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▶ To cite this version:

Igor Silva, François Gentils, Pascal Rain. Investigating the I-V characteristics of an HTV silicone rubber for MVDC electrical insulation. IEEE 2022 International Conference on Dielectrics (ICD 2022), Jul 2022, Palerme, Italy. hal-03727583

HAL Id: hal-03727583 https://hal.science/hal-03727583v1

Submitted on 19 Jul2022

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Investigating the I-V characteristics of an HTV silicone rubber for MVDC electrical insulation

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Abstract- High-temperature vulcanized silicone rubbers (HTV SiR) have been using for a long time as insulation of MVAC components. This work aims to investigate the suitability under DC of such HTV SiR. Measurements of current were carried out with control of electric field (1, 2, 3, 4, and 5 kV/mm), temperature (20, 40, 60, and 80°C), relative humidity (1%, 50%, and 90%), and electrode system (gold, silver and copper sputtering, and conductive silicone tape). Regarding the electric field, a non-linear current density was observed above values of fields between 1 and 2 kV/mm. An approach by Schottky injection presented a good fit with data points, and clarified the impact of electrode systems, but proved less accurate when material properties were calculated from the model. The influence of temperature followed an Arrhenius-like behavior, which is in accordance with many others conduction mechanisms. Moisture increased current in at least 1 order of magnitude, but the correlation with the water uptake is still under examination. In conclusion, the current density showed a strong dependence on all parameters selected for the study.

I. INTRODUCTION

High-temperature vulcanized silicone rubbers (HTV SiR) have been widely applied in medium-voltage alternatingcurrent (MVAC) electrical applications for decades due to special properties as low permittivity, high dielectric breakdown, hydrophobicity, heat resistance, and formation of a non-conducting ash when a breakdown occurs [1]. Recently, because of new applications identified in the field of highvoltage direct-current (HVDC) and medium-voltage direct current (MVDC) studies have been carried to validate the use of dielectrics applied in AC conditions to DC ones, the last mainly validated through DC conductivity and space charge measurements. Some phenomena, like anomalous water uptake [2]-[3], influence of electrode material [4]-[5], and fielddependent conductivity [6] have been explored for roomtemperature SiR and liquid SiR, but the impact of such factors over conduction mechanisms and conductivity is still not clear.

The aim of this work is to investigate the effect of water uptake, temperature, electric field, and electrode systems on DC conduction of an HTV SiR.

II. MATERIALS AND METHODS

A. Material description and sampling

The SiR employed is an HTV molded into 1 mm thick plates and then cut into 70 mm diameter discs. Scanning electron microscopy (SEM) of the surface topography, as seen in Fig. 1a, shows an irregular flatness. Analysis of a cross sectional cut done after a cryo-fracture failure in Fig. 1-b displays a rough structure. The images are in accordance with literature [7] about SiR filled with alumina tri-hydrate (ATH). The energydispersive X-ray spectroscopy (EDS) analysis of cross sectional cut confirms the presence of ATH.

B. Conditioning of samples

All samples have been previously dried at 70° C under vacuum conditions for at least 2 days, and stored in the same conditions. Aiming to evaluate the impact of water uptake, dried samples have been exposed to controlled fog environment inside climatic chambers with relative humidity (RH) set from 20% to 90%. The gravimetric method was applied to determine the quantity of water absorbed. Fig. 2-a shows that at 40°C the sample weight stabilized after 1 h and increased of 1% due to water absorption.

The same procedure of sample wetting was applied for different values of RH, the weight of specimens tested after 1 day of conditioning are illustrated in Fig. 2-b. It shows clearly a non-linear behavior between weight gain and RH. Fitting was conducted following a power law. Although the hydrophobicity [8], the specimens can absorb 1.2% of water at high RH environments. For all data points in Fig. 2-b, samples have been previously dried before conditioning.



Fig. 1. Scanning electron microscopy of the surface topography (a) and a cross sectional cut of the sample (b). Sample was cut in cryogenic conditions.



Fig. 2. Water uptake as function of time during exposition to fog conditions at 40°C and 50% RH (a), and water uptake at stabilization at 40°C for various RH (b). The last point at 100% RH was measured after immersing samples in demineralized water at 40°C for 1 day.

C. Preparation of electrode systems

A thin layer 200 nm thick was applied to both sides of the samples by metal sputtering (copper, silver or gold). Sputtering was conducted using a turbomolecular pumped coater Quorum Q150T ES.

D. Experimental system and settings

Measurements of current were performed with a device developed by Zavattoni et al [9] displayed in Fig. 3-a and Fig. 3-b, that consisted of a metallic vessel in which plane-plane electrodes arrangement can be enclosed. The electrode system installed in the vessel is made of conducting silicone (SiR + carbon black) of 10 Ω resistance in series with the specimen, as seen in Fig. 3-c. Samples are squeezed between main electrodes and guard electrode. The vessel is placed inside a climatic chamber and connected from one side to a high-voltage source (PS365 SRS High Voltage Power Supply) and on the other side to an electrometer (Keithley 6517B Electrometer/High Resistance Meter). All measurements were done at isothermal conditions (20°C, 40°C, 60°C and 80°C). Electric fields vary between 1 to 5 kV/mm.

For measurements in dry conditions, the vessel is sealed. First, the air is pumped out, then the vessel is filled with dry air under pressure of 2 relative bars; this cycle is repeated at least 3 times to achieve a low value of RH (normally 1%). For measures in fog conditions, and at field levels between 1-3 kV/mm, the cell is maintained open inside the climatic chamber that was previously set at the same parameters of sample conditioning. To avoid breakdown into the air at higher field (4-5 kV/mm), cell is closed and filled with dried air under 2 relative bars without previous draining in order to preserve sample humidity. Chosen RH are 50% and 90%, and some complementary tests were performed at 25%.



Fig. 3. Scheme of electrical tests (a); external view of test cell (b); view of electrodes arrangement (c).

Once isothermal condition is achieved, at each electric field (starting from 1 kV/mm), the test protocol consists of 1 h charging followed by 1 h discharging. The sum of charging and discharging currents at the end is taken as the conduction current, i.e. the current that does not take into account polarization effects.

III. EXPERIMENTAL RESULTS

A. Dried samples

Fig. 4 presents the isothermal curves of current density as a function of electric field for dried samples. Two distinct domains are proposed:

- In the first one, at lower fields, the dot line with a slope equal to 1 representing a linear relationship matched with measurements. A linear relationship represents an ohmic behavior in which $J = \sigma E$, with σ being the conductivity that doesn't depend on the field. From an ohmic conduction, one can also presume an ohmic contact between sample and electrodes, i.e. a low-resistance junction.

- In the second one, a non-linear behavior recognized by a line with a slope a greater than unity. The slope decreases with temperature from 3.6 at 20 °C to 2.5 at 80 °C. The field thresholds separating these domains (represented by a black arrow) also decrease with temperature, suggesting that the mechanism responsible for the non-linearity is also temperature-dependent.

Aiming to investigate the influence of electrode materials, measurements similar as those made with gold sputtering were carried with silver or copper metallized electrode and carbon black electrode (without metal sputtering). All results are illustrated in Fig. 5 that highlights the effect of electrode material. In similar conditions, current density can change up to 1 decade from one electrode system to another. Among different temperatures, the hierarchy of electrode systems remained generally the same except for carbon black that progressed differently than others.



Fig. 4. Current density as a function of electric field for dried samples. RH = 1-3%. Gold sputtering. Measurements were done 3 times, each time with a different sample. Dots represent the average and error bars the standard deviation.



Fig. 5. Influence of electrode material on variations of current density with electric field, at four different temperatures; dried samples. RH = 1-3%.

B. Wet samples

Fig. 6 shows the results for wet samples previously conditioned in moist environments (50% RH and 90% RH) and tested at equal conditions at low fields (1-3 kV/mm) or under 2 relative bars of dry air at higher fields (4-5 kV/mm). As for dried samples, two domains (linear and non-linear, or ohmic and non-ohmic) are suggested. As a whole, for 50% RH, current density increased 1 decade in comparison with dried specimens.

For tests done at 50% RH, one can notice that:

- The field thresholds are similar than for dried samples, despite the increase of current density.

- The slopes are higher than those of dried ones, as seen in Fig. 4, and they decrease more slowly with temperature. It indicates a more field sensitive process at high temperatures.

- An Arrhenius-like behavior seems to be reproduced between 20° C and 60° C, but not for data between 60° C and 80° C.

For tests at 90% RH, it is worth to mention that:

- Most values of current density are similar to those at 50% RH unless values at 20°C for lower fields.

- The black arrow looks more bent, which point out a higher sensibility with field.

- No values at 80° C and 90% RH were measured due to instabilities of test cell at this condition.

IV. DISCUSSION

A. Investigating the mechanism occurring in dried samples

Based on Fig. 5 that shows the effect of electrodes, one can suppose that the electrode system (interfacial mechanisms) plays a crucial role on charge transport, possibly as the *phenomena* limiting it. It is known that current can be injected at the interfaces through Schottky or Fowler-Nordheim (FN) effects. Among them, a correlation between Schottky has been found in a first moment. Schottky injection assumes that a gradually changing barrier between the metal of electrode and the insulating material is created due to thermo-activated electrostatic attraction [10]. Equation (1) presents the Schottky effect, with A a constant equal to $1.2.10^{6}A/m^{2}K^{2}$, ϕ the energy barrier, β_{s} the Schottky constant equal to (2), k the Planck constant, E_{c} the electric field at the electrode-dielectric contact, T temperature, q the charge, ϵ_{0} vacuum permittivity, and ϵ_{r} the relative permittivity of insulating.



Fig. 6. Current density versus electric field at four temperatures for wet samples at 50 and 90% RH.

$$J = AT^{2} \exp\left[-\frac{\phi - \beta_{s}\sqrt{E_{c}}}{kT}\right]$$
(1)

$$\beta_s = \frac{q^{\frac{3}{2}}}{(4\pi\epsilon_0\epsilon_r)^{1/2}} \tag{2}$$

A first evidence of Schottky effect should be a good fit between the logarithm of current density and the square root of electric field for constant temperatures, as presented in Fig. 7 [10]. Using just the mean field instead of E_c , a good fitting has been found between 2 kV/mm and 5 kV/mm. The coefficients have been extracted, with values showed in Table 1.

It is noted that:

- The ϵ_r are lower than common values for SiR (between 3-4)[1], and do not represent the real property of material. However, considering space charge close to electrodes, the contact field must be modified, which would affect the permittivity calculated. Complementary measures of space charge should enable the attainment of E_c .

- β_s is calculated from the slope, and it can be seen that the slope decreases when temperature increases, indicating a leading role of temperature effects over electric field. As β_s is used to obtain ϵ_r , it is expected that the temperature affects ϵ_r , as showed at table 1.

- The energy barrier ϕ is lower than the work function of each metal [11], which is coherent since the energy barrier represents the work function lowered by the insulator affinity.

A second evidence of Schottky injection would be an Arrhenius-like relation between J/T^2 and 1/T, illustrated in Fig. 8 for dried samples with gold sputtering. As it can be seen, for all temperatures, J/T^2 is proportional to exp (-1/T). The *phenomena* would be thermo-activated, as presumed in Schottky injection.

It is, indeed, worth to mention that many other mechanisms (as ionic conduction or Poole-Frenkel effect) reproduce an Arrhenius-like behavior. This article does not explore other mechanisms since they are not able to explain the variability of current density as function of electrode material.



Fig. 7. Current density as function of the square root of electric field for 4 electrode materials at 4 temperatures; dried samples tested at 1% RH.

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PROPERTIES EXTRACTED FROM SCHOTTKY FIT COEFFICIENTS													
T (°C)	Gold			Silver			Copper			Carbon Black			
	β/1E-24	٤r	Φ(eV)	β/1E-24	٤r	Φ(eV)	β/1E-24	٤r	Φ(eV)	β/1E-24	٤r	Φ(eV)	
20	7.6	0.64	0.93	4.9	1.5	0.87	5.6	1.2	0.88	4.3	2	0.85	
40	7.2	0.71	0.95	4.3	2.0	0.88	5.2	1.4	0.88	4.2	2.1	0.86	
60	6.5	0.87	0.96	4	2.3	0.89	4.6	1.7	0.89	4.2	2	0.88	
80	6.4	0.89	0.98	3.9	2.4	0.9	3.9	2.5	0.88	5.1	1.4	0.93	

B. Impact of water uptake

The analysis of wet samples is complex because of many events occurring at the same time: water can dissociate into ions H^+ and OH^- , interact with ATH and diffuse through the volume and surface. Fig. 9-a highlights the exponential increase of current density as a function of RH up to 50% of humidity. At higher values of RH, current density remains practically constant. The current density as a function of water uptake, as Fig. 9-b, reveals a non-linear behavior in which the first amounts of water absorbed provided a small contribution to the increase of current density, in comparison with larger amounts.



Fig. 8. J/T² (A/m²K²) vs 1000/T (1000/K) for 5 electric fields; dried sample with gold sputtering and tested at 1% RH and 40°C.



Fig. 9. Current density as a function of the relative humidity (a) and as a function of water uptake (b) for specimens tested at 40°C.

V. CONCLUSIONS AND PERSPECTIVES

Designing tools for DC applications requires the knowledge of the insulating properties, which is not evident when a nonlinear behavior occurs. Intending to explore the non-linearity of HTV SiR, data from I-V measurements was obtained taking into account a wide range of electric field (1-5 kV/mm), temperature (20-80°C), and RH (1-90%). For the HTV SiR studied in this work, it can be concluded that:

- The thresholds from linear to non-linear domain are located at low fields (1 to 2 kV/mm) for all conditioning methods.

- For dried samples, current density presented a strong dependence on the nature of electrode system. Here, the mechanism that better matched the results was the Schottky injection, although concerns about the coefficients of the fitting.

- Wetting samples increased values of current density up to 1 order of magnitude.

Further studies should be carried out to take into account the impact of ATH and other fillers on these mechanisms. Besides these, complementary space-charge measurements should be conducted to clarify the approach by injection mechanism.

ACKNOWLEDGMENT

We thank the technical team Mr. Flury, Mr. Pollet, and Mr. Cuilla, as well as Dr. Lesaint for the support on test-cell set-up.

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