Effect of polarity-reversal voltage on charge accumulation and carrier mobility in silicone rubber/silicon carbide composites

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Funding information
Chinese National Natural Science Foundation, Grant/Award Numbers: 51537008, 51707133, U1966203

Abstract
Polarity-reversal voltage in high-voltage direct current transmission system would cause serious electric field distortion, accelerating charge injection. The effect of polarity reversal on charge transport characteristics in silicone rubber (SiR)/silicon carbide (SiC) composites with nonlinear conductivity at different temperatures is studied. The charge transport characteristics of silicone rubber/silicon carbide composites under unipolarity voltage and polarity-reversal voltage at 30°C, 70°C and 90°C are obtained by the isothermal surface potential decay method, and the carrier mobility and trap characteristics of the composites are further calculated. Studies have shown that a large amount of hetero-charge remains in silicone rubber materials under polarity-reversal voltage, which in turn leads to intensive charge neutralisation and severe electric field distortion. The silicon carbide doped composites exhibit excellent performance in suppressing the accumulation of residual charges and weakening the charge neutralisation under polarity-reversal voltage. In addition, high temperature enhances the conductivity of silicone rubber/silicon carbide composites and reduces the accumulation of residual charges, which restrains the distortion of the local electric field under polarity-reversal voltage. This research shows that the application of non-linear conductivity material is an effective measure to solve the problem of cable insulation under polarity-reversal voltage.

1 | INTRODUCTION

Cable accessories with complicated structure are one of the weakest components in high-voltage direct current (HVDC) transmission system [1, 2]. In HVDC cable system, due to the effect of DC electric field, charge accumulation occurs in the accessory body insulation, which results in the enhancement of local electric field [3]. Excessive local electric field will cause partial discharge and accelerate insulation ageing, eventually leading to insulation failure [4]. Therefore, it is essential to solve the problem of charge accumulation in cable accessories. For non-linear conductivity material, the increase in local electric field leads to the increase in conductivity, which in turn weakens and homogenises the local electric field [5]. The non-linear conductivity composites are commonly applied in DC cable accessories as control layers [1, 6], which can uniformly the DC electric field [7, 8].

Line-commutated converter-based HVDC (LCC-HVDC) transmission with the advantages of simple design and low line loss [9], is essential for large-capacity transmission over long distance and the integration of synchronous AC networks [10]. However, in LCC-HVDC transmission, the polarity-reversal voltage is required in the case of power flow reversal. The charges accumulated before reversal of voltage polarity are difficult to dissipate, leading to an electric field induced inside the insulation [11]. In addition, the newly applied electric field after polarity reversal will be superimposed with the internal electric field induced by the accumulated charges. The superimposed electric field will result in serious electric field distortion, accelerating charge injection and even causing partial discharge or breakdown [12]. Especially in the actual operation of HVDC transmission system, the cable insulation is subjected to the electro-thermal field [13], and the insulation problem is more serious, threatening the safe operation of the power system. It

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was believed that the acceleration of insulation ageing in DC cable under polarity-reversal voltage was related to space charge [14]. Under polarity-reversal voltage, the space charge distribution of full size cross-linked polyethylene (XLPE) power cable could not follow the polarity reversal synchronously, resulting in an increase in the electric field of the insulation [15]. The space charge distribution and field strength distortion of XLPE under polarity-reversal voltage were studied, and a method of filler addition was proposed to suppress electric field distortion [16]. It is considered that the application of dielectrics with non-linear conductivity is an effective means to restrain charge accumulation and electric field distortion. However, it is not clear how the non-linear conductivity materials affect the charge transport characteristics under polarity-reversal voltage.

This paper studies the charge transport characteristics of silicone rubber/silicon carbide (SiR/SiC) composites under unipolarity voltage and polarity-reversal voltage at different temperatures. The carrier mobility and trap characteristics of SiR/SiC composites are further analysed based on the isothermal surface potential decay (ISPD) process. Besides, the effect of temperature and SiC content on the polarity-reversal process in SiR/SiC composites is studied.

2 | EXPERIMENTAL ARRANGEMENT

2.1 | Sample preparation

Polydimethylvinyl SiR with molecular weight of 550,000 and vinyl content of 0.12 mol% is used in this study. Compared with nano-composites, the non-linear conductivity of micro-composites is stronger due to the easier formation of conductive paths. Therefore, α-SiC particles with an average particle size of 8 μm are selected. This work uses the SiC volume fractions of SiR/SiC composites which are 0, 29.2 and 38.2 vol%, respectively. Firstly, SiO₂, hexamethyldisilazane and polyvinyl silicone oil are separately added to the SiR matrix as reinforcing agent, structure controlling agent and tear resistant agent, respectively. The mixture is mixed in the twin-roll mixer until the components are evenly mixed. Secondly, 2,5-dimethyl-2,5-di-(tert-butylperoxy) hexane is mixed to the SiR matrix as the curing agent. Finally, the mixed materials are vulcanised in a mould with a diameter of 70 mm and a thickness of 0.3 mm at 160°C under 15 MPa for 10 min. Besides, polyethylene terephthalate (PET) films with smooth surface are placed on both sides of the mixed materials and mould to avoid air bubbles.

2.2 | DC conductivity measurement

The temperature-dependent DC conductivity measurement device is based on the three-electrode system. An electrometer Keithley 6517B is applied on this device with the measuring range of 1 fA~20 mA. The samples are placed inside a sealed heating tank. The temperature can be controlled by the external control panel of the heating tank. The maximum operating temperature of HVDC cables is 70°C, and the heating is more serious during the actual operation. Therefore, this work sets the temperatures to 30°C, 70°C and 90°C, respectively. Before the measurement, samples are short-circuited in the drying oven for 12 h. A typical I-t curve is shown in Figure 1. In the initial stage of measurement, there is a rapidly decaying polarisation current in the sample. As time increases, the sample exhibits stable conduction current [17]. Each measurement lasts for 30 min, and this work selects the average of the current measured in the last 100 s as the conduction current of the sample. The conductivity is calculated as follows:

\[
\sigma = \frac{I}{U} \cdot \frac{4L}{\pi (d + g)^2}
\]

where \(I\) is the conduction current, \(U\) is the DC voltage, \(L\) is the sample thickness, \(d\) is the diameter of the main electrode and \(g\) is the earth guard ring gap.

2.3 | Surface potential measurement

The surface charge characteristics of the samples are measured by the surface charge measurement system shown in Figure 2(a) [18]. This work uses DC voltage sources with opposite polarities, and the voltage is increased to 5 kV with a ramp rate of 1 kV/s. Firstly, a voltage of specific polarity is applied to the sample for 300 s. After the polarity is reversed, a voltage of opposite polarity is applied for 300 s. As a control, positive and negative voltage without polarity reversal is applied to the sample for 300 s. The unipolarity negative voltage is used to compare with positive-to-negative voltage, and the unipolarity positive voltage is used to compare with negative-to-positive voltage. After the corona charging, surface charge transport characteristics of SiR/SiC composites under unipolarity voltage and polarity-reversal voltage are measured. In addition, the
average ISPD rate is defined as

\[ R = \frac{V_1 - V_2}{t_1 - t_2} \]  

(2)

where \( R \) is the average ISPD rate; \( V_1 \) is the surface potential at \( t_1 \); \( V_2 \) is the surface potential at \( t_2 \).

When the thickness of the samples is thin enough, the ISPD process under the DC electric field can be used to analyse the carrier mobility and trap characteristics of the samples. The carrier mobility of the composites can be calculated according to the following equation:

\[ \mu = \frac{L^2}{t_T V_0} \]  

(3)

where \( t_T \) is the transit time and represents the time for charges to pass through the sample. \( V_0 \) is the initial surface potential.

When the time interval between \( t_1 \) and \( t_2 \) approaches 0, \( R \) equals \( \frac{dV}{dt} \) and represents the ISPD rate at a certain time.

A typical \( \frac{dV}{dt} \) curve with time in bilogarithmic coordinates is shown in Figure 3. When \( t \) is less than \( t_T \), \( \frac{dV}{dt} \) is approximately constant; when \( t \) is greater than \( t_T \), \( \frac{dV}{dt} \) is a linear function. The inflection point of the curve is \( t_T \) [4].

The relationship between the trap density \( N_t(E) \) and the trap-level \( E_t \) is as follows [19]:

\[ N_t(E) = \frac{4\varepsilon}{\alpha L^2 k^2 T^2 \ln(\nu)} \left| \frac{dV}{dt} \right|, \]  

(4)

where \( \varepsilon \) is the dielectric constant, \( e \) is the electronic charge, \( k \) is Boltzmann constant, \( T \) is the temperature and \( \nu \) is the electronic escape frequency of trapped electrons. The characteristics of deep and shallow traps are studied by means of peak separation.

3 | EXPERIMENTAL RESULTS

3.1 | Sample characteristics

SiR/SiC composites are characterised by scanning electron microscopy (SEM, ZEISS Gemini 500). SiR/SiC composites are placed in liquid nitrogen for brittle fracture, and the cross section is observed by SEM. As shown in Figure 4(a), it can be seen that the diameter of α-SiC particles is relatively uniform in each direction. In Figure 4(b), the SiO₂ particles are uniformly distributed in the 0 vol% sample. Figure 4(c) and (d) shows that the SiC particles are uniformly dispersed in the 29.2 and 38.2 vol% samples without obvious particle agglomeration, and the SiC particles in the 38.2 vol% sample are more densely distributed.

Figure 5 shows the relation between the conductivity and the electric field of SiR/SiC composites at different temperatures. It can be seen that the conductivity of SiR/SiC composites is
The conductivity of the SiC-doped samples is significantly enhanced under high electric field, and the threshold electric field is less than 2 kV/mm. When the applied electric field is low, the carriers with irregular thermal motion are blocked by the barrier in the interface region between SiR matrix and SiC particles, and there is no significant directional migration of carriers. When the applied electric field is enhanced, the barrier is inclined along the direction of the electric field and its height is reduced, making it easier for carriers to cross the barrier. Besides, the carriers can pass through the barrier directly under the action of high field due to the quantum tunnelling effect. In addition, according to percolation theory, the conductive network is formed inside the 29.2 and 38.2 vol% samples, which greatly increases the conductivity. At 30°C and 70°C, the conductivity of the 0 vol% sample hardly changes with the electric field, but increases significantly with the increase in electric field at 90°C. This is because most of the carriers in the 0 vol% sample are in the forbidden band, which are difficult to be driven by the electric field. When the temperature rises, the intense thermal motion provides these carriers more energy to hop into the conduction band, making it easier to migrate. However, the conductivity of the 29.2 and 38.2 vol% samples begins to increase rapidly at 2 kV/mm, indicating that the conductivity is significantly affected by the electric field. Besides, as the temperature goes up, the slope of the conductivity versus electric field in the 0 vol% sample becomes larger, while the slope in the 29.2 and 38.2 vol% samples is reduced, especially for the 38.2 vol% sample. This is mainly because the intense lattice vibration of SiC caused by high temperature increases the probability of carrier scattering, resulting in temperature even playing a negative role in increasing the conductivity of SiR/SiC composites [20].

3.2 Charge accumulation

Figure 6 shows the initial surface potential of SiR/SiC composites under unipolarity voltage and polarity-reversal voltage at 30°C. As shown in Figure 6(a), the initial surface potential of the 0 vol% sample reaches −3321 V under unipolarity negative voltage. The initial surface potential of the 29.2 and 38.2 vol% samples is lower than that of the 0 vol% sample, and the 38.2 vol% sample has the lowest initial surface potential among the three samples. This is due to the low conductivity of the 0 vol% sample, and the charges are easy to accumulate during corona discharge. The 29.2 and 38.2 vol% samples have higher conductivity and more charges will be released to the ground electrode during corona discharge. Under positive-to-negative voltage, the initial surface potential of the 0 vol% sample is still negative. It can be deduced that the original positive charges on the surface of sample are neutralised, and the negative charges further accumulate. The 29.2 and 38.2 vol% samples show the same pattern, but the initial surface potential is lower than the 0 vol% sample, due to the high conductivity. Comparing the initial surface potential under positive-to-negative voltage and unipolarity negative voltage, it is found that the initial surface potential of the 0 vol% sample decreases most, followed by the 29.2 and 38.2 vol% samples. This is due to the large accumulation of residual-positive charges in the 0 vol% sample before polarity reversal from positive to negative.

As shown in Figure 6(b), the initial surface potential of SiC/SiR composites under unipolarity positive voltage is similar to that under unipolarity negative voltage, but the initial
The initial surface potential is lower. This phenomenon also appears in the research of other scholars [21]. When the needle electrode is positively charged, the negative charges generated by air ionisation are neutralised by the needle electrode. A large number of positive charges accumulate near the electrode, which weakens the electric field near the needle tip. Eventually, less positive charges migrate to the surface of the sample slowly. When the needle electrode is negatively charged, the positive charges generated by ionisation accumulate near the needle tip, which increases the local electric field, and more negative charges move to the surface of the sample quickly. Therefore, the negative voltage is more likely to cause the surface charge accumulation under the same voltage level. The initial surface potential becomes positive under negative-to-positive voltage, which is similar to the surface charge accumulation under positive-to-negative voltage. The negative charges on the surface of sample are neutralised, and the positive charges further accumulate. Likewise, the initial surface potential of the 29.2 and 38.2 vol% samples is also lower than that of the 0 vol% sample, due to the higher conductivity. Comparing the results under negative-to-positive voltage and unipolarity positive voltage, the initial surface potential of the 0 vol% sample decreases most, which is caused by the large accumulation of residual negative charges before polarity reversal.

It can be seen from Figure 3 that the ISPD rate after corona discharge decays with time. In order to reflect the influence of polarity-reversal voltage on the ISPD process intuitively, the average ISPD rate of SiR/SiC composites within the initial 30 s under unipolarity voltage and polarity-reversal voltage is calculated. As shown in Figure 7(a), the 0 vol% sample has the lowest ISPD rate. With the increase in SiC content, the conductivity of SiR/SiC composites increases, which makes the surface potential of the 29.2 and 38.2 vol% samples decay rapidly. The average ISPD rate of the 29.2 and 38.2 vol% samples is higher than that of the 0 vol% sample because of higher conductivity. Comparing the average ISPD rate under positive-to-negative voltage and unipolarity negative voltage, it is found that the average ISPD rate of the 0 vol% sample increases most, followed by the 29.2 and 38.2 vol% samples. Obviously, the ISPD process of each sample is promoted by polarity reversal. The average ISPD rate under negative-to-positive voltage and unipolarity
positive voltage is shown in Figure 7(b), which is similar to that under positive-to-negative voltage and unipolarity negative voltage. The average ISPD rate under unipolarity positive voltage is lower than that under unipolarity negative voltage. As the SiC content increases, the increase in the average ISPD rate after polarity reversal is suppressed. Section 4.1 discusses the charge transport process under polarity-reversal voltage.

The charge transport process at different ambient temperatures of the composites under negative-to-positive voltage and positive-to-negative voltage has obvious similarities. Therefore, this work uses positive-to-negative voltage as an example to illustrate the influence of ambient temperature on the charge transport characteristics. According to the previous research [18], it can be found that a lower initial surface potential occurs at 70°C than that at 30°C. At 90°C, the initial surface potential of the sample is further reduced. This is because the conductivity of the sample increases as the temperature goes up. Similarly, at all temperatures, the initial surface potential of the 29.2 vol% sample is lower than that of the 0 vol% sample, and the 38.2 vol% sample has the lowest initial surface potential among the three samples. According to the previous content, the initial surface potential of SiR/SiC composites decreases after polarity reversal. Figure 8 shows the decreasing value in initial surface potential of the composites under positive-to-negative voltage at different temperatures compared to that under unipolarity negative voltage. Comparing the initial surface potential under positive-to-negative voltage and unipolarity negative voltage, it is found that the initial surface potential of the 0 vol% sample decreases maximum at 30°C, followed by the 29.2 and 38.2 vol% samples. At 70°C, the decrease in the initial surface potential under positive-to-negative voltage compared to that under unipolarity negative voltage of each sample is smaller than that at 30°C, and it is further reduced at 90°C. This phenomenon may be caused by the residual-positive charges inside the composites.

In order to further explain the effect of temperature on the charge dissipation process, the increased value in average ISPD rate within the initial 30 s of the composites under positive-to-negative voltage compared to unipolarity negative voltage at different temperatures is shown in Figure 9. It can be seen that the average ISPD rate can be enhanced by both temperature and polarity reversal. The increased value in average ISPD rate of the 0 vol% sample at 70°C is less than that at 30°C, and the increased value in average ISPD rate is further limited at 90°C. Besides, the increased value in average ISPD rate of the 29.2 and 38.2 vol% samples is lower than the 0 vol% sample at the same temperature. Consistent with the previous text, the increase in average ISPD rate of the composites is suppressed as the conductivity increases. This phenomenon may be caused by the residual-positive charges in the composites. Besides, the conductivity of the sample increases at 70°C and 90°C, which dominates the ISPD process, bridging the gap of average ISPD rate between the positive-to-negative voltage and the unipolarity negative voltage. Section 4.2 discusses the charge transport process at different temperatures.

3.3 Carrier mobility

Based on the ISPD process of the composites under positive-to-negative voltage and unipolarity negative voltage, the carrier mobility is calculated as shown in Figure 10. The conductive network is formed inside the 29.2 and 38.2 vol% samples, which improves the possibility of directional migration of carriers, thereby increasing the carrier mobility. Moreover, there are only a few carriers provided by SiR matrix in the 0 vol% sample, and the carrier mobility is smaller than other samples. The apparent carrier mobility of the composites can be improved by polarity reversal. Besides, the carrier mobility of each sample is also positively related to the temperature. This is because the
thermal movement of the carriers is intensified by the high temperature, making the carriers easier to participate in dissipation. In addition, as the temperature rises from 70°C to 90°C, the increase in the carrier mobility in the 29.2 and 38.2 vol% samples becomes insignificant, which can be explained by the low conductivity temperature coefficient [20].

Figure 11 shows the density and energy level of the deep and shallow trap in SiR/SiC composites under positive-to-negative voltage and unipolarity negative voltage at different temperatures, which is used to further characterise the charge transport process under polarity-reversal voltage. It should be noted that the traps under positive-to-negative voltage are apparent traps, that is, traps can be measured during the surface charge decay process. It can be seen that as the SiC content increases, the energy level of shallow trap decreases and the density increases, while the energy level and density of deep trap are reduced. This is consistent with the conclusion that the surface charge dissipates rapidly in samples with high SiC content. Besides, considering the influence of ambient temperature on the trap characteristics, the 0 vol% sample has a deeper trap levels at a higher temperature. This is because the high temperature endows the carriers with higher energy, which makes the carriers trapped by deeper trap at low temperature possibly take part in dissipation through thermal movement. Although the trap energy level increases as the temperature goes up, the surface charge dissipation can be promoted by the increase in the proportion of shallow traps [22]. The relation between the trap characteristics and the temperature in SiC-doped samples is similar to that of the 0 vol% sample. At 30°C, the shallow trap levels of the 38.2 vol% sample under positive-to-negative voltage and unipolarity negative voltage are 0.803 and 0.822 eV, respectively. It is shown that most of the carriers in the 38.2 vol% sample exist in the traps with low energy levels, which are easy to hop out and participate in dissipation. Overall, the conductivity, carrier mobility and trap characteristics of SiR/SiC composites have obvious consistency.
4 | DISCUSSION

4.1 | Effect of polarity reversal on charge transport characteristics

The charges remaining inside the composites before polarity reversal have a significant effect on the charge transport characteristics [23]. Due to the low conductivity of the 0 vol% sample, massive positive charges accumulate in the process of positive voltage polarisation. The residual-positive charge creates its own electric field, which will be superposed with the newly established electric field after polarity reversal from positive to negative, resulting in serious electric field distortion and consuming lots of injected negative charges during surface charge neutralisation after polarity reversal [11, 15]. While the accumulation of residual-positive charges can be effectively suppressed in the SiC-doped samples, and the migration of the injected negative charges is fast after polarity reversal, thereby weakening the distorted electric field and suppressing the charge neutralisation. Thus, the initial surface potential of the 0 vol% sample decreases the most after polarity reversal from positive to negative. Due to the high conductivity, the charge neutralisation process of the 29.2 and 38.2 vol% samples is weaker than the 0 vol% sample, and the initial surface potential of the composites decreases less. This indicates that under positive-to-negative voltage, the accumulation of residual-positive charges is suppressed and the charge neutralisation is effectively weakened due to the doping of SiC. Similarly, a large number of negative charges accumulate in the 0 vol% sample when the negative voltage is applied, resulting in serious electric field distortion and large negative charge consumption in the charge neutralisation after polarity reversal from negative to positive. Besides, the charge accumulation and neutralisation process under negative-to-positive voltage are inhibited by the addition of SiC.

After polarity reversal from positive to negative, the residual-positive charges on the surface of sample are neutralised. Most injected negative charges accumulate in the centre of the sample and gradually spread around. The conductivity of the 0 vol% sample is relatively low, and the charge migration to the ground electrode is slow. As a result, when the negative charge accumulation on the surface of sample is serious, there are still large numbers of residual-positive charges unneutralised inside the sample. On the one hand, strong local electric field is formed between the residual-positive charges inside the sample and the injected negative charges on the surface of sample, which promotes the charge migration. On the other hand, the neutralisation of hetero-charge also accelerates the dissipation of negative charges. All of these reasons make the average ISPD rate of the 0 vol% sample increase greatly. For the 29.2 and 38.2 vol% samples with high non-linear conductivity, the accumulated charges diffuse rapidly under unipolarity negative voltage, resulting in a larger average ISPD rate. Besides, there is less residual-positive charge before polarity reversal from positive to negative, which has little effect on enhancing ISPD rate after polarity reversal. Therefore, the increase in the average ISPD rate of the 29.2 and 38.2 vol% samples after polarity reversal from positive to negative is less than that of the 0 vol% sample. Besides, a large number of residual negative charges accumulated in the 0 vol% sample under negative-to-positive voltage have a similar effect on the charge neutralisation process and local electric field.

4.2 | Effect of ambient temperature on charge transport characteristics

In the process of polarity reversal, it is necessary to consider both transport and neutralisation of charge, which are greatly affected by the ambient temperature [4]. The lower temperature corresponds to the lower conductivity. Before polarity reversal, the residual charges dissipate slowly and accumulate in large amounts. After polarity reversal, the injected hetero-charges will be largely neutralised by the residual charges accumulated before. There is a positive correlation between the ambient temperature and the conductivity of SiR/SiC composites. With the increase in temperature, the charge dissipation is accelerated, which leads to less residual charge accumulation before polarity reversal. The hetero-charges injected after polarity reversal are only neutralised by a small amount.

At 30°C, the initial surface potential of the composites drops the most after polarity reversal. This is due to the low conductivity which causes the accumulation of residual charges, consuming a large amount of injected hetero-charge. The increase in temperature leads to the enhancement in the conductivity of the composites, which reduces the residual charges consumed during the charge neutralisation process. Therefore, at 70°C and 90°C, the initial surface potential of the composites decreases less after polarity reversal. Likewise, the ISPD rate of the composites after polarity reversal increases the most at 30°C. The changes inside the composites migrate quickly at high temperature, due to the high conductivity. Before polarity reversal, only a small amount of residual charge exists in deep traps of the composites [24]. These residual charges have little effect on accelerating the dissipation of the injected hetero-charges. Moreover, at 70°C and 90°C, the ISPD rate of the composites under unipolarity voltage is also at a high level due to the effect of high conductivity. Therefore, when the temperature is higher, the ISPD rate of the composites increases less after polarity reversal.

5 | CONCLUSIONS

The temperature-dependent charge transport characteristics of SiR/SiC composites under polarity-reversal voltage are obtained by ISPD method. The main conclusions are as follows:

1. Compared with unipolarity voltage, the initial surface potential of SiR/SiC composites decreases and the ISPD rate increases under polarity-reversal voltage. Besides, the intensity of this change is inversely related to the conductivity of SiR/SiC composites.
2. Under polarity-reversal voltage, a large amount of residual charge and injected hetero-charge in the 0 vol% sample will lead to intense charge neutralisation and severe electric...
field distortion. As the doping amount of SiC increases, the conductivity of the composites enhances, which effectively inhibits the accumulation of hetero-charges, thereby weakening charge neutralisation.

3. The ISPD process of each sample are accelerated with temperature. As the temperature goes up, the ISPD rate increases due to the higher conductivity. Besides, the residual charge accumulation before polarity reversal decreases, bridging the gap between the unipolarity voltage and the polarity-reversal voltage.

ACKNOWLEDGEMENT
This work was supported by the Chinese National Natural Science Foundation under the grants 51537008, 51707133 and U1966203.

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How to cite this article: Du B, Han C, Li Z, et al. Effect of polarity-reversal voltage on charge accumulation and carrier mobility in silicone rubber/silicon carbide composites. IET Sci Meas Technol. 2021;15:184–192. https://doi.org/10.1049/smt2.12020