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Dielectric Strength Improvement of Natural Ester Insulation Oil via Mixed Antioxidants: Taguchi Approach

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ABSTRACT

Recently, natural ester insulation (NEI) oils are found to be the best candidates to replace mineral-based insulation oils for oil-immersed transformer applications. However, NEI oilsare prone to oxidation due to their poor oxidative stability which can be improved by adding antioxidants into the oils. Latest studies have also shown that the use of selected antioxidants improves the AC breakdown voltage (BdV) of NEI oils. However, the experiments in previous studies were designed using the conventional one-factor-at-a-time (OFAT) method, which requires a large number of samples to be tested in order to determine the optimum response. Thus, a Taguchi-based designed experiment is introduced in this study in replacement of the OFAT method. It is found that this method is capable of determining the optimum concentrations of propyl gallate (PG) and citric acid (CA) which will maximize the AC BdV and improve the oxidative stability of the NEI oil. An AC breakdown voltage test is conducted in accordance with the ASTM D1816 standard using Megger OTS60PB portable oil tester, in which the electrode gap distance is kept fixed at 1 mm. The results indicate that the addition of PG and CA antioxidants increases theAC BdV of the rapeseed-based NEI oil. It is found that the optimum concentrations of PG and CA antioxidant is 0.05 and 0.25 wt.%, respectively. Lastly, the model developed in this study is analysed using analysis of variance (ANOVA). Validation test is also conducted on the optimized NEI oil to determine its dielectric strength and oxidative stability.

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1. INTRODUCTION

Transformers are one of the key components in power network and their main function is to convert voltage and transfer energy. The reliability of power transformers is largely determined by the condition of their insulation [1]. In liquid-filled transformers, the insulation liquid has two important functions: (1) it provides electrical insulation and (2) it serves as a coolant by absorbing heat when there is temperature increase in the transformer windings and core. For more than a hundred years, liquid-immersed transformers are typically filled with mineral insulation oils due to their wide availability, good properties and low cost. However, due to environmental issues resulting from the use of mineral insulation oils, it becomes more important than ever to use insulation oils thatare highly biodegradable and environmentally friendly. The recent availability of natural ester fluids based on vegetable oils has provided a new alternative for transformer insulation liquids [2]. Nowadays, studies on the viability of natural ester-based insulation (NEI) oils as an alternative to replace mineral oils in power transformers are gaining much attention due to their excellent biodegradability, higher fire point and good dielectric properties such as AC breakdown voltage (BdV). In addition, NEI oils have the potential to increase the lifespan oftransformers because of their superior hydrophilic properties compared to conventional mineral oils [3], [4]. However, NEI oils have lower pour point, which make them less suitable for use in cold climate regions. Moreover, NEI oils are prone to oxidation due to their lower oxidative stability [5], [6]. For these reasons, scientists and researchers are actively working on improving the properties of NEI oils for use as transformer insulation oils.

It has been proven that the poor oxidative stability of NEI oils can be improved by the addition of antioxidants into the oils [7], [8]. Antioxidants are essentially compounds that delay or slow down the oxidation process of transformer oils [9], [10]. However, interestingly, recent studies have shown that the incorporation of selected antioxidant mixtures into NEI oils also improves their AC BdV [7], [11], [12]. Therefore, in this study, the effect of antioxidant mixtures on the oxidative stability of NEI oil is assessed based on theAC BdV. In addition, previous researchers have only implemented the one-factor-at-a-time (OFAT) method as their experimental design approach to determine the optimum ratios of antioxidants that will improve the dielectric performance of insulation oils. However, the OFAT method requires a large number of test runs or experiments in order to estimate the effect that can possibly give better outputs. The large number of test runs required is highly undesirable since it consumes a considerable amount of time and cost to execute all of the experiments [13], [14]. Hence, in this study, a design of experiments (DoE) approach by means of the Taguchi method is introduced to replace the OFAT method. The method proposed in this study is advantageous since it reduces the number of experiments, time and overall experimental cost [15-17]. The Taguchi method is also used to determine the optimum concentrations of antioxidants which will maximize the AC BdV of the NEI oil.

2. EXPERIMENTAL DESIGN

2.1. Design of Experiments

The Taguchi design (L_8 orthogonal array) was used in this study consisting of eight rows (corresponding to the number of tests) and two columns for two levels. This array has seven degrees of freedom (DOF), whereby twoDOFs were assigned to two factors (*i.e.* each factor has one DOF) and five DOFs were assigned to errors. In order to observe the degree of significance of the design parameters in terms of wt.% contributions, two factors (each at two levels) are highlighted in Table 1. Table 2 shows the list of test runs according to the L_8 orthogonal array generated by Minitab statistical software.

Table 1. Level of conditions for propyl gallate and citric acid antioxidants

Level	Propyl gallate (wt.%)	Citric acid (wt.%)
1	0.05	0.05
2	0.25	0.25

Table 2. L_8 (2^2) orthogonal array

Run no.	Factors	
	Antioxidant *A* Propyl gallate (wt.%)	Antioxidant *B* Citric acid (wt.%)
1	0.05	0.05
2	0.05	0.05
3	0.05	0.25
4	0.05	0.25
5	0.25	0.05
6	0.25	0.05
7	0.25	0.25
8	0.25	0.25

Once the series of test runs shown in Table 2 was conducted, the AC BdV response for each test run was analysedbased on the signal-to-noise (S/N) ratio. The S/N ratio is important in the Taguchi method. The goal of the S/N ratio is to analyse the response by reducing the sensitivity of the system to the sources of variation, which in turn, produces better results [16], [17]. There are three categories of S/N ratio, namely,“smaller is better”, “nominal is best” or “larger is better”. In this study, the S/N ratio for “larger is better” given by Equation (1) was used to attain a higher AC BdV response for the NEI oil added with antioxidants.

$$\frac{S}{N} = -10\ log \left| \frac{1}{n} \sum_{i=1}^{n} \frac{1}{y_i^2} \right| \tag{1}$$

In this equation, n is the number of repetitions under the same design parameter conditions, $y_i$indicates the measured results and subscript i indicates the number of design parameters arranged in the orthogonal array. Following this, ANOVA was used to determine the contribution of each main factor which affects the AC BdV of the NEI oil. The optimum concentrations of antioxidants can be predicted based on the S/N ratio and ANOVA analysis.

2.2. Sample Preparation

Rapeseed-based oil was chosen as the NEI oil in this study. Pre-processing was first carried out by removing foreign particles and moisture from the NEI oil. The oil samples were filtered using a quantitative filter paper (pore size: 0.02 μm) and then heated in a vacuum oven for 48 hours at a temperature of 70°C. This process was carried out to ensure that the base NEI oil samples have a dielectric strength more than 20 kV/mm in accordance with the specification given in the ASTM D6871 standard test method [18]. Propyl gallate (PG) and citric acid (CA) were selected as the primary and secondary antioxidant, respectively. These antioxidants were purchased from NacalaiTesque, Inc. (Japan). Theantioxidants were mixed with 500 ml of NEI oil according to the number of test runs given in Table 2. In order to achieve maximum effectiveness, the mixtures were uniformly dispersed using a hot plate magnetic stirrer set at a stirring speed of 750 rpm [7]. The temperature of the hot plate was set according to the melting point of each antioxidant.

2.3. AC Breakdown Voltage Test

The oil samples were tested to determine their dielectric strength using Megger OTS60-PB insulating oil tester in accordance with the ASTM D1816 standard test method (Figure 1) [19]. The dielectric strength represents the potential difference in which electrical failure occurs in an electrical insulation material under AC test conditions. Based on the ASTM D1816 standard test method, the electrode configuration consisted of two VDE (Verband Deutscher Elektrotechniker, Specification 0370) with a gap distance of 1.0 mm. The rate of voltage rise was kept fixed at 0.5 kV/s. At least two samples were prepared for each set of experiments to ensure a high confidence level. In this study, 25 AC BdV tests were performed for each sample and the average value for each set of tests was determined.

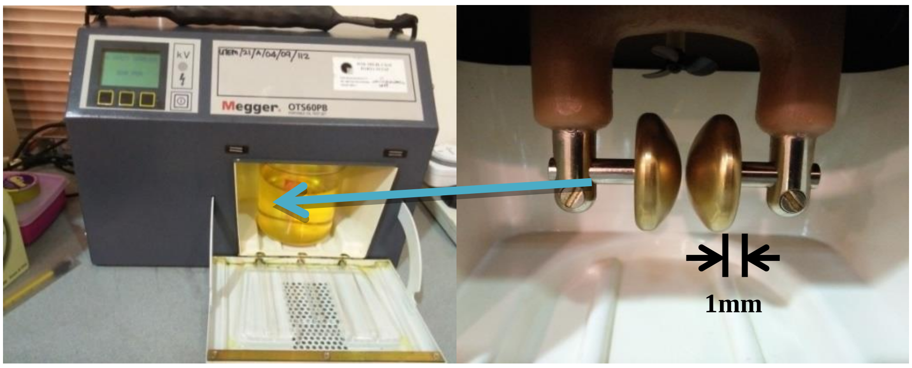

Figure 1. Megger OTS60-PB AC BdV portable oil tester with an electrode gap distance of 1 mm

3. RESULTS AND DISCUSSION

3.1. Analysis of the S/N Ratio

The AC BdV values measured from the tests and their corresponding S/N ratio values are shown in Table 3. It can be seen from the results that a higher S/N ratio results in higher AC BdV for the NEI oil. The response table for the S/N ratio of each antioxidant is given in Table 4, which shows the rank of each antioxidant on the AC BdV of the NEI oil. PG is found to have higher impact on the AC BdV compared to CA. Figure 2 shows the main effect plot ofthe S/N ratio on the AC BdV of the NEI oil. It can be seen that the optimum concentrations of antioxidants which will maximize the AC BdV of the NEI oil are obtained from the points above the reference line of the S/N ratio. It is found that the optimum concentration of PG and CA that results in the highest AC BdV is 0.05 and 0.25 wt.%, respectively.

Table 3. Response of the AC BdV, mean and S/N ratio of each test run

Run no.	AC BdV (kV/mm)	Mean (kV/mm)	S/N ratio (dB)
1	50	50	33.98
2	50		
3	53	53.5	34.57
4	54		
5	39	39.5	31.93
6	40		
7	46	46.5	33.35
8	47		

Table 4. Response table for the signal-to-noise (S/N) ratio of each antioxidant

Level	Propyl gallate (wt.%)	Citric acid (wt.%)
1	34.27	32.95
2	32.64	33.96
Delta	1.63	1.00
Rank	1	2

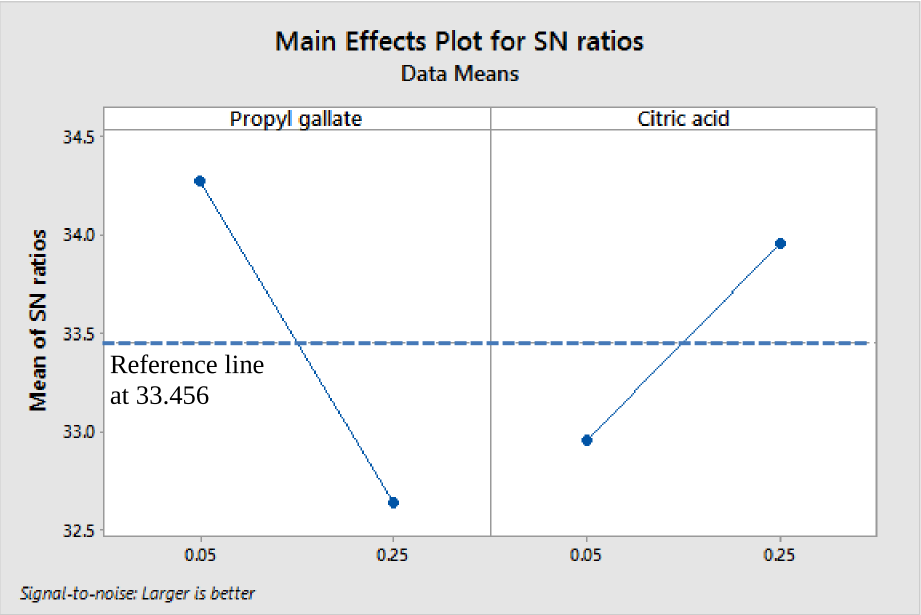

Figure 2. Main effect plot of the S/N ratio on the AC BdV of the NEI oil

3.2. ANOVA Analysis

ANOVA was used to examine and model the relationship between a response variable with one or more predictor variables. The main benefit of ANOVA is that it tests the significance of all factors by comparing the mean square against an estimate of the experimental errors at a specific confidence level. Firstly, the total sum of squared deviations (SS_T) from the total mean S/N ratio (n_m) was calculated using Equation (2).

$$SS_T = -\sum_{i=1}^{n}(n_i - n_m)^2 \quad (2)$$

where n is the number of experiments in the orthogonal array and n_m is the mean S/N ratio for the experiment. The percentage of contribution (P) was calculated using Equation (3).

$$P = \frac{SS_d}{SS_T} \quad (3)$$

where SS_d is the sum of the squared deviations. The ANOVA results are summarized in Table 5. The F-value represents the ratio of the mean square error to the residual error and it is traditionally used to determine the significance of a factor. The p-value represents the significance level. A p-value less than 0.050 ($p < 0.050$) indicates that the factor is significant and likewise, a p-value greater than 0.050 indicates that the factor is not significant for optimization. Based on the ANOVA results, it can be deduced that both factors contribute to the increase in AC BdV of the NEI oil, whereby PG and CA has a contribution of 70.93 and 25.54%, respectively.

Table 5. ANOVA results for the factors involved in improving the AC BdV of NEI oil

Factor	Degrees of freedom, Df	Sum of squares, SS	Mean squares, MS	F-value	p-value	Percentage of contribution (%)
Source						
Propyl gallate	1	153.125	153.125	100.41	0.000	70.93
Citric acid	1	55.125	55.125	36.15	0.002	25.54
Error	5	7.625	1.525	16.33	0.016	3.53
Lack-of-fit	1	6.125	6.125			
Pure error	4	1.500	0.375			
Total	7	215.875				100

3.3. Dielectric Strength and Oxidativ Estability Analysis

A validation test was also carried out using the optimum concentrations of antioxidants in the NEI oil (*i.e.* propyl gallate: 0.05 wt.%, citric acid: 0.25 wt.%). Weibull breakdown probability plot was used to obtain the most probable value of AC BdV at a probability of 63.2%. Figure 2 shows the Weibull probability plots for the fresh and optimized NEI oil samples. The AC BdV is found to be 24.79 and 53.89 kV/mm for the fresh and optimized NEI oil, respectively, at a Weibull breakdown probability of 63.2%. The increase in the AC BdV of the rapeseed-based NEI oil is nearly twice the value for fresh NEI oil. This increase may be attributed to the gassing tendency characteristics of the antioxidants. According to Walker et al. [20] and Bolliger et al. [21], additives containing aromatic compounds have the tendency to produce gas absorbing characteristics. This in turn, results in higher and AC BdV for insulation oils. The PDIV is defined as the voltage in which the partial discharge (PD) is initiated above the threshold of 20 pC. Zaky et al. [22] also attributed this behaviour to the addition of additives containing aromatic compounds into the insulation oil.

The oxidative induction time (OIT) is a relative measure of thedegree of oxidative stability of the material evaluated at an isothermal temperature of the test. The OIT value is compared between one material and the reference material in order to determine the relative oxidative stability. The presence, quantity or effectiveness of the antioxidants is determined using this method. In this study, the OIT was measured using Perkin Elmer Jade differential scanning calorimeter in accordance with the ASTM E1858 standard test method [23]. The OIT was determined fromthe time at which the base line and tangential line of the rising exotherm intersect. The temperature was set at 40°C/min and isothermal test temperature was set within a range of 170–210°C. The oxygen flow rate was 50 ml/min and the specimen mass range was 3.00–3.30 mg. Three samples were prepared for each test run and the average value was determined for each OIT test. The enhancement of the OIT for fresh and optimized NEI oil samples is shown in Figure 4 and 5, respectively. It is found that the oxidative stability of the NEI oil improves by roughly 60% using the optimum combination of PG and CA antioxidants in the rapeseed-based NEI oil. Both of these antioxidants complement each other since each antioxidant serves a different purpose – PG is a radical scavenger whereas CA acts as a synergist and hydro-peroxide scavenger.

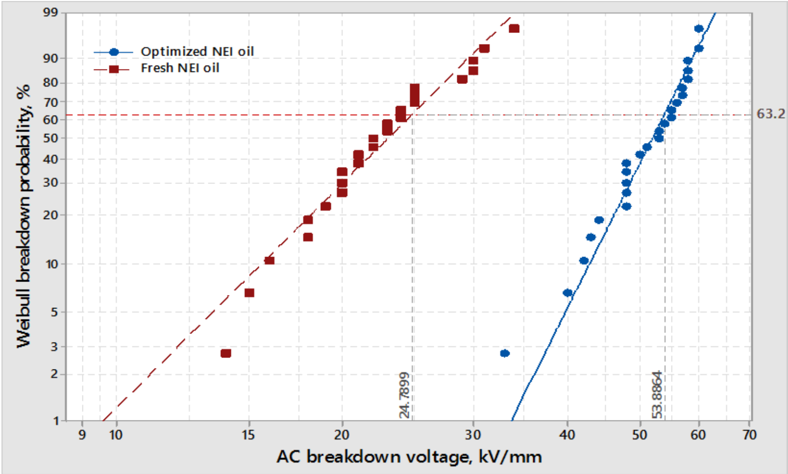

Figure 3. Weibull probability plot of the fresh and and optimized NEI samples. The most probable value of the AC BdV is taken at a probability of 63.2%

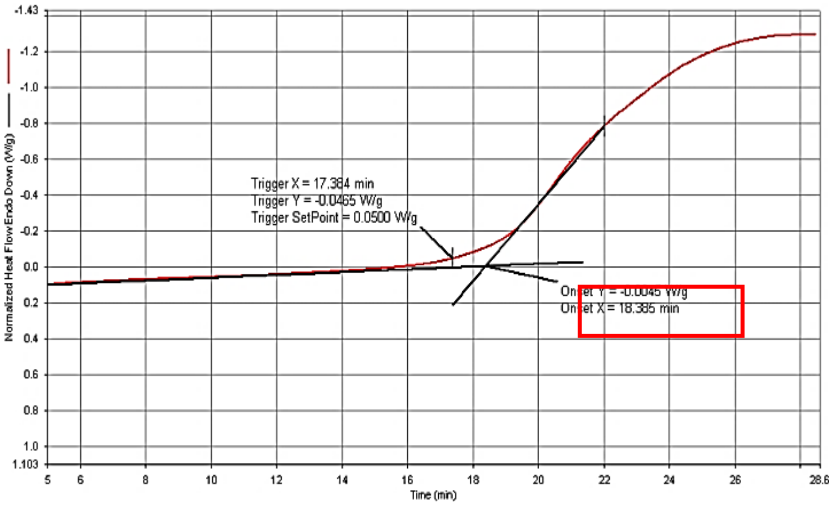

Figure 4. OIT response of the fresh NEI oil. The OIT is 18 minutes

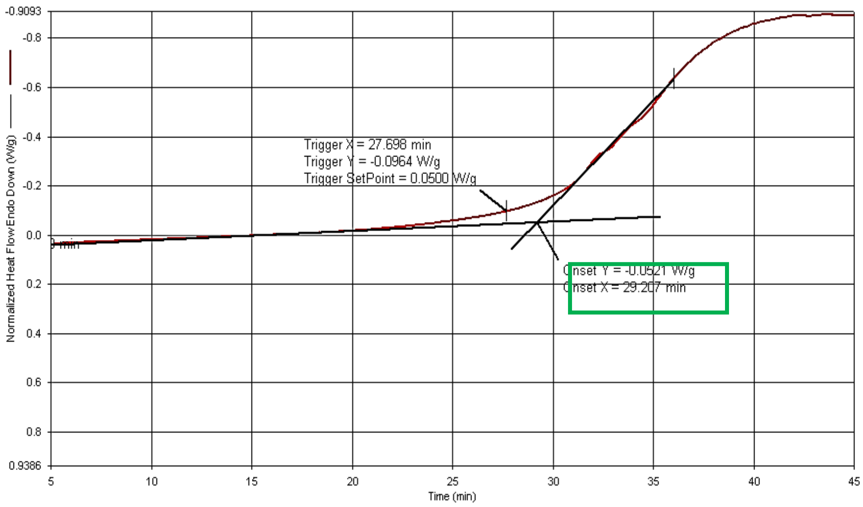

Figure 5. OIT response of the optimized NEI oil. The OIT is 29 minutes.

4. CONCLUSION

In this study, it is proven that Taguchi-based designed experiment is a useful technique to determine the optimum concentrations of propyl gallate and citric acid which will enhance the dielectric strength and oxidative stability of rapeseed-based NEI oil. This enhancement is due to the fact that the propyl gallate and citric acid antioxidants have reached a synergistic level and gas absorbing tendency characteristics. The main advantage of the Taguchi method is that one can determine the factors which will have a significant effect on the AC BdVof the NEI oil from fewer experiments, 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. It is found that the optimum concentration of propyl gallate and citric acid is 0.05 and 0.25 wt.%, respectively for the rapeseed-based NEI oil.

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