. 10, pp. 3727–3733, Oct. 2015.

[48] C. Li, J. He, and J. Hu, "Surface morphology and electrical characteristics of direct fluorinated epoxy-resin/alumina composite," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 23, no. 5, pp. 3071–3077, Oct. 2016.
[49] C. Li, J. Hu, C. Lin, B. Zhang, G. Zhang, and J. He, "Surface charge migration and dc surface flashover of surface-modified epoxy-based insulators," *J. Phys. D, Appl. Phys.*, vol. 50, Feb. 2017, Art. no. 065301.
[50] J. J. Bai, J. Wu, and J. F. Wang, "Influence of non-linear filler on electrical parameter of polyimide," *Aerosp. Mater. Technol.*, vol. 1, no. 3, pp. 53–55, Feb. 2010.
[51] S. Boggs and J. Kuang, "High field effects in solid dielectrics," *IEEE Elect. Insul. Mag.*, vol. 14, no. 6, pp. 5–12, Nov. 1998.
[52] Y. Y. Wei, H. Zheng, D. Zhou, and Z. H. Li, "Performance simulation of nonlinear dielectric under AC condition," *J. Heilongjiang Hydraulic Eng. College*, vol. 33, no. 4, pp. 75–77, 2006.
[53] Y. Zhang, "Research on application technology of nonlinear insulating materials in high voltage cable," (in Chinese), M.S. thesis, College Elect. Electron. Eng., Harbin Univ. Sci. Technol., Harbin, China, 2011.
[54] J. Hu and J. Xie, "Simulation analyse of modification effect of nonlinear composites on nonuniform electrical fields," *High Voltage Eng.*, vol. 3, no. 40, pp. 741–750, 2014.
[55] B. Z. Han, F. L. Ma, W. M. Guo, Y. J. Wang, and J. Hui, "Effect of magnetic field treatment on the electrical conductivity of low-density polyethylene and its composites with CNTs," *Carbon*, vol. 56, no. 1, p. 393, May 2013.
[56] K. P. Donnelly and B. R. Varlow, "Nonlinear dc and ac conductivity in electrically insulating composites," *IEEE Trans. Dielectrics Electr. Insul.*, vol. 10, no. 4, pp. 610–614, Aug. 2003.
[57] H. Lin, W. Lu, and G. Chen, "Nonlinear DC conduction behavior in epoxy resin/graphite nanosheets composites," *Phys. B, Condensed Matter*, vol. 400, no. 1, pp. 229–236, Nov. 2007.
[58] T. Imai *et al.*, "Effects of nano- and micro-filler mixture on electrical insulation properties of epoxy based composites," *IEEE Trans. Dielectrics Electr. Insul.*, vol. 13, no. 2, pp. 319–326, Apr. 2006.
[59] J. C. Xie, J. Hu, J. L. He, L. Gao, and Z. Guo, "Nonlinear dielectric and conductivity properties of ZnO varistor/silicone rubber polymer composites," *High Voltage Eng.*, vol. 41, no. 2, pp. 446–452, 2015.
[60] T. Imai *et al.*, "Influence of temperature on mechanical and insulation properties of epoxy-layered silicate nanocomposite," *IEEE Trans. Dielectrics Electr. Insul.*, vol. 13, no. 2, pp. 445–452, Apr. 2006.
[61] J. Li *et al.*, "Nonlinear conductivity and interface charge behaviors between LDPE and EPDM/SiC composite for HVDC cable accessory," *IEEE Trans. Dielectr. Elect. Insul.*, vol. 24, no. 3, pp. 1566–1573, May 2017.
[62] H. Liang *et al.*, "Effects of nonlinear conductivity on charge trapping and de-trapping behaviors in epoxy/SiC composites under DC Stress," *IET Sci. Meas. Technol.*, vol. 12, no. 1, pp. 83–89, May 2018.
[63] B. X. Du *et al.*, "Temperature dependent surface potential decay and flashover characteristics of epoxy/SiC composites," *IEEE Trans. Dielectr. Elect. Insul.*, vol. 25, no. 2, pp. 631–638, May 2018.
[64] B. X. Du and J. Li. "Effects of ambient temperature on surface charge and flashover of heat-shrinkable polymer under polarity reversal voltage," *IEEE Trans. Dielectr. Elect. Insul.*, vol. 23, no. 2, pp. 1190–1197, Feb. 2016.
[65] C. Liu *et al.*, "Discussion on non-linear conductivity characteristics with space charge behavior of modified epoxy for spacecraft," *IEEE Trans. Nuclear Sci.*, vol. 63, no. 5, pp. 2724–2730, Feb. 2016.

CHENYANG LIU was born in Henan, China, in 1991. He received the master's degree from Northeastern University, Shenyang, China, in 2013, and the Ph.D. degree from the State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, Xi'an, China, in 2017. He is currently a Postdoctoral Fellow with Xi'an Jiaotong University and also an Electrical Engineer with Xuji Group Corporation. His research interests include high-voltage test technology, space radiation, electrical insulation, and UHVDC transmission technology.



YING ZHENG was born in Shannxi, China, in 1987. She received the bachelor's degree from Xi'an Jiaotong University, in 2010, where she is currently a Teacher.

BO ZHANG was born in Yan'an, China, in 1991. He received the B.S. and master's degrees from Xi'an Jiaotong University, in 2013 and 2014, respectively, where he is currently pursuing the Ph.D. degree. His current research interest includes nanodielectric and surface charge of polymer insulating materials.

XIAOQUAN ZHENG was born in Shanxi, China, in 1954. He received the Ph.D. degree in electrical engineering from Xi'an Jiaotong University, Xi'an, China, in 2000, where he has been a Professor. His work research interest includes antistatic material of spacecraft.

SIQUAN HU was born in Henan, China, in 1976. He received the bachelor's degree from North China Electric Power University. He is currently the General Manager of State Grid Xuji Flexible Transmission System Corporation.

KUN HAN was born in Jiangsu, China, in 1983. He received the master's degree from Zhejiang University. He is currently with State Grid Xuji Flexible Transmission System Corporation.