IRREGULARITY IN CHARACTERISTICS OF HIGH FREQUENCY BREAKDOWN VOLTAGE OF POLAR LIQUIDS

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In the field of study on electric breakdown phenomena, there are comparatively few reports on liquid dielectrics in high frequency field. Previously Schlegelmilch,[1] R. Becker,[2] Hirano[3] and Masaki[4] studied in this field and reported that the high frequency breakdown voltage of liquid dielectrics decreased with increasing frequency, and so, high frequency breakdown of liquid was utterly the so called *thermal breakdown* owing to the dielectric loss. The author has continued to study in this field these years and has reported some data[5] on the irregular phenomena which were seen only in polar liquids.

Recently, some of the causes of that irregular phenomena have been made clear enough to be reported in this paper.

I. Apparatus

High frequency voltage is generated by a vacuum tube oscillator with RT-356, maximum output is 2 KW, and stepped with a tuned secondary circuit up to 10 KV in max., its frequency can be varied from 1 to 30 megacycles. The voltage is measured with a cathode ray oscillograph which is calibrated with a *conductor-rod-pendulum* voltmeter.

Both of sphere electrodes and needle-to-plane electrodes have been used. But in consideration of the fact that sphere electrodes need very high field intensity to bring about breakdown and yet the frequency characteristics with sphere electrodes are entirely similar to that with needle-to-plane electrodes, most of the following experiments have been done with needle-to-plane ones.

Gap length has been measured by a micro-meter of accuracy of 1/100 mm., this accuracy seems to be sufficient for my purpose with the consideration of the fact that gaps mostly used were 0.5 to 2.0 mm..

II. Materials

Measurement has been made on dielectrics, both of polar and of non-polar liquids. As the representative materials for non-polar liquids, carbon-tetrachrolide, normal hexane, benzene and transformer oil have been used, and for polar ones, nitrobenzene, ortho-nitrotoluene, toluene and ethyl-alcohol. These materials were purified by chemical process and distillation in the laboratorium.

III. Measurement on non-polar liquids

At first, the frequency characteristics of high frequency breakdown voltage of ordinary non-polar liquids will be shown. They are in Fig. 1 to Fig. 4. Full lines in Fig. 1 are the curves of transformer oil with needle-to-plane electrodes in the frequency range of 1 to 8 megacycles. Schlegelmilch previously measured transformer oil in the same frequency range with "Spitze-Kalotte" electrodes, his data of 1.0 mm. gap are plotted in broken line in Fig. 1. It is clear that his curve shows good coincidence with my data. In Fig. 2 there are three curves of carbon-tetrachrolide, benzene and normal-hexane. These materials including transformer oil are all non-polar, and show nearly equal characteristics with frequency, which have monotonous decrease with increasing frequency. The fact that the inclinations of these characteristics of breakdown voltages are nearly equal to inverse proportion of square root of frequency will be intelligible when these data are plotted in logarithmic co-ordinates. These curves in logarithmic scales are written in Fig. 3, they are nearly parallel to the broken line of $V_d \propto 1/\sqrt{f}$. Thereby, we can suggest that on the above measured materials the so called "*thermal breakdown*" occurs, and the principal part of heat will be generated by high frequency dielectric heating owing to the existence of dielectric loss in materials.

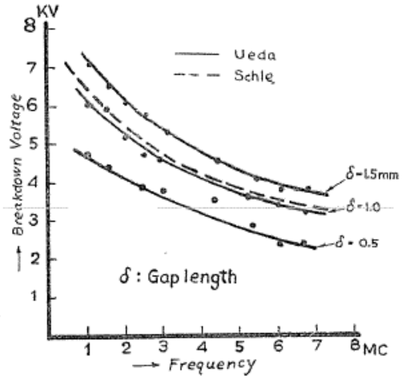

FIG. 1. Frequency characteristics of breakdown voltage of pure transformer oil.

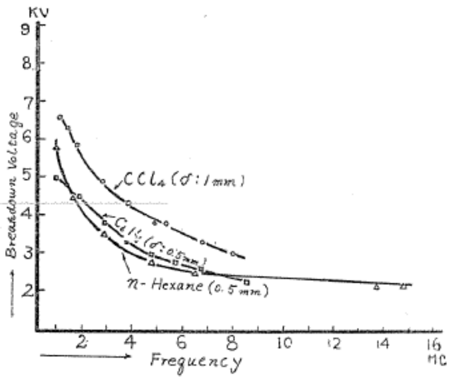

FIG. 2. Breakdown voltage of three non-polar liquids.

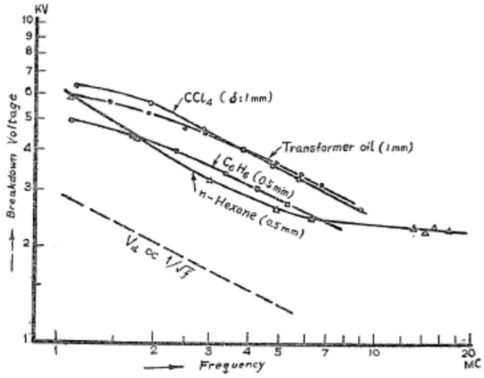

FIG. 3. Breakdown voltage of four non-polar liquids in *log-log* scales.

Since heat generation by dielectric loss is proportional to frequency, dielectric constant, dielectric loss factor and square of field intensity, it can be written in the next form ;

$$Q = E^2 \omega \varepsilon \tan \delta \qquad (1)$$

here, Q is the rate of heat generation and ω is angular frequency. Let Q' be the rate of heat diminution from the particular domain, where breakdown occurs at first, and maybe by convection of liquid to electrodes, the temperature of the particular domain will reach the value that Q and Q' come into equilibrium. The rate of heat diminution by convection depends on viscosity of liquid and shape of measuring vessel and electrodes, and is a function of temperature of liquid, therefor, it is difficult to determine the final temperature for various frequencies. Here, we suppose a characteristic temperature of liquid, let us name it "*breakdown temperature*," it will mean in some liquids boiling temperature and in others it will be such temperature that either the long chain or some bonds of organic molecule are broken or chopped. Then, breakdown condition will be completed when the temperature of equilibrium becomes higher than the "*breakdown temperature*" of the liquid in the given situation. According to this, as far as the gap length in settled vessel is constant, we can consider Q' to be constant and Q, at the *breakdown temperature*, also to be constant.

So, the breakdown condition is :

$$E^2 \omega \varepsilon \tan \delta = \text{const.} \qquad (2)$$

In this formula, ε and $\tan \delta$ are not constant but functions of frequency and temperature. But, by the same reason, we need not think over their dependence upon temperature. Frequency dependence we can consider to be little in the used frequency range with respect to the above mentioned four liquids.

Therefor, frequency characteristics of the breakdown voltage of liquid where the *thermal breakdown* occurs are simply written in the next form :

$$V_d^2 \cdot f = \text{const.} \quad \text{or} \quad V_d \propto 1/\sqrt{f}. \qquad (3)$$

Curves in the above written three figures have an inclination nearly equal to this formula.

IV. Measurement on polar liquids

Now, the frequency characteristics of breakdown voltage on polar liquids are to be shown. They are in Fig. 4 to Fig. 7.

Fig. 4 shows curves of ethyl-alcohol and Fig. 5 of toluene. Curves of ethyl-alcohol are quite different from ordinary curves of non-polar liquids ever shown in the frequency range of 1 to 3 megacycles, i.e., in the neighborhood of 2 MC, they are not smooth but have small peaks. Such tendency is clearer on curves of toluene shown in Fig. 5. Evidently, toluene has the highest breakdown voltage at 2 MC so far as this order of frequency is concerned.

Furthermore, such a phenomenon that breakdown voltage does not decrease with increasing frequency has been discovered on ortho-nitrotoluene and nitrobenzene as shown in Fig. 6 and Fig. 7 respectively by full lines. And it has been

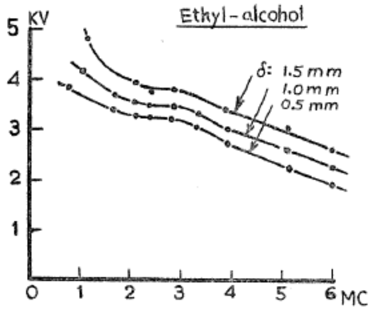

FIG. 4. Breakdown voltage of ethyl-alcohol.

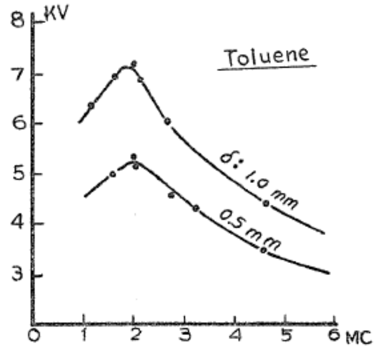

FIG. 5. Breakdown voltage of toluene.

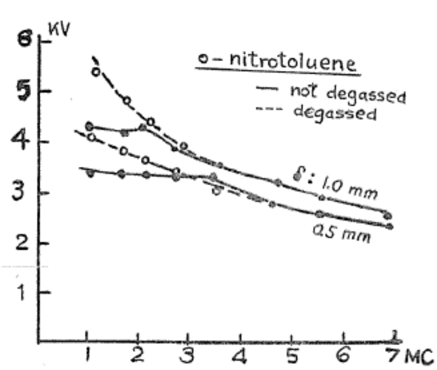

FIG. 6. Breakdown voltage of ortho-nitrotoluene, both of degassed and not degassed.

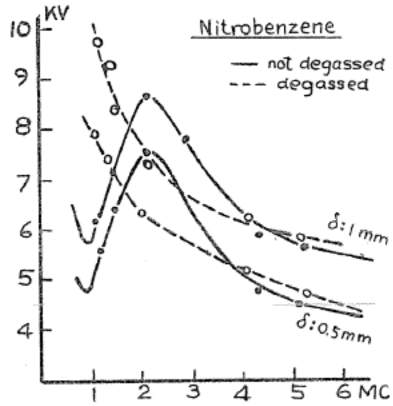

FIG. 7. Breakdown voltage of nitrobenzene, both of degassed and not degassed.

made sure that the frequency at which the highest breakdown voltage appears does not vary by varying gap length or shapes of electrodes. Moreover, the nitrobenzene which has been treated by very carefull purification including distillation and degassing sometimes does not show this irregular characteristics. Now it has been made clear that the entirely degassed liquid has not any kind of irregularity on its frequency characteristics of high frequency breakdown voltage.

Broken lines in Fig. 6 and Fig. 7 are curves of ortho-nitrotoluene and nitrobenzene, respectively, entirely degassed in bell-jar after carefully distilled.

The origin of those irregular characteristics has been throughly investigated with nitrobenzene, and it has been clear that moisture has the greatest effect in absorbed gasses. This experiment has been carried by means of the following process and the results are shown in Fig. 8.

The process and symbols are as follows:

(*a*):—entirely degassed nitrobenzene after carefully distilled.

(*b*):—let (*a*) in the air for a long time, removing from it CO_2 and H_2O by soda-lime and P_2O_5 respectively.

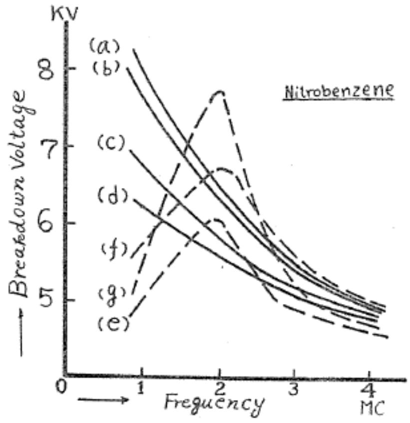

FIG. 8. Difference of frequency characteristics of breakdown voltage of nitrobenzene, treated by means of various methods.

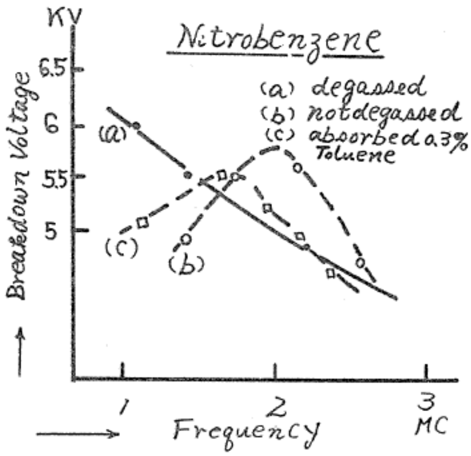

FIG. 9. Difference of frequency characteristics of breakdown voltage of nitrobenzene, absorbed different gasses.

(*c*):—let (*a*) in CO_2 gas for 3 hours.
(*d*):—let (*a*) in CO_2 gas for 20 hours.
(*e*):—moisture throughly absorbed into (*d*).
(*f*):—let (*a*) in the air of 60% humidity for 6 hours.
(*g*):—carefully distilled nitrobenzene, but not degassed.

It is clearly shown in Fig. 8 that, however much gasses absorbed, with the exception of moisture, the characteristic curves of nitrobenzene show only monotonous decrease with frequency and there is no peak of dispersion of breakdown voltage, while, of course, the breakdown voltage becomes lower than in pure state, uniformly in all range of used frequencies. But moisture, even if its content is very little, produces very different characteristics.

According to moisture absorption, the breakdown voltage of nitrobenzene, though in the range beyond 3 MC, it becomes uniformly lower, but, at the neighbor of 2 MC, strange to say, becomes higher than that of dry nitrobenzene, and at 1 MC, it becomes quickly lower. Similar tendency will be seen in ortho-nitrotoluene. Entirely degassed toluene and ethyl-alcohol can not be tested owing to their own low boiling points.

It is remarkable that the frequency range having irregular characteristics by moisture is nearly identical for the measured four polar liquids. They are very different from each other at various points of properties, i.e., molecular weight, boiling point, viscosity and dipole moment etc.. Therefore, that value of frequency or period of field alternation can not be peculier to the solvent, but is decided by molecule of water itself or by interaction between water and molecules of polar solvent. It is well known that water molecule has big dipole moment and sometimes it attracts many other molecules around it.

For the purpose of investigating if another polar molecule absorbed into polar liquid does not move the frequency of irregularity, nitrobenzene having absorbed toluene in place of moisture has been tested.

Fig. 9 shows this result that also by toluene absorption the irregular characteristics have appeared, but the frequency of the highest breakdown voltage is not identical to the case of moisture and has moved to a little lower region. Further investigation on the last point is now being continued.

V. Discussion

There will be no question on the characteristic curves of non-polar liquids which show "thermal breakdown" in the range of 1 to 10 megacycles. But it is a difficult problem how to decide the *breakdown temperatures* of various materials. Generally, it can be said that with an ordinary simple liquid which has a comparatively lower boiling point the breakdown temperature is identical to its boiling point.

There is one point to be noted, that in Fig. 3 the breakdown voltage of normal-hexane decreases with frequency nearly parallel to the broken line showing $E \propto 1\sqrt{f}$ up to 8 MC, and thereafter its inclination becomes less, in another words, the breakdown voltage of normal-hexane varies very little between 8 and 18 MC. This phenomenon lets us suggest that beyond a particular frequency some terms contributing to breakdown condition have changed with very high frequency. I have also investigated on this point, but it will be not concerned in this paper.

With polar liquid the behavior is complicated when it includes another molecules. Of course, even polar liquid shows simple thermal breakdown when it is sufficiently pure.

In recent years, the behavior of liquid molecules under high frequency electric field has been studied and it has been made clear that there is the so called dielectric dispersion. The frequency of dispersion can be calculated from molecular diameter, viscosity and other known data of liquid. With nitrobenzene the dispersion frequency is near 1000 MC, and it is widely different from the frequency now to be discussed. Therefore we must suggest another mechanism for the kind of dispersion reported in this paper. If one suggest rotation of large particles in such low frequency dispersion, the particle must have a diameter of 10 fold of nitrobenzene molecule. With nitrobenzene well known molecular association consists of two molecules, and with ethyl-alcohol three molecules. But water molecule is known to make sometimes large *clustered* particle, so, it will not be so strange that water molecule absorbed in polar solvent may possibly make very large particle to reach the above said diameter. Besides, water molecule has very big dipole moment, and in polar liquid adhesive force for rotation of particle can be increased more than that of solvent alone, this will contribute to reduce dispersion frequency.

Then, at an available frequency the energy of electric field will be consumed to effectively rotate the *clustered* particles, and since these particles include much water, its specific heat is very large the energy consumption is so great that the solvent liquid is obstructed to reach "breakdown temperature" by dielectric heating.

According to this suggestion, it is not unreasonable that the breakdown voltage of a little wet polar liquid can become higher than that of dry one at a certain particular frequency, and besides, the frequency can move by different gas absorption.

The fact that there is the most available frequency to supply electric energy into mixture of polar liquids, for example some alcoholic liquors, and that frequency is of the order of 1 MC, has been found in these several years by Prof. K. Noguchi,[6] Nagoya University. His purpose has been to make alcoholic liquor ripen, and he has given an explanation for his result that the molecules of water and alcohol, both intense polar, make long chains and are not easy to diffuse into each other, but in the electric field of the most suitable frequency the energy is given to the chains so effectively that the chains are broken and the molecules are course to be able to mix with each other.

My result reported in this paper seems to come in agreement with Noguchi's result in a certain sense. But to completely solve such an irregular phenomenon of polar liquid, molecular behavior in high frequency field must be furthermore investigated in detail, and the author has begun the investigation which results will be reported successively.

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