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Study of the electrical properties of HVAC EPDM and HVAC silicone rubber under DC constraint

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ABSTRACT

EPDM and Silicone Rubber (SiR) are elastomeric materials used in HVAC cable accessories due to their good mechanical and electrical properties. In this paper one EPDM and one SiR, are characterised for their electrical properties under DC constraint. Bulk DC conductivity and space charge measurements using the pulsed electroacoustic method (PEA) were carried out at different electric fields and temperatures. The electrical behaviour of silicone rubber under DC stress is found to be largely governed by space charges while this effect is more limited for EPDM.

KEYWORDS

EPDM, SiR, DC conductivity, space charge, PEA, HVDC.

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INTRODUCTION

For an effective transition from HVAC to HVDC transmission systems, the materials used in cable accessories must be adapted towards DC stress [1] [2]. An important step is to understand the intrinsic limitations of candidate materials, and therefore investigate the impact of the material structure on the electrical properties subjected to DC stress. In this study, we have chosen standard HVAC systems to set a first comparison between EPDM and silicone rubber.

To validate an accessory design subjected to DC stress, all materials conductivity must be adapted so the electric field is always concentrated in the most suitable material, and it should be carefully checked that in no condition does the electric field become critical in any material. Therefore, the most critical parameter is the DC bulk conductivity and its evolution over the application temperature and electric field range [2]. In addition to the electric field predicted by DC conductivity, space charge accumulation in dielectric materials creates an electric field enhancement that could lead to a breakdown in the material, or contribute, along with thermal stress, to the premature ageing of the material [3].

These two aspects of the DC stress specificity should be considered together when investigating the impact of material structure, since space charges may significantly impact DC conductivity. The aim of this paper is to investigate the effects of thermal and electrical stress on DC conductivity and space charge accumulation in EPDM and SiR and to establish a connection between space charge's behaviour and the evolution of DC conductivity of insulation materials. These two techniques are complementary to investigate conduction mechanisms in insulation materials.

EXPERIMENTAL

Samples

SiR is an LSR (Liquid Silicone Rubber) typically used in HVAC cable accessories. It has been cured at SuperGrid Institute at 160 °C for 15 min and post-cured at 200 °C for 4 h. Thickness ranges from 1.03 to 1.15 mm.

EPDM plates were provided by IMERYS. For DC conductivity measurements, the sheets were kept as received with a thickness of approximately 2.3 mm. For PEA, the samples were sliced in the volume resulting in samples with a thickness ranging from 0.96 to 1.00 mm. During measurements, sliced surfaces of the samples were put in contact with the high voltage electrode.

DC conductivity

Bulk DC conductivity measurements were performed on a test bench designed at SuperGrid institute. Prior to the measurements, the samples were metallised with sputtered gold to ensure a perfect electrical contact with the electrodes. A guard ring, connected to the ground in the test bench, was added to avoid any unwanted current from reaching the ground electrode. The measurements protocol is based on the polarisation and depolarisation method. Steps of polarisation –under positive electric field ranging from 1 to 25 kV/mm– and of depolarisation of 8 h each, were performed. The test was repeated at three different temperatures (30, 50 and 70 °C).

The DC conductivity is calculated using Equation 1 and Equation 2.

$$I_{DC} = |I_{pol} + I_{Depol} - 2 \cdot I_{Offset}|$$
Equation 1
$$\sigma_{DC} = \frac{I_{DC} \cdot e}{E \cdot S}$$
Equation 2

The samples were tested in a dry setting. The test bench chamber was filled with dry air and the temperature stabilised in the chamber for 8 h before the first voltage is applied.

Space charge measurements

Space charge measurements were conducted using the pulsed electroacoustic method in a commercially available PEA cell for flat samples. The samples are placed in the cell between one aluminium electrode – the cathode for a positive applied electric field– and one semiconductive electrode – the anode. The semiconductive material is a standard semiconductive silicone rubber used in HVAC

accessories. Two different sets of measurements were carried out for each material. For the first, 2 h of polarisation (volt on) and 2 h of depolarisation (volt off) are performed, at the same temperature of either 30 or 70 °C under 5 and 20 kV/mm. For the second, the polarisation is at 70 °C and the depolarisation at 25 °C. For that, after 2 h of volt on at 70 °C, the temperature is lowered to 25 °C and then the voltage is switched off. At every temperature and for each sample, a calibration is performed to remove the distortion from the obtained signal using a software provided by the PEA cell manufacturer. For both DC conductivity and space charge, all the measurements were repeated at least twice.

RESULTS

DC conductivity behaviour SiR vs EPDM

Figure 1 presents the DC conductivity as a function of temperature and electric field of SiR. As transient current did not present stabilisation, even after 48 h of polarisation, measurements were stopped after 8 h and therefore the value of conductivity can be referred to as apparent conductivity. While apparent DC conductivity of SiR increases as a function of temperature, a negative tendency is observed with increasing electric field up to a certain value. This value is known as the high electric field threshold, which in this case is approximately 18 kV/mm. Above this threshold, the behaviour of the DC conductivity changes. Although the measurement range is too limited to identify the behaviour above the threshold, the conductivity value can be considered constant between 18 and 30 kV/mm at 50 and 70 °C.



Figure 1: DC conductivity of SiR as a function of temperature (top) and electric field (bottom).

A parametric fitting gives the conductivity formula as function of field and temperature as presented in Equation 3 for fields below the high electric field threshold value.

$$\sigma_{SiR} = 1.5 \times 10^{-16} e^{0.04T - 0.14E}$$
 Equation 3

Above this threshold, there are not enough points to extrapolate an additional equation fitting the DC conductivity.

Figure 2 presents the evolution of the DC conductivity of EPDM. The evolution of the conductivity with temperature is more important compared to the one of silicone rubber, resulting in higher temperature coefficient in the extrapolated equation (commonly referred to as alpha). The evolution of DC conductivity of EPDM as a function of the electric field, however, differs significantly compared to that of SiR. While the DC conductivity decreases below the high electric field threshold in the case of SiR, only an increase in conductivity is observed for EPDM, resulting in a positive electric field coefficient (commonly referred to as beta).

In the range of the applied electric fields and temperatures, Equation 4 is the extrapolated DC conductivity of EPDM.

$$\sigma_{FPDM} = 1.18 \times 10^{-17} e^{0.086T + 0.11E}$$
 Equation 4

By comparison, at all temperatures and electric fields, DC conductivity of SiR is higher than that of EPDM despite the negative tendency below the high electric field threshold for SiR and a stronger temperature dependency for EPDM.



Figure 2: DC conductivity of EPDM as a function of temperature (top) and electric field (bottom).

The former equations presented can be used in a first approximation for the design of DC accessories. The compatibility of these materials with DC applications should be based on a global study including other materials with which they share an interface [4] as well as the geometry of the accessory.

The evolution of DC conductivity of both SiR and EPDM suggests that injected charges - in addition to intrinsic charges - contribute to the conduction for both materials [5]. Therefore, the injection threshold is supposed to be lower than the measurement range. In the case of EPDM, these injected charges result in an increase of the current flowing through the sample [5]. In the silicone rubber, an opposite phenomenon is observed, implying that an addition of charges in the material would result in a limitation of the current. This phenomenon has already been observed elsewhere and attributed to an accumulation of charges in the material close to the electrodes, which results in a local decrease of the electric field. The injected charges therefore form a so-called blocking electrode [6]. This would raise the question of the persistence of these accumulated charges, whether located in shallow or deep traps. Such hypothesis should be confirmed with space charge measurements. It is also interesting to note that the behaviour of DC conductivity in silicone changes at about 18 kV/mm, which could be linked to a modification of the space charge behaviour.

Space charge behaviour

Silicone Rubber SiR

Figure 3 presents the space charge distribution in SiR subjected to 5 kV/mm at 30 and 70 °C during polarisation (volt on) and depolarisation (volt off).



Figure 3: Charge density profiles of SiR at 30 °C (top) and 70 °C (bottom) subjected to 5 kV/mm.

Charge density profiles, as a function of the thickness, outline the position and polarity of the space charges accumulated in the specimens. The electrodes are highlighted with vertical lines, the cathode is the detection electrode at the ground and the anode is the high voltage electrode. The temperatures of 30 °C and 70 °C correspond to the limit of the DC conductivity measurements. Two hours of polarisation and 2 h of depolarisation were performed, and for depolarisation, no significant change in the signal was observed after 20 min at 30 °C or 5 min at 70 °C. At 30 °C, below the high electric field threshold of DC conductivity, 5 kV/mm is the minimum electric field, above which the signal to noise ratio is high enough for a calibration.

In Figure 3 (top), at 30 °C, the main observation when considering the charge density curves (on the left), is that the peak at the cathode decreases in intensity and shifts towards the bulk of the sample (1), and simultaneously negative charges appear to accumulate both in the bulk of the sample and next to the cathode (2). At the anode, the peak shifts towards the bulk of the material and after 20 min of polarisation, the peak decreases in intensity (3). The patterned charge profile (on the right) shows that the more positive homocharges move towards the opposite electrode, the fewer negative charges are detected. The first depolarisation charge profile shows remaining charges in the bulk that cannot be seen after 20 min of short-circuit. This could be explained by homocharges injection and accumulation close to the electrodes: the presence of homocharges decreases the electric field locally at both electrodes and therefore decreases the peak associated to the charges at the interface, while being detected in the bulk. A similar behaviour has been previously observed by Wang et al. on LSR silicone rubber and been attributed to the accumulation of homocharges as well [7].

As shown in Figure 3 (bottom), at 70 °C and 5 kV/mm, positive homocharges are observed at the anode and spread into the bulk. For the first 20 min of polarisation, those charges seem to have crossed the material and can be identified as heterocharges at the cathode (1). The presence of heterocharges increases the peak at the cathode (2). These positive heterocharges then decrease continuously during polarisation and negative homocharges can be observed (3), which indicates a recombination process. Indeed, only homocharges are observed in the first depolarisation charge profile and the concentration of homocharges is higher close to the anode compared to the cathode (4). The mobility of the charges especially positive charges - seem to increase with temperature, which could also explain a faster depolarisation of the material.



Figure 4: Charge density profiles of SiR at 30 °C (top) and 70 °C (bottom) subjected to 20 kV/mm.

Figure 4 presents the charge profile of SiR polarised at 20 kV/mm at 30 and 70 °C. At 30 °C positive charges accumulate in the bulk and close to the anode (1). Positive homocharges move towards the bulk for the first 20 min and then gradually decrease. This could be explained by negative homocharges injection at the cathode and a recombination of positive with negative charges (2). At the start of the depolarisation, accumulated positive charges can be measured in the sample (3), which is then fully discharged after 5 min. It is interesting to note that increasing the electric field from 5 to 20 kV/mm leads to similar phenomenon that occurs when increasing the temperature from 30 to 70 °C: an increase in the positive charges' injection and mobility in the bulk.

At 70 °C, the intensity of the peaks at both electrodes slightly decreases with time (1) (2). Both positive and negative homocharges accumulate in the vicinity of the electrodes (3) (4). At the beginning of the depolarisation, charges remain in the material and after 5 min into the depolarisation the material is fully discharged. In this case, the transport of charges and recombination cannot be clearly observed. High temperature and electric field may have accelerated the charge dynamic so that the equilibrium between injection and recombination can be reached faster.

Charge profiles measured with PEA confirm that injection occurs on the whole range of DC conductivity measurement, meaning that the injection threshold is below the measurement range. Injected charges are expected to increase conductivity, although in this case, the conductivity was found to decrease with electric field. This phenomenon has already been described in literature and attributed to the presence of injected charges close to the electrode, acting as blocking electrode [6]. Indeed, the electric field at the high voltage electrode is decreased due to the presence of homocharges, which in turn limits the injection of additional charges. In this study, negative homocharges at the cathode were found to make recombination possible with injected charges from the anode. Therefore, it can be assumed that relatively high conductivity at low electric field benefits from this concentration of negative charges available for recombination. The limiting step would be the charge injection at the anode. When electric field is increased the injected charges at the anode become majority and the current seems to decrease due to the limited charge injection from the cathode. Therefore, all injected charges cannot contribute to the current and conductivity decreases. As observed at 70 °C and 20 kV/mm, temperature and electric field promote the kinetic of charge injection and transport so injected charges can contribute to the conduction and the negative tendency of the conductivity with electric field is no more observed.

The presence of charges in the material that can be measured starting from relatively low electric field compared to other materials [8] [9] may be a matter of concern for HVDC application. However, after switching the voltage off, the charges are evacuated from the sample in a matter of minutes indicating that the charges are in fact in shallow traps and that the charge accumulation during polarisation is a reversible phenomenon. In this case, the space charge would not be a consequence of a material degradation.

EPDM

Measurements were conducted on EPDM samples at 30 and 70 °C subjected to 5 and 20 kV/mm. Only the charge profiles recorded at 20 kV/mm are displayed in Figure 5. The charge profiles observed at 5 kV/mm were found similar to the ones at 20 kV/mm at the same temperatures although with lower definition and lower signal to noise ratio. During polarisation at 30 °C, positive homocharges are injected from the anode and accumulated charges are observed next to the electrode (1). The presence of positive charges in the sample influences the interface charge density at the cathode and a slight increase of the peak is observed (2). The positive homocharges injected and accumulated during polarisation remain in the sample, even after 2 h of depolarisation, indicating that the charges are accumulated in deeper traps. At 70 °C, the charge profile is not modified during the polarisation. The depolarisation however shows homocharges as emphasised in Figure 5. Moreover, the evolution of DC conductivity with electric field at 70 °C suggests that charges are injected in the material, and it is consistent with the detection of charges at 30 °C. Therefore, the absence of detected charges during polarisation at 70 °C may result from a high mobility of the injected charges, contributing to the conductivity. In this hypothesis, the mobility of the charges would depend on the temperature.



Figure 5: Charge density profiles of EPDM at 30 °C (top) and 70 °C (bottom) subjected to 20 kV/mm.

Temperature dependence

The temperature dependence of the charges' mobility in SiR and EPDM is further investigated in Figure 6, showing the depolarisation charge profiles of SiR and EPDM at 25 and 70 °C for samples polarised at 70 °C. It should be noted that in this protocol, polarisation is maintained during the temperature decrease to $25 \,^{\circ}$ C after 2 h at 70 °C. This additional polarisation time only slightly affects the charge profile at the first depolarisation signal, except for the charges at the anode for SiR, where a higher density of homocharges is found after further polarisation until $25 \,^{\circ}$ C (1).



Figure 6: Depolarisation charge density profiles of SiR (top) and EPDM (bottom) at 25 and 70 °C after 2 h of polarisation at 70 °C subjected to 20 kV/mm.

For SiR, at the cathode, negative homocharges are observed in both samples (2) – depolarised at 70 and 25 °C. Depolarisation is clearly faster when temperature remained at 70 °C, although the evacuation of charges is almost complete after 5 minutes in both cases. These results are consistent with the charge profiles shown in Figure 3, with a fast evacuation of the positive charges at 70 °C in 5 min, and a slower evacuation of negative charges at 30 °C.

Figure 6 (bottom), shows the charge profiles of EPDM during depolarisation at 25 and 70 °C. Again, negative homocharges are observed at the cathode (1). In this case, the additional polarisation during the cooling of the sample seems to have limited impact on the charge profile. The depolarisation seems to be completed after 5 min at 70 °C, and about 30 min at 25 °C.

For both materials, the depolarisation dynamics depend on temperature and a complete depolarisation can be observed, indicating that the charges are in shallow traps. Therefore, the polarisation of these elastomeric materials leads to the accumulation of charge carriers at the electrode and in the bulk but not permanently, and the integrity of the material does not appear to be affected over the time frame of this study.

DISCUSSION

The evolution of DC conductivity as a function of the electric field is influenced by the mobility of charges - intrinsic or not - in the bulk of the material. For SiR, there is a competition between the injection of charges at the high voltage electrode (anode), the transport in the bulk of the material and the recombination with injected charges at the cathode. At low temperature and electric field, transportation is not clearly observed and homocharges are observed at both electrodes with a majority of negative homocharges. The increase of the electric field leads to an increase in the injection of charges that move progressively towards the cathode and recombine. At higher temperature, the positive charges' mobility is higher so, the positive charges are detected at the cathode from the beginning of the polarisation before the recombination with negative charges can be observed. When the injected negative homocharges start to recombine with positive heterocharges, the concentration of charges in the bulk decreases. At a higher field and temperature, a lower density of accumulated homocharges is observed. When there is a high concentration of accumulated homocharges, the electric field at the electrodes decreases, blocking the injection of additional charges. Above the high electric field threshold observed in DC conductivity, the electric field increases the injection of negative charges that recombine eliminating the strong accumulation of homocharges at the anode.

The charge dynamic in EPDM is governed by injection of positive homocharges with a high mobility and recombination rate at the opposite electrode. Only at low temperature can accumulation be observed indicating deep traps.

Other studies on different grades of EPDM and silicone rubber show a large diversity of conductivity values and evolution with electric field, indicating that the charge dynamic is not a direct consequence of the polymer matrix, but that the whole formulation should be considered [10]. In particular, the nature and content of the fillers may play an important role in the conduction. The modification of the fillers has been identified as a way to adapt the conductivity and chose DC compatible materials [11].

The complex charge dynamic in silicone rubber results in DC conductivity behaviour that depends on temperature and electric field. The main phenomenon is the blocking electrode, which results in a decrease of the apparent conductivity below the high electric field threshold. This emphasises the necessity of measuring both DC conductivity and space charges on the whole application range when designing a new product.

Conclusion

In this paper, we have compared the DC behaviour of two materials: a silicone rubber and an EPDM. For both materials, a charge injection has been observed on the whole measurement range and therefore space charges play an important role, even at low electric field. The charge dynamic has an impact on the DC conductivity value for both materials but with different effect.

In the case of the silicone rubber, at low electric field injected charges contribute significantly to the conduction current, resulting in a relatively high conductivity. With increasing electric field, a high accumulation of injected charges close to the electrode reduces this phenomenon in proportion to the electric field, resulting in a decrease in the apparent conductivity. Finally, above a threshold, the high electric field promotes the recombination of most injected charges, reducing the blocking effect of homocharges. In EPDM, same as for SiR, the injection of charges is observed on the whole measurement range. A relatively higher kinetics of charges transport and recombination is assumed, consistent with an increase of the conductivity with the electric field.

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