

Article



The Effect of Electronic Scavenger Additives on the AC Dielectric Strength of Transformer Mineral Oil

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Received: 6 September 2018; Accepted: 27 September 2018; Published: 30 September 2018



Abstract: This paper is devoted to the influence of two types of electronic scavenger additives/compounds, namely, carbon tetrachloride (CCl₄) and methyl iodide, which is also called iodomethane (CH₃I), on the dielectric strength of transformer mineral oil. The tests are achieved in a sphere-sphere electrodes arrangement under AC voltage according to the IEC 60156 standard. The investigated additive concentrations range from 0 to 600 ppm. The verification of the conformity of the experimental results with normal and Weibull probabilistic distributions as well as the estimation of the breakdown voltage with risk probabilities of 1%, 10%, and 50% are also performed. It is shown that there is an optimum concentration of each type of electronic scavenger compound at which the dielectric strength of the mineral oil is significantly improved (i.e., it reaches a maximum value). This improvement is of 98% with 500 ppm of CH₃I and 93% with 200 ppm of CCl₄. It is also shown that the breakdown voltage values of all of the investigated samples with and without additives conform to a Weibull distribution but not to a normal distribution. The obtained results are discussed with regard to the possible mechanisms that may be responsible, particularly the two phases of inception and propagation of the streamers.

Keywords: AC dielectric strength; insulating oils; electronic scavenger additives; statistical analysis; Weibull distribution; normal distribution

1. Introduction

The research for additives that can improve the dielectric strength of insulating oils has received the attention of investigators for many decades. Different types of additives have been considered. More specifically, these last three decades, many studies have focused on the addition of nanostructured particles. Thus, it has been observed that several specific nanoparticles such as Fe_3O_4 significantly enhance the dielectric strength of mineral oils [1–5]. Nevertheless, the results reported in the literature on the influence of aromatic additives on the breakdown voltage of insulating oils are contradictory [6].

Thus, Zaky and Hawley [6] reported that weak concentrations of aromatic additives enhance the breakdown voltage of insulating oils and that there is an optimum concentration of additives that results in a maximum increase of the breakdown strength. Evangelou et al. [7] reported that the breakdown voltage varies with the additive concentration in a complex manner. The characteristics obtained for a very wide range of additive concentrations indicated that there could be more than one optimum concentration.

2 of 12

Contrary to Zaky and Hawley [6], Mathes and Rouse [8] reported that the addition of a small concentration of poly-aromatic compounds significantly reduces the dielectric strength of naphthenic oil in a point-to-sphere electrode arrangement under impulse voltage. This reduction in the breakdown voltage has been attributed to the streamer development and especially to the increase in the velocity of the streamers' propagation: the faster the streamer, the lower the dielectric strength. Since these poly-aromatic compounds have both low ionization potentials (Vi) [9] and large electronic-trapping sections (Ee) [10], it is difficult to distinguish which property of these additives is responsible for the streamer development.

Thus, in their pioneering work, Devins et al. investigated the influence of each of the additives' properties separately (electronic scavenger and low ionization potential) and highlighted the importance of electronic processes in the streamer propagation phenomena [11]. They observed that: (1) the addition of electronic scavengers such as sulfur hexafluoride (SF₆: Vi = 15.9 eV and Ee = +1.5 eV) or ethyl chloride (C₂H₅Cl: Vi = 10.97 eV and Ee = 1.4 eV) to naphthenic oil (Marcol 70) or to 2,2,4-trimethyl-pentane renders the negative streamers more filamentary and increases their velocities. With 0.05 mol/L of SF₆ or C_2H_5Cl , the streamer velocity can reach five times its initial value. There is no detectable effect on the positive streamers in these liquids. They also observed that: (2) the addition of a low ionization potential compound such as N,N-dimethyl-aniline; an organic chemical compound and it is substituted derivative of aniline; (DMA: Vi = 7.14 eV and Ee = -2.1 eV) does not change the negative streamer velocity, whereas it increases (two- to three-fold) compared to that of the positive streamers in naphthenic oil and 2,2,4-trimethyl-pentane. In both cases, there is a concentration (about 0.05 mol/L) above which a saturation is observed [11,12]. Chadband et al. [13] confirmed this result in n-hexane. In contrast, Hebner et al. [14] reported that with 0.1 mol/L of DMA, the positive streamer in n-hexane changes from a filamentary structure to a more or less hemispherical shape and that its velocity reduces slightly.

In order to confirm and to extend the above results, Beroual et al. [15,16] considered compounds having either a high electronic affinity or a very low ionization potential. They observed that: (1) the addition of small concentrations of an electronic scavenger such as carbon tetrachloride (CCl₄: Vi = 11.47 eV and Ee = +2.1 eV in the gas phase [10]) to cyclohexane increases the negative streamer velocities: 0.04 mol/L of CCl₄ increases the velocity by a factor of 10 and the streamer becomes filamentary. Above 0.04 mol/L, the increase in the velocity is not as significant. They also observed that: (2) the addition of a low ionization potential compound (0.05 mol/L) such as tetramethyl-para-diphenylamine (TPMD) (TMPD: Vi = 6.7 eV [17]) to cyclohexane leads to a moderate increase in the negative streamer velocity (by a factor lower than two) and its shape remains practically unmodified. The velocity of the positive streamer is, in the same conditions, multiplied by a factor of three and the streamer becomes still more filamentary.

A similar effect was observed with DMA [16,18], thus confirming the results reported by Devins et al. [11]. There is also a concentration (about 0.06 mol/L) at which saturation is reached. The influence of the electronic scavenger and low ionization potential compounds is comparable in other liquids such as phenylxylylethane (PXE) and mono-dibenzyltoluene (M/DBT) [19]. Note that the influence of both the electronic scavengers and the low ionization potential compounds on the fast negative streamers is absolutely the same for their positive polarity correlates.

The theory according to which the negative streamers are bush-like and the positive streamers are fast and filamentary is not true. The negative streamers can also be filamentary and fast, as reported in References [16,20,21].

From the above, it appears that the addition of electronic scavenger compounds to dielectric liquids accelerates the streamer propagation and thus reduces the dielectric strength. This acceleration is more significant for slow (bush-like) negative streamers.

More recently, several investigators have shown that the addition of given amounts of specific electronic scavenger compounds increases the inception threshold voltage of the streamers, resulting in an increase in the dielectric strength. Ingebrigtsen et al. [22] observed that in a point-plane electrode

arrangement, the 1-methylnaphthalene additive increases the initiation threshold voltage of the point anode streamers in cyclohexane. By adding carbon tetrachloride (CCl₄) or Iodobenzene (C₆H₅I) to mineral oil and tetra-ester oil, Beroual and Aka [23] observed an increase in the inception threshold voltage of the streamers. According to Beroual [24,25], if the inception threshold voltage is higher, the streamer will be more energetic and move faster. Thus, the question that is posed is whether the addition of electronic scavenger compounds: (1) accelerates the streamers, resulting in the decrease of the dielectric strength of the liquid or (2) increases the inception threshold voltage of the streamers and thence leads to an increase in the dielectric strength of the liquid.

The goal of this paper is to investigate the effect of two types of electronic scavenger compounds, namely, carbon tetrachloride (CCl_4) and methyl iodide, which is also called iodomethane (CH_3I), on the dielectric strength of transformer mineral oil. An analysis of the conformity of the experimental results with Weibull and normal distributions is also performed. Our aim is also to clear up the different contradictory results reported in the literature.

2. Materials and Procedure

The characteristic parameters of the basic mineral oil used are given in Table 1. Note that the mineral oil we used is somewhat aged and that the water content was 39 ppm. Two types of electronic scavenger additives/compounds, namely, carbon tetrachloride (CCl₄: Vi = 11.47 eV and Ee = +2.1 eV in the gas phase [3]) and methyl iodide, which is also called iodomethane (CH₃I: Vi = 9.54 eV) were used. These were provided by Nexgen Chemical, India and BDH Chemicals Ltd. Poole, England, respectively.

Mineral oil-based electronic scavenger samples were prepared by adding the scavenger in concentrations ranging from 200 to 600 ppm by micro-pipit into 500 mL of dry oil. The liquids were mixed with the aid of a magnetic stirring process at ambient temperature and at a speed of 1100 rpm for about 15 min. Then, the electronic scavenger samples were placed in the ultrasonic homogenizer for 30 min to obtain a homogeneous dispersion fluid. Then, the samples were kept under a vacuum of 0.16 MPa for 24 h to eliminate humidity and internal bubbles.

The dielectric breakdown (BDV) measurements were performed with a Foster Oil Test 90 type with a test cell of 500 mL according to the IEC 60156 standard [26]. The electrode arrangement consisted of two brass hemispheres 12.5 mm in diameter and an electrode gap of 2.50 ± 0.05 mm. An alternating voltage ramp rate of 2 ± 0.2 kV/s was applied continuously from zero to breakdown. The breakdown voltage is the average of 30 successive measurements and the time delay between the successive measurements was 2 min. For the Weibull distribution, we performed another series of measurements. It is to have a power of 2 ($2^4 = 16$) and to deduce the slope of Weibull plots. Sixteen is a reasonable number for a Weibull analysis.

Property	Mineral Oil
Density at 25 °C (g/mL)	0.85
Kinematic viscosity at 40 °C (cSt)	9
Pour point (°C)	-40
Flash point (°C)	150
Fire point (°C)	160
Total acid number (mg KOH/g)	< 0.5
Antioxidant content	<0.3%
Water content (ppm)	39
Gassing characteristics (mm ³ /min)	-35 to $+30$
Interfacial tension (mN/m)	40-60
Resistivity ($\Omega \cdot m$)	$>3 \times 10^{9}$
Dissipation factor at 90 °C	0.1-0.5%

3. Experimental Results

Figures 1 and 2 depict the variation in the breakdown voltage (U_{BDV}) of the mineral oil when adding CCl₄ and Figures 3 and 4 depict that of the mineral oil when adding CH₃I. We observe that the addition of 200 ppm of CCl₄ increases the BDV of the mineral oil by 93%, while with the addition of 300 ppm; there is only a 31% improvement. Beyond this amount of CCl₄, the BDV decreases to reach 17.8 kV for a concentration of 600 ppm, i.e., a BDV reduction of 54%. While adding the CH₃I, the BDV of the mineral oil increases with the amount of CH₃I up to 500 ppm. There is a 98% improvement at this concentration. Beyond this concentration, the BDV decreases, but remains higher than the breakdown voltage of the mineral oil. There is an 80% enhancement at 600 ppm. Thus, there is an optimal concentration of these specific additives at which the dielectric strength of the mineral oil is significantly improved. On the other hand, CH₃I appears to be a very effective additive for the improvement of dielectric strengths, as shown in Figure 5.



Figure 1. The breakdown voltage for various concentrations of the mineral oil (MO)/CCl₄ scavenger.



Figure 2. Comparison between the average AC breakdown voltage of the MO and those of the MO with different CCl₄ concentrations.



Figure 3. The breakdown voltage for various concentrations of the MO/CH₃I scavenger.



Figure 4. Comparison between the average AC breakdown voltage of the MO and those of the MO with different CH₃I concentrations.



Figure 5. Comparison between the CCl₄ and CH₃I additives in the MO.

4. Statistical Analysis of Experimental Data

In this section, we analyze the conformity of the experimental data with two of the main probabilistic functions that are the most frequently used to study the breakdown voltage of dielectric materials, namely, the normal and Weibull distribution laws [27–29]. For that purpose, we applied the Shapiro–Wilk [30] and Anderson–Darling [31] tests, respectively, by considering a test level, α , to be significant if it was equal to 5% ($\alpha = 0.05$) and by using R software [32]. Note that the hypothesis was accepted or rejected depending of the calculated *p*-value with respect to the value of α . For *p*-values > α , we accepted the null hypothesis according to which the sample data belongs to a statistical distribution [30,32].

4.1. Weibull Probability of the AC Breakdown Voltage of the Investigated Samples

Figures 6 and 7 depict the Weibull plots of the AC breakdown voltages of the mineral oil with different concentrations of CCl_4 and CH_3I . We observe that the experimental data from all of the investigated samples conforms to a Weibull distribution function, as shown in Table 2.



Figure 6. Weibull plot of the breakdown voltage of the mineral oil with various scavenger additives of CCl₄.



Figure 7. Weibull plot of the breakdown voltage of the mineral oil with various concentrations of the CH₃I scavenger additive.

Where the shape parameter is equal to the slope of the line in a probability plot, it affects the shape of the curve. The scale parameter is related to the scattering of the data and indicates the degree of failure. The Anderson–Darling (AD) value is the Anderson–Darling measure of the area between the fitted line and the empirical distribution function, which is based on the data points. N is the number of breakdown voltage data points. The *p*-value is a probability that measures the evidence against the null hypothesis.

Table 2. The *p*-value of the mineral oil with various concentrations of CCl4 and CH3I scavenger additives.

Concentration of the Additives	<i>p-</i> Value MO/CCl ₄	<i>p-</i> Value MO/CH ₃ I	Conformity to a Weibull Distribution
Pure (MO)	0.1	123	Accepted
200 ppm	0.250	0.186	Accepted
300 ppm	0.250	0.250	Accepted
400 ppm	0.250	0.250	Accepted
500 ppm	0.250	0.250	Accepted
600 ppm	0.250	0.250	Accepted

4.2. Histogram and Normal Distribution of the AC Breakdown Voltage of the Investigated Samples

Figures 8 and 9 give the histograms of the distribution of the breakdown voltages of the tested samples with CCl_4 and CH_3I , respectively. The blue lines refer to normal distribution of the tested sample measurements. Contrary to a Weibull distribution, we observe some anomalies, as depicted in Table 3. The experimental data does not obey a normal distribution.



Figure 8. Cont.



Figure 8. Frequency of the breakdown voltage of: (a) the MO; (b) the MO/CCl₄ (200 ppm); (c) the MO/CCl₄ (300 ppm); (d) the MO/CCl₄ (400 ppm); (e) the MO/CCl₄ (500 ppm); (f) the MO/CCl₄ (600 ppm).



Figure 9. Frequency of the breakdown voltage of: (**a**) the MO; (**b**) the MO/CH₃I (200 ppm); (**c**) the MO/CH₃I (300 ppm); (**d**) the MO/CH₃I (400 ppm); (**e**) the MO/CH₃I (500 ppm); (**f**) the MO/CH₃I (600 ppm).

Concentration of the Additives	<i>p</i> -Value	<i>p</i> -Value	Conformity to a Normal Distribution			
Concentration of the Additives	MO/CCl ₄	MO/CH ₃ I	MO/CCl ₄	MO/CH ₃ I		
Pure (MO)	0.	593	Accepted			
200 ppm	0.012	0.034	Not Accepted			
300 ppm	0.291	0.023	Accepted	Not Accepted		
400 ppm	0.266	0.457	Accepted			
500 ppm	0.457	0.198	Accepted			
600 ppm	0.020	0.542	Not Accepted	Accepted		

Table 3. The *p*-value of the mineral oil with various concentrations of the CCl₄ and CH₃I electronic scavenger additives.

4.3. Estimation of the the Main Breakdown Voltage Probabilities

Table 4 gives the AC breakdown voltage probabilities at 1%, 10%, and 50% of the Weibull distribution for the investigated samples. In addition, it shows the incremental percentage of the mineral oil with various concentrations of electronic scavenger additives. The breakdown voltage at a cumulative probability of 50% is an indication of the median value. The breakdown voltage at a cumulative probability of 1% gives an indication about the reliability of the oil. This is fundamental for determining the nominal voltage for the equipment design (i.e., the safety coefficient).

We observe that the U_{BDV} (1%) is also optimum at 200 ppm of CCl_4 , with an improvement of 126%, and that it is optimum at 500 ppm of CH_3I with an improvement of 126% as well with respect to the U_{BDV} (1%) of the mineral oil. The improvement with respect to the AC average breakdown voltage of the mineral oil (U_{BDV} = 38.5 kV) is 82%. These interesting results can be very useful when designing high-voltage oil-filled apparatus.

y (%)	_	ives	200 ppm		300 ppm		400 ppm		500 ppm		600 ppm	
BDV Probabilit	BDV Probabilit BDV (kV) Type of Addit	Type of Addit	BDV (kV)	Increment (%)	BDV (kV)	Increment (%)	BDV (kV)	Increment (%)	BDV (kV)	Increment (%)	BDV (kV)	Increment (%)
1.0	31	CCl ₄ CH ₃ I	70 49	62.04 13.43	40 62.5	$-7.41 \\ 44.68$	26 62.6	-39.81 44.91	18 70.2	-58.33 62.50	13 56	-69.91 29.63
10.0	35	CCl ₄ CH ₃ I	73 55.5	40.38 6.73	46 66	-11.54 26.92	30 66.5	-42.31 27.88	23 74	-55.77 42.31	16 64	-69.23 23.08
50.0	39	CCl ₄ CH ₃ I	75 63.5	24.17 5.13	52 71	-13.91 17.55	34 72	-43.71 19.21	28 76	-53.64 25.83	18 70	-70.20 15.89

Table 4. The AC breakdown voltage at different breakdown probabilities for the mineral oil with CCl_4 and CH_3I electronic scavenger additives at various concentrations in the mineral oil.

5. Discussion

It appears from the above results that electronic scavenger additives enable the improvement of the dielectric strength of mineral oil and that for each type of additive, there is an optimal concentration allowing to the achievement of the optimum breakdown voltage value. Beyond this concentration, the effect of the electronic scavenger additives drops. It is also possible to have different optimum concentrations. This somewhat confirms the idea reported by Evangelou et al. [7].

As indicated in Section 1, many investigators attributed the reduction or the enhancement of the dielectric strength to the streamer development. What is the underlying mechanism?

To explain the influence of the additives on the streamer propagation, Devins et al. [11] proposed a model in which they assume that field ionization occurs in the liquid. They use Zener's theory of tunneling in solids to calculate the concentration of the positive and negative carriers contained in a cylindrical conducting channel. According to this model, the positive streamer velocity is constant. Concerning the negative streamers, the propagation occurs in two stages: an electron injection and trapping followed by ionization within the liquid. This produces a plasma similar to that produced with positive polarity. The negative streamer velocity is determined by the time spent in one or the other stage, i.e., the injection or the trapping.

Even if this model is questionable, because some assumptions need to be justified [15], it is supported by two interesting facts. The first is that the addition of electron scavengers reduces the trapping distance and the time (t_1) spent in the first step and thus increases the negative velocity. The second fact is that the addition of low ionization potential additives increases the rate of ionization and reduces the time (t_2) spent in the second step, and thus still increases the velocity. The velocity computed according to this model is constant. This is not true. The propagation of the streamers is done in jumps.

In recent work, a new model perfectly describing this step propagation and the influence of additives was proposed [24,25]. This model, based on considerations related to energy, allows us to explain the pre-breakdown processes and to evaluate the propagation velocity of the streamer. According to this model, the electronic scavenger additives increase the current pulse frequency and reduce the time duration between consecutive discharges, and then increase the electrical charge that assists the streamer in its propagation. This results in an increase in the average propagation velocity of the streamer. The streamer becomes more energetic, then more filamentary, and thence rapid [24,25]. Thus, if the streamer is more rapid, the time to breakdown will be shorter. However, this does not necessarily mean that the dielectric strength (breakdown voltage) of the liquid will be low/reduced. As reported by Beroual [23] and Beroual et al. [25], electronic scavenger additives act on the initiation of the streamer will be more energetic and become faster. As long as the amount of additive has not reached the saturation threshold, all of the injected charges are captured. Beyond this saturation threshold, the electronic scavenger additive as resulting in the drop of the dielectric strength of the oil.

6. Conclusions

This work shows that the dielectric strength of liquids can be improved by adding a certain concentration of an electronic scavenger compound. Electronic scavenger additives act on the initiation of streamers by reducing the injected charge carriers. This results in an increase in the threshold voltages of the streamers and thence in an increase of the dielectric strength of the oil. The breakdown voltage increases up to a certain optimal concentration of additives. Beyond this optimal amount of additive, which can be considered the saturation level, the electronic scavenger additive accelerates the streamers, resulting in the drop in the dielectric strength of the oil.

Such a result is of great importance for industrial applications and especially for high-voltage oil-filled apparatuses.

Author Contributions: Conceptualization, U.K. and A.B.; Methodology and Software, U.K.; Validation, A.B.; Formal Analysis, Investigation, Resources, Data Curation, A.B. and U.K.; Writing-Original Draft Preparation, A.B.; Writing-Review & Editing, A.B. and U.K.; Project Administration and Funding Acquisition, U.K.

Funding: This research was funded by King Saud University grant number ISPP#47.

Acknowledgments: The authors extend their appreciation to the International Scientific Partnership Program (ISPP) at King Saud University for funding this research work through ISPP#47.

Conflicts of Interest: The authors declare no conflicts of interest.

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