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FORECASTING THE CURRENT-VOLTAGE BEHAVIOR OF MULTILAYERS DIELECTRICS BASED ON MONOLAYERS MEASUREMENTS

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Abstract

Equipment connections in MVDC networks may combine different materials. We consider here the assembly of an epoxy and a silicone rubber composite. As the apparent conductivities of the two materials are highly dependent on temperature, water content and electric field, situations may arise where the voltage applied to the bilayer insulation is mainly applied to one of the two layers, either epoxy or silicone, or is divided over the two sheets. Current densities-voltage (J-V) measurements were carried out on the 2 mm thick bilayer samples either at 40°C under three relative humidity conditions (dry, 50% and 90% relative humidity) or under dry conditions at 40, 60 and 80°C. The voltages applied were between 0.25 and 10 kV. These measurements are compared to the currents calculated from measurements made on each monolayer sample under similar T and RH conditions. Under conditions of T and RH where the apparent conductivities of the two materials are very different, as expected, the currents measured in the bilayer samples were close to the currents flowing in the more resistive material. When the currents are of the same order of magnitude, the calculated currents were found to be relevant for evaluating the measured current in the bilayer samples.

1 Introduction

Following recent developments in HVDC transport, the interest in MVDC distribution has recently increased. Some successful cases include applications in railways, shipboard systems, distribution grids and offshores [1]. As a branch of these studies, the validation and improvements of insulating materials have been imposing itself as a mandatory step to ensure the reliability and the safety of such systems.

With this change from alternating to direct current, the property responsible for the distribution of the electric field within the insulator is no longer permittivity (ϵ) but conductivity (σ). This is all the more important when the insulators are arranged in series. This situation can be encountered in connections between cables and equipment bushings, for example a circuit breaker, or in connections between equipment. Under DC voltage, the electric field is distributed along those insulators according to their conductivities. Because the conductivity of each polymer insulator will present a particular dependence as a function of temperature and electric field, the variation of these parameters can disturb the field distribution inside a multilayer insulator much more than under AC.

The field distribution of multilayer samples have been investigated primarily for cable connections. For a bilayer of cross-linked polyethylene (XLPE) and ethylene propylene rubber (EPR) tested at fixed voltage levels and various temperatures, Bodega [2] observed that the electric field will be concentrated in the most resistive layer just after a certain

time is achieved (Maxwell-Wagner time constant, calculated from the ratio σ/ϵ). Vu [3], [4] performed current measurements of XLPE monolayer and ethylene propylene diene monomer rubber (EPDM) monolayer at many voltage levels and temperatures, with conductivity values applied to predict the charge build-up and electric field distributions of bilayer configurations.

Here we are considering the assembly of an epoxy resin and a silicone rubber (SiR), hereafter referred to as the bilayer sample. The J-V characteristics have been measured and studied in both materials as a function of temperature, electric field and water content [2-3]. The objective of this paper is to measure the J-V characteristics of the bilayer sample and to investigate to which extent it can be deduced from the data of each material without taking into account space charge build up. Hereafter, the two materials will be compared, one being more or less conductive, or resistive, than the other one. This relies on their apparent conductivities, but not on the real I-V regime. In other words, this does not mean that the material follows a linear regime.

Andersson [5] has already associated AC electrical tests of epoxy and SiR bilayers with ageing. He has found that the contact tends to degrade, with presence of droplets as a consequence of acidic by-products decomposition on the epoxy side, and cracking patterns due to oxidative crosslinking on the silicone side. Later on [6], DC resistivity measurements of a bilayer at room conditions revealed that the behaviour of an unaged sample followed the pattern of the SiR since this monolayer presented a higher resistivity than the epoxy

monolayer. However, as pointed out by the author, the tested SiR was more sensible to water uptake, the monolayer of boiled SiR samples had a decrease of resistivity up to two decades. The boiled SiR/Epoxy bilayer, therefore, followed the pattern of the boiled epoxy monolayer.

2 Materials and Methods

2.1 Materials description

Anhydride-based epoxy resins (EP) filled with microsilica have been reticulated and shaped into plates of 1 mm of thickness. A view from Scanning Electron Microscope (SEM) through the thickness, as the one in Figure 1, underlines a good dispersion and distribution of particles in the matrix, so as the variability of filler sizes (from mesoparticles to 100 μm). From these plates, disks of 70 mm of diameters are obtained.

The Silicone Rubber (SiR) employed is an HTV rubber molded into 1 mm thick plates and then cut into 70 mm diameter discs. It also contains silica.

2.2 Sample preparation

Prior to electric tests, all samples were dried at 70°C under vacuum conditions for at least 2 weeks until stabilization in weight. The weight is considered stable when the same value is measured for three consecutive days. Once dried, samples are installed inside climatic chambers under controlled temperature and relative humidity. The water uptake is also computed based on the gravimetric method, and samples are considered stable when their weights have not been changing for at least three consecutive days. Dried samples are tested without this conditioning step.

After conditioning, the samples' surfaces are sputter-coated using a turbomolecular pumped coater Quorum Q150T ES. A 200 nm disc-shaped gold layer was deposited on both sides, with a diameter of 50 mm on one side and 54 mm on the other. Since the samples can lose some water during the process, they are conditioned again for four hours after installation into the test cell.

The bilayer samples are assembled by pressure with two monolayer samples without any added product at the interface. The same procedures of drying and metallization as for the

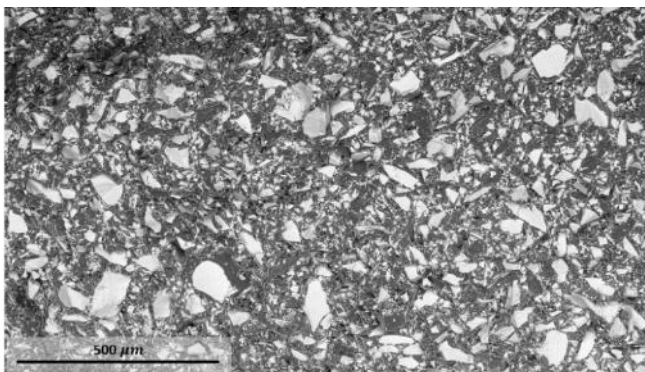


Fig. 1 Scanning electron microscope of an epoxy sample obtained from cryogenic cut.

monolayers are followed. Possible air pockets between the two dielectric sheets are not excluded.

2.3 J-V Measurement

Measurements of current were performed with a device developed by Zavattoni et al [7] displayed in Fig. 2-a and Fig. 2-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. 2-c. Samples are squeezed between main electrodes and guard electrode. The vessel is equipped with temperature and humidity sensors allowing a continuous control. When closed, vacuum or gas filling may be operated. The vessel is placed in 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 have been done at isothermal conditions. The RH inside the climatic chamber is controlled to maintain the same value as during the conditioning of the materials. Voltages were applied in the range from 0.25 to 10 kV. At each voltage level, the voltage has been applied 1 hour, following by 1 hour of discharge. The current density is calculated based on the conduction current obtained via the sum of the current at the end of polarization and depolarization steps. The entire program was repeated for both polarities, and results are presented as the average of them. All current densities are given in relative values J/J_0 , J_0 being an arbitrary value which is the same for all the graphics.

2.4 Calculation of the bilayer current

The expected currents in the bilayer samples were also calculated from the J-V characteristics of the monolayer samples measured under identical conditions of temperature and relative humidity. Each monolayer J-V plot was interpolated with a power law of the form $J = a + bV^c$. For better accuracy, plots were sometimes split into two parts with a linear part ($a=0, c=1$) and a non-linear part, giving in total four set of equations (two linear equations, one for each material, and additional two power equations), and two threshold voltages (V_{th}), marking the transition between the linear and the non-linear behaviour. Inversion of these interpolations provided $V(J)$ relations. Remark that these power laws have been used to get a good fit of the measured values, without any physical consideration.

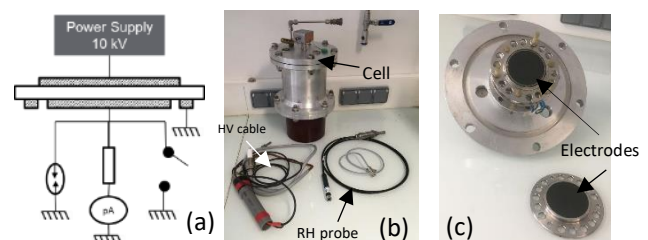


Fig. 2 Scheme of the electrical test cell (a); external view of test cell (b); view of electrodes arrangement (c).

In the steady-state regime, the same current density flows through the two sheets of the bilayer sample. Assuming negligible effect of possible space charges at the interface between the two dielectrics, the applied voltage on the bilayer sample is the sum of the voltages across each material. Using the $V(J)$ relationships for each material, the equation is derived: $V=[V(J)]_{\text{epoxy}}+[V(J)]_{\text{silicone}}$. Numerical solving of this implicit equation leads to the expected calculated bilayer current in any T and RH condition. For each voltage level, the program identifies the regime of each layer (linear or non-linear) based on the given V_{th} , picking the most appropriate equation for resolution.

3 Results

3.1 Monolayer Samples

In previous studies [8], [9], current-voltage (J - V) measurements were performed for each individual 1 mm thick component, over a range of temperatures (20-80°C) and voltages (250 V to 10 kV), with controlled water content on the samples (1-3; 50; 90% RH). For the epoxy resin, we observed a classical situation with a linear regime at low field and a quadratic behaviour above a voltage threshold. Silicone rubber showed a non-linear J - V behaviour over the whole range of fields studied.

Figure 3 compares the currents measured at 3 kV/mm in the two materials. It shows that the currents, and then the apparent conductivities, can cross each other. Thus, depending on the T and RH conditions, one of the two materials may be more "resistive" than the other.

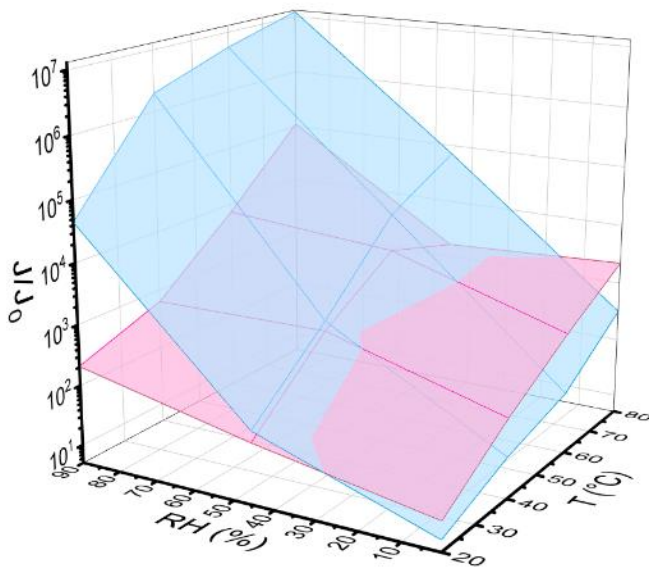


Fig. 3 Comparison of current densities in the epoxy (blue) and silicone (pink) samples under 3 kV depending on temperature and RH.

3.2 Bilayer Samples

3.2.1 Results at 40°C as a function of the RH: Current densities for the bilayer sample have been measured at 40°C with 3 different water contents resulted from equilibrium with air at RH of 1-3%, 50% and 90%. In Fig. 4, Fig. 5 and Fig. 6-b, respectively, the experimental values obtained from real bilayer samples are compared with the forecast issued from the measurements of the monolayers. J - V plots of the monolayers are also given either in the same graph (Fig. 4 and Fig. 5) or in a dedicated graph (Fig. 6-a) to avoid misunderstanding.

For dried samples at 40°C (Fig. 4), the epoxy is more resistive than the silicone rubber. The measured current through the bilayer is close to the current in the epoxy and in good accordance with the calculated plot. The same observation can be made for wet samples at 40°C (Fig. 5), but in a reverse situation: at 90% RH, the epoxy is more conducting and the measured current through the bilayer is close to the current in the silicone rubber.

At 40°C and under 50% RH, current densities in the two monolayers are of the same order of magnitude and almost similar in the range from 0.25 to 3 kV (Fig. 6-a). Then, the same currents at twice the voltage are expected in the range 0.5 to 6 kV, if the voltage is simply divided by two in the two materials of "equivalent conductivities". For example, the relative currents in the monolayers are measured at 3-4.10² under 2 kV, which is close to the current measured in the bilayer under 4 kV (Fig. 6-b). As concerns the agreement between the measured and calculated currents, the same remark as for Fig. 5 applies. In all case, the two values are within a factor of 2.

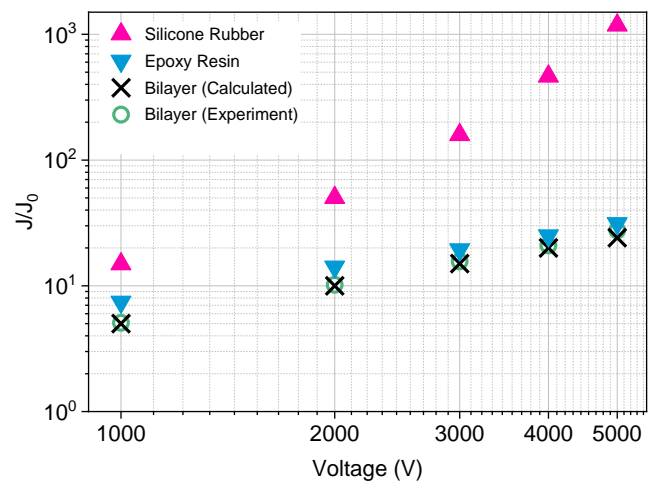


Fig. 4 Current density as a function of the applied voltage for SiR monolayer, EP monolayer, calculated SiR/EP bilayer based on monolayer results, and experimental SiR/EP bilayer samples; 40°C, 1-3% RH.

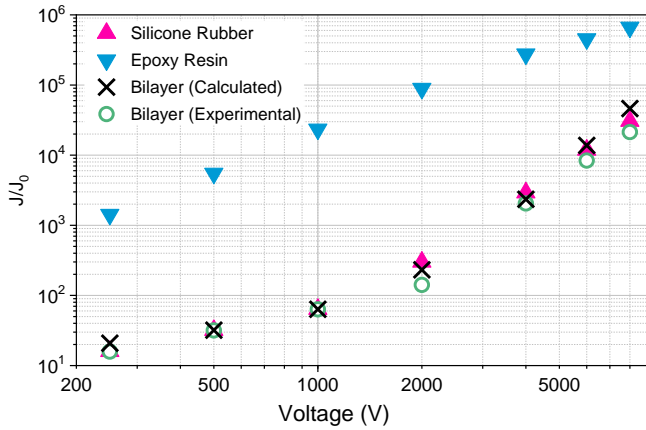


Fig. 5 Current density as a function of the applied voltage for SiR monolayer, EP monolayer, calculated SiR/EP bilayer based on monolayer results, and experimental SiR/EP bilayer samples; 40°C, 90% RH.

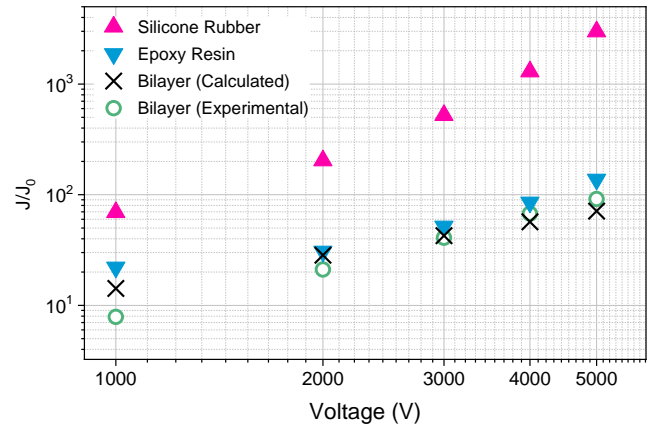


Fig. 7 Current density as a function of the applied voltage for SiR monolayer, EP monolayer, calculated SiR/EP bilayer based on monolayer results, and experimental SiR/EP bilayer samples; 60°C, 1-3% RH.

3.2.2 Results for dried samples as a function of the temperature: Current densities in dried bilayer samples have been measured for 3 different temperatures, 40°C in Fig. 4 above, 60°C and 80°C in Fig. 7 and Fig. 8. Not surprisingly, the measured currents reproduced the values of the more resistive material, which is here the epoxy composite.

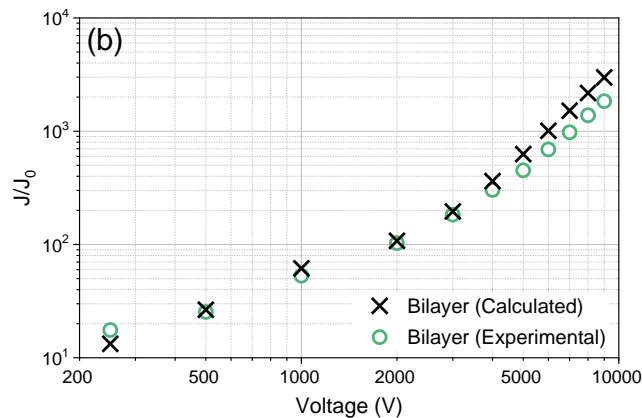
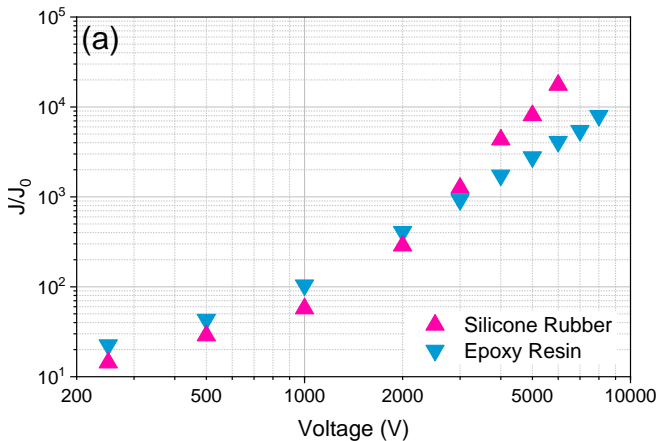


Fig. 6 Current density as a function of the applied voltage for: (a) monolayer of SiR, and EP; (b) experimental and calculated SiR/EP bilayer; 40°C, 50% RH.

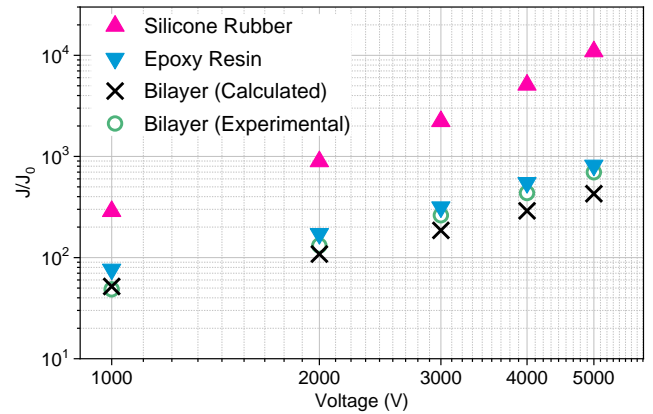


Fig. 8 Current density as a function of the applied voltage for SiR monolayer, EP monolayer, calculated SiR/EP bilayer based on monolayer results, and experimental SiR/EP bilayer samples; 80°C, 1-3% RH.

4 Discussion

The numerical model calculated from monolayers seems to describe with reasonable precision the behaviour of bilayers, even if it has no physical meaning, and no further exploration of space charge build up. Similar results have been found for a model based on an exponential law instead of power law for the non-linear regime. In spite of it, the following points deserved our attention.

4.1 The absence of polarity effects:

The absence of polarity effects has already been noted for monolayers, but it also appeared true for bilayer samples; that is why the values presented in this paper are an average of both polarities. Nevertheless, this behaviour was not intuitive in the case where the silicone rubber is the most resistive layer, as at 40°C and 90% RH, because the respective monolayer current measurements seemed to be affected by the electrode system,

that is to say the nature of the electrode metallization, as well as the polarity applied to each electrode [10].

4.2 Why showing $J(V)$ instead of traditional $J(E)$?

Since the electric field will be concentrated in the most resistive layer, the effective thickness applied to calculate the electric field may change case by case, being particularly challenging in the case the resistivities cross each other, as in Fig. 6-a. Because of these effects, the authors believed that showing the $J(V)$ plot was more appropriate than $J(E)$, all the more space charges may accumulate at the interface.

4.3 Is it a trustworthy method that doesn't take into account space charges?

Even if space charge weren't taken into account to calculate the bilayer J , they are partially considered when non-linear current densities were obtained. In the long-term, the following effects of charge build-up in the interface, as its consequences in the conduction and charge density profile have to be further explored.

5 Conclusion

Current-voltage characteristics of a bilayer assembly of an epoxy composite and a silicone rubber composite have been measured in a wide range of temperature, relative humidity and electric field.

When the current densities within the monolayers were clearly different, the measured current densities of the bilayers logically followed the current of the most resistive material, being epoxy or silicone.

In most cases, calculations from the interpolations of the J - V characteristics of the monolayer samples revealed in good accordance with the measured values. Thus, this method is considered convenient to estimate the currents in the bilayer samples and the conditions of our investigations.

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