The Electrical Performance of Microvaristor Composites at Different Filler Loadings

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Abstract. Resistive field grading materials is deliberately gaining attention since it shows better performance in steering uniformly electrical properties of high voltage equipment compared to conventional machinery techniques. The key point of these extraordinary materials because its electrical properties that can be tuneable according to material characteristics, composition of chemical elements in the host matrix and significantly influenced from fabrication process. In this research, microvaristor composite was added with silicone rubber at different weight percentage (wt. %) with dimension 50 mm x 50 mm x 5 mm. The selection of this material because it is more robust and less sensitive to a variety parameters such as pressure and humidity. The samples were examined under power frequency voltage at 50 Hz. The experimental results were analysed using Point on Wave (POW) technique and electrical properties for all samples were evaluated at highest applied voltage, 5 kV.

1. Introduction
For the past half-decade, the incorporation of filler in host matrix has improved the reliability of equipment for long term applications at minimum costs and high flexibility of designs. In conjunction to this interest, broad research have been conducted to investigate the effect of nano and micro fillers in various fields as presented in few publications [1]–[3]. Significant responses from industries and academia towards this research field have intensified the importance of smart material over conventional methods. In power system industries, the resistive field grading material was introduced in cable accessories, insulators, and bushings which can mitigate the high localization of electrical fields and controlling the over voltages. Non-linear material such as SiC that compounded with polymer matrix are generally used in this area, however few drawbacks have been highlighted during its applications. The dependence of non-linearity contacts, the difficulty to control the process of production, and the high risk of degradation attack under voltage overload have tuned the direction towards ZnO microvaristor. Doped ZnO microvaristor exhibits high nonlinear resistivity either in powder or micro composite form. The production process for this material is also simple and very economical. This paper explains the process of ZnO microvaristor powder dispersed in silicone rubber matrix at certain weight concentrations.
The alternating current test of 50 Hz was applied to the sample with dimension of 50 mm x 50 mm x 5mm in order to investigate the breakdown voltage and the non-linear resistivity of micro composite sample. Meanwhile, the Point On Wave (POW) technique [13] [14] was used to discriminate the leakage current to two main components; resistive and capacitive currents. This current discrimination is important because there is high possibility of degradation to the electrical properties due to continuous power frequency voltage application. Moreover, the results from this technique will also determine the electrical properties of micro composite sample itself.

2. Methodology
Microvaristor filled silicone rubber sample is fabricated in High Voltage Laboratory, Cardiff University. In this research, there were a few techniques that applied which commonly applied as such in the publication [4]-[5]. Generally, the insulating materials such as oils, ethylene-propylene rubber (EPR), thermoplastic elastomer, glass, silicone gel and silicone rubber are used as the host matrix for filler loadings. The properties of silicone rubber and microvaristor powder are shown in Table 1.

Table 1: The material properties of host matrix (silicone rubber) and microvaristor powder

| Material     | Properties     | Inspection method | Value          |
|--------------|----------------|-------------------|----------------|
| Silicone rubber | Permittivity  | IEC 60250         | 2.9            |
|              | Density        | ISO 1183-1 A      | 1.13 g/cm³     |
|              | Volume resistivity | IEC 60093     | 10x10¹⁵ Ω.m    |
| Microvaristor | Density        | -                 | 5.6 g/cm³      |

In this material preparation process, the microvaristor powder are in different sizes is chosen as fillers. A profile of microvaristor dimension is shown in Figure 1.

![Figure 1: The variation sizes of the microvaristor powder.](image)

From this graph, the distribution sizes of microvaristor particles mainly in range of 10 μm. The sizes of particles has a great influence to the electrical properties of micro composite as discussed in [6]. This will give advantages to the microvaristor as it comprises electrical conductive particles in
different coarse and fine fraction, hence able to transfer its strong electrical nonlinearity directly to the composite. Different sizing of micro particles provides a good bimodal distribution that exclusively improved nonlinear electrical properties, increased power absorption and minimizes the breakdown field strengths [7][8].

The influence of moisture on the microvaristor powder was also studied. It is very important to understand the characteristic of microvaristor powder before adding this filler with silicone rubber matrix. The efficiency of material to operate will be gradually decreased if there is high moisture trapped in the particles. Therefore, the microvaristor powders were pre-heat for at least 1 hour at 50°C in an oven which has possibility to reduce the formation of moisture in the micro-particles. On top of that, it benefited for high microvaristor loadings when the silicone rubber composition contains more than 30 wt%. Therefore, the high temperature from pre-heating process will increase the solubility of microvaristor powder in the silicone rubber during mixing process. In the first step, the silicone rubber was weighed with the microvaristor powder. The weights of percentage of microvaristor were then compounded into the base polymer by using a high shear mixer.

This mechanical mixing was used to disperse the micro-particles uniformly in the polymer matrix and to minimize any agglomeration in the micro-composite. According to Catalin Picu et.al, the good dispersion of micro-meter-size fillers has been identified an important factor for achieving properties of micro composite even though there are few uncertainties parameters such as filler distribution which may affect the overall micro composite properties. Therefore, in this study, a mechanical mixing (Ross model HSM-100LSK) was used to disperse the micro composite particles into silicone matrix at starting speed of 400 until 2000 rpm. This process was known as agitation process [9] and this range of speed was applied until the mixture wetted out. The second process was mixing stage followed by dispersion stage.

At the third stage, the speed was slowly increased until 6000 rpm for at least 5 minutes and this dispersion speed is practically applied for high shear laboratory machine. It is important to justify the suitable shear speed for microvaristor powder; hence a specific calculation was adopted from [10] to determine the relationship between shear mixer speeds with the size of fillers. This high shear mixer has 16 mm blade diameter, meanwhile the peripheral velocity for mixing blade of micro particles is 5 m/sec [11] to obtain sufficient stir for uniform microvaristor dispersion. It has been identified that the mixing process was the critical method during fabricating micro-composite sample. There was a high possibility of agglomeration structure to occur during the mixing process. Because of this reason, the temperature and the material reaction during micro composite process should be controlled [9]. The optimum time for mixing was set for 7 minutes with the temperature rising approximately to 45°C. This concept was adopted from theory that at high temperature, the coalescence of micro-particle was fast and may minimize the fractal of agglomeration [12]. If the mixture goes beyond the maximum temperature, it is necessary to give a rest to the mixture in room temperature instead of placing in the cooling vessel.

A monitoring of solidification was done in order to understand the reaction of mixture after transition from mixing process to casting process. The solidification reaction was occurred after the mixture was added with the hardener in vacuum casting machine and underwent the degassing process. Figure 2 shows a solidification time for different microvaristor loadings. From the figure below, the solidification of mixture is rapidly occurring at the highest microvaristor concentration. Because of this condition, all the processes in the sample preparation shall be carry out very quickly to minimize the agglomeration in the micro composite structure.
Figure 2: The solidification of microvaristor filled silicone rubber at different concentrations.

The mixture was then injected into aluminum mould and allowed to cure in vacuum casting machine for 1 hour. Then, the sample was placed in room temperature in the oven at 50 °C for 4 hours. A set of 10 wt. %, 30 wt. %, 60 wt. % and 70 wt. % samples were fabricated, in order to investigate the effect of microvaristor contents to the non-linearity characteristics of field grading materials. The dimension for all samples is 50 mm x 50mm x 5mm.

3. Results and Discussion

The AC analysis is an important test to investigate the electrical properties and voltage and current characteristic of microvaristor filled silicone rubber under normal frequency operation. The test is conducted in High Voltage Laboratory, Cardiff University. This test is particularly to evaluate the inter-contact of microvaristor grains and polymer matrix which will determine the non-linearity behavior of samples. The test is conducted accordingly to the standard BS 60060-1: 2010. In this test, the maximum voltage supply is 10 kV and it is controlled by voltage regulator. A voltage divider at ratio 1000:1 is used to measure the actual input voltage across the test sample. Meanwhile, a shunt resistor of 1 kΩ is used to determine the instantaneous current during the test. Both of voltage and current are simultaneously displayed using a Lecroy digital storage oscilloscope. In the meantime, the data for this experimental works are also saved in data acquisition (DAC) system. This data system is constructed through Labview applications that specifically designed for analysis studies. The justification of the experimental results in this system is through a comparison between the results on digital signal oscilloscope and the output from DAC system. Any discrepancies of data can be traced immediately; hence the improvement action will be taken.

The test electrode system was specifically designed and constructed for this A.C test. The electrodes are equipped with guard ring. The guard rings were made from insulating material (PTFE), used in controlling the current flow from high voltage terminal to ground terminal. Because of the electrode physical design, there was no contact between electrode holder and counter electrode. Therefore the uniformity of electric field distribution around the micro composite surface can be achieved. The design of electrode system is shown in Figure 3. The diameter of the counter electrode was 10 mm with a gap of 0.5mm between the guard ring 0.5 mm. Meanwhile the radius of electrode holder was 15 mm.
The results of micro composite samples at concentration of 10 wt. %, 30 wt. %, 60 wt. % and 70 wt. % were evaluated by using point on wave analysis. The results show the electrical properties for all samples which consist the resistivity, capacitance, dielectric constant and the current profile.

It shows at 10 wt. % of microvaristor loading, the composite has highest reading of $14 \times 10^5$ ohm.m and the trend goes down when the concentration of fillers is become higher. This evaluation is done when the applied voltage was at 5 kV. At this voltage rating, the electrical field that distribute along the sample was achieved 10 kV/cm. Meanwhile, the sample with the highest microvaristor concentration shows the lowest resistivity profile as the sample has the minimum content of silicone rubber. Therefore, the sample acts as perfect conductor when reached its threshold voltage.
The capacitance profile is governed by the resistivity value. Thus, the capacitance is decreasing when the microvaristor concentration is increased from 10 wt. % to 70 wt. %. The dielectric constant of each sample is measured accordingly as shown in Figure 6.

The results of 10 wt. %, 30 wt. % and 60 wt. % microvaristor filled silicone rubber show small changes of dielectric constant value even at low voltage to high voltage. Interestingly, the sample of 70 wt. % shows a non-linearity response as the dielectric value gradually decreases when sample reached applied voltage of 5 kV.
Figure 7: The current components profile for all samples. The measurement was done at the same voltage level of 5 kV.

The characteristic of currents are analyzed as shown in Figure 7. It is clearly can be seen the dominant of resistive current compared to capacitive current when the filler is added into the sample. The result of 70 wt. % sample shows the highest resistive current which is corresponding to the effect of non-linear resistivity. From this result, there is a clearly agreement with [5][15] that highlighted at certain microvaristor composition (40 vol. %), the nonlinearity behavior of micro composite can be obtained. Moreover, the research from [16]-[17] also highlighted the behavior of nonlinear composites can be revealed when the suitable filler content is composed with the host matrix.

4. Conclusions
This paper has discussed the technique for fabricating samples of microvaristor filled silicone rubber. A set of samples at different compositions (wt. %) were fabricated and analyzed. Exclusively, the non-linear resistivity of ZnO microvaristor is different corresponding to the microvaristor composition in the samples. When sufficient amount of microvaristor loaded in silicone rubber, the composite will change the behavior from insulator to conductor which responding to good inter-linkages of micro fillers. The conductivity of ZnO microvaristor is depending on built-in grain boundaries, therefore the highest microvaristor contents (wt. %) will increase the leakage current flow in the sample. As the nonlinearity characteristics of composite is related to the particle contacts; the high fillers content will improve considerably the electrical performance of nonlinear electrical properties. An AC test was conducted to examine the performance for all samples. From the observations, the sample at 70 wt. % shows non-linearity behavior as compared to other samples. With attention to determine the current components from total leakage current flow; the Point On Wave (POW) technique was performed. Therefore, resistive and capacitive currents can be evaluated. From this technique, the electrical properties such as resistivity, conductivity, dielectric constant are obtained.

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References
[1] Zhonglei Li, Zhuran Yang, and Boxue Du, “Surface Charge Transport Characteristics of ZnO/Silicone Rubber Composites Under Impulse Superimposed on DC Voltage,” IEEE Access, vol. 7, 2019.
[2] Xiao lei, Zhao Xiao, Yang Jun, Hu Qi, Li Jinliang He, “Globally reinforced mechanical, electrical, and thermal properties of nonlinear conductivity composites by surface treatment of varistor microspheres” in Science Direct. Volume 175, 3 May 2019, pages 151-157

[3] O. Mejia and O. Fernando, “Micro And Nano Composites Composed Of A Polymer Matrix,” Thesis, 2007.

[4] K. Lau, “Structure and electrical properties of silica-based polyethylene nanocomposites,” Thesis, 2013.

[5] S. Ishibe, M. Mori, M. Kozako, and M. Hikita, “A New Concept Varistor With Epoxy/Microvaristor Composite,” IEEE Transactions on Power Delivery, pp. 1–6, 2014.

[6] A. Gustafsson and L. Palmqvist, “Electrical properties of field grading materials Influenced by the Silicon Carbide Grain Size,” ICSD'01 International Conference on Solid Dielectrics, pp. 43–45, 2001.

[7] A. C. Angelo Austermuhl, “Modified silicone gels for medium and high voltage insulation,” in Conference on silicone insulation, 2013.

[8] J. Schulte-fischedick, F. Lehretz, P. M. Clemens, and M. Bornowski, “Electric field grading using insulators with microvaristor filled silicon rubber,” IEEE International Conference on Solid Dielectrics, Bologna, Italy, 2013, pp. 0–3.

[9] C. R. Son, “Installation, Maintenance and Operating Manual.”

[10] S. Bian, “A Study of the Material Properties of Silicone Nanocomposites Developed by Electrospinning,” Thesis, 2013.

[11] J. E. White and L. Jackson, “Polymer microfiller composites”, US Patent, No. US 2010/0137478 A1, 2010.

[12] P. V. B. Pulickel M.Ajayan, Linda S.Schadler, Nanocomposite science and technology, 1st edition. Wiley-VCH, 2004, p. 94.

[13] C. Spellman, “A Technique for on-Line Monitoring of ZnO Surge Arrester,” Proceedings of the 10th International Symposium on High Voltage Engineering, 1997.

[14] D. W. A.Haddad, Advances in High Voltage Engineering. The Institution of Electrical Engineers, London, 2004, pp. 230–231.

[15] J. Pitha, “Non-linear resistance material,” US Patent 3,291,759, pp. 1–3, 1966.

[16] L. Donzel, F. Greuter, and T. Christen, “Nonlinear resistive electric field grading Part 2: Materials and applications,” in IEEE Electrical Insulation Magazine, volume: 27, issue: 2, March-April 2011.

[17] G. R. Ruschau, S. Yoshikawa, R. E. Newnham, and G. F. L. Fiuschau, “Resistivities of conductive composites,” Journal of Applied Physics 72, vol. 953, no. 1992, pp. 10–17, 2005.