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A Statistical Approach to Assess the Filler Dispersion of Silicone Rubber Composites for HV Outdoor Insulators

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Abstract- **This paper investigates the use of a statistical analysis for assessing the filler dispersion in silicone rubber composites used for high voltage outdoor insulation applications. The study particularly examines the use of the Median Absolute Deviation as a robust measure to evaluate the variability, and thus repeatability, of thermogravimetric analysis test outcomes for different silicone rubber composites at different temperature rise rates. Different Alumina-trihydrate filled silicone rubber composites are examined in the study. The results suggest a correlation between the Median Absolute Deviation and the degree of filler dispersion observed under Scanning Electron Microscopy. This correlation in outcomes is particularly evident at 25 °C/min. The composites are further tested using the dry-arc resistance test as per ASTM D495. The variability in the erosion resistance outcomes of the dry-arc resistance test correlate with the Median Absolute Deviation outcomes. These outcomes collectively suggest the potential of the proposed statistical approach as a quick assessment tool to supplement microscopic imaging towards understanding the effect of the degree of filler dispersion on the erosion resistance of silicone rubber for use in high voltage outdoor insulation applications.**

***Keywords*- High voltage outdoor insulator, silicone rubber composites, filler dispersion, Median Absolute Deviation.**

I. Introduction

Outdoor insulators play a crucial role as backbone components in electrical power transmission and distribution systems. Polymeric outdoor insulators with polymeric housing materials, such as silicone rubber (SiR), are considered advantageous over ceramic outdoor insulators in terms of the significant weight reduction they provide. The use of SiR outdoor insulators is particularly beneficial for silicone hydrophobicity which naturally resists the formation of wet contaminant films and, subsequently, hinders the formation of eroding dry-band arcing which is the critical ageing mechanism of SiR outdoor insulators [1]. Research primarily concluded that the inclusion of inorganic fillers in silicones to create SiR composites is beneficial towards improving the erosion resistance of these materials for use in high voltage alternating and direct current (HVAC and HVDC, respectively) outdoor insulation applications. Alumina-trihydrate (ATH) and silica are examples of such commonly used inorganic fillers in SiR composites [2].

Effective dispersion and distribution of the filler particles in the silicone matrix is essential to obtain the utmost benefit of the filler in improving the erosion resistance of SiR. Research indicates that the proper dispersion of the filler particles improves the thermal conductivity of the SiR composite which in turn improves the erosion resistance of the composite against eroding dry-band arcs. Du et al. in [3] illustrated the effect of proper filler dispersion on the thermal conductivity of boron nitride filled-SiR and the impact of that improvement on thermal conductivity on the DC erosion resistance of the composites. The proper dispersion of the fillers was preserved using mechanical mixing with a sonication technique. The impact of filler dispersion of silica fillers on the erosion resistance of SiR composites was further outlined in the work of [4] by illustrating the superior erosion resistance of composites fabricated by electrospinning technique as compared to conventional mixing technique. In similar research, Khanum et al. in [5] indicated the improvement of the thermal conductivity of SiR composites by means of using electrostatic dispersion technique to ensure effective dispersion of boron nitride filler particles in SiR. In other research [6], the impact of filler dispersion on the thermal decomposition properties of the SiR composites was also highlighted whilst indicating the impact of filler calcination and surface treatment on the filler dispersion.

The role of filler interface on the erosion resistance of SiR composites was highlighted in previous studies such as in [7-9]. The interaction between the filler and the silicone plays a crucial role in determining the thermal decomposition properties during the erosion of the SiR material under dry-band arcing. Increasing the effectiveness of the filler-polymer interface role would essentially require a good dispersion and distribution for the filler particles within the silicone matrix. Thus, examining the filler dispersion in the prepared SiR composites is a key factor towards predicting the thermal decomposition properties and the erosion resistance of the composites under dry-band arcing. Microscopy is typically used to qualitatively asses the dispersion of the filler in SiR composites. However, setting up the samples for imaging in

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tools such as Scanning Electron Microscopy (SEM) could be difficult and time consuming in terms of the effort that has to be invested in obtaining the images that could properly indicate the degree of filler dispersion in the prepared composite. The presented paper proposes a complementary and relatively quantitative approach which utilizes the Median Absolute Deviation (MAD) as a measuring statistic to assess the variability in thermogravimetric analyses (TGA) test outcomes. The proposed method examines the TGA plots and provides an insight of the degree of filler dispersion in the SiR composites. Two different types of ATH-filled SiR composites have been selected for the study, one of which is a commercial composite while the other is prepared in the laboratory. The dry-arc resistance test is used to further asses the erosion resistance outcomes of the composites, and investigate whether the variability in these outcomes could correlate with those of the MAD.

II. Materials and Methods

Table I shows the details of the SiR composites used in the study along with their associated properties. The composites used in this study are ATH-filled SiR composites with different ATH filler levels and particle sizes. The 5 percent by weight (wt%) nano-ATH filled SiR composite, or A05, was prepared in the laboratory using a two-part room temperature vulcanized (RTV) SiR, RTV 615, from Momentive®. Details of the preparation procedure of the composite can be further found in [7-9]. The 60 wt% micro-ATH filled SiR composite, or A60, is a commercial composite for HV outdoor insulation applications. The composites have been investigated in prior research in [7, 11] and selected for this study based on the prior knowledge of the degree of dispersion of the filler in the SiR composite.

TABLE I
SiR Composites Used in the Study and Assoiated Filler Properties

Composite Sample Code	Inorganic Fillers	Type of SiR	Filler Particle Size (µm)	Filler wt% in samples prepared
A05	Alumina Trihydrate	RTV	1×10^{-3}	5
A60		HTV[a]	NP[b]	60

[a] High Temperature Vulcanized, [b] Information not provided

The prepared composites are tested using the TGA. Each composite formulation was tested five times in nitrogen for a given temperature rise rate. Two temperature rise rates of 25 °C/min and 100 °C/min have been selected for the study to represent the slow and fast temperature rise rates respectively. The temperature span was set from 80 to 800 °C with the sample under test weighing between 10 to 20 mg. For a given set of five TGA outcomes, the MAD was computed for each temperature point T (ranging from 80 to 800 °C) using the equation,

$$MAD_T = median(|W_{T_i} - \widetilde{W}_T|) \quad T=[80,800] \quad i=[1,5] \qquad (1)$$

where MAD_T is the computed median value for set of five absolute differences between the TGA percentage weight W_{T_i} (at the i^{th} TGA test outcomes) and the median TGA percentage weight $\widetilde{W}_T$ of the five W_{T_i} values at a given temperature point T. The final outcome of using (1) is to eventually produce a single MAD plot for the given set of five TGA plots at a given temperature rise rate for the temperature span from 80 °C to 800 °C. The higher the MAD_T value for a given temperature point T, the higher is the variability and inconsistency amongst the five TGA weight percentage values for that same temperature point. To have a reference for the filler dispersion and distribution in the composites, SEM was used in the study. The samples were prepared for SEM imaging through blade cutting, followed by gold sputter coating.

Fig. 1 shows the setup for the dry-arc resistance test used in the study. The dry-arc test is set as per the ASTM D495 standard [10]. For this study, the dry-arc resistance test is primarily used to assess the erosion resistance of the composites and their variability through multiple test runs, while ensuring a stable arc pattern during the tests. Such a condition is important in order to minimize the effect of the arc instability on the variability of the erosion resistance determine, either in terms of erosion depth or volume. Accordingly, variability in the erosion resistance obtained would be rather attributed to the effect of the filler dispersion. The test subjects the composites under test to an arcing current mimicking the dry-band arc. The magnitude and duration of the arc changes in scheduled Current Step cycles (CS) which are defined in [10]. For this study, only the first four cycles (CS1 to CS4) are used. Five tests were conducted for each composite in the study to asses the variability and repeatability in the erosion resistance outcomes.

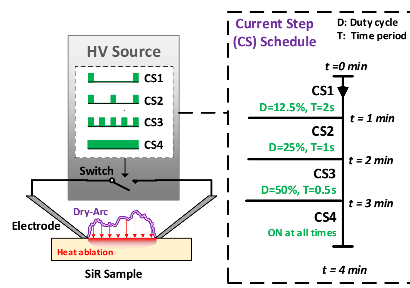

Fig. 1. Dry-arc resistance test used in the study.

II. Results and Discussion

Fig. 2 and Fig. 3 show the SEM images obtained for the A05 and A60 composites. A more dispersed and distributed pattern for the filler particles can be seen in A60 as compared to A05 composites. Though the filler loading level is 12 times higher in A60 than in A05, the filler particle distribution and dispersion is inconsistent in the later composite as observed in the SEM image. The A05 SEM image shows agglomerates of the filler particles and inconsistency in the filler particle sizes and inter-particle spacing as compared to the A60. This observation is further evident in terms of the much higher magnification level of the image of the A60 as compared to A05.

The superior filler dispersion and distribution in the A60 particle could be highly dependent on the fabrication method used to tear apart the ATH filler particles and mix them with the silicone matrix. The composite preparation method can highly influence the degree of filler dispersion as was explained in [4,6], and for a commercial composite like that of A60 a surface treatment of the ATH filler could have been used to improve the filler dispersion during the fabrication process. As for the

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nano ATH filler used in A05, previous research indicated poor dispersion of nano ATH in SiR which was attributed to its unfavorable interaction with SiR at the filler-polymer interface resulting in the formation of filler agglomerates during high shear mixing [7].

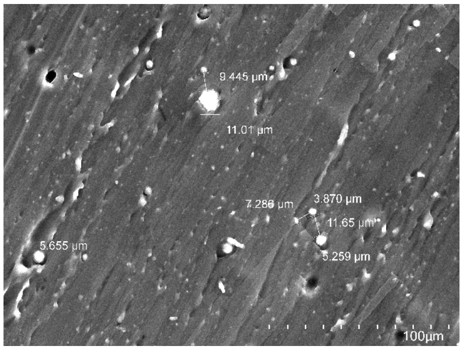

Fig. 2. SEM image obtained for A05 at x500 magnification.

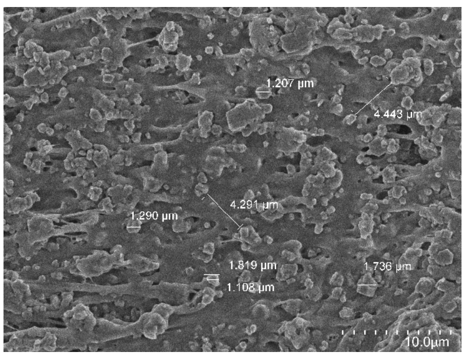

Fig. 3. SEM image obtained for A60 at x3000 magnification.

Fig. 4 and Fig. 5 show the TGA plots obtained for the A05 and A60 SiR composites respectively. Each figure shows two different temperature rise rates, 25 °C/min and 100 °C/min, with five TGA tests plotted for a given temperature rise rate. As was explained in [11], the evident thermal decomposition near 200 °C represents the dehydration of the ATH filler which is more visible in the A60 composite as compared to the A05 composite given the higher ATH filler loading in the former. Further detailed analysis of the decomposition mechanisms in ATH filled SiR composites under TGA can be found in [7]. As can be clearly observed in the Fig. 4 and Fig. 5, a distinct characteristic could be observed for the A60 composite in terms of the consistency of the test outcomes at both temperatures rise rates. The A05 composite, however, seems to show more variable TGA curves for both temperatures rise rates.

Fig. 6 and Fig. 7 show the MAD plots obtained for the TGA outcomes of Fig. 4 and Fig. 5. As was explained earlier, the MAD provides a robust measure for the variability of the TGA test outcomes for a given temperature rise rate. As can be clearly seen in Fig. 6, the A60 composite shows consistently lower MAD values compared to the A05 composite at temperatures beyond 400 °C at the slow 25 °C/min temperature rise rate. As was explained in [13], this 400 °C temperature point represents the depolymerization of SiR under inert atmosphere. This could accordingly suggest lower variability in the TGA outcomes of the A60 composite which in turn represents high repeatability in these test outcomes. This repeatability could be primarily attributed to the well dispersed and distributed ATH filler in A60 as compared to A05 as was evident in SEM images of Fig. 2 and Fig. 3. In other words, each 10-20mg sample taken from the A60 composite for TGA testing is consistent in terms of the weight fraction of the filler available in the sample under test. For the A05 composite, each TGA sample under test could contain different weight fraction of the filler as a result of the poor filler dispersion and distribution which could significantly impact the thermal decomposition characteristics as is observed in Fig. 4.

At the fast temperature rise rate in Fig. 7, the MAD plot seems to show a difference in the trend of variance between the A60 and A05 composites. Though the MAD values for the A60 composite are higher for temperatures less than 600 °C, the MAD trend changes at higher temperatures to be similar to Fig. 6 between the two composites. This inconsistency in the MAD trend could be a result of the faster temperature rise rates which impacts the thermal decomposition properties of the composites during the test run. It was explained in [12] that radical based crosslinking is introduced in SiR at elevated temperatures beyond 600 °C, which competes with the depolymerization of SiR during the erosion process. This radical-based crosslinking mechanism was also reported to be influenced by the temperature rise rate which is set during the TGA test run [13]. Further investigation would be required in order to understand the effect of the competing nature of the two thermal degradation mechanisms on the corresponding MAD in the TGA.

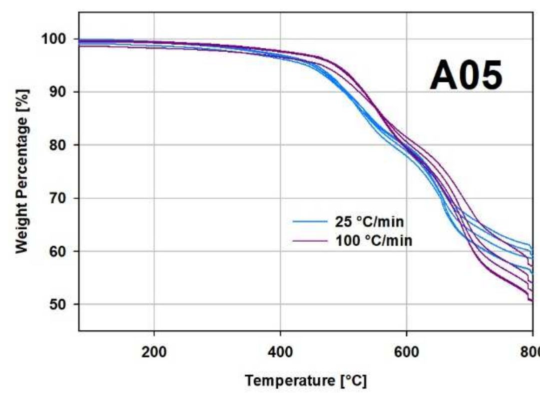

Fig. 4. TGA plots for the A05 composite under 25 °C/min and 100 °C/min.

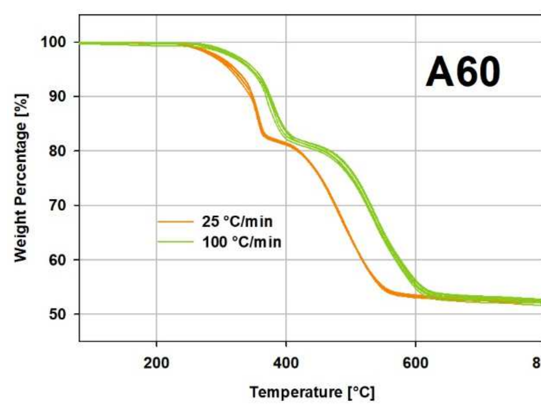

Fig. 5. TGA plots for the A60 composite under 25 °C/min and 100 °C/min.

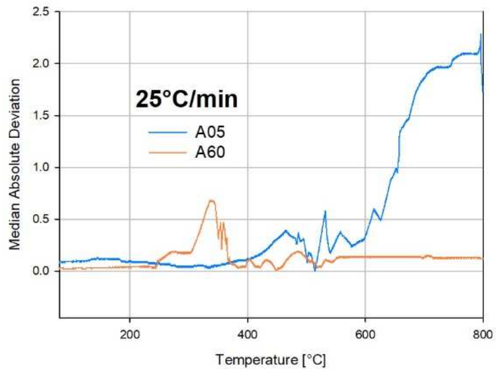

Fig. 6. MAD plots for TGA outcomes under 25 °C/min.

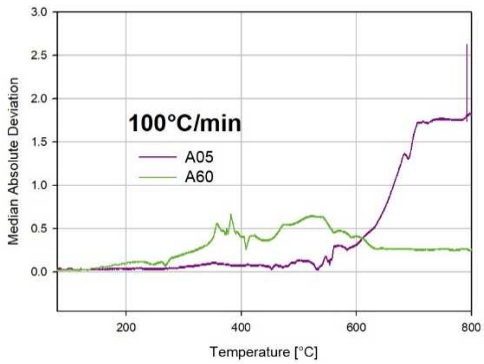

Fig. 7. MAD plots for TGA outcomes under 100 °C/min.

Fig. 8 shows the dry-arc resistance test outcomes for the composites of the study which are also reported in [11]. The results indicate the average erosion depth and volume obtained for five tests from each composite along with the associated uncertainties in the measurements. The dry-arc resistance test outcomes primarily indicate negligible uncertainties, and thus insignificant variability, in the erosion resistance outcomes obtained for A60. As for A05, however, the higher uncertainty in the measurements of both the eroded depth and volume indicate a high inconsistency in the erosion resistance outcomes which could be attributed to the poor dispersion and distribution of the filler in the composite as observed in Fig. 2. Accordingly, the degree of variability in the erosion resistance outcomes correlates with the MAD outcomes obtained in Fig. 6.

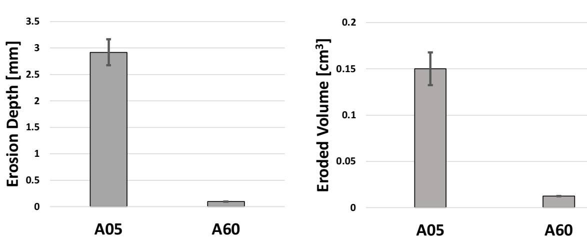

Fig. 8. Dry-arc resistance test outcomes for the composites of the study which were previously obtained in [11].

These aforementioned outcomes collectively suggest the feasibility of using the MAD as a statistical approach in processing TGA test outcomes towards assessing the filler dispersion of SiR composites. This approach could act as a supplemental test to SEM for assessing the filler dispersion, which ultimately is used to aid the erosion resistance outcomes obtained by the dry-arc resistance test for developing SiR composites for outdoor insulation applications.

CONCLUSION

The presented paper introduced a statistical approach for assessing the filler dispersion in SiR composites using the MAD. The outcomes of the MAD plots suggest a more dispersion and distributed pattern for the filler particles in the A60 commercial composite as compared to the A05 laboratory prepared composite which goes in correlation with the SEM images obtained in the study. Moreover, the MAD outcomes for the two composites correlate with the variability in the erosion resistance outcomes of the dry-arc resistance test. The proposed approach could therefore be used as an additional tool complementing the dry-arc resistance test for assessing the SiR composites for use in HV outdoor insulation applications.

REFERENCES

[1] K. N. Mathes and E. J. Mcgowan, "Surface electrical failure in the presence of contaminants: The inclined-plane liquid-contaminant test," in *Trans. of the Amer. Inst. of Electr. Eng., Part I: Comm. and Electron.*, vol. 80, no. 3, pp. 281-289, Jul. 1961.

[2] L. Meyer, R. Omranipour, S. Jayaram and E. Cherney, "The effect of ATH and silica on tracking and erosion resistance of silicone rubber compounds for outdoor insulation," in *2002 IEEE Intern. Symp. on Electr. Insul.*, Boston, MA, USA, 2002, pp. 271-274.

[3] B. X. Du and H. Xu, "Effects of thermal conductivity on dc resistance to erosion of silicone rubber/BN nanocomposites," in *IEEE Trans. on Dielectr. and Electr. Insul.*, vol. 21, no. 2, pp. 511-518, Apr. 2014.

[4] S. Bian, S. Jayaram and E. A. Cherney, "Erosion resistance of electrospun silicone rubber nanocomposites," in *IEEE Trans. on Dielectr. and Electr. Insul.*, vol. 20, no. 1, pp. 185-193, Feb. 2013.

[5] K. K. Khanum and S. H. Jayaram, "Improved Thermal Properties and Erosion Resistance of Silicone Composites With Hexagonal Boron Nitride," in *IEEE Trans. on Ind. Appl.*, vol. 58, no. 5, pp. 6583-6590, Sep. 2022.

[6] K. K. Khanum, A. M. Sharma and S. Jayaram, "Impact of Dispersion Processes and Surfactant on Performance of Silica-Silicone Nanocomposites," in *IEEE Trans. on Dielectr. and Electr. Insul.*, vol. 29, no. 1, pp. 22-29, Feb. 2022.

[7] A. Y. Alqudsi, R. A. Ghunem and É. David, "Analyzing the Role of Filler Interface on the Erosion Performance of Filled RTV Silicone Rubber under DC Dry-band Arcing," in *IEEE Trans. on Dielectr. and Electr. Insul.*, vol. 28, no. 3, pp. 788-796, Jun. 2021.

[8] A. Y. Alqudsi, R. A. Ghunem and É. David, "The Viability of the Filler Barrier Effect During the DC Dry-Band Arcing on Silicone Rubber," in *IEEE Trans. on Dielectr. and Electr. Insul.*, vol. 29, no. 5, pp. 1873-1881, Oct. 2022.

[9] A. Y. Alqudsi, R. A. Ghunem, and E. David, "A Novel Framework to Study the Role of Ground and Fumed Silica Fillers in Suppressing DC Erosion of Silicone Rubber Outdoor Insulation," in *Energies*, vol. 14, no. 12, p. 3449, Jun. 2021.

[10] Standard Test Method for High-Voltage, Low-Current, Dry Arc Resistance of Solid Electrical Insulation, ASTM D 495-14.

[11] A. Y. Alqudsi, R. A. Ghunem and E. David, "Ranking the DC Erosion Resistance of Silicone Rubber Composites with the Dry-Arc Resistance Test," in *2021 IEEE Conf. on Electr. Insul. and Dielectr. Phenom. (CEIDP)*, Vancouver, BC, Canada, 2021, pp. 328-331.

[12] S. Kumagai, X. Wang and N. Yoshimura, "Solid residue formation of RTV silicone rubber due to dry-band arcing and thermal decomposition," in *IEEE. Trans. Dielectr. Electr. Insul.*, vol. 5, no. 2, pp. 281-289, Apr. 1998.

[13] G. Camino, S.M Lomakin and M. Lageard, "Thermal polydimethylsiloxane degradation. Part 2. The degradation mechanisms,"b in *Polym.*, vol 43, no.7, pp. 2011-2015, Mar. 2002.