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OPTIMIZATION OF SURFACE RESISTIVITY AND RELATIVE PERMITTIVITY OF SILICONE RUBBER FOR HIGH VOLTAGE APPLICATION USING RESPONSE SURFACE METHODOLOGY

Silicone Rubber (SiR) is considered as one of the most established insulator in High Voltage (HV) industry. SiR possess a great function ability such as its lighter weight, great heat resistance and substantial electrical insulation properties. Dynamic research were performed all around the world in order to explore the unique insulating behavior of SiR but very little are done on the optimization of SiR in term of their processing parameters and formulation. In this work, four materials and processing factors were introduced; A: Alumina Trihydrate (ATH), B: Dicumyl-Peroxide (DCP), C: mixing speed and D: mixing time in order to analyze its contribution towards improving the surface resistivity and relative permittivity of SIR rubber. The factors range were set based on prior screening and are defined as; ATH (10 – 50 pphr), Dicumyl Peroxide (0.50 - 1.50 pphr), speed of mixer (40 – 70 rpm) and mixing period (5 – 10 mins) which were then varied accordingly to produce an overall 19 samples of SiR blends. The testing results were analyzed using statistical Design of Experiment (DOE) by applying two level full factorial from Design Expert Software (v10) to discover the inter-correlation between the factors studied and benefaction of each factor in improving both surface resistivity and relative permittivity responses of produced SiR blends. The model analysis on surface resistivity shows the coefficient of determination R^2 value of 88.72% while the one for relative permittivity shows R^2 value of 82.34 %. Combination of both dependent variables had yielded an optimization suggestion for SiR formulation and processing strategy of ATH: 50 pphr, DCP: 0.50 pphr, mixing speed: 70 rpm and mixing period: 10 mins with the desirability level of 0.835. The optimized formulation had resulted in the production of SiR blend with the characteristic of surface resistivity of 1.02039×10^{14} Ω/sq and relative permittivity of 4.0231, respectively. In conclusion, it can be said that the materials formulation and processing parameters had significantly influenced the performance of SiR blends and thus, having the optimized material composition and processing parameters is required in producing an insulator with great function ability for high voltage application.

Keywords: Silicone Rubber, surface resistivity, relative permittivity, mixing parameters; optimization; desirability; high voltage; Design of Experiment.

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1. Introduction

Silicone Rubber (SiR) composites concept had been established in USA as early as in the year of 1948 [1]. Ever since, SiR was denoted as one of the most favorable insulation due to its excellence compatibility, high heat resistance, great weather ability and most importantly, great electrical insulation properties [2], [3]. However, SiR possesses low tensile and tear strength in its pure state, which restrains its performance as an outdoor insulation. To

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overcome this, SiR was required to be modified by filler addition and should undergoes proper vulcanization process to reinforce and increase their mechanical strength and at the same time enhancing its performance as High Voltage (HV) insulator [3], [4]. There are few commonly used fillers i.e., calcium carbonate, glass, alumina trihydrate, mica flake and kaolin [5]. Alumina Trihydrate (ATH) is one of the preferable filler that had always been used in reinforcement of SiR due to its chemically inert properties and fire retardant abilities [6], [7]. In fact, alumina trihydrate were also identified as a significant factor that contributes towards dielectric strength output response which highlights its importance as filler in polymeric materials [8]. However, despite of their contribution, it was also discovered that the filler is also capable in prompting unscrupulous effect to the electrical properties of SiR. The amount of reinforced filler added should be limited to certain concentration as excessive loadings will result in the appearance of crack and hole on the surface of prepared SiR based composites due to filler agglomeration factor, which weaken the matrix-filler interphase interaction [9]. Imprudent filler content will also increase the leakage current value and hence reduced the SiR function ability as an insulator for high performance applications [10].

In order to increase performance of SiR, it is also important to have certain amount of vinyl in SiR material. This is because, vinyl enhances peroxides vulcanization and thus yield a better crosslinking during the vulcanization process [11]. In fact, it was also mentioned previously that mixing SiR together with filler and different vinyl concentration allows the composites to have a better tensile strength [12]. Hence, the presence of vinyl can be considered as one of the key ingredients in improving SiR functions ability as HV insulator. Besides of filler and curing agent, the processing parameters is also one of the crucial details that need to be focused on while developing a polymeric blends even though there are scarcely any work done regarding the optimization of processing conditions. In both reference [13], [14] the importance of mixing parameters such as the mixing time and speed were underlined as proper mixing parameters allows for good dispersion of particles within polymeric blends. Miscibility of polymeric blends also prevents the occurrence of filler agglomeration and hence allows a lower susceptibility of polymer against electrical stress. An insulator should be able to demonstrate pronounced electrical properties such as a great surface resistivity. Study on surface resistivity had failed to gain serious attention from researchers in spite of its importance in evaluating the endurance level of SiR against leakage current which has been highlighted in [15]. Thus, high surface resistivity is particularly essential in insulator as it allows SiR to stand against both environmental and electrical stress. Moreover, a great surface resistivity does not only act as a shield that protects the surface of SiR against leakage current, but it also indicates that the SiR have a larger contact angle and hence a better hydrophobicity traits [6]. Hydrophobicity helps in protecting SiR against humidity and pollutions which also contributes in restricting the formation of conducting paths that will limits the leakage current on SiR [16]. Basically, an insulator with high surface resistance and low leakage current is preferable as it was proven that high leakage current enhances tracking and erosions [17].

On the other hand, permittivity (ε) is also one of parameter that can be used to characterize the performance of an insulator. Permittivity is obtained by determining the ratio of charge stored by a material while being sandwiched with two metallic plate. A higher ability of storing charge is represented by a higher value of permittivity which makes a good capacitance and hence insulator. Generally, for a normal insulator used in separating

electrical network and ground, it is preferable to have a lower permittivity consistent with acceptable mechanical, chemical and heat resisting materials. However, on the other hand, higher permittivity is also recommended so that the size of insulator can be reduced [18]. Reduction in insulator size is also beneficial as a smaller insulation will inflict a lesser load on the supporting structure and hence lower installation cost. Fundamentally, the permittivity of SiR depends on the amount of dielectric molecule inside its material and the overall polarizability where an increase in both will cause increase in permittivity of SiR [19]. Insulation materials with higher permittivity will also have a higher dielectric strength and thus higher breakdown voltage [20], [21]. Basically, in the permittivity test performed, the results will be obtained in terms of relative permittivity which can be directly analyzed or multiplied by vacuum permittivity to get value of permittivity (F/m).

Previously, the study done on SiR in [17], [22] had specifically conducted to determine the right amount of filler that should be added into SiR but there is barely any work that focus on studying the optimization of SiR based on both filler and vulcanizing agent. Besides, the trend of performing optimization are currently overwhelmed even in other field of studies [23], [24]. Hence, this paper provides emphasis on optimizing the SiR performance in both surface resistivity and relative permittivity through Design of Experiment (DOE) approach of two level factorial strategies using the latest *Design Expert Software* (v10). This involves the variation of independent variables with four important factor (2^4) which are the amount of ATH as filler, the amount of Dicumyl Peroxide (DCP) as vulcanizing agent, the mixing speed and mixing time of mixer used during the SiR filled ATH composites preparations. The relations between the four factors and the prepared SiR blends will be analyzed through permittivity and surface resistivity responses. Through this study, the factor that affects the optimization of SiR in both surface resistivity and relative permittivity will be revealed and most importantly, the best combination of mixtures that will give out the best sample with highest surface resistivity and relative permittivity will be deduced. In addition, this study also contributes to the understanding of the correlations between all four factors which further emphasize the connection between the ATH filler and DCP concentration with mixing processing parameters.

2. Samples Preparation

2.1 Materials

The main ingredients used in producing SiR blends in this study are, HTV SiR, heat stabilizer, Alumina Trihydrate (ATH) and Dicumyl Peroxide (DCP). The HTV SiR (Elastosil 401) and Aux Heat Stabilizer are both obtained from Immortal Greens Industrial Sdn Bhd (Malaysia). The filler used was ATH as purchased from HmbG Chemicals (Germany), while the curing agent used was DCP. All materials were used as is without further purification.

2.2 Samples preparation

Firstly, the range of concentration and parameters to be used for each factors is decided as listed below. The decision made was based on preliminary trial run, machine capability, screening test, manufacturer recommendation and after careful literature searches.

    i.   ATH concentration   : 10 – 50 *pphr*
   ii.   DCP concentration   : 0.50 – 1.50 *pphr*
  iii.   Mixing Speed   : 40 – 70 *rpm*
  iv.   Mixing Time   : 5 – 10 *mins*

Then the level for each concentration of each factors were decided by dividing it to three types of concentration that is lowest concentration, medium concentration and highest concentration as shown in Table 1. The value of ATH was chosen based on reference [22] which found that 30 wt.% of micron sized ATH filler were induced higher resistance abilities during dry band discharge with lesser loss in weight. Then, the DCP values was varied through the recommended value given in the guideline provided by the manufacturer [25]. The mixing parameters were decided after considering the machine capabilities and related past literature that are using a similar set-up [13], [14], [26].

Table 1: The Level used in SiR Blends Preparation

ATH content (A: *pphr*)	DCP Content (B: *pphr*)	Mixing Speed (C: *rpm*)	Mixing Time (D: *mins*)
10 (-1)	0.50 (-1)	40 (-1)	5 (-1)
30 (0)	1.00 (0)	55 (0)	7.5 (0)
50 (+1)	1.50 (+1)	70 (+1)	10 (+1)

Table 2 : SiR samples with different levels of concentration for 2^4 factorial design

Samples	ATH	DCP	Mixing Speed	Mixing Time
1	-1	+1	+1	-1
2	-1	+1	-1	-1
3	+1	-1	+1	-1
4	+1	+1	+1	+1
5	+1	-1	+1	+1
6	-1	-1	-1	-1
7	-1	+1	-1	+1
8	-1	-1	+1	-1
9	-1	-1	-1	+1
10	-1	-1	+1	+1
11	+1	+1	+1	-1
12	+1	-1	-1	+1
13	-1	+1	+1	+1
14	+1	+1	-1	-1
15	+1	+1	-1	+1
16	+1	-1	-1	-1
17	0	0	0	0
18	0	0	0	0
19	0	0	0	0

Then, by using the specified factor range and 2^4 factorial DOE strategies, the experimental method for all nineteen samples were created by using the *Design Expert Software* (v10) as listed in the following Table 2.

The SiR, ATH filler, heat stabilizer and vulcanizer were weighted accordingly before being blended using *Haake Polylab OS Rheo Drive 16* internal mixture. Firstly, to produce a SiR sheet of 2mm, the blends of each sample need to be weighted roughly around 30 grams before being placed into a mold of 100 mm x 100 mm x 2 mm. It should be noted that the SiR blend need to be placed in between a couple of *mylar* sheets before being further sandwich in between a pair of metal plates to allow an easy release of SiR sheet from the mold after the vulcanization. Then, the samples together with mold is hot pressed for vulcanization with a compression molding machine under a temperature of 170°C and pressure of 50 bar for a period of 10 mins. Lastly, each samples is taken out from the mold to be further post-cured in forced air oven for 12 hrs under 130°C [27].

2.3 Surface resistivity

Surface resistivity is obtained by measuring the resistance of insulating material against current leakage along the surface which was performed using Pico ammeter. The measurement of surface resistivity is done along the surface of materials and is denoted by the unit of Ohm/sq, [18], [28]. A higher value of surface resistivity indicates a better insulation property of materials. The measurement is done using Monroe Portable Surface Resistivity (Model 272A) which follows the ASTM Standard D 257. The tests were conducted to all 19 samples for a minute under room temperature with varying humidity range from 60 - 70%.

2.4 Relative permittivity

In this experiment, the relative permittivity of all samples were measured using a Vector Network Analyzer (VNA) under frequency range of 0.50 GHz until 5 GHz. The value taken to be used in the work is the value of relative permittivity obtained at the frequency of 3GHz.

3. Result and Discussion

3.1. Graph obtained from Vector Network Analyzer

The relative permittivity versus frequency range plots for several selected samples were presented as in Figure 1. This test was conducted under the frequency of 0.50 – 5.00 GHz.
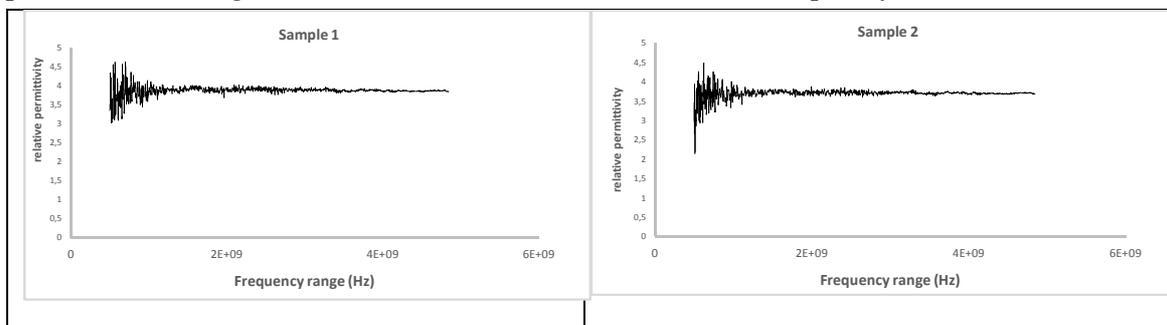
Figure 1: Relative permittivity versus frequency range plots for sample 1 and sample 2

Through the analysis, it could be seen that the result of relative permittivity for all 19 samples shows instability at lower frequency which in this case falls in the range of 0.5 to 1.3 GHz. The permittivity values are waving significantly at first but started to settle down at frequency higher than 1.3 GHz. Previously, in reference [29], the same unusual behaviour regarding permittivity and frequency were highlighted. It was stated that permittivity value does not shows significant changes under high frequency but fluctuates at low frequency. To be specific, it was found in recently published publication [30], the peculiar increment of permittivity value in low frequency occurs at frequency below 10 Hz especially in low filled and unfilled SiR. It was further explained that the increase phenomena was due to the Maxwell-Wagner type polarization. Basically, the Maxwell-Wagner polarization is the charge build up that occurs in dielectric material that are inhomogeneous or at interface of materials. The materials will show dependency on frequency and hence effect the permittivity reading of the materials at lower frequency. It was explained in [31] that lower frequency induced higher permittivity due to the mechanism of charge transport under thermal and electrical constrains.

To explain more regarding this interesting phenomenon, depolarization theory is referred. Permittivity could be complex to be explained if the matter is in the solid state. Basically solid matter such as SiR blends which had been further modified with added filler and curing agent will have a different polarization theory as the electric field applied is no more equal to the electric field in the local material, as stated in Equation 1. The local field of material is the combination of both macroscopic and internal electrical field. The macroscopic consist of external field and the depolarization that occurs on dielectric surface due to generated charges while the internal electric field is gained from the reaction of dipoles in immediate surroundings within the materials. The internal electrical field will also affect the permittivity of the materials as now it involves molecules. Normally, the permittivity of solid matter is obtain through addition of both ionic and electron polarization as shown in Equation 2. Now, when the dielectric is exposed under frequency, its permittivity varies due to the ionic polarization which is not counted in high frequency due to incapability of ionic bond to cope with frequency variation at higher frequency. As for material which consist of only covalent bond like polymeric SiR, not much different is observed for the permittivity in high frequency compared to those in lower frequency as the permittivity is totally based on electronic polarization [19], [32], [33].

It was mentioned in [34] that a polar covalent bond can also occupy some ionic character and so do an ionic bond which can also acquire a covalent character. It can be said that in this case, composites of SiR is not stable at lower frequency (0.3-1.3 GHz) and hence, for modelling purposes, it is preferred to choose a value of relative permittivity which fallen into medium range, to be used in the simulation process.

$$E_{applied} \neq E_{local} \tag{1}$$

ε_r is the relative permittivity, ε_0 is vacuum permittivity, N is the number of dielectric molecules per unit volume and α is the total polarizability.

$$P = P_i + P_e \tag{2}$$

P represent the total dielectric polarization, P_i is the ionic polarization and P_e is the electron polarization.

3.2 Result of both Surface Resistivity and Relative Permittivity

The results of surface resistivity and relative permittivity for all 19 samples are summarized as in Table 3.

Table 3 : The result of surface resistivity and permittivity for different SiR samples

Sample	Surface Resistivity(Ω/sq)	Relative Permittivity
1	1.2 x10^14	3.8496
2	1.0 x10^14	3.6988
3	1.5 x10^14	3.7360
4	1.6 x10^14	4.1377
5	2.0 x10^14	4.0499
6	1.0 x10^14	3.8967
7	1.2 x10^14	3.4898
8	1.2 x10^14	4.0095
9	1.1 x10^14	4.1229
10	1.2 x10^14	3.7753
11	1.6 x10^14	4.1936
12	1.5 x10^14	3.9370
13	1.2 x10^14	3.7573
14	1.4 x10^14	3.7657
15	1.6 x10^14	3.7129
16	1.1 x10^14	3.9722
17	1.5 x10^14	3.8183
18	1.5 x10^14	3.8599
19	1.4 x10^14	3.9402

3.3 Evaluation on Surface Resistivity of SiR Blends

The result of surface resistivity shows that the highest surface resistivity was 2.0 x10^14 Ω/sq possessed by samples 5 and the lowest was 1.0 x10^14 Ω/sq which was gained by sample 2. Samples 5 contains the highest ATH filler and maximum processing parameters of 70 *rpm* and 10 *mins* time but with the lowest DCP content of 0.50 *pphr*. The sample 2 was produced using the highest DCP content at 1.50 *pphr* and minimum ATH filler addition and processing parameters. Through the optimization performed and as reported in the previous conference proceeding [35], it was proven that the surface resistivity was much influenced by the mixing speed and ATH content. The most optimum surface resistivity of 1.92039 x 10^14 could be obtained if the SiR is mixed with the highest ATH loadings at 50 *pphr*, lowest DCP content at 0.50 *pphr*, highest mixing speed at 70 *rpm* and the highest mixing time at 10 *mins*. This combination has the highest desirability level of 0.959. The interaction between three factors of ATH loadings, DCP content and the mixing speed with constant mixing time of 10 *mins* is represented in cubical form as available in the Figure 2. Overall, it can be deduced that the surface resistivity of SiR depends on both ATH content and mixing parameters [35].
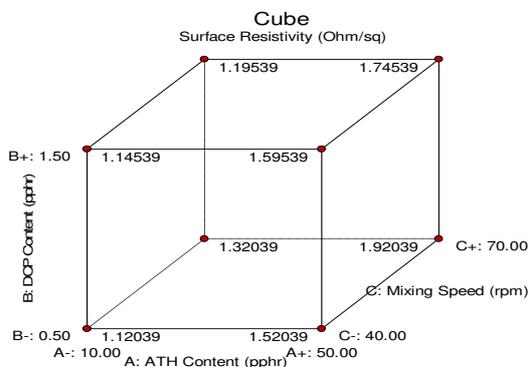
Figure 2: Cubical interaction of surface resistivity dependency on variables factors

3.4 Evaluation on Relative Permittivity of SiR Blends

The highest relative permittivity response was recorded by sample 11 with response value of 4.1936 while the lowest was recorded by sample 7 with response value of 3.4858. Roughly, the sample with higher surface resistivity could also have the higher relative permittivity excepts for few samples that shows an exceptional values such as for sample 1, sample 8, sample 9 and sample 16. The analysis were performed by using two level factorial DOE method in order to decide the inter-correlation and level of contribution of each and multiples factor in determining the relative permittivity value. It should be noted that the dielectric constant or permittivity are swayed by many factors such as frequency, temperature or even the chemical composition of a dielectric sample [36]. The frequency factor was solved by taking the relative permittivity value in medium range, the temperature is also maintained as the experiment was done in room temperature and hence only the chemical composition of the SiR is left to affect the performance of measured surface permittivity response. The chemical composition of SiR is affected by its mixing parameters and the concentration of ATH filler and DCP added. SiR blends with higher relative permittivity value is required as higher permittivity could help in stress reduction for SiR composite which ensure a longer life span if it is used for HV applications [37].

3.5 The Analysis on Percentage of Contribution

The analysis on effect lists of four factors towards relative permittivity of all 19 samples of SiR were first analysed by determining their percentage of contribution as in Table 4.

Table 4 : Effect list of each factor on relative permittivity of SiR blends

Term	Studentized Effect	Sum of Squares	(%) Contribution
A	0.11	0.051	8.18
B	-0.11	0.050	7.99
C	0.11	0.052	8.33
D	-0.017	1.213E-003	0.19
AC	0.068	0.019	2.98
AB	0.14	0.079	12.61
AD	0.060	0.014	2.29
BC	0.20	0.17	26.51
BD	-0.085	0.029	4.63
ABCD	-0.12	0.054	8.63

By focusing on the percentage of contribution, the highest contribution was recorded by the BC interaction with 26.51% while the second highest was achieved by AB interaction with 12.61% of contribution. AB interaction term is referring to the interaction between ATH loading and DCP content while the BC interaction refers to the interaction between DCP content and mixing speed. This findings emphasize the major role of DCP content in effecting the relative permittivity response which was amplified when it interacted with A and C independent factors. The single factor of A, B, C and D which referring to the ATH loadings, DCP content, mixing speed and mixing period, were recorded a contribution of 8.18%, 7.99%, 8.33% and 0.19%, respectively. Overall contribution of all the four factors interaction (ABCD) was relatively low but still noticeable with a value of 8.63. The overall ABCD interaction terms are also indicates a significant contribution, which means that the

correlation of all four factors does contributes in affecting the relative permittivity response of produced SiR blends.

3.6 The Analysis of Variance (ANOVA) of Relative Permittivity Response

Next ANOVA was used for the response analysis of relative permittivity and the results were shown in Table 5. The ANOVA results highlight the significant contributor in this selected model. In order to be categorized as significant, the *p-value* obtained by the factor must be ≤ 0.05 and if the *p-value* obtained is higher than 0.100, it is categorized as non-significant. The overall model was classified as significant with *p-value* of 0.0375, representing the accuracy of the selected model. Through the result, it can be seen that AB and BC recorded the lowest *p-value*. The *p-value* obtained by AB (0.0438) and BC (0.0085) represents their significant contribution in affecting the relative permittivity response of SiR blends. The higher significant value is recorded between AB model term represents the relationship between ATH filler and DCP content. Prior analysis on surface resistivity response had found that the relationship between A and B model terms are significant.

Fundamentally, for relative permittivity evaluation, the entire bulk specimen is practically involved compared to surface resistivity which only covers the specimen's surface. Obviously, relation between DCP content and ATH filler loading is important as both filler and cross-linking agent helps to reinforce the produced SiR blends [11]. The correlation of both ATH filler and DCP content are important in ensuring a good bonding between the particles and SiR matrices, while BC interaction represents the correlation between DCP and the mixing speed. The DCP used is a peroxide cross-linker which was in the form of solid-crystal. Hence, by having a higher mixing speed allows a proper dispersion of DCP within the polymer matrix and yielded a proper and homogeneous crosslinking of SiR. It was further mentioned in [38] that to formulate a good resistance of tracking and erosion of SiR blends, it involves a balance between high enough filler content and its proper dispersion, including good bonding characteristic between the particle and the matrix. To further emphasis, a good bonding is unachievable without proper mixing and could be impossible without an efficient vulcanizing agent. Thus the correlation between both AB and BC is particularly important in influencing the permittivity of produced SiR blends.

The A, B, C terms and ABCD interaction term shows a *p-value* smaller than ≤ 0.100 with 0.0903, 0.0937, 0.0881 and 0.0834 respectively, which indicates that it is not categorized as insignificant. Thus, it can be said that individually, the factor of ATH, DCP and mixing speed also contributes in affecting much to the relative permittivity function of SiR blends. The 0.0834 value obtained by ABCD interaction terms highlights that the four factor do somehow correlates with each other and the relationships between all four factors are important in influencing the relative permittivity value of SiR blends.

In addition, the ANOVA done on the model also gives out R^2 value of 0.8234 which highlighted that the model chosen has the higher reliability percentage of 82.34 %. This high value of R^2 proves that all four factors and inter-correlation between them are attributed towards affecting the relative permittivity response of produced SiR blends.

Table 5 : ANOVA of SiR blends that shows the *p-value* of each and in between factor

Source	Sum of Squares	Mean Square	F-Value	p-value
Model	0.52	0.052	3.75	0.0373
A-ATH Content	0.051	0.051	3.73	0.0903
B-DCP Content	0.050	0.050	3.64	0.0937
C-Mixing Speed	0.052	0.052	3.79	0.0881
D-Mixing Time	1.213E-003	1.213E-003	0.088	0.7745
AB	0.079	0.079	5.75	0.0438
AC	0.019	0.019	1.36	0.2790
AD	0.014	0.014	1.05	0.3377
BC	0.17	0.17	12.08	0.0085
BD	0.029	0.029	2.11	0.1856
ABCD	0.054	0.054	3.93	0.0834
Residual	0.096	0.014		
Lack of Fit	0.040	7.992E-003	0.28	0.8885
Pure Error	0.056	0.028		

3.7 Regression Equation of Relative Permittivity Response

Besides, the model can also be represented with final regression equation in terms of coded factor as shown in the following Equation 3.

$$
\begin{aligned}
Relative\ Permittivity = \ &+3.87 \ + 0.057 * A - 0.056 * B + \ 0.057 * C - 8.706E \quad (3)\\
&-003 * D \ + \ 0.070 * AB \ + \ 0.034 * AC \ + \ 0.030 * A\\
&+ 0.10 * BC \ - \ 0.043 * BD - 0.058 * \ ABCD
\end{aligned}
$$

The derived regression equation can be used in making prediction about the response studied. Basically, high level of factors are coded as +1 which alternatively underline the high relative impact of the factors while the -1 highlights the low levels with low relative impact of factor by comparing the factor coefficient. The high relative impact factors are A, C, AB, AC, AD and BC while factor B, D and their interaction terms had low relative impact towards the relative permittivity response. For the single factor, it can be deduced that the factor of ATH filler loading and mixing speed are both important to improve the response studied while addition of DCP content should be minimized with proper control of their mixing period. This situation relates with the importance of having better dispersion of ATH filler while ensuring efficient peroxide vulcanization by having minimum content of DCP added.

3.8 Response Surfaces Plots Evaluation using 3D Graph for Relative Permittivity

The 3D response surface plots is used to illustrate the relationships between two factor in affecting the relative permittivity value of SiR blends samples. The first response surface plots are shown in Figure 3 describing the relationship between variable factor A (ATH filler loading) and factor B (DCP content) and their contribution towards the relative permittivity values. The two factors are plotted against relative permittivity with another two factor of C and D are being maintained on medium speed of 55 *rpm* and mixing time at 7.5 *mins* as shown in Figure 3. The plot shows that the higher the ATH content, the higher the relative permittivity response while as for the vulcanizer an increase in the DCP content would lowering the relative permittivity results. The lowest relative permittivity value recorded was at the intersection of highest DCP content and lowest ATH filler loading. Thus, it can be said that besides of enhancing the mechanical property and tracking resistance of SiR [39], [40] the ATH filler loadings are also proven able to improve the relative permittivity of SiR blends.

However, the DCP peroxide type vulcanization agent are found to reduce the relative permittivity of SiR blends with every increase of its content even though the main purpose of curing agent was actually to improve SiR strength by inducing the crosslinking formation. In fact, it was also stated that the crosslinking improves the mechanical property of SiR such as its ability in withstanding a wider range of temperature changes [41]. However, too much vulcanizer added is detrimental for the SiR rubber blends since excessive amount of DCP used in SiR blends might had allowed higher amount of untreated peroxide to be present in the samples. It was explained in [42] that unreacted peroxide had caused significant dropped in the fire retardant ability of SiR blends. In addition, it was also mentioned that thermal stability of SiR is exceptionally sensitive towards impurities such as residual curing agent even if it is only present in a very small quantity. Thus, it is also possible for the excess vulcanizer to affect the performance of SiR in terms of their relative permittivity values. Besides, peroxides based vulcanizer also had another weakness of introducing by-product into SiR blends during cross-linking which is usually being eliminated out during the post-curing period. There are three possibilities of by-product that will be produced from DCP, that is acetophenone, 2-phenylispropanol and methane [43]. Too much DCP is perilous as it might cause the establishment of higher by-product leftover in the SiR samples even after the post-curing process is performed. In another paper [44], it was discovered that the by-product of peroxide in LDPE support carrier transport which promotes conductivity and hence reduced the relative permittivity of produced samples. Perhaps, the same thing is happening here in SiR where the by-product is taking its toll on SiR and had caused the sample with excess DCP to have lower relative permittivity results.

Nevertheless, there are still a slight increase observed in the relative permittivity values with every increase of DCP content but it is only achievable at higher ATH filler loading at 50 *pphr*. This denotes that there is an important connection between DCP content and ATH filler loading and perhaps the amount of DCP used should increase slightly as the amount of ATH loadings increases as to match the reaction between them. This reaction between AB is already highlighted that resulted higher percentage of contribution and *p-value*.
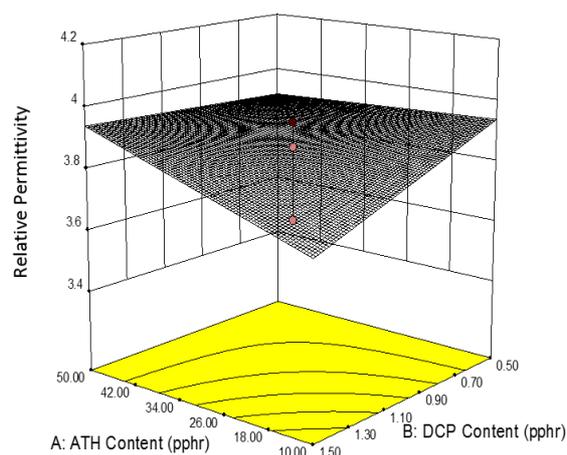
Figure 3: 3D surface plot representing the interaction between factor A and B towards the relative permittivity response

The analysis on relationship between factor B (DCP content) and factor C (mixing speed) towards the relative permittivity response is shown as in the Figure 4 for both top and side view of the interaction. Previously, the interaction term of BC also shows an amazingly higher contribution and a significant *p-value* which highlights its contribution in affecting the response. Through the plot in Figure 4, it can be seen that by maintaining the ATH and

mixing time, an increase in DCP content will cause a decrease in relative permittivity value while increase in mixing speed will consequently increase the relative permittivity. However, if the DCP is at higher concentration (1.5 *pphr*), an increase in mixing speed would be followed by increase in relative permittivity. This scenario underlined the importance of matching a proper concentration with a proper mixing speed during blends. Supposedly, a mixture consisting of higher concentration should be mixed faster so it can be well dispersed. A proper dispersion prevents any accumulation of substances. In reference [14], the case of mixing ethylene propylene diene rubber (EPDM) with natural rubber shows that the lower solubility of vulcanizer in EPDM phase had reduced the tendency of crosslinking formation which subsequently contributes to uneven distribution of vulcanizer and hence immiscibility in the polymer blends. Besides, it was stated that crosslinking may affect the electrical characteristic of blends material due to the physical changes induced and also due to by-product residual which accumulated in the materials [45]. The solid-crystal form of DCP used in the experiment is unlikely to be soluble and will only start to melt at 39°C while the mixing of SiR blends were conducted in room temperature condition. Thus, there is actually a higher probability of DCP vulcanizer to face difficulties in being properly dispersed if the mixing speed is inappropriate especially when the amount of DCP used is higher. The importance of having a properly mixed polymer blends is also emphasized by other researchers in [46] which had introduced electrospinning method to allow a proper dispersion of polymeric blends.

The importance of having appropriate mixing speed in accordance to ATH and DCP is further discussed based on both Figure 4 and Figure 5. The 3D plot of 10 *pphr* as shown in Figure 4, displayed the highest permittivity value at intersection of 0.5 *pphr* of DCP and 40 *rpm* of the mixing speed. This shows that at lower ATH content, a lower DCP and mixing speed is sufficient in producing a SiR with good permittivity value. Adding too much DCP or having too high mixing speed would not be beneficial in mixing SiR with low filler content, in fact, it may contributes to a waste of source. On the other hand, as for higher ATH content, it was vice versa. The 3D plot for high concentration of DCP (50 *pphr*) is shown in Figure 5. The higher ATH concentration requires a higher matching of DCP content and higher mixing speed in order to gain higher permittivity result in produced SiR blends. Having less DCP and lower mixing speed causes the permittivity value to reduce drastically.

However, the highest dielectric constant was recorded by the sample with the highest ATH content of 50 *pphr* compared to those of 10 *pphr* which emphasizes the importance of ATH concentration in increasing the dielectric constant of SiR blends. Nonetheless, if the mixing period is increased, from 7.5 to 10 *mins* as shown in Figure 6, there is an increment in the value of relative permittivity which occurs at the intersection of highest DCP content at 1.50 *pphr* and the lowest mixing speed at 40 *rpm*, in which the previous value was denoted by the red dotes whereby the difference in the highest and lowest DCP content is almost constant. This means that, a longer period of mixing had enabled a proper compounding to be achieved even if the DCP concentration used is higher.
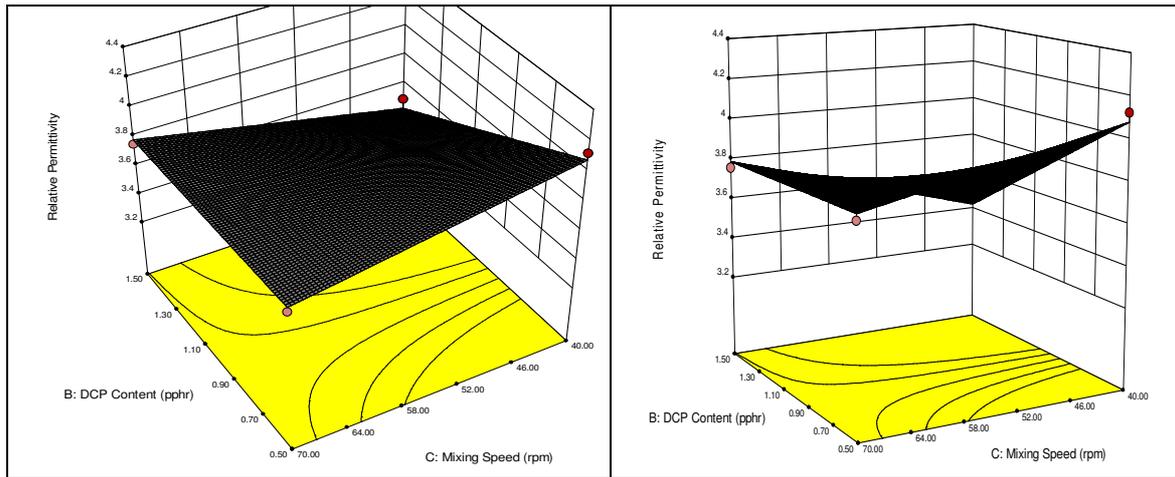
Figure 4 : 3D response surface plot showing the interaction between factor B and C against relative permittivity with 10 *pphr* of ATH filler loading
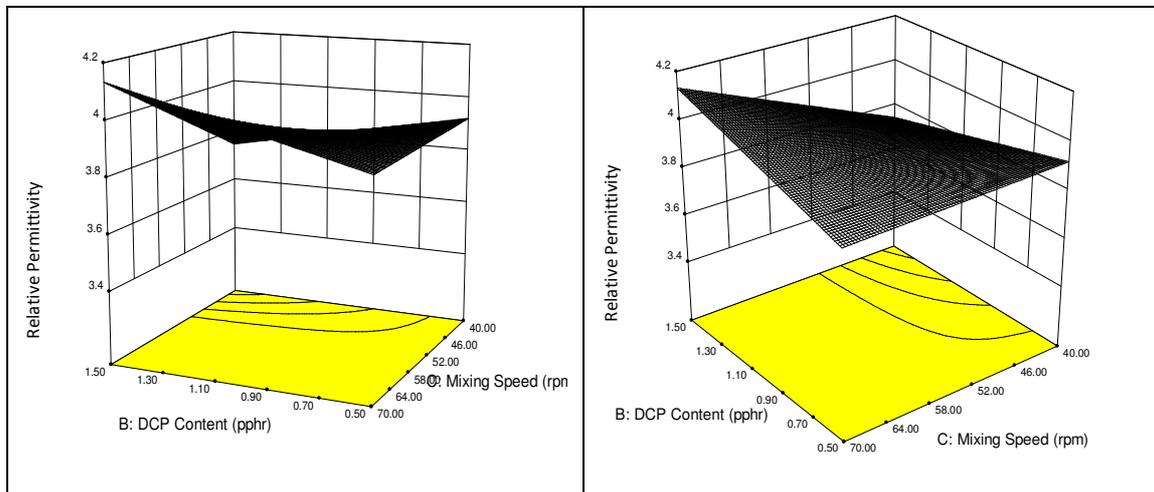
Figure 5 : 3D surface plot for the interaction between factor B and C against relative permittivity with the highest ATH filler loading of 50 *pphr*
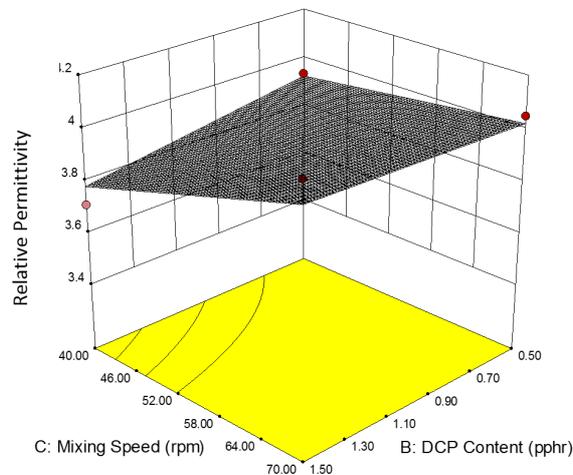
Figure 6: 3D surface plot for the interaction between factor B and C against the relative permittivity response with the highest ATH filler content and mixing period

4. Optimization of SiR Blends

The discussion on SiR blends regarding both responses of surface resistivity and relative permittivity had been done separately at the previous section. However, in this part, the optimization of materials was performed using both parameters of surface resistivity and relative permittivity. It means that the optimized blends that are having maximum surface resistivity and relative permittivity output was proposed together with their suggested combination of variable factors. Prior to the optimization step, all the factors were set to be in-range while the surface resistivity and permittivity is towards maximum target. The summary of an optimization strategy was depicted as in Figure 7 where it can be seen that the ATH filler loading was purposed to be added as at the highest amount of 50 *pphr*, while the curing agent of DCP content was suggested at minimal with the amount of 0.50 *pphr* and mixing speed and mixing time factor were recommended as at the highest value of 70 *rpm* and 10 *mins*, respectively.

Based on the optimization results, it was suggested that the parameters of ABCD could yielded an optimum value of surface resistivity with 1.92039 x 10^14 Ω/sq and relative permittivity of 4.0231. Through the optimization the overall desirability obtained is 0.835 or 83.5% which is relatively higher. Thus, it can be proposed that increase of surface resistivity will occurs simultaneously with the increase in their relative permittivity at higher frequency region, particularly in between of 1.3 GHz and 5 GHz. Earlier, it was also known that there are several factors that might affects the current conductivity of polymer that is the molecular weight, density, distribution, crystallinity and its morphology. Normally, electrical conductivity will decrease as the molecular weight of polymer were increases. Hence, the polymer that had been cured and being added with filler will practically have higher molecular weight and thus, resulting a lower conductivity output. In addition, increment of the intermolecular forces and viscosity of materials as the consequence of the increment in their molecular weight had causes the increase of surface resistivity output [47]. Overall, it can be deduced that the highest ATH filler loading, mixing speed, mixing time and the lowest DCP content will results in the highest surface resistivity and relative permittivity of SiR blends samples. The optimization output was further represented using the cubic form representation as shown in Figure 8, whereby an overall reaction between ATH filler loading, DCP content, the mixing speed and mixing period for both surface resistivity and relative permittivity responses are shown.
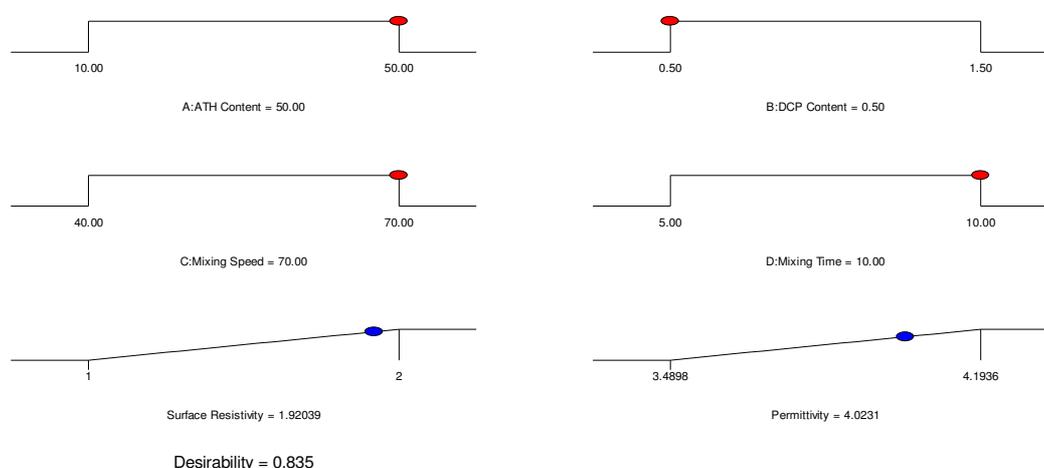
Figure 7 : The optimization strategy of SiR blends preparation for both the independent and dependant variables
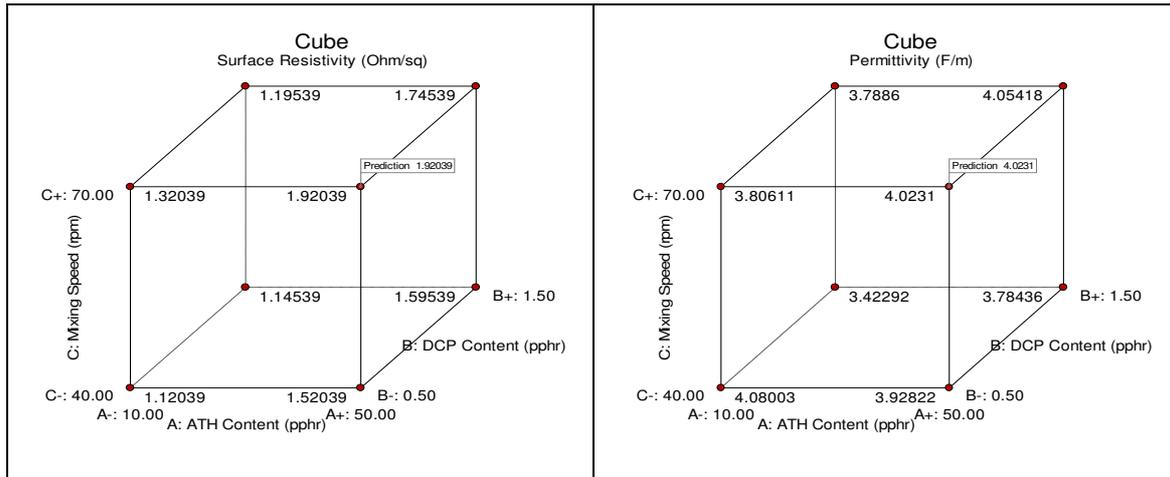
Figure 8: Overview of optimization in the form of cube that involves all four factors

5. Conclusion

Optimization on surface resistivity and relative permittivity responses was performed in this work by using the Design Expert Software of two-level full factorial design method. The design had analysed the effect of varying four factors which consist of ATH filler loading, DCP curing agent and mixing parameters comprising of mixing speed and mixing time. The analysis also provide an optimum value that is predicted to gives out the highest surface resistivity and relative permittivity with value of $1.02039 \times 10^{14}\Omega/sq$ and 4.0231, respectively. The suggested optimized factors are ATH: 50 *pphr*, DCP: 0.50 *pphr*, mixing speed: 70 *rpm* and mixing time: 10 *mins* with an overall desirability of 0.835. All in all, it is concluded that both responses of surface resistivity and relative permittivity are not only affected by the filler and vulcanizer added but in fact, it was also affected by the processing parameters such as mixing speed and compounding period. A combination of correct processing parameters with suitable content of both filler and vulcanizing agent are important in producing better quality of SiR blends insulator for HV applications.

Acknowledgment

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