Theoretical analysis and application of polymer-matrix field grading materials in HVDC cable terminals

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Abstract: The distortion and uneven distribution of electric field in high-voltage direct current (HVDC) terminal insulator is the key problem for stable running of the power transmission system. Semiconducting flexible materials filled polymer-matrix composites with non-linear conductivity have also been considered to modify the distribution of electric field in cable terminal. Several factors having influence on the non-linear conductivity will be analysed in this review.

1 Introduction

With the development of power transmission technology, high-voltage direct current (HVDC) has been put into use in long-distance transmission for its lower dissipation, larger transmission capacity, higher operation stability, less restrict in the transmission distance and the convenience to interconnect asynchronously power grids, comparing with high-voltage alternating current. While the cable terminal is one of the most important parts in the transmission line, as well as the weakest part due to an electric field distortion. The electric field commonly has distortion and nonuniform distribution inside HVDC cable terminals, which is the main reason for their destruction.

It is well known that the electric distribution under AC voltage depends on the permittivity of dielectric materials, which is stable for most of the insulating materials. As for the insulating materials under DC, the electric distribution is mainly depended on the conductivity of materials [1–3]. The conductivity of materials applied in HVDC cable terminal does often show a strong temperature dependence, which has a crucial effect to adjust the electric field distribution and suppress the electric field distortion.

2 Theoretical research

2.1 Classical theories in dielectric physics

A lot of researches have been done with the dielectric physics, but it is necessary to introduce some basic theories related to non-linear conductivity.

2.1.1 Hopping conduction theory: The electrons will go through band gap when they are thermally activated, forming free electron and hole. The free electron moves along the field direction. While the temperature is high enough to activate the electron overcoming the energy difference, the electron will transfer into its adjacent point. Mott [4] presented, however, electron may more likely transfer to further point with similar energy, when the temperature is low enough. It can be described by Arrhenius formula.

\[ J = J_0 \exp(-W/kT) \] (1)

where \( J_0 \) is a pre-exponential factor, \( W \) is an activation energy which can be estimated from the slope in the \( J–1/T \) curve, \( k \) is Boltzmann’s constant and \( T \) is the absolute temperature.

2.1.2 Fowler–Nordheim theory: The Fowler–Nordheim theory can also be called tunnelling theory, which describes the electrons from metal as wave-mechanical tunnelling through a triangular potential barrier without energy loss. It usually happens under high field when the barrier is very thin without distinct dependence on temperature. One of its main usage in nowadays focus on the conductivity theory about carbon-based materials [5, 6], which is a kind of non-linear conductive material we will discuss at next section.

2.1.3 Schottky theory: Different from ohmic contact, the interface barrier between metal and semiconductor is not low, band bending lead to the Schottky barrier. The carrier injection from electrode is governed by the rectification, which can be described by

\[ J = A T^2 \exp \left\{ -(\Phi - \beta q^{1/2}E/kT) \right\} \] (2)

\[ \beta = (e^3/(4\pi \varepsilon_0 \varepsilon_r)^{1/2} \] (3)

where \( A \) is a constant independent of electric field \( E \) and the temperature \( T \), \( \Phi \) is the effective work function between the Fermi level of the metal and the conduction band formed with insulator and metal, \( k \) is Boltzmann’s constant, \( \varepsilon_0 \) is the permittivity of free space, \( \varepsilon_r \) is the relative dielectric permittivity and \( e \) is the electronic charge. Fig. 1 shows the conduction mechanisms for a reverse-biased (n-type) Schottky barrier.

2.1.4 Poole–Frenkel effect: For the insulator under electric field, electrons can transfer through the insulator when they get out of localised state with the energy introduced by random thermal fluctuations to conduction band. However, for the high electric field, it will be easier for the electron to transfer into the conduction band with less energy from thermal fluctuation, which will also make the transfer more frequently [8–11]. The standard expression for Poole–Frenkel effect is as follows

\[ J \propto E \exp \left\{ -q(\phi_h - \sqrt{qE/(\pi e)}) / k_b T \right\} \] (4)

where \( J \) is the current density, \( E \) is the applied electric field, \( q \) is the
elementary charge, $\Phi_b$ is the voltage barrier (in zero applied electric field) that an electron must cross to move from one atom to another in the crystal, $\varepsilon$ is the dynamic permittivity, $k_B$ is Boltzmann’s constant, $T$ is the temperature.

2.1.5 Space-charge-limited current: When the concentration of injected charge carrier is predominant, it will determine the distribution of electric field [12]. At low voltages, the current is given by Ohm’s law. The interface charge is excited at different surface, for the different electric current density. The space-charge-limited current (SCLC) can be described by Child–Langmuir formula [13].

$$J = \frac{9}{8} \mu_e \varepsilon_0 \left( \frac{V^2}{d^3} \right)$$

where $\mu$ is the carrier mobility, $V$ is the applied voltage and $d$ is the thickness of the insulator, $\varepsilon_0$ and $\varepsilon_r$ are the relative permittivity and the vacuum permittivity, respectively, $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m.

Under that condition, the charge carrier, electric field and current compete and limit with each other. The carrier passing by space-charge area is restricted by the space charge. Auckland [14] fitted data to the formula and illustrated that charge conduction change from ohm to SCLC as the field increasing.

2.2 Theories in conductive polymer

2.2.1 Percolation theory: Percolation phenomena is easy to be observed at grain filled polymeric composites. When the filled concentration reaches to a certain level, some physical properties of the composites change remarkably. It is more obvious for the conductivity change in conductive particle filled polymer. The composite can transfer from insulator to conductor when the concentration of filler increases to a critical level with orders of magnitude change of conductivity. The corresponding critical value of filler concentration is called percolation threshold, which is presented as $\Phi_c$. It is directly related to the formation of conductive network throughout the composite, depending on the properties of matrix and processing conditions. Percolation threshold is an important parameter in conductive polymer materials.

Broadbent and Hammersley [15] were the first to explore percolation processes and research about comparing percolation process with the diffusion process. Mead [16] continued to study the percolation relationship with temperature. Percolation threshold is a crucial concept in understanding disorder and non-linear system, which is also the important theory to understand the conductive polymer filled with conducting or semiconducting fillers [17].

2.2.2 Fowler–Nordheim tunnelling conduction theory: When the de Broglie wavelength of microparticles is close to the barrier height which is the quantum barrier, microparticles will tunnel through the barrier with the wave-like behaviour. Fig. 2 is the three typical quantum potential barrier tunnelling.

The interface barrier of metal-insulator-semiconductor (MIS) can be seen as triangular barrier when the electric field is high as shown in Fig. 3. The barrier width is related to the electric field $E$. The tunnelling happen under that condition is called Fowler–Nordheim tunnelling.

2.2.3 Field emission theory: The electron in solid is restricted by the attraction of nucleus. While the external electric field strength is high enough, the electron will be pulled by the field to the surface, which is called field emission phenomenon. This theory considers the tunnelling as a particular case of field emission only two particles are close enough to interact with each other and form charge transport. This theory is widely used to explain some non-linear phenomenon.

2.2.4 Effective medium theory: Assuming that the property of a single-phase medium is same with the macro average of a multiphase medium, the single phase here is the effective medium of the multiphase. It is popular with four types as Maxwell–Garnett theory, Bruggeman theory, differential effective medium theory and Ping Sheng theory. Effective medium theory
is widely used in composite materials and can be used to explain and forecast the conductivity for the conducting polymer composite materials.

2.3 Models and theories used in non-linear conductivity of field grading materials

Based on the basic theories above, researchers have given several assumptions, theories and models to describe the non-linear conductivity. Oonby et al. [18] proposed a model based on their data about SiC/ethylene-propylene-diene monomer (EPDM) composite with the grain size changing from micrometre to submicron, which indicated that number of contacts through the thickness of the sample and the resistance of that contact is proportionate with the volume resistivity of the composite. The voltage of each contact is assumed to the percolation threshold and charge transformation completes through the shortest path. It can be described as [18]

\[ V_c = \frac{\text{applied voltage}}{\text{number of contacts}} = E_{AG} \]

where \( E_A \) is the applied electric field, and \( g \) is the particle diameter. The amount of electrical path can be affected by the filler distribution and volume concentration, which is different from the conduction under percolation. While it is not so effective for non-linear conductivity and a more complicated one should be taken in the future.

Surface band bending model is also used to explain the non-linear conductivity of semiconductor filled polymer composites. It is believed that the thermal motion of carriers in semiconductor come into being the inner electric field, which is the main reason for the non-linear conductivity of that kind of composite. While this theory is mostly used for the solar cell, further research may work on it for high-voltage field area.

As for the three-dimensional network model, it is proposed by Mårtensson and Gäfvert [19] to simulate the field depended behaviour of polymer composites, which covers the distribution of fillers, conductivity of fillers and some intrinsic properties of fillers. They also suggest that the contact between particles is controlled by double Schottky barrier when the filler content is far beyond percolation. This model fits the experimental data well. In Fig. 4, the contact between particles is considered as an resistance-capacitance circuits. There are two kind of contacts, one is face-to-face contacts for faceted particles, which is double Schottky barrier. The other one is edge contact, Fig. 4b. Considering the edge contact is indirect contact and the current need to be transmit through the matrix when the contact area is small, it is defined as a compound conductivity as shown in (7), which indicates the geometric mean conductivity of the whole part [20].

\[ \bar{\sigma}_c = \sqrt{\bar{\sigma}_t \bar{\sigma}_e} \]

3 Fillers used in non-linear conductive composite

According to the definition of Donzel [21], field grading fillers can be divided into two kinds based on the intrinsic property of the fillers, which can also be used to classify the fillers. For the first kind is that the filler does not have any non-linear conductivity, generally conductor. The composite’s non-linearity stems from the particle–particle contacts. The filler content has to be above the percolation threshold [22]. Another kind is that the non-linearity is the intrinsic property of the filler itself.

The ohmic \( j(E) \) behaviour at different voltage has a non-linear relationship under electrical fields lower than the breakdown field \( E_B \).

\[ j \sim E^\alpha \]

where \( j \) is the current density in materials, \( E \) is the applied electric field and \( \alpha \) is the non-linear coefficient.

In this review, we will classify the fillers of non-linearity materials. The carbon-based material is singled out because its unique and unknown properties.
3.1 Conducting materials

Using this kind of materials as fillers to modify the composite for field grading is the earlier research [23–25]. The common features of this kind of material are that they are irreversible and low breakdown strength. What is more the resistivity being sensitive to some subtle change [25]. When the filler content is well below the percolation threshold, it is believed that the average distance between conducting particles is large and no conducting paths can be established through the whole composite, which means that even tunnelling will not appear.

If the mean distance between filler particles is very close each other at percolation threshold, the tunnelling can occur with the help of electric field, as insisted by Van Beek [26]. Equation (9) describes the relation between electric field and the tunnelling current [26].

\[
J_{\text{Tunneling}} = A \cdot E^{n} \cdot \exp\left(\frac{B}{E}\right)
\]

where the \( B \) is a measure value of the energy barrier between the polymer and the filler. The \( \exp(-B/E) \) describes the transition probability of charge carriers between the filler and the polymer. With different mechanism, Frenkel [27] suggests a hopping mechanism for the charge carrier transport as

\[
J_{\text{hopping}} = A_{R} \cdot T^{2} \cdot \exp\left(\frac{K \cdot E^{1/2} \Phi}{k_{B} \cdot T}\right)
\]

As for the high loading, particles will be in close contact. The conduction of charge carriers occurs throughout the whole composite. The conductivity is mainly determined by the contacts between fillers.

The example of this mechanism can be seen at the article [28], the highly non-linear behaviour had not previously been observed in conductive composites before. They thought the polymer coated on the particle to prevent the direct contact from each other, which lead to a greatly high resistance even with the filler content above the percolation threshold. Simmons [29] proposed a universal equation based on quantum theory named tunnelling conduction equation. Several researchers used this equation to analysis the conductivity about polymers and the results are in good consistency with the equation [30–32].

Beek [26, 33] used (9) to study the conducting materials filled with carbon black (CB). Equation (10) was also used to studies the charge carrier transportation in CB filled polymer. The matrix and the CB particles determine the charge carrier transport. He thought a lot of factors have weight about the conducting, including the surface properties of the filler particles and the interfacial area [34]. Actually, as the carbon-based polymer composites have good thermal conductivity and electrical conductivity, it is more widely used in the positive temperature coefficient material, the conductivity is more related to temperature. When CB surface is coated with oxygen as the temperature increased, which phenomenon we mentioned at the beginning of this section, the conductivity of the composite will display a sharp drop [35]. It is the evidence for CB can be used in non-linear composites.

Some researchers doped carbon-based materials with other fillers to make non-linear polymer composites. For example, Martensson [23] doped SiC with CB and found that the conductivity would increase and the onset voltage would decrease as the CB content increased.

Generally speaking, for the conducting fillers, the filler content has a direct influence on the conductivity of the composites. The percolation threshold of this kind of non-linearity materials is often low. It is also difficult to observe the non-linear coefficient in this kind of conducting-filler polymer composites.

3.2 Semiconducting materials

Most of metal will be coated with the oxygen naturally without any protecting process, which will decrease the conductivity. Some metallic oxide semiconductors possess the varistor characteristics, which can be seen as a microvaristor [36, 37]. Some kind of this material has been used to regulate electric field like in the DC cable terminal since about 10 years ago and the effectiveness is better than the conducting materials filled polymer composites [21, 38].

To produce DC cable terminal, microvaristor is usually used as functional filler by mixing with polymer, such as silicone rubber and EPDM. The conductivity of such composite materials are determined by the physical–chemical characteristics and the content of fillers, which we will discuss at the next section.

The mostly used microvaristors are ZnO and SiC with semiconducting characteristic. ZnO are heterogeneous ceramic materials. Its varistor property is from its typical grain boundary structure, which is decided by the processing. When the conductivity turns into non-linear area, the conduction band of grain decreases to the value under valence band of the grain interface, the electron in hole will form current through tunnelling [39, 40]. The different manufacturing process can be seen at the article wrote by Einzinger [41–44]. Thanks to its stronger degree of non-linearity and high resistivity below their switching voltage and the non-linear coefficient of ceramic ZnO varistor can be higher than 40. It can build more compact accessories, while heat development remains under control [7, 45–50]. An exception example of the metallic oxide that do not have non-linearity is Al2O3 filled polymer composite. Being different from the inherent electrical sensitive characteristic, though the free electron and hole can be activated. The experiment result did not show that the conductivity of filled polymer composite had the non-linearity [47].

Another mainly used semiconductor is SiC, which is normally used electrical grade material [14, 51]. The dominating conduction mechanism in SiC is tunnelling by field emission, amplified by pre-avalanche multiplication. Fig. 5 is the model of back-to-back Schottky barrier to describe the conductive mechanism in SiC [51]. It means that the conductive feature is mainly controlled by the reverse bias barrier when the voltage is higher than the barrier height, while the positive barrier is considered highly conducting and related to pressure drop, which is lower than the applied field. At that condition, the conductivity will vary non-linearly.

When the content of semiconductor fillers is very high, contact voltage or low-temperature tunnelling can dominate the current, the conductivity of the materials will show the non-linear conductivity [18], and diagram of this mechanism can be seen at Fig. 1. In general, the semiconducting microvaristor does possess low conductivity before the threshold and high conductivity after the threshold, which is stronger than the conductor filled materials. It may be used as functional fillers for high-voltage cable accessory solving the problem about heat generation and distribution, which cannot be done well comparing with the conductor filler materials.
4 Characteristics and influencing factors of fillers

4.1 Morphology and distribution of fillers

According to percolation theory, with the increasing content of fillers, the conductivity of non-linear composite materials will meet a sudden change after a percolation threshold. Combining it with (11) to describe the percolation of composite.

\[ \sigma \propto (\phi - \Phi_c)^t \]  

where \( \Phi \) is the volume concentration, \( \Phi_c \) is the percolation threshold, \( t \) is the power law constant, depending on the system morphology, which is also the structure function of fillers’ morphology, dispersion degree and the connectivity between fillers [52, 53].

Lots of sphere fillers are not perfectly distributed in the matrix and the deformation during the processing will affect the morphology and occupying space of the fillers in composite. Balberg [54] introduced a concept of excluded volume to predicate the influence. When the volume fraction of a high aspect ratio filler is equal to the ratio of the actual volume of the filler over the excluded volume, the percolation is there. For the fillers with a high aspect ratio, the percolation threshold can be drastically reduced for particles with an aspect ratio larger than one [55]. We can make the carbon nanotube as an example [20, 56, 57]. In addition, Nettelblad [58] found that there are two percolation thresholds in the angulated SiC grain, while the polished SiC grain only has one. It also demonstrates the morphology has some influence on the \( \Phi_c \).

Another model to describe the morphology influence about the conductivity is the three-dimensional percolation model, which is first proposed by Bruggeman [59] with introducing a form factor \( f \), which is dependent on isotropic or anisotropic, the filler shape and particle distribution and so on. Tavernier [60] did some further research about it.

\[ \sigma_{\text{fmp}} = \frac{1}{((1 - \delta)/\sigma_m) + (\delta/\sigma_f)} \]  

where \( \sigma_m \) is the conductivity of the matrix, \( \sigma_f \) is the conductivity of the filler, \( \delta \) is the filler’s volume fraction, \( \sigma_{\text{fmp}} \) is the lower limit for the conductivity of the composite, which is the series connection. The upper limit is the parallel connection.

\[ \sigma_{\text{imp}} = (1 - \delta)/\sigma_m + \delta/\sigma_f \]  

The real conductivity should be some value between \( \sigma_{\text{fmp}} \) and \( \sigma_{\text{imp}} \), and the form factor \( f \) is 0 ≤ \( f \) ≤ 1.

\[ \sigma_{\text{f}} = f \sigma_{\text{fmp}} + (1 - f)\sigma_{\text{imp}} \]  

Assuming the evenly distribution of fillers in matrix is the common way to do the theoretical research and data analysis. It has been done to control the orientation and distribution through some specific processing method to change the electrical property of the composite materials. Electric or magnetic fields are often used to control the orientation of fillers to obtain the anisotropic conductivity during the processing [61–63].

4.2 Size of filler

A little different from the traditional tunnelling theory, the electrical percolation will not occur if the coated oxide layer or polymer layer separate the fillers from each other [64–67]. The distance between fillers decrease as the filler particle size decreases. Therefore, the smaller size of filler will lead to lower percolation threshold [68].

Onneby [18] proposed an assumption that the volume resistance is proportion to the number of contacts through the thickness of the sample and the resistance of that contact, every contact voltage reached the percolation threshold and the transmit path is the shortest. The voltage per grain-to-grain contact was obtained by assuming that a conductive path consists of a straight column of particles through the sample, the voltage across each contact can
be estimated by

\[ V_C = \frac{U_N}{N} = U_R \frac{g}{d} = E_A g \]  \hspace{1cm} (15)

where \( U_N \) is the applied voltage, \( E_A \) is the applied electric field, \( N \) is the number of contacts, \( g \) is the SiC grain size and \( d \) is the thickness of the sample. The resistivity of the material mixture is significantly reduced as the voltage across each contact is increased.

Onneby accomplished it in the SiC-filled EPDM composite with the filler size distribution from micronimetre to submicron as shown in Fig. 6. The onset field value of non-linearity and the resistivity decrease while increasing the filler size.

Glatz-Reichenbach [69] and Ruschau [70] did the theory analysis and considered that the contact resistance decreases with the increasing filler size. As it can be seen in Fig. 7, the same phenomenon is with the SiC-SiR of different filler size [71].

### 4.3 Hardness of filler

The hardness here can also be understood as the physical properties of the filler which has influence on the particle interface area. The harder the filler particle, the smaller is the contact area according to its distortion. Some particles such as silicones, borides and carbides are all hard fillers. For the particles like boron oxide, we can count it as soft fillers. The advantage of the hard particles is that they are easy to separate.

Glatz-Reichenbach [72] had some discuss about the contact spot area and considered the soft fillers would lead to smaller constriction resistance. Considering the effect of average force of thermal expansion, a thermoelastic mean-field model was used. For most of the researches, the filler contents just work well at low filler content [72, 73].

### 5 Engineering application in HVDC cable terminal

For field grading materials, the non-linearity of microvaristor composite materials have distinct advantage over the conductor filled ones, as we talked above, which have the disadvantages of that fillers’ content is limited to the percolation threshold [24] and the non-linearity of composite is not reproducible [74].

This kind of materials has been used in medium voltage cable terminal commonly for several years. One of the obstruction of applying in high field for several years was the heat generation and distribution, which may be harmonised by using microvaristor filled polymer composites. In the HVDC terminal applications, Fig. 8 cited from article [75] shows a DC cable joint where a field grading material is used.

The research about application in medium voltage has continued for several years [76], and with the demanding of high-voltage field, lots of improvement should be done to apply to it [77]. The companies including of Asea Brown Boveri Ltd. (ABB) and Tyco are the two main producers to the commercial product [77, 78] and their researchers [21, 38, 79, 80] have done plenty of research work about it. Other researchers have also done studies about it with experimental research and simulation one [21, 38, 79–85].

The space charge is also another key point influencing the non-linear conductivity in HVDC cable accessories. Researchers [21, 80, 86–89] have been studying the composite materials with field-dependent conductivity for several years. They focus on the space-charge formation and dissipation in composites, and suggest the composites with non-linear conductivity are helpful to improve the electric field grading property through raising the carrier mobility and suppressing the accumulation of space charge.

### 6 Conclusion

The conductive theory and non-linear conducting model is significant to manipulate non-linear conducting materials for HVDC cable terminal. Conductor filled material may have non-linearity but the non-linear coefficient is not high enough for high-voltage application. The semiconducting microvaristor filler has better non-linear conducting property and does well at HVDC cable terminal insulator, which is the main fillers we should focus on. For future development of material used in HVDC cable terminal, semiconductor with high thermal conductivity, beside the non-linear electrical conductivity, should be paid more attention.

### 7 Acknowledgments

This work was financially supported by NSF of China (grant nos. 51425201 and 51377010), Ministry of Education of China through Doctor Project (grant no. 20130006130002) and National Basic Research Program of China (973 Programme, 2014CB239503).

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