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Published version information

Citation: B. A. Orton, N. M. Chalashkanov and S. J. Dodd, "Investigating Properties of a Composite Dielectric using Broadband Dielectric Spectroscopy," 2023 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), East Rutherford, NJ, USA, 2023, pp. 1-4,

DOI: https://doi.org/10.1109/CEIDP51414.2023.10410571

Investigating Properties of a Composite Dielectric using Broadband Dielectric Spectroscopy

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***Abstract* - The dielectric properties of a complex composite insulator based on a Methyl Nadic Anhydride (MNA) cured Diglycidyl Ether of Bisphenol-A (DGEBA) epoxy has been examined using broadband dielectric spectroscopy (BDS). Epoxy samples containing each of the individual composite components – mica and glass fibre – as well as the composite and the pure epoxy, have been studied in order to establish their individual contributions to the overall response above and below the glass transition temperature. Samples with and without sputter coated gold electrodes have been used to isolate and identify interfacial electrode effects. Two classes of process have been observed in the dielectric response, with signatures consistent with a bulk charge transport and dielectric loss peaks associated with bulk or interfacial polarization. Additionally, the glass transition temperature has been verified using differential scanning calorimetry (DSC).**

I. INTRODUCTION

The ISIS neutron and muon source is located at the Rutherford Appleton Laboratories in Oxfordshire, UK. It provides neutrons for use in high energy physics experiments by accelerating protons up to 800 MeV [1] in a synchrotron accelerator.

To constrain the high energy protons substantial electromagnetic fields are required. These fields are generated by electromagnets, primarily driven with AC voltages of a few kV, but also with a small DC voltage component of around 100V. The largest of these magnets, the Dipole Bending Magnets (DBM's) – each 4m long – are made up of series of copper conductors arranged in six stacked "racetrack" coils, each wrapped in mica backed tape and glass tape and then embedded in an epoxy resin by vacuum impregnation to form a "composite" dielectric. The DBM's are subjected to multiple stress factors during normal operation and occasionally fail, taking the source offline, with consequent interruption to the scientific research programmes. This is costly - both financially and temporally – and often results in a considerable amount of remedial action. The primary stress factor in this case is the AC / DC electric field, and secondary stress factors are: temperature cycling, mechanical vibration at various frequencies (predominantly 50Hz), moisture ingress, ionising radiation and physical ageing. With all these stresses there comes about a change in the physical and chemical properties, a manifestation of which can be observed in changes in the electrical characteristics of the material: for example, a change in the permittivity and conductivity or in the extreme, electrical breakdown.

The definition of dielectric failure is when a conductive path is created between the conductors that the dielectric is designed to separate [2]. There are various ways in which this can happen, but ultimately it is always the movement of charge that constitutes failure. All dielectrics allow a certain amount of leakage current to pass even in normal operation; determining the mechanisms by which this happens is the key to understanding failure. The composite dielectric contains several component materials, each of which has differing conduction characteristics – charge is able to flow via different mechanisms in each of these components, each of which exhibits a unique dielectric response. According to the theory of superposition [3] each of these component mechanisms contributes to the overall conduction of the whole composite. The intention of this work is to use dielectric spectroscopy to identify which processes – charge transport or dipolar relaxation – occur in each of the insulator components. This paper presents the data collected and the initial analysis that will be used to construct and validate an equivalent circuit model of the composite dielectric.

Previously it has been demonstrated that both a quasi-DC (QDC) bulk charge transport process and interfacial processes were identifiable in the dielectric spectra for similar epoxy dielectrics [4].

In this preliminary work the variables were the frequency of the applied electric field and temperature, ranging from below to above the glass transition temperature, Tg, of the epoxy resin. The other aforementioned stress factors were held constant in order to ensure they had a negligible impact on results.

II. EXPERIMENTAL

A. Sample preparation

The insulation system in the DBM's consisted of four components: vacuum impregnated epoxy (principally consisting of DGEBA epoxide resin and MNA hardener), a VIDATAPE AS (amino silane coated) glass fibre tape and a KREMICA.POR® K3015-14 Mica tape.

Four types of samples were manufactured: catalysed epoxy resin only, catalysed epoxy resin with a single layer of glass fibre, catalysed epoxy resin with a single layer of mica, and catalysed epoxy resin with single layers of glass fibre and mica. Samples measured 75 x 75 mm and ranged in thickness between approximately 200 μm and 500 μm. Initially, single layers of the tapes were ½ lapped around plates – either, mica, or glass or both stacked one layer on top of another. Next, the DGEBA and the MNA were mixed by weight in the ratio 100:80 and degassed for 16 hours at 40 °C in a vacuum ($2.3x10^{-1}$ mBar). The accelerator was then added and the solution was returned to the degasser for an hour to ensure any additional trapped volatiles - mostly water vapour - were removed. This was then removed from the degasser and

flowed, under vacuum, into the moulds containing the mica and glass tape wrapped plates, which were then gelled in a between hot plates mounted in a large press at 95 °C for 16 hours. They were then removed from their moulds and placed on curing plates in an oven for a further 15 hours at 122 °C.

Several samples of each of the four sample types were manufactured, and one of each of these was sputter coated with a uniform gold layer underneath the corresponding brass electrodes.

B. Differential scanning calorimetry (DSC) measurements

DSC measurements were carried out on a dried epoxy only sample. The instrument used was a NETZSCH DSC 200F3. Two test runs were carried out in the temperature range 20 °C to 150 °C at a heating rate of 10 °C/min.

C. Dielectric measurements

Broadband dielectric spectroscopy (BDS) was carried out as a function of temperature in the range 70 – 130 °C using a Solatron 1260A Impedance Analyser with a 1296 dielectric interface. Samples were clamped in a bespoke measurement cell incorporating a three electrode configuration. The entire assembly was contained within a Genlab OV150F oven.

A 5-hour dwell time was incorporated into the control program at each temperature to ensure the samples had attained thermal equilibrium.

Measurements were carried out in the frequency range 1 mHz - 67 kHz. In order to minimise noise 20 points were taken per decade in the frequency range 65 kHz to 1 Hz with a 10 s integration, and 9 points per decade with a 5-cycle integration from 10 Hz to 1 mHz due to time constraints. The test voltage was 1 V_{rms}. There was no dc offset voltage applied in this case.

III. RESULTS

A. DSC results

The onset of glass transition, Tg, was 75.3 °C, the midpoint 85.0 °C and the end point 94.6 °C.

B. Dielectric measurements – comparison of sputter coated and non-sputter coated samples

The dielectric repsonse of each of the four sample types was examined as a function of frequency over a range of temperatures. To isolate contact and interfacial issues; related to imperfect electrical contact between the sample and the measurement electrodes, two versions of each of the samples were prepared – one with gold sputter coated electrodes (SC), and one with non sputter coated electrodes (NSC). For brevity only the comparison of epoxy samples with and without sputter coating are compared here, shown in Fig. 1. In this plot the real and imaginary parts of the relative permittivity are shown plotted against frequency on log-log axes, which enables the Kramers - Kronig relationships between the real and imaginary parts to be assessed. The imaginary and real parts are delineated by open and closed markers repectively, and the different temperatures are reperesented by different colours, as detailed in the captions. In these plots, labels, DP_1, DP_2 etc. are given to identify different dispersion processes.

Considering the dielectric spectra above Tg for both the SC and NSC samples, the real part associated with process DP_1 shows greater increment with temperature at low frequencies, coupled with a similar trend in the imaginary part, as shown in Fig. 1. An increase in the magnitude of the real part in the mid frequency range is also observed for both SC and NSC samples which corresponds to a very broad dielectric dispersion, DP_3. However, the associated loss peak is hardly observable in the imaginary part and the real part appears to shift to higher frequency with temperature. An additional dispersion process, DP_2, was found in the NSC epoxy sample. This process was also found in all NSC samples containing mica tape, glass tape and the combination, but absent in the SC samples – with the exception of the mica containing SC sample, to be shown later.

At a test temperature within the glass transition region (around 90 °C) there is a departure from the flat response of the real part in both the SC and NSC samples. Correspondingly, the imaginary part also increases with decreasing frequency, which is compatible with Kramers - Kronig and represents the beginnings of a low frequency dielectric process. At higher frequencies a loss peak appears centred around 5 Hz (as shown in the SC epoxy) which is not apparent for the data for the NSC sample.

For both the SC and NSC samples, the 70 °C scans, taken below the glass transition temperature show that the real part of the response is flat, as shown in Fig. 1. The imaginary part in both samples at the lowest frequencies has a slope << 1 and so it cannot be attributed to a bulk electrical conductivity [3]. In the mid to high frequency ranges, below Tg, the real part of both the NSC and SC response remains flat, however the imaginary part initially shows a decreasing trend with increasing frequency, which then begins to increase with increasing frequency. The onset is shifted to higher frequencies in the NSC sample compared to that of the SC sample.

C. Dielectric measurements – comparison of insulation components

The dielectric spectra of the sputter coated glass tape, mica and composite are shown in Fig. 2. Similarly to the epoxy sample, as shown in Fig. 1b, the glass sample exhibits a flat response in the real domain at 70 °C at low frequencies (Fig. 2a), and the imaginary response has a very similar slope of << 1 in same frequency range. There is a flattening of the imaginary response at the lowest frequencies - in the region 1 mHz to 10 mHz. Again, as with the epoxy sample, the real response is flat in the mid and high frequency ranges but the imaginary response has an upward trend with increasing frequency above 500 Hz, although it is not as pronounced as with the epoxy sample, and in fact the nadir of the imaginary part has shifted up in frequency from 15 Hz in the epoxy to 500 Hz in the sample with the glass fibre.

During the onset of the glass transition at 90 °C the epoxy glass fibre sample exhibits the same tendencies in the real part as the epoxy only sample, viz an increase in the real response with decreasing frequency and a corresponding increase in the imaginary part, however unlike the epoxy only sample there is a flattening off of the imaginary part – similar to that seen at 70 °C in the glass fibre – although this has shifted up in frequency by around half a decade. The real part of the glass sample response is almost indistinguishable from the epoxy at mid and high frequencies. However, the imaginary part is different; the loss peak seen centred around 10 Hz in the epoxy

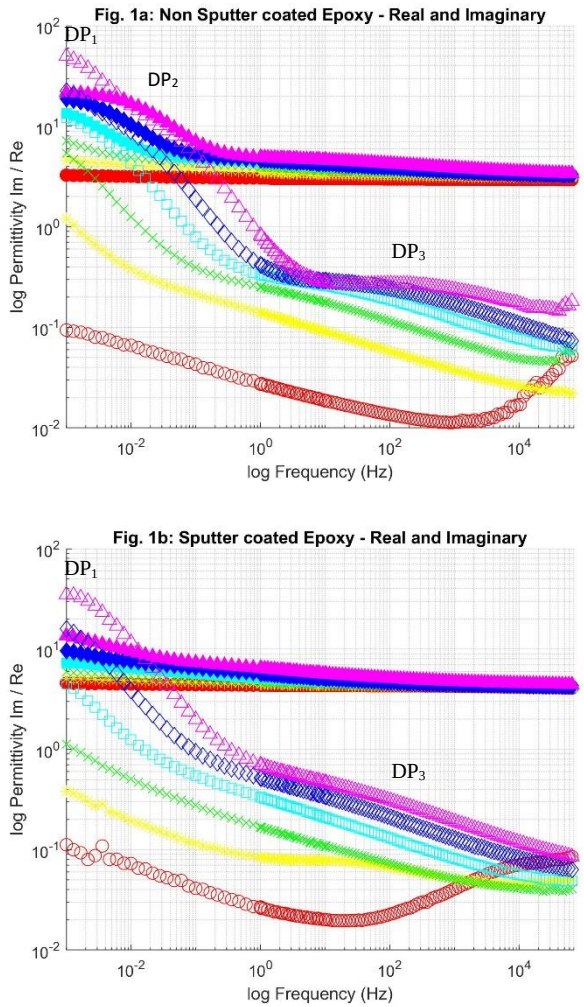

Fig. 1: Real (solid markers) and Imaginary (open markers) parts of non-sputter coated Epoxy (a) and sputter coated Epoxy (b) at temperatures 70 °C (●), 90 °C (*), 100 °C (X), 110 °C (■), 120 °C (◆), 130 °C (▲)

only SC data, has now disappeared to be replaced with a slight uptick from around 350 Hz (as seen in the 70 °C glass data). For the temperatures above the glass transition temperature, the responses in both the real and imaginary domains are almost identical to those of the pure epoxy sample. The only observable difference being the gradients of the imaginary spectra below and above the crossover frequency of approximately 1 Hz. In the sample containing glass the gradients are less than those found for the pure epoxy samples.

The dielectric response of the sample containing mica is shown in Fig. 2b. The dielectric spectra at 70 °C exhibits a slight upward trend in the real component with decreasing frequency particularly in the lowest frequency range (1 mHz – 10 mHz). In this lowest frequency range, there is a corresponding increase in the imaginary part with decreasing frequency with a slope of < -1, which is therefore not a conduction process. In the mid frequency range, there is a loss peak centred about 10 Hz. At 90 °C there is a significant increment in the real part at low frequencies. A significant increase is also seen in the imaginary part, and again the slope is < -1. There is a mid-frequency loss peak – the onset of DP_3 – observable in the imaginary part. This is not seen in the real response due to the overlapping of the spectra on the graph. At temperatures above Tg a large dispersion in the real can be seen at low frequencies, and the large broad dispersion seen at lower temperatures in the imaginary response shifts to higher frequencies by several decades. The slope of the imaginary response at the lowest frequencies is approximately -0.5, which could indicate a possible diffusion process (Fig. 2b). Two loss peaks also appear in the data at these temperatures, the lower frequency one, DP_2, being centred around 15 mHz, and a higher frequency one, DP_3, centred around 1.5 kHz.

Fig. 2c depicts the composite sample response. This contains an amalgamation of the responses of all three previous sample types. At 70 °C the real part of the response is flat across the measured frequency range. The imaginary response is described by an upwards trend in permittivity with decreasing frequency in the low frequency, 1 – 100 mHz, range, and the same flattening between 1 mHz and 10 mHz as seen in the glass sample. In the mid frequency range, the imaginary response is very similar to that of the mica, however, the loss peak occurs at lower frequencies. At the very highest frequencies (of 10 kHz and above) there is the beginning of an upwards trend in the imaginary permittivity, as is seen in all other sample responses. At 90 °C there is the clear onset of a dispersion in the real part of the response at low frequencies, and the mid and high responses are fairly flat – albeit with a gradual increase in permittivity with decreasing frequency. DP_3 is still present, however it is partially obscured by the shift towards higher frequencies of DP_1. Here DP_1 and DP_3 scale differently with temperature and they therefore represent processes with different activation energies.

As the temperature increases above Tg the real response becomes more pronounced at low frequencies, corresponding to the shift towards higher frequencies of DP_1 seen in the imaginary response, which completely obscures DP_2 and partially obscures DP_3.

IV. Discussion

It is evident from the data that there is a clear distinction between the responses of the NSC epoxy, mica and composite and that of the SC epoxy and glass. This indicates that the gross change in spectra is due to the presence of interfaces – whether they be mica / epoxy or electrode / epoxy. Comparing the dielectric spectra of the epoxy and the epoxy glass samples demonstrates that the glass tape has no significant impact on the dielectric properties. This is because the glass tape consists of woven fibres, and as such does not form an effective barrier to charge transport.

The interfacial features concern the presence of DP_2 which is dependent on the type of epoxy – electrode contact or the presence of mica. The magnitude of both the peak and the onset frequency of DP_2 depends on the actual interface concerned. In the glassy state in the epoxy and also the glass tape there appears to be an upward trend in the loss as a function of frequency at higher frequencies. The origin of this process is as yet undetermined, however in samples that contain mica it is not as apparent as in the epoxy and epoxy glass samples. This is possibly due to the higher dielectric losses in the samples containing mica. The dispersion processes, DP_1 and DP_3, appear to be associated with the epoxy resin component as they are observed in all samples, including the epoxy only samples and are independent of electrode contact. Both of these processes appear around and

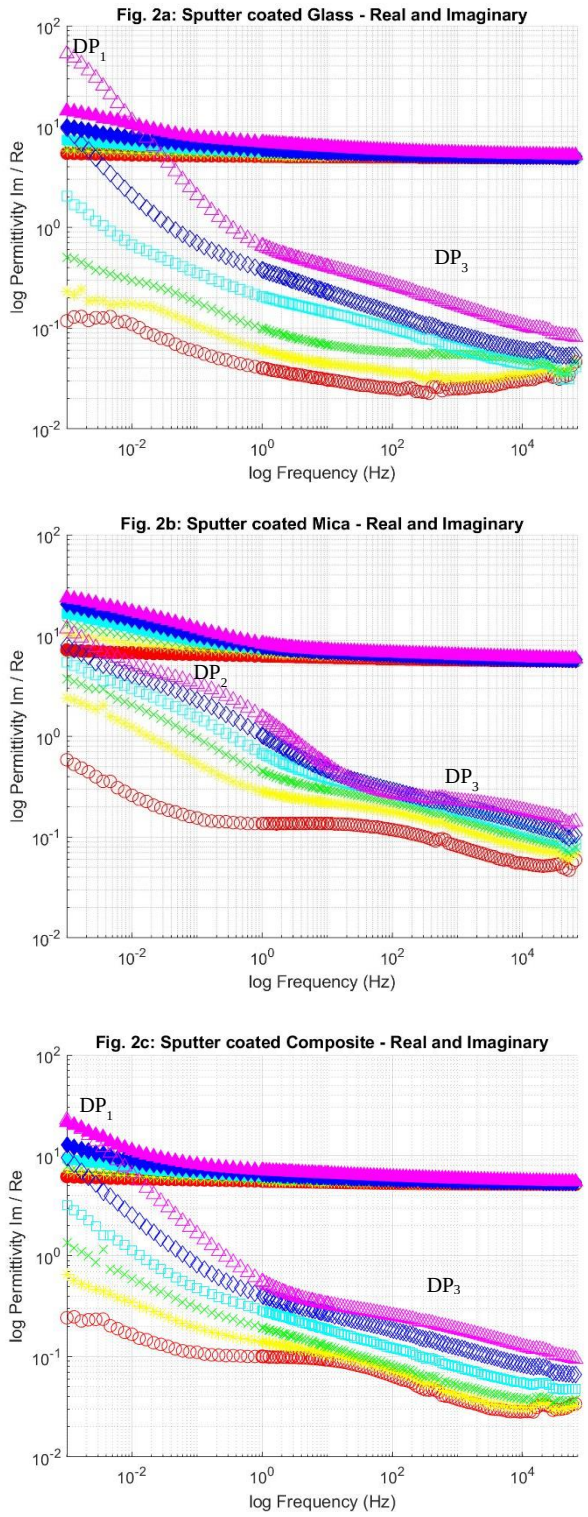

Fig. 2: Real (solid markers) and Imaginary (open markers) parts of sputter coated Glass (a), sputter coated Mica (b) and sputter coated Composite (c) at temperatures 70 °C (●), 90 °C (*), 100 °C (X), 110 °C (■), 120 °C (◆), 130 °C (▲)

above the glass transition temperature. This suggests that they are most likely associated with the alpha relaxation. Process DP_3 can probably be attributed to the alpha relaxation of the resin since it appears at frequencies below 10 kHz, and considering the broadness of the peak, the likely cause is larger scale molecular chain motions. However, process DP_1 is almost certainly a charge transport process as previously demonstrated in [4] where it was identified as a quasi – DC (QDC) conduction mechanism.

V. Conclusion

A dielectric study has been carried out on the individual components of the composite dielectric found in the DBM's at ISIS. Various dielectric features have been identified that relate to both charge transport and to dipolar relaxations that can form the basis for construction of equivalent circuits representing the DBM's insulation system. The presence of mica seems to have higher impact on the dielectric properties than the glass tape. The glass mats do not form an effective barrier to charge transport, while the mica layers seem to provide an effective barrier identified by the presence of an additional interfacial feature in the dielectric spectra.

Acknowledgment

B.A.O would like to thank the technology department at RAL for their diligent preparation of samples and DSC testing, specifically Joel Hodder, Steve Robertson and Simon Canfer.

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