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DIELECTRIC STRENGTH IMPROVEMENT OF NATURAL ESTER INSULATION OIL: OPTIMIZATION OF MIXED ANTIOXIDANTS VIA TWO-LEVEL FACTORIAL DESIGN

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ABSTRACT

Recent studies have shown that the addition of selected antioxidants into mineral oils or natural ester insulation (NEI) oils have improves the AC breakdown voltage (BdV), viscosity, flash point and fire point of these oils. The results of previous studies show that the addition of propyl gallate (PG) and citric acid (CA) antioxidants increases the AC BdV of NEI oils. However, previous researchers have implemented one-factor-at-a-time (OFAT) method as their experimental design approach and they overlooked the possibility through combination of antioxidants at optimum ratios that will yield a better result. In addition, a large number of test runs are required to investigate the effect of these antioxidant mixtures on the performance of insulation oils. Hence, in this study, two-level (2^k) factorial design of experiments is used to determine the optimum concentration of PG and CA which will maximize the AC BdV of NEI oil. The results show that the optimum concentration of PG and CA that yields the highest AC BdV of NEI oils is 0.05 and 0.25 wt.%, respectively. A regression model is also developed to predict the AC BdV of NEI oils as a function of PG and CA concentrations.

Keywords: insulation oil, antioxidants, optimization, design of experiments, dielectric strength, natural ester.

1. INTRODUCTION

Insulation oils are dielectric liquids which are able to insulate power equipment such as cables, capacitors, bushings, circuit breakers, tap changers and transformers against electrical stresses. Insulation oils are specially formulated for use in transformers and they are generally used for the following purposes: (1) as a heat transfer medium (*i.e.* the oil is circulated to regulate heat throughout the transformer), (2) as an electrical insulator (*i.e.* the oil provides protection against turn-turn winding faults) and (3) as a health monitoring agent (*i.e.* the oil is used to assess the condition of the transformer) [1].Liquid-immersed transformers are typically filled with mineral insulation oils. These oils have been in use for more than a hundred years since they are widely available in the market and moreover, they are inexpensive and they possess good thermal and electrical insulation properties [2]. However, much effort is being made nowadays to replace mineral insulation oils with products that are biodegradable and environmentally friendly due to environmental issues associated with these oils [3, 4]. The availability of natural esters derived from vegetable oils in recent years has made them a promising alternative to replace conventional mineral insulation oils for transformers. Many studies are being carried out to determine the feasibility of natural ester insulation (NEI) oils for both power and distribution transformers [5–7]. This is indeed unsurprising since NEI oils are highly biodegradable. In addition, NEI oils have a higher fire point as well as good dielectric properties. NEI oils can also help extend the lifespan of transformers because of their superior hydrophobic properties compared to mineral insulation oils [8, 9].

However, NEI oils are not without drawbacks. Firstly, NEI oils have lower pour point such that they lose their flow characteristics at low temperatures, which make them less suitable for use in cold climates [10]. In addition, these oils are prone to oxidation due to their low oxidation stability [11]. Researchers are now focused on improving the properties of NEI oils for use in transformers. In order to improve the oxidation stability of NEI oils, antioxidants are added into the oils to prevent the oils from further oxidation. The antioxidant which seems to be preferred choice for transformer oils since the early 1970s is 2,6-ditert-butyl-4-methylphenol (DBPC) [12]. Wilhelm *et al*. [13], [14] and Xu *etal*. [15] also added this antioxidant into NEI oils at a concentration of 0.3 wt.%. Abdelmalik *et al*. [16] formulated an improved version of NEI oil, whereby palm kernel oil alkyl ester (PKOAE) was mixed with 3 wt.% tertiary butyl hydroxyquinone (TBHQ). In addition, Raymon *et al*. [17] and Thanigaiselvan *et al*. [18] also used various combinations of antioxidants in order to enhance the properties of NEI oils. However, it shall be noted that the studies were only focused on evaluating the properties of NEI oils such as the breakdown voltage, viscosity, fire point and flash point whereby the oils were mixed with antioxidants using the following mass ratios in grams: 0.5:0.5, 1:1, 1.25:1.25 and 2.5:2.5. These studies are in line with the recommendations given in the Cigre WG D1.30 report [19], which emphasizes on improving the oxidation stability of mineral insulation oils and NEI oils using the standard practice of lubricant technology. According to the

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report, one should consider adding more than two antioxidants into the insulation oils in order to exploit their favorable properties.

In this study, propyl gallate (PG) and citric acid (CA) antioxidants are added into NEI oil in order to improve the oxidation stability of the insulation oil. The AC breakdown voltage of the NEI oil is assessed to determine the effect of these antioxidants on the oxidation stability of the oil. The PG and CA antioxidants are chosen for this study since they have been proven to improve the AC breakdown voltage, viscosity, fire point and flash point of both NEI and mineral insulation oils [20]. However, the optimum concentrations of these antioxidants which will maximize the AC breakdown voltage of the NEI oil is not known and therefore, they are determined using two-level factorial design of experiments. This technique is suitable for multi-factor experiments and it is capable of determining the optimum combination of factors with a fewer number of samples. In addition, this technique is inexpensive and less time-consuming [21-23]. The results obtained from this technique are then verified using analysis of variance (ANOVA). A regression model is then developed in order to predict the AC breakdown voltage of the NEI oil as a function of the PG and CA concentrations and adequacy of the model is verified using ANOVA.

2. METHODOLOGY

i. Sample preparation

The PG and CA antioxidants were purchased from Sigma-Aldrich. The oils samples were prepared by mixing these antioxidants with rapeseed-based NEI oil within a concentration range of 0.05 to 0.25 wt.%. The concentration of each antioxidant was chosen such that, so the total concentration of both antioxidants in the mixtures was in range of 0.3 to 0.4 wt.% [18-19]. The oil samples were uniformly dispersed using a magnetic stirrer integrated with a hot plate at a stirring speed of 750 rpm. The temperature of the hot plate was set according to the melting point of each antioxidant [7-8, 11].

ii. AC Breakdown voltage test

The AC breakdown voltage was measured using Megger OTS60-PB portable oil tester according to the ASTM D1816 standard test method. The electrode configuration was in accordance with the VDE 0370 specification published by Verband der Elektrotechnik with a gap distance of 1.0 mm. The voltage was increased gradually at a rate of 0.5 kV/s. Five breakdown voltage tests were performed for each sample. Mean breakdown voltage is used to quantify the dielectric strength of sample.

iii. Design of experiments

Two-level (2^k) factorial design of experiments was carried out using the Design Expert software version 10.0 (Stat-Ease, Inc., Minneapolis, USA) in order to screen a significance effect between main and interaction of antioxidants towards improvement of AC breakdown

voltage in NEI oil. Following this, the optimization process was carried out using a response surface model which can determine the optimum concentrations of PG and CA which will maximize the AC breakdown voltage of the NEI oil.

iv. Screening process

The 2^2 factorial design with two independent variables and three replications at the center points was used to screen the factors which will have a significant effect on the AC breakdown voltage of the NEI oil. Table 1 show the level of the PG and CA variables, whereby low, medium and high is indicated by -1, 0 and +1, respectively. The 2^2 factorial design matrix used to screen the factors is shown in Table 2, and it can be seen that there are seven test runs. The AC breakdown voltage tests were carried out according to the 2^2 factorial design matrix. The effects of the PG and CA antioxidants (factor 1 and factor 2, respectively) on the AC breakdown voltage of the NEI oil were determined using a half-normal graph and effect list.

Table-1. Level of variables.

Factor 1 Propyl gallate (PG)A: PG (wt.%) Type: Numeric	Factor 2 Citric acid (CA)B: CA (wt.%) Type: Numeric
0.05 (-1)	0.05 (-1)
0.15 (0)	0.15 (0)
0.25 (+1)	0.25 (+1)

Table-2. Factorial design matrix used to screen the factors.

Experiment (Test run)	Variable code	
	A: PG (wt.%)	B: CA (wt.%)
1	-1	+1
2	+1	-1
3	0	0
4	0	0
5	+1	+1
6	0	0
7	-1	-1

v. Optimizationprocess

A regression model was then developed to predict the AC breakdown voltage of NEI oils as a function of the PG and CA concentrations. Analysis of variance (ANOVA) was used to determine the adequacy and statistical significance of the regression model. The results obtained from the 2^2 factorial design of experiments were used to fit the regression model. The regression model is composed of a list of coefficients multiplied by its associated factor levels. The regression model is given by the (1):

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$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{12} x_1 x_2 \qquad (1)$$

In (1), β_n represents the coefficient associated with factor n, whereas x_1 and x_2 represent the factors of the model. The product $x_1 x_2$ represents the interaction between the individual factors. β_0 is the intercept of the model whereas $\beta_1 x_1$ and $\beta_2 x_2$ represent the individual effects of x_1 and x_2. $\beta_{12} x_1 x_2$ represents the two-factor interaction between x_1 and x_2. The regression analysis was done in coded units and the coefficients were based on these coded units. ANOVA was then used to determine the sum of squares (SS), mean squares (MS), F-value, p-value, coefficient of determination (R^2) and correlation coefficient ($|R|$). Response surface plot was used to determine optimum concentrations of PG and CA antioxidants which will maximize the AC breakdown voltage of the NEI oil.

3. RESULTS AND DISCUSSIONS

i. AC Breakdown voltage test results
The AC breakdown voltage values for all seven test runs are presented in Table-3.

Table-3. AC breakdown voltage for each test run.

Experiment (Test run)	Variable code		Mean AC breakdown voltage(kV/mm)
	A: PG (wt.%)	B: CA (wt.%)	
1	-1	+1	52
2	-1	-1	51
3	0	0	45
4	+1	+1	41
5	0	0	46
6	0	0	46
7	+1	-1	45

ii. Results of the screening process
Figure-1 and Table-4 shows the half-normal plot and effect list, respectively, which are obtained from the 2^2 factorial design of experiments. It can be observed from Figure 1 that factor A (propyl gallate) is positioned at a distance far away from the straight line. Likewise, factor B (citric acid) and interaction AB are also positioned at a distance away from the straight line – though the distance is not as marked as that for factor A. This indicates that factors A and B as well as interaction AB are all significant model terms. The effect list which shows the sum of squares and percentage contribution for all model terms is shown in Table 4. The results indicate that factor A is the most significant factor with a percentage contribution of 84.29% while the sum of squares for this factor is 72.25. In contrast, factor B only has a contribution of 2.63%, which clearly shows that this factor has the lowest contribution among all factors. The sum of squares for factor B is 2.25. Finally, interaction AB has a

contribution of 7.29% while its sum of squares is 6.25. Based on the results, it is evident that factor A (propyl gallate) has a significantly higher contribution on the AC breakdown voltage compared to factor B and interaction AB. However, the contribution of other factors is still considered important.
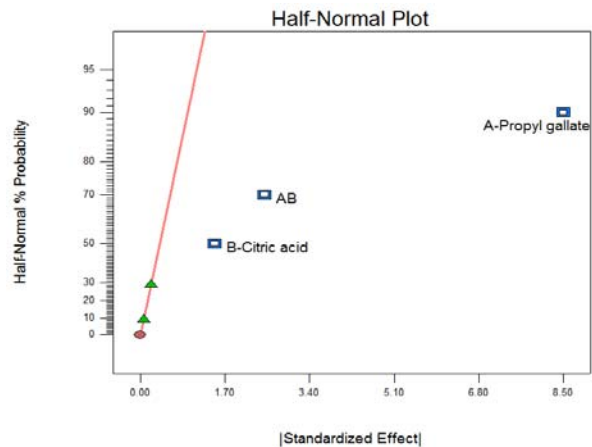

Figure-1. Half-normal plot obtained from the 2^2 factorial design of experiments.

Table-4. Effect list obtained from the 2^2 factorial design of experiments.

Model term	Standardized effects	Sum of squares (SS)	Percentage contribution (%)
A	-8.50	72.25	84.29
B	-1.50	2.25	2.63
AB	-2.50	6.25	7.29

iii. Results of the optimization process
The optimization process is carried out using RSM to determine the optimum concentrations of PG and CA which will yield the highest AC breakdown voltage for the rapeseed-based NEI oil. A regression model is developed according to (1), is shown in (2) which indicates the AC breakdown voltage of the NEI oil as a function of the propyl gallate concentration (variable: x_1; unit: wt.%) and citric acid concentration (variable: x_2; unit: wt.%).

$$y = 46.57 - 4.25 x_1 - 0.75 x_2 - 1.25 x_1 x_2 \qquad (2)$$

The AC breakdown voltage for each experimental point predicted using (2) is summarized in Table-5, along with the observed values obtained from experiments. The mean and standard deviation of the AC breakdown voltage obtained from the regression equation is found to be 46.57 and 1.29 kV/mm, respectively. Table-6 shows the sum of squares (SS), degrees of freedom (df), mean squares (MS), F-value, p-value, coefficient of determination (R^2) and correlation coefficient ($|R|$) for the regression model terms

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determined using ANOVA. According to Mohamed *et al*. [22] and Aman *et al*. [24], a *p-value* that is less than or equal to 0.05 indicates that the model (or model term) is statistically significant. The results show that the overall regression model is significant since the p-value is 0.0123. Factor A (propyl gallate) and interaction AB are significant model terms since the *p-value* is less than 0.05 (0.0046 and 0.0494 for factor A and interaction AB, respectively). In contrast, the *p-value* for factor B (citric acid) is 0.1217 (which is more than 0.05) and therefore, this model term is not significant.

Based on the ANOVA results, it can be inferred that factor B (citric acid) will not have a pronounced effect on the AC breakdown voltage if it is used alone as an antioxidant in the NEI oil. In other words, factor B (citric acid) needs to be combined with factor A (propyl gallate) in order to enhance the AC breakdown voltage of the NEI oil by a significant margin. It can also be deduced that the regression model developed in this study is adequate since the R^2 value is 0.8824, which indicates the model explains 88.42% of the total variation of the AC breakdown voltage due to variation of the independent variables (*i.e.* PG and CA concentrations). In addition, the *p-value* of the overall regression model is 0.0123 (which is less than 0.05), indicating that the model is significant. The correlation coefficient $|R|$ shows there is a correlation between the observed and predicted AC breakdown voltage values. In general, if $|R|$ is closer to 1, this indicates that there is a strong correlation between the observed and predicted values. The $|R|$ value of the regression model is found to be 0.94, which shows that there is a strong correlation between the predicted AC breakdown voltage values and those obtained from experiments.

Figure-2 is the response surface plot which shows the variation of the AC breakdown voltage of the NEI oil when the concentrations of PG and CA are varied. The beauty of this plot is that the interaction between the factors which influence the AC breakdown voltage of the NEI oil can be visualized and interpreted easily. It can be seen from Figure 2 that the AC breakdown voltage of the NEI oil increases gradually as the CA concentration is increased from 0.05 to 0.25 wt.% while the PG concentration is decreased from 0.25 to 0.05 wt.%. The

maximum AC breakdown voltage of 52 kV/mm is attained when the PG concentration and CA concentration is 0.05 and 0.25 wt.%, respectively

In general, a higher concentration of CA is recommended for insulation oils since the price of this antioxidant is cheaper compared to that for PG. However, the high cost of PG antioxidant is compensated by the fact that only a low concentration of PG is needed to maximize the AC breakdown voltage of the insulation oil. Both of these antioxidants are needed since each antioxidant serves a different purpose - PG is a radical scavenger whereas CA acts as a synergist. In other words, CA is a hydro-peroxide scavenger. The enhancement of the AC breakdown voltage for the NEI oil upon the addition of PG and CA antioxidants is associated with their mono-aromatic molecule structures. According to Evangelou *et al*. [25], Zaky *et al*. [26], and Walker *et al*. [27], these aromatics will alter the gassing tendency of the insulation oil to more 'gas absorbing' rather than 'gas evolving'. The gas absorbing properties will enhance the AC breakdown voltage of the insulation oil, as well as increase its resistance against partial discharge [28].
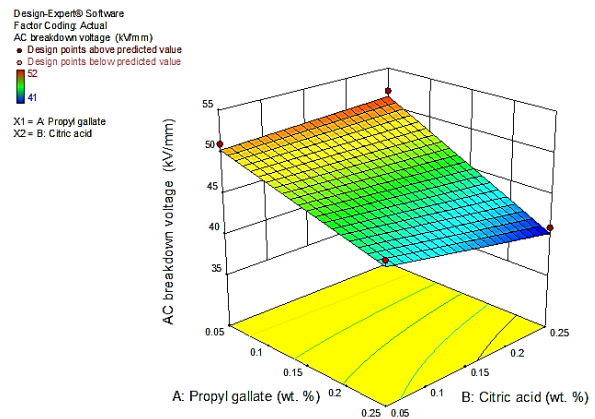

Figure-2. Response surface plot which shows the effect of the propyl gallate and citric acid concentrations on the AC breakdown voltage of NEI oil.

Table-5. Observed and predicted AC breakdown voltage.

Experiment (Test run)	**Observedvalue, y** (kV/mm)	**Predicted value, y'** (kV/mm)	**Residual|y-y'|**
1	52	52.00	0.000
2	51	51.00	0.000
3	45	45.67	0.670
4	41	41.00	0.000
5	46	45.67	0.330
6	46	45.67	0.330
7	45	45.00	0.000

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Table-6. ANOVA results for the regression model with factorial response surface fitting.

Source	SS	df	MS	F-value	p-value	R^2
Overall model	80.75	3	26.92	80.75	0.0123	0.8842
A-Propyl gallate	72.25	1	72.25	216.75	0.0046	
B-Citric acid	2.25	1	2.25	6.75	0.1217	
AB	6.25	1	6.25	18.75	0.0494	
Residual	4.96	3	1.65			
Lack of fit	4.30	1	4.30	12.89		
Pure error	0.67	2	0.33			
Total correlation	85.71	6				

4. CONCLUSIONS

In this study, it is proven that two-level factorial design of experiments is useful technique to determine the optimum concentrations of propyl gallate and citric acid antioxidants which will enhance the AC breakdown voltage of rapeseed-based NEI oil. The main advantage of the two-level factorial design of experiments approach is that the factors which will have a significant effect on the AC breakdown voltage of the NEI oil can be determined from fewer test runs, as indicated by the percentage contribution of each factor. This considerably reduces time and cost, which is the typically an issue with conventional experimental techniques. The results obtained from the 2^2 factorial designs reveal that propyl gallate has the most pronounced effect on the AC breakdown voltage of NEI oil, with a percentage contribution of 84.29%. The response surface plot generated reveals that the optimum concentration of propyl gallate and citric acid which will yield the highest AC breakdown voltage is 0.05 and 0.25 wt.%, respectively. A regression model is also developed in this study, and it is found that the model is adequate to predict the maximum AC breakdown voltage of the NEI oil as a function of propyl gallate and citric acid concentrations, whereby the R^2 and *p*-value of the model is 0.8842 and 0.0123, respectively.

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