

ELECTRIC BREAKDOWN PHENOMENA OF DIELECTRIC LIQUIDS IN HIGH FREQUENCY FIELD

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Introduction

Constructions of gases and solids are fairly good understood, but on the liquid state our knowledge is very little. In fluid state, in spite of that the concentration of molecules is not so different from it of solid, the intermolecular force is not so strong to construct the grids and the molecules are able to have free motions each other. Therefore, analysis by the X-ray deflection is difficult and a statistical method is not fully successful. Numerical data obtained by the experiments on liquids can not be connected to the proper constants of the media (as the molecular weight, density, molecular volume).

In the field of electrical engineering liquid dielectrics have very important places as transformer oil, circuit breaker oil, condenser oil and cable insulator or the Kerr-cell shutter medium. But on these liquid insulators we can not say the concretely decided numerical data of leakage conductivity or electric breakdown strength. These data obtained in the past by many researchers do not agree mutually by the difficulties of purification and observation.

The author studied on such difficult problems especially on characteristics of liquid under the high frequency electric field. His results are not so enough to make clear the all electric properties of liquids, but in consideration of stage of to-day they seem to have in some extent succeeded to solve the properties.

Chapter I. Measurements and Theories in the Past

§ 1.1. Electric conduction and breakdown of liquids generally

Prior to the subject, high frequency breakdown of liquids, the general breakdown phenomena of liquid dielectrics must be explained.

Under dc. electric field, conduction current in insulating liquid has the characteristic shown in Fig. 1¹⁾. In the region of A the current is very small and can not be measured by a galvanometer. It is said the in the region the current has a saturation characteristics. But the current increases in B region, and at the field E_d the current makes rapid progress to go into breakdown. In the region B there are two groups of

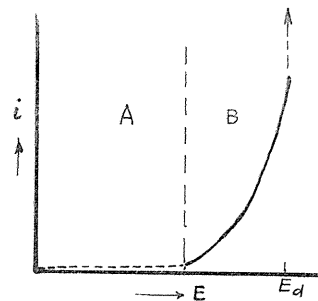


FIG. 1. Increasing process of conduction current.

theories, one is to say $i \propto \exp\{E\}$, and the other $i \propto \exp\{\sqrt{E}\}$. To-day it is said that the latter is more reasonable.

At first view the mechanism of breakdown of liquids will be considered to be the result of great progress of the mechanism of conduction, but there can be exist such a consideration that the entirely different mechanism grows up suddenly in the field. Some experiments to research this point have been made by many peoples³⁾ and the author³⁾, but now the concrete theory is not established.

The famous theories on the conduction and breakdown of liquids in the past are as follows.

(i) Theory of accumulation of ions by collision. (W. Schumann⁴⁾, A. Nikuradse⁵⁾): Criterion is, as in gases, decided by the ionization factor α and factor of electron emission of a cathode γ . On the mechanism of electron emission at the cathode has been corrected afterward by Baker and Boltz⁶⁾, LePage and Du Bridge⁶⁾, Edler⁷⁾, Zeier⁸⁾ or Dornte⁹⁾.

(ii) Theory that the concentration of ion increases by desociation by the field. (Onsager¹⁰⁾, Wien¹¹⁾, Reiss¹²⁾ and Plumley¹³⁾): The faults of this theory is that the desociation constant of pure liquid insulators is not so great that can explain the measurable conduction current. But the insulating oil for practical use can not be entirely purified as the grade of experimental use, so this theory is important practically.

(iii) Streamer theory. (Toriyama²¹⁾, Shinohara¹⁴⁾): The conductive paths present on the dust figures in oil are named streamers and seem to be traces of accumulated ions made by electron collision.

(iv) Theory that the gaseous breakdown in bubbles of vaporized liquid anticeeds. (Wagner¹⁵⁾, Becker¹⁶⁾, Güntherschultze¹⁷⁾, Inge and Walther¹⁸⁾).

(v) Theory that the gaseous breakdown in bubbles made of the absorbed gas in the liquid anticeeds. (Clark¹⁹⁾, Koppelman²⁰⁾): Hirano²²⁾ showed that the breakdown characteristics of entirely degassed liquid are very different from common oils.

It is difficult to decide which theory is the most valuable in dc, or ac, breakdown. For, in any cases whenever the breakdown occurs by thermal mechanism or pure electric (or electronic) processes, factor of electron emission, factor of desociation or electron and ion mobilities depend on the medium temperature. And the other important factor of space charge effect and the problem of boundary between the liquid and the electrode material are very difficult point to measure.

In these several years many investigations with pulse techniques have been made, as Edwards²³⁾, Salvage²⁴⁾, Sharbough, Crowe and Bragg²⁵⁾, Goodwin and Macfadyen²⁶⁾, Meksiejewski and Tropper²⁷⁾, Green²⁸⁾, Suita and Yamanaka²⁹⁾. Komellkov³⁰⁾ observed photographically the streamer in liquid. Nagao³¹⁾ also made a photograph of same trace by smoke chamber method.

At measurement by pulse method, there are few irregularities, effect of impurities and thermal affection, so, the results seem to be purely electrical and the breakdown strength by available pulse is called "Intrinsic electric strength". Dependence of the "intrinsic strength" of some liquid and its physical properties is one of the most important problem in the field of breakdown phenomena.

§ 1.2. Past data on the high frequency breakdown of liquids

There were not so many investigators who measured the breakdown strength

by high frequency field. The reason seems to be that the high frequency breakdown is more simple than the dc. or ac. breakdown. In the high frequency field, liquid and solid insulators produce fairly much heat by dielectric loss. And the field distribution is made ununiform by the temperature rise and the variation of ϵ and $\tan \delta$. Therefor, so called thermal breakdown seemed to be very easy to grow up. Naehrer³²⁾, Sorge³³⁾, Dräger³⁴⁾ researched at rather low frequency (near commercial frequency). The most famous experiment was done by Schlegelmilch³⁵⁾. He measured with transformer oil and xylene under the frequency between 4×10^5 to $1,2 \times 10^7$ c.p.s. The results are shown in Fig. 2 and Fig. 3. The breakdown strength decreases by the increasing frequency. And he observed that the effect of impurity was less than under commercial frequency.

Fig. 4 is the same data of Schlegelmilch plotted by $\log f$ vs $\log V_d$. Curves

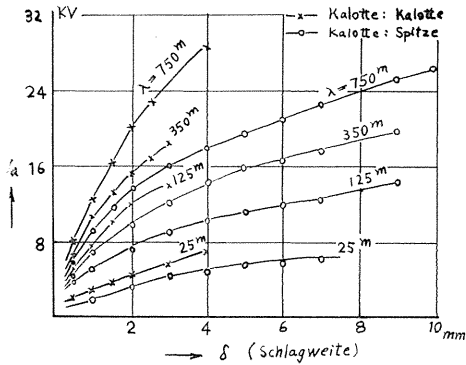


FIG. 2. Transformer oil.

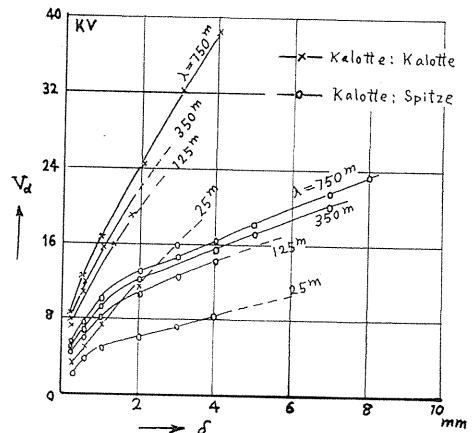


FIG. 3. Xylene.

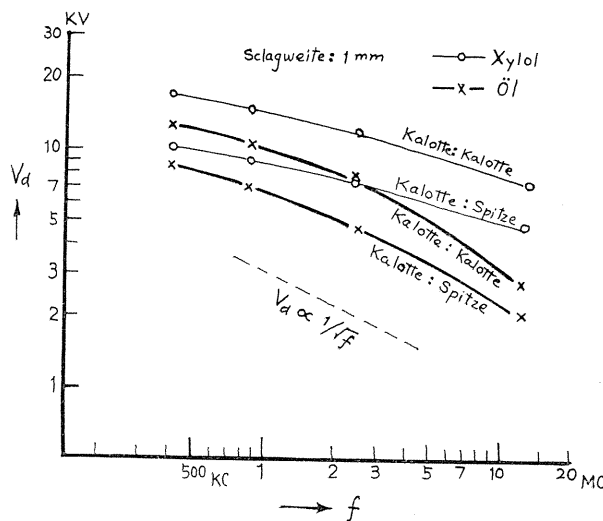


FIG. 4. Results of Sch. in log-scales.

have the inclination of $1/\sqrt{f}$. He says that $\tan \delta$ under these frequency region increases with frequency, so the breakdown strength on Fig. 4 is inverse proportional to the heat generation by the dielectric loss, and then the mechanism of breakdown is purely thermal.

Hirano and Nemoto³⁶⁾ reported such a high frequency breakdown and Masaki also studied by solid and liquid and explained the breakdown by the thermal degradation. By Masaki³⁷⁾ "the thermal degradation" was declared that differs from thermal breakdown. Under the frequency now discussed heat generation by the dielectric loss can not be neglected. But the different factors, as the accumulation of ions, desociation or space charge effect were not neglected. But the different factors, as the accumulation of ions, desociation or space charge effect were not neglected. With this point the author will explain afterwards.

§ 1.3. Consideration of mechanism of high frequency breakdown of liquid

The past theories on the high frequency breakdown of liquid dielectrics are so called "thermal breakdown". The criterion of completion of breakdown is given by the limit of local temperature rise in a small domain near the centre of the electrode separation. And the vaporation of liquid itself or absorbed gas introduces the medium to the breakdown. Ordinarily frequency characteristics and the temperature dependence of oils seem to be fully explained. But by the fine consideration about the past experiments the kinds of liquids were not so enough and the measured frequency was not continued but selected to point to point. According to the author's experiment, that the frequency has been varied continuously and the very many kinds of oil were used, some different points were discovered. For example, one of the most difficult point to explain by the past so called "thermal breakdown" theory is the temperature dependence of the breakdown voltage.

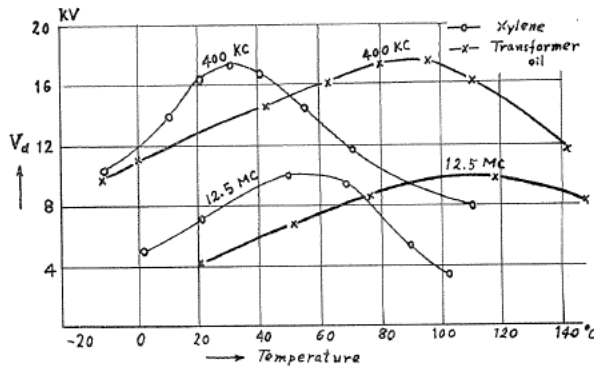


FIG. 5. Temperature dependence (Sch.).

Fig. 5 shows the representative example³⁵⁾. The breakdown voltage increases with increasing temperature but there is a maximum value and thereafter the value decreases with the increasing temperature. The oils in this figure are xylene and transformer oil, and by the transformer oil the author also gets the entirely same results. The initial part of the curve seems to be increased by the fact that the value of $\tan \delta$ of the oil decreased with the temperature rise

and the necessary field strength to form breakdown becomes higher, and the final part of the curve shows that the vaporation of the liquid becomes easy for the temperature becomes near to the boiling point.

But here is a very different figure obtained by the author³³⁾. Tested liquid is silicone oil, 800 cs* of viscosity, and the used frequency are 1.4 and 6.5 MC. There is a very different character between the 1.4 and 6.5 MC. The former shows nearly same shape of the transformer oil, but the effect of temperature on the ϵ and $\tan \delta$ of silicone oil is very small, so we can not explain the increase of breakdown voltage in the Fig. 6 by the decrease of $\tan \delta$. (The available explanation will be presented in the later part of this paper). Besides, the latter curve does not possibly seem to be explained by the variation of heat generation. It needs to introduce the increasing of ion mobility by the decreasing viscosity.

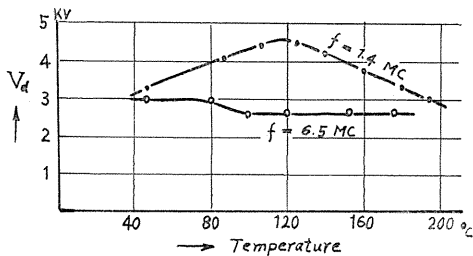


FIG. 6. Temperature dependence of high frequency breakdown voltage of 800 cs silicone oil.

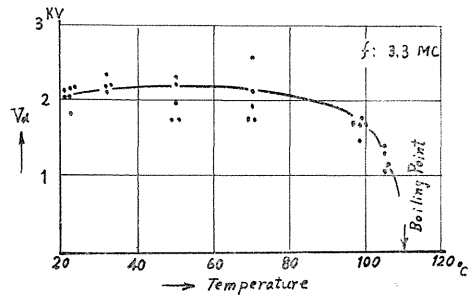


FIG. 7. Temp. dep. toluene.

Another example is the temperature dependence of high frequency breakdown of toluene, i.e. Fig. 7³³⁾. There is no temperature dependence up to 80°C, and in the very narrow range between 90°C and 110°C the curve goes downward very rapidly. At the temperature range near the boiling point, most liquids show the characteristics of the "thermal breakdown", but at the range of low temperature we must search for a new theory.

Next consideration is about the "breakdown temperature".

In the region where the "thermal breakdown" goes we must suppose a value of "breakdown temperature". When the temperature of any location of medium becomes higher than the breakdown temperature, the electric breakdown will grow up. In the ordinary liquid we can easily consider this temperature is equal to the boiling point, as Fig. 6 shows. But with the silicone oils, there are not boiling temperature. By heating their long chains were cut off and the molecular construction is changed. With silicone oil, i.e. poly-dimethylsiloxane, we can discover production of silica by higher temperature than 200°C. By the electric breakdown also we can see the isolation of silica. This fact shows that the energy of electric discharge is not given at first to the methyl base and thereafter to the bond of -Si-O-, but there is same probability that the electric energy is given to the bond of -Si-O- at first. By this fact, it is difficult to decide the breakdown temperature. In this case the breakdown temperature

* Centi-stokes at room temperature.

means some wide region of temperature, and then it becomes clear that the curve of silicone oil with 1.4 MC goes down slowly at the region of higher temperature.

Chapter II. New Measurements on the High Frequency Breakdown of Liquids

§ 2.1. Equipment

1. Source of high frequency

Source and the measuring circuit are shown in Fig. 8. There are not so difficulties. Frequency range is from 1 to 30 MC. At the special measurement the large source of 20 kW out put was used.

2. Measurement of voltage

High frequency voltages were measured by a cathode ray oscilloscope, calibrated by the pendulum voltmeter. The frequency dependence of measuring circuit is always checked and sometimes they were reconstructed for the purpose of check of frequency dependence of all circuit.

3. Vessels and electrodes

Vessels were made of pyrex glass and have volume of 3 to 4 cc. Electrodes are needle to plane, and the needle is moved by a micrometer.

4. Tested liquid

i) Non-polar liquids:—Transformer oil, aliphatic hydrocarbons (C_5H_{12} , C_6H_{14} , ... $C_{16}H_{34}$), carbon tetrachloride, benzene etc.

ii) Dipolar liquids:—Pure water, alcohols (methyl alcohol, propyl alcohol, amyl alcohol), toluene (weak dipolar), nitrobenzene, nitrotoluene, chlorobenzene, dichlorobenzene, xylene etc.

iii) Artificial higher polymer liquids: Silicone oils (viscosities of 20 cs. to 30,000 cs.). They are included in dipolar liquids from their molecular construction but their character is same as non-polar ones.

These liquids were purified and degassed in laboratory from commercial highest class.

§ 2.2. Frequency characteristics generally

1. Frequency characteristics of high frequency breakdown voltage of transformer oil

Fully degassed mineral oil are measured under the continued high frequency voltage. The result in case of needle-plane gap is shown in Fig. 9. Full lines are obtained by the author at three different gaps and the broken line is the result of Schlegelmilch³⁵⁾ with the Kallotten-electrodes of 1.0 mm gap. They show very good agreement. This shows that the values of high frequency breakdown voltage are very reproducible under good arranged purifying condition.

Fig. 10 shows same curves which are replotted by logarithmic co-ordinates. Broken line shows inclination of $V_d \propto \sqrt{f}$. This result says that the breakdown voltage of transformer oil at higher frequency than 2 MC goes down nearly parallel to the broken line. At the region near 1 MC the inclination is

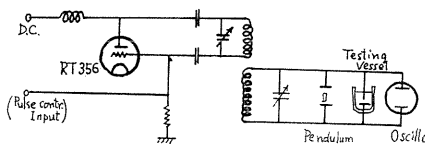


FIG. 8

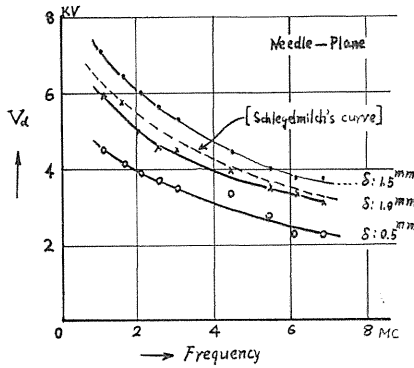


FIG. 9. H. F. breakdown of transformer oil.

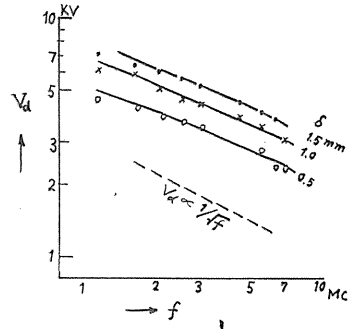


FIG. 10. Breakdown voltage vs frequency in log-scales.

rather small. In the frequency range of $10^5 \sim 10^6$ cps. dielectric loss factor of transformer oil is not constant, and the fact that the curves in Fig. 10 have inclination a little different from the line of $1/\sqrt{f}$ may agree in some quantities to this variation of loss factor.

2. Effect of absorbed gasses

In case of transformer oil, effects of absorbed gasses on the high frequency breakdown is less than that on the dc. or ac. breakdown voltage. Fig. 11 shows these results for comparison with the case of dipolar liquid, which is explained afterwards.

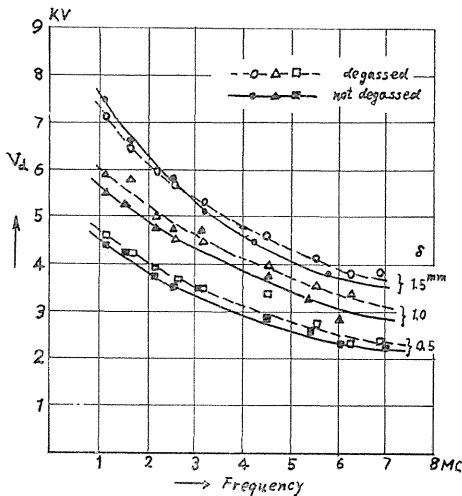


FIG. 11. Effect of degassing of transformer oil.

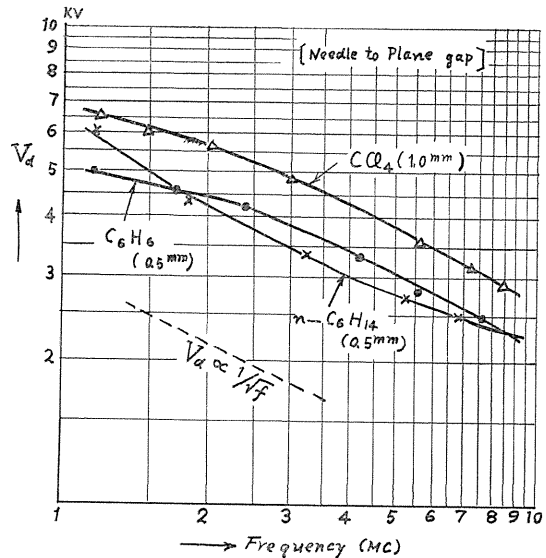


FIG. 12. H. F. breakdown voltage of pure liquids.

3. Non-polar liquid except transformer oil

CCl_4 , C_6H_6 , $n-C_6H_{14}$ etc. have nearly equal characteristics to transformer oil. These are shown in Fig. 12. In Fig. 13 curves of silicone oil (20 cs), i.e. TS-

951, Toshiba, polydimethylsiloxane. From the molecular construction, silicone oils must have some dipolar moment but they have very long chain and for the effect of configuration they show nearly same character to non-polar liquids. Someone will find that the silicone oil, which is believed to have very excellent dielectric characteristics, has unexpectedly low breakdown strength in the frequency range of 1 MC to 10 MC.

4. Phenyl-ethanolamine etc.

In Fig. 12 we can see one noticeable slope in the curve of normal hexane. This curve crosses to the curve of benzene at 8 MC, in other words, at the both frequency range of lower part than 8 MC and higher part than 8 MC, the order of values of breakdown voltage of hexane and benzene are reciprocal. And in higher frequency region, the curve of benzene goes decreasing, but it of hexane goes nearly flat. Resembled characteristics are seen with ortho-dichlorobenzene and phenyl-ethanolamine. These results are shown in Fig. 14. In this figure a curve of CCl_4 is plotted for comparison, and it goes nearly parallel to the line of $V_d \propto 1/\sqrt{f}$ at almost measured frequency range. The other three curves go nearly parallel to $1/\sqrt{f}$ line at lower frequency region, but they become flat at higher frequency region. Boundary frequencies of the both regions for ortho-dichlorobenzene, hexane, phenylethanolamine are read to be 3 MC, 8 MC and 3 MC respectively. Since in these three media hexane is non-polar and the other two are dipolar liquids, molecular polarity seems to be independent on the above mentioned phenomenon. For the question what is the

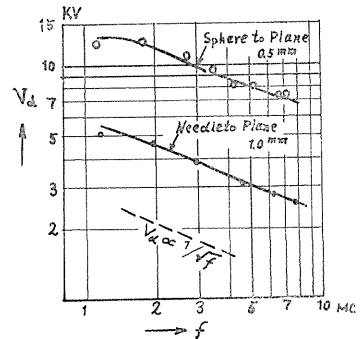


FIG. 13. H. F. breakdown voltage of 20 cs silicone oil.

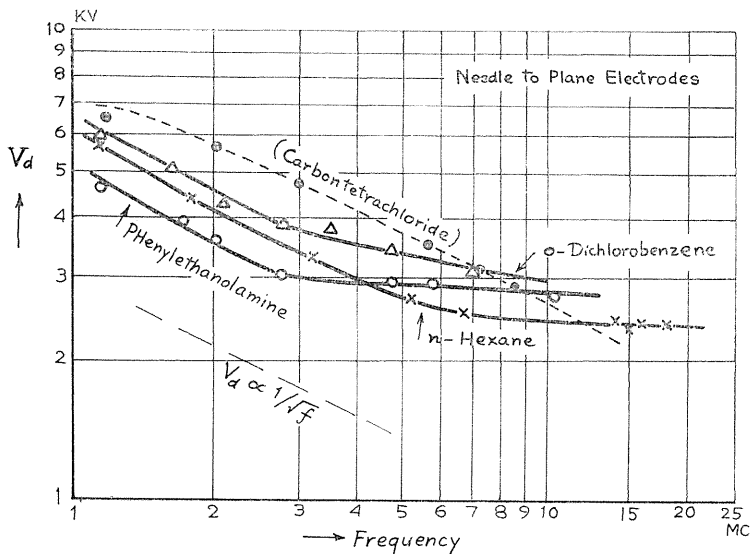


FIG. 14. H. F. breakdown voltage of phenylethanolamine etc.

mechanism of the frequency independent breakdown strength, the author will explain one opinion afterwards. By simple consideration we can say that the media which have the frequency independent breakdown strength are more or less viscous ones.

5. High frequency breakdown strength of silicone oil

The frequency range in which hexane or phenyl-ethanolamine have frequency independent breakdown strength is seen clealy with silicone oils. Fig. 15 shows the curves of high frequency breakdown voltages of silicone oils of 20, 100, and 3,000 cs. with the sphere-plane electrodes. In this figure the curve of 20 cs resembles to that of transformer oil, but the curve of 100 cs becomes flat at higher region than 5 MC and the 3,000 cs goes almost flat at the whole measured frequency range. In other words, at the frequency between 4 to 5 MC the order of values of breakdown strength of silicones become reciprocal. In lower frequency range, the lower the viscosity, the higher is the strength, but in higher frequency range, the order of viscosities is same to the order of strength. Since the substancial constants as dielectric constant, dielectric loss factor or breakdown temperature may not be varied by these frequency, we must recognize that a different mechanism grows up in higher frequency range.

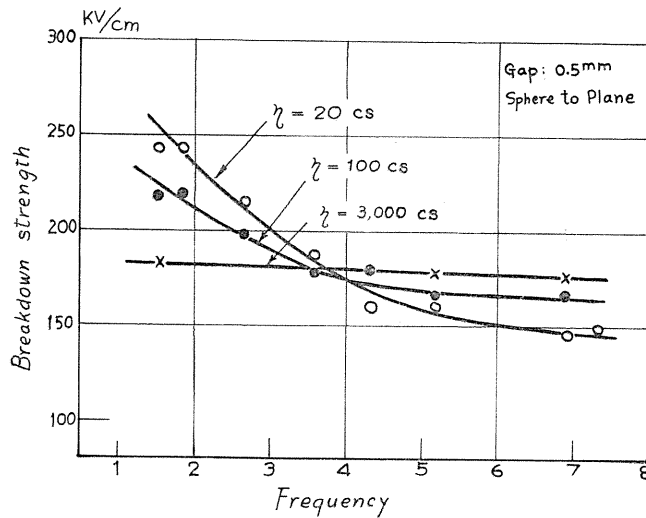


FIG. 15. H. F. breakdown strength of silicone oils.

§ 2.3. Temperature dependence

1. Results of measurement

Some data were already shown in § 1.3, but now more detailed data will be discussed. Fig. 16 is the same figure to Fig. 7. Fig. 17 and Fig. 18 are the temperature dependence of nitrobenzene and xylene respectively. Both curves are independent of temperature up to the temperature which is lower than boiling point by 30°C, and then they become rapidly lower in the range of 30°C up to the boiling point.

2. Condition of breakdown

Here the formula of thermal equilibrium up to the breakdown under the

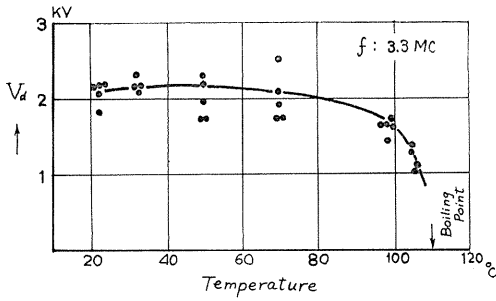


FIG. 16. Temperature dependence of toluene.

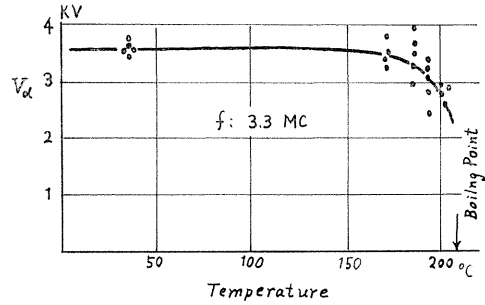


FIG. 17. Pure nitrobenzene.

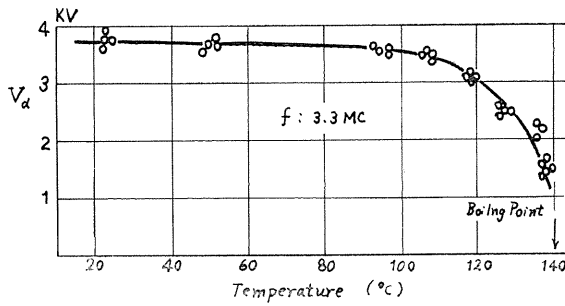


FIG. 18. Xylene.

uniform field condition as Fig. 19 must be considered. Thermal equilibrium in local small volume dx is

$$Qdx = -\lambda d(d\theta/dx) \tag{1}$$

or

$$Q = -\lambda(d^2\theta/dx^2) \tag{1'}$$

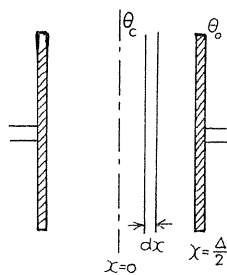


FIG. 19

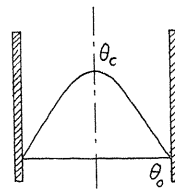


FIG. 20

here, Q is rate of heat generation, λ is factor of heat conduction of medium. The solution is simple when Q and λ are constant,

$$\theta = \theta_c - \frac{1}{2} \frac{Q}{\lambda} x^2 \tag{2}$$

here, θ_c is the temperature of central part. We put θ_0 the temperature of the

electrodes,

$$\theta_0 = \theta_c - \frac{1}{2} \frac{Q}{\lambda} \left(\frac{d}{2} \right)^2 \quad (3)$$

or

$$\theta_c = \theta_0 + \frac{1}{2} \frac{Q}{\lambda} \left(\frac{d}{2} \right)^2 \quad (4)$$

We can consider the breakdown occurs when θ_c is equal to the breakdown temperature θ_d , then

$$\theta_d = \theta_0 + \frac{1}{2} \frac{Q}{\lambda} \left(\frac{d}{2} \right)^2 \quad (5)$$

If Q is proportional to the square of field intensity E , for example

$$Q \propto fE^2 \epsilon \tan \delta \quad (6)$$

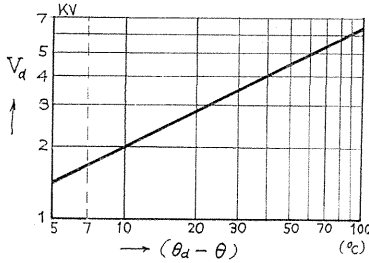


FIG. 21. Temperature characteristic of thermal breakdown model.

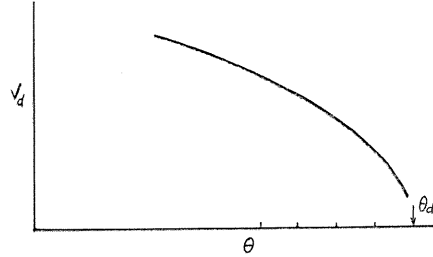


FIG. 22. Temperature characteristic of thermal breakdown model.

then breakdown field E_d becomes proportional to $\sqrt{\theta_d - \theta_0}$. This curve is shown in Fig. 21 and Fig. 22. In liquid dielectrics the heat conduction will be not without convection. But if we consider the factor of convection to neighbour location to be proportional to the temperature gradient of this part, the formula will be same to eq. (5). In spite, we must consider different compensation. That is the fact that heat generation Q , given by the following formula

$$Q = (5/9)E^2 f \epsilon \tan \delta W / \text{cm}^3, \quad (7)$$

is not constant, for ϵ and $\tan \delta$ are functions of temperature. $\tan \delta$ varies nearly proportional to an exponential function of temperature:

$$\tan \delta = \tan \delta_0 e^{\alpha \theta} \quad (8)$$

With transformer oil, if we put $\tan \delta: \phi$ 0.0005, then α becomes 0.041, and the eq. (7) becomes

$$Q = (5/9)E^2 f \epsilon \tan \delta_0 e^{\alpha \theta} = Q_0 e^{\alpha \theta} \quad (9)$$

and the equation of heat conduction becomes

$$d^2 \theta / dx^2 = - (Q/\lambda) e^{\alpha \theta} \quad (10)$$

then we can get the following solution:

$$\frac{d\theta}{dx} = \pm \sqrt{\frac{2Q_0}{\alpha\lambda} e^{\alpha\theta_c} \cdot \sqrt{1 - e^{-\alpha(\theta_c - \theta)}}} \quad (11)$$

From the value of α , above mentioned, in the range of $\theta_c - \theta < 5^\circ\text{C}$, $\alpha(\theta_c - \theta)$ is less than 0.2, and $e^{-\alpha(\theta_c - \theta)}$ is nearly equal to $1 - \alpha(\theta_c - \theta)$, then;

$$\frac{d\theta}{dx} \doteq \pm \sqrt{\frac{2Q_0}{\alpha\lambda} \cdot e^{\alpha\theta_c} \cdot \sqrt{\alpha(\theta_c - \theta)}} \quad (12)$$

In the range of $\theta_c - \theta > 40^\circ\text{C}$, $\alpha(\theta_c - \theta)$ is larger than 1.6, and nearly:

$$\sqrt{1 - e^{-\alpha(\theta_c - \theta)}} \doteq 1 - \frac{1}{2} e^{-\alpha(\theta_c - \theta)} \doteq 1,$$

then

$$\frac{d\theta}{dx} \doteq \pm \sqrt{\frac{2Q_0}{\alpha\lambda} e^{\alpha\theta_c}} : \text{nearly constant} \quad (13)$$

So, the temperature distribution between electrodes is presented by Fig. 23. At the central part the curve is nearly parabola and the other part nearly linear.

The solution of eq. (11) is:

$$\pm x = \frac{1}{\alpha \sqrt{\frac{2Q_0}{\alpha\lambda} e^{\alpha\theta_c}}} \cdot \log [e^{\alpha/2(\theta_c - \theta)} - \sqrt{e^{\alpha(\theta_c - \theta)} - 1}] \quad (14)$$

here, when $\theta_c - \theta > 40^\circ\text{C}$ next equation is get;

$$\sqrt{\frac{2\alpha Q_0}{\lambda} e^{\alpha\theta_c}} \cdot x = \frac{\alpha}{2} (\theta_c - \theta) + \log 2 \quad (15)$$

Put the electrode temperature θ_0 , i.e. $x = \Delta/2 \rightarrow \theta = \theta_0$

$$\sqrt{\frac{2\alpha Q_0}{\lambda} e^{\alpha\theta_c}} \cdot \Delta = \alpha(\theta_c - \theta) + 2 \log 2 \quad (16)$$

With the equation there may be such a condition that here is no definite value of θ_c when all value except θ_c were given. Clearly such condition exists when Q_0 is very large or λ is very small, and such a condition is unexpective in here treated liquid dielectrics. In other words, the "thermal breakdown" defined by Masaki³⁷⁾ does not consist in liquid dielectrics.

From eq. (16), by varying θ_0 required value of Q_0 for θ_c to be equal to a value of θ_d is given. It shows that $\sqrt{Q_0}$ is linear with $\theta_c - \theta_0$, and the inclination of the line is calculated by followings, for transformer oil:

$$\left. \begin{aligned} \alpha &= 0.04, \theta_c = 200^\circ\text{C}, \Delta = 0.5 \text{ mm}, \\ \lambda &= 0.1 \text{ KCal/m.h.deg.} = 10^6/0.86 \text{ KW/cm.deg.} \end{aligned} \right\} \quad (17)$$

$$\text{there} \quad e^{\alpha\theta_c} = 3,000, \quad \frac{\partial \sqrt{Q_0}}{\partial (\sqrt{\theta_c - \theta_0})} = 5.4 \times 10^{-4} \quad (18)$$

And from the relation

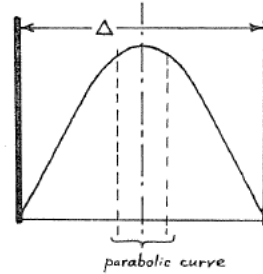


FIG. 23

$$Q_0 = (5/9)fE^2\varepsilon \tan \delta \times 10^{-9} KW/cm^3 \tag{19}$$

$\tan \delta = 0.0005, \varepsilon = 2,$

$$\frac{5}{9} f\varepsilon \tan \delta \times 10^{-9} = 0.55 \times 10^{-6} \tag{20}$$

then
$$\frac{\partial E}{\partial(\theta_c - \theta_0)} = 0.75 \tag{21}$$

This is very small quantity. Since if the temperature varies by 100°C, the necessitive electric field varies less than 100 V/cm. The author says finally that the electric field which makes the temperature of central part to become a decided value does not vary in the temperature range lower than boiling point by 40°C, and it decreases proportional to square root of $(\theta_d - \theta_0)$ at the range near boiling point. This is shown in Fig. 24.

In above written calculation the variation of a factor of heat conduction with variation of temperature is not considered. In liquid dielectrics λ is a factor of convection and is decided by viscosity. Viscosity of liquid is known to vary exponentially with temperature, so we can write λ in shape of $e^{\alpha\theta}$. Then in the right side of the eq. (10) instead of $e^{\alpha\theta}$ we may put $e^{(\alpha-\beta)\theta}$. So the

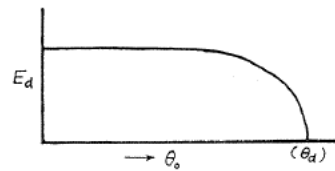


FIG. 24

above mentioned result does not vary essentially. The viscous the fluid, the larger is the variation of viscosity with temperature. So in some range by the increasing temperature the increase of heat conduction by decrease of viscosity is larger than increase of heat generation, and the breakdown strength increases by increasing temperature. Silicone oil of 800 cs in Fig. 6 shows clearly these characteristic.

With the substances which $\tan \delta$ decreases by increasing temperature, the breakdown voltage increases by increasing temperature, said by Schlegelmilch, is not correct. The effect of decrease of $\tan \delta$ upon the increase of breakdown strength is negligible. For increase of breakdown strength it needs the fairly large decrement of viscosity.

§ 2.4. Effects of electrode materials

1. Results

With the needle-plane electrodes, materials of the needle (curvature of 0.5 mm) were silver, copper and iron, and the plane copper and silver. Media are normal hexane and 800 cs silicone oil. The results are shown in Table 1 to Table 4.

TAELE 1. H. F. Breakdown voltage of *n*-hexane

Rod-electrode materials→		Silver	Copper	Iron
Freq. ↓				
Plane-electrode Copper	1.3 MC		8.5 KV	7.8 KV
	1.9	7.9 KV	7.0	6.8
	2.7	7.05	7.1	7.1

TABLE 2. H. F. Breakdown voltage of *n*-hexane

Rod-electrode materials→ Freq. ↓		Silver	Copper	Iron
Planeelectrode Silver	1.2 MC	11.5 KV	12.1 KV	12.0 KV
	3.3	9.3	8.85	8.9
	5.5	8.1	7.8	7.9

TABLE 2. H. F. Breakdown voltage of 800 cs silicons oil

Rod-electrode materials→ Freq. ↓		Silver	Copper	Iron
Planeelectrode Copper	3.7 MC	6.45 KV	6.0 KV	6.2 KV
	5.3	6.25	6.2	6.3

TABLE 4. H. F. Breakdown voltage of 800 cs silicone oil

Rod-electrode materials→ Freq. ↓	Silver		Copper		Iron		
	Mean	Dev.	Mean	Dev.	Mean	Dev.	
Plane-electrod Silver	1.2 MC	8.9 KV	1.7 KV	8.0 KV	2.2 KV	7.7 KV	1.6 KV
	1.9	7.6	1.7	7.1	1.4	6.7	1.0
	3.7	6.4	0.7	6.6	0.4	7.7	1.4

With both of hexane and silicone oil the order of the breakdown voltage is as $Ag > Cu > Fe$ on needle materials and $Ag > Cu$ on plane materials. This sequence is same to the order of materials for dc. or ac. breakdown strength. This sequence had been said to be the order of work function or heat conductivity. But 1942 Hirano⁽⁴³⁾ said this was the order of gas absorbability of metals. The order of above used metals is entirely same by work functions, heat conductivities or gas absorbabilities. So from only these results which sequence is correct is not decided. Now we plot the data in frequency vs V_d in Fig. 25. These curves become very close each other at higher frequency range. It seems that with the frequency being higher, the effect of electrode materials becomes less.

In § 2.2 the author has shown that the breakdown characteristic of viscous silicone oil at the frequency range higher than 4 MC is more or less different from a simple thermal breakdown. Here also the author has shown that in these frequency range the effect of electrode materials also diminishes. These facts show that the cooling effect by the heat conduction of electrodes becomes less and more or less different breakdown mechanism grows up. Even if the effect of electrode material is not the effect of cooling, but effect of work function or gas absorbability, the fact that the effect diminishes shows existence of electrodes to become less effective on the breakdown.

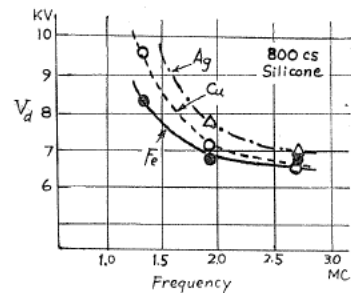


FIG. 25. Effect of electrode materials.

§ 2.5. Measurement on dipolar liquids

1. Frequency dependence

Representative dipolar liquids, ethyl alcohol, nitrobenzene and nitrotoluene, and a weak dipolar liquid: toluene were used and the results are shown in from Fig. 26 to Fig. 29. When one compares the curves of Fig. 26, Fig. 29, full lines of Fig. 27 and Fig. 28 to the curves in Fig. 9 and Fig. 12 very different characteristics between dipolar liquids and non-dipolar liquids will be discovered. These dipolar liquids have very abnormal characters in the range of 1 MC to 3 MC, i.e. here is a maximum value of breakdown voltage at near about 2 MC. There may be some doubt in these experiments since these were measured with a needle-plane gap, i.e. very ununiform field. In order to make sure the facts experiments with sphere-plane gap (6.35 mm ϕ) were made and the results for nitrobenzene are shown in Fig. 30. In this figure is also a peak of breakdown voltage near 2 MC.

In Fig. 27 and Fig. 28 there are dotted lines which are measured by nitrobenzene and nitrotoluene fully degassed after distilled. These curves have no peak between 1 to 3 MC and they resemble to curves of non-polar liquids. That is to say, these two liquids have no irregularities when they are entirely degassed. Ethyl-alcohol and toluene have rather low boiling point, and so they can not be entirely degassed. Nitrobenzene in Fig. 27 has a very strange character that near 2 MC the gas-absorbed medium has higher breakdown strength than gas-free one. Generally it is well known that impurities in liquid insulators must make the electric strength become lower by the ununiformity of electric field at the part of impurities or increase of ability of ion generation of impurities. It is very difficult to understand that impurities make the strength of medium become higher. In the past Toriyama and Sakamoto reported that breakdown strength of alcohol including little Na was higher than that of pure

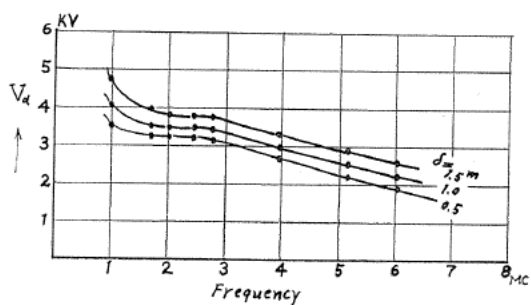


FIG. 26. H. F. Breakdown voltage of ethylalcohol.

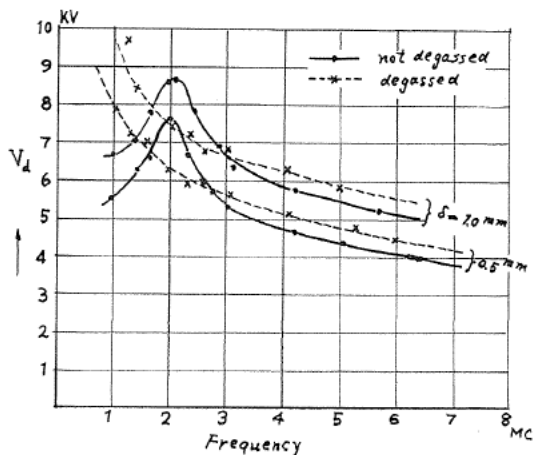


FIG. 27. Nitrobenzene.

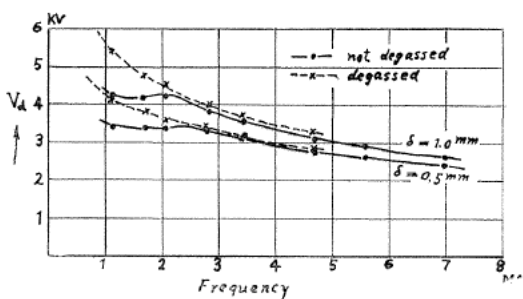


FIG. 28. H. F. Breakdown voltage of o-nitrotoluene.

one, but the reason was unknown. Here the author notice the weak dipolar liquid, as toluene, also has a clear increase of strength.

2. Detailed experiments on the effect of degassing

With nitrobenzene which component is a leading role in absorbed gasses is investigated. Signatures in Fig. 31 present the following treatments:

- (a): Pure medium; i.e. distilled and entirely degassed.
- (b): Pure medium put in air which is dried and CO_2 removed by P_2O_5 and sodalime; i.e. absorbed O_2 and N_2 only.
- (c): Put in CO_2 gas 3 hours.
- (d): Put in CO_2 gas 20 hours.
- (e): Put in air of 60% humidity.
- (f): Medium distilled only, without degassed.
- (g): Water saturated nitrobenzene.

From the results, not only the entirely pure nitrobenzene, but also

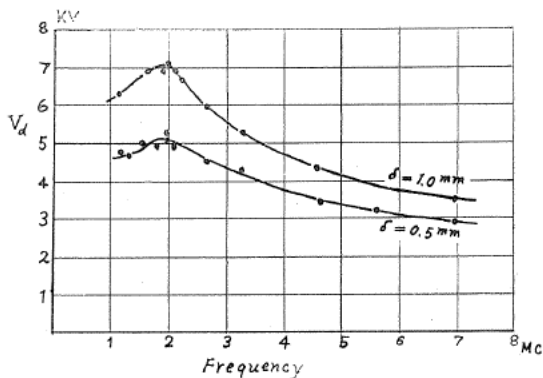


FIG. 29. Toluene.

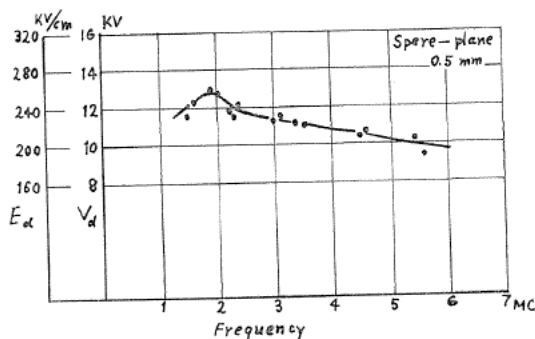
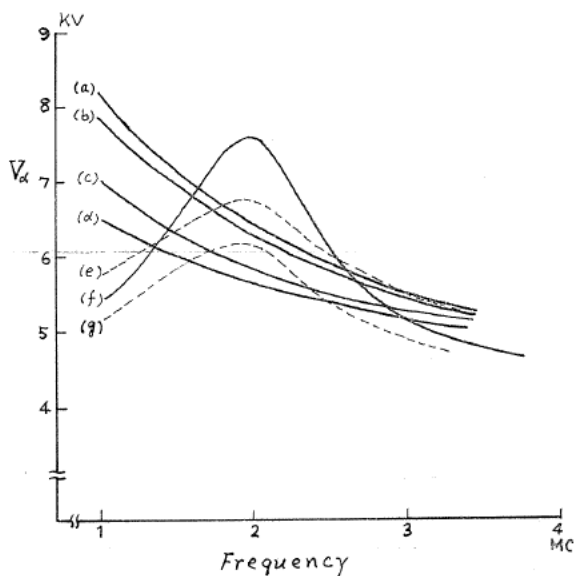


FIG. 30. Breakdown strength of nitrobenzene.



- (a) Pure medium, entirely degassed.
- (b) Absorbs O_2 and N_2 only.
- (c) Absorbs CO_2 .
- (d) Absorbs a lots of CO_2 .
- (e) Absorbs gasses including H_2O .
- (f) Same to the above.
- (g) Saturated by water.

FIG. 31. Dependence of kinds of including gasses on H. F. breakdown voltage of nitrobenzene.

N_2 and O_2 including one has a monotonous decreasing frequency characteristic. The value of breakdown strength is a little lower than pure one. And the curve (c) and (d) show that the medium including CO_2 has also non irregularity except the value being fairely low. On the contrary, the curve (e), it includes a little water, has a peak at near 2 MC. The values of breakdown strength at the both ranges of lower than 1 MC and higher than 4 MC are lower than that of pure medium but at near 2 MC the value is higher than that of pure one. The curve (f) must include more water than (e), and the peak is clearer and higher. The curve (g) has much more water than (f), and this curve is lower than (f) at whole frequency range, but his shape resembles to (f).

From this experiment, it was made clear that the irregularity of the frequency characteristic of nitrobenzene is made by the effect of water. With nitro-toluene we can easily expect the same result. The reason why we can not get the monotonous curves by degassed ethyl alcohol and toluene is now very clear. The difficulty of separation of water from ethyl alcohol is well known, and the boiling point of toluene is near $100^\circ C$.

Water as impurity in dielectrics was decided to have a very different character. But, why in transformer oil, hexane and benzene does he not show such a irregularity? We can assume that water shows some irregularities only in dipolar liquid dielectrics. With non-dipolar liquids, any impurities are included, the breakdown strength becomes lower uniformly in whole range. Besides, even with dipolar liquids, when they include non-polar molecules as N_2 or O_2 , the curve of breakdown strength goes down uniformly in whole frequency range and its shape does not vary essentially. Toluene as well known has small dipolar moment, and clearly presents irregularity by including water, i.e. now the value of dipole moment is not a problem, whether the liquid has or has not dipole moment is essential.

Another point to be noted is that the frequency at which the irregularity occurs is lower range than 3 MC, and in higher range than 4 MC such tendency is not seen. When the medium includes non-irregularity-making gasses as CO_2 , the breakdown strength becomes lower, but the decreament of the strength at higher frequency than 4 MC is rather little. Then we can assume that in fairly higher frequency range the effect of impurities is little and the breakdown strength is nearly the same to the value of the medium itself.

3. High frequency breakdown of pure water

In the former section it has been clear that the role of water in dipolar dielectrics is very distinguished. There, one will assume that water itself may have such a strange breakdown characteristic that has a peak value near 2 MC. For testing this point very pure water was made and measured. DC conductivity of here made water is up to $0.2 \mu\Omega$. The breakdown characteristic of this water under a needle-plane electrodes is in Fig. 32. Contrary to one's expectation, frequency character of water is not irregular. His breakdown

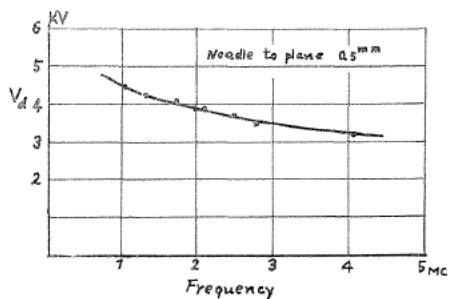


FIG. 32. H. F. Breakdown voltage of pure water.

strength decreases monotonously with increasing frequency. Here, the author notices that water has unexpectedly high breakdown strength under high frequency field.

No matter how both of solvent and solute, i.e. nitrobenzene and water, have no irregularities, the solution of them has very irregular character and has a higher strength than both of them at 2 MC.

4. Kinds of absorbed molecules

The fact that dipolar dielectric liquids show a peak value of high frequency breakdown between 1 to 3 MC seems to be made by the molecular action between dipolar molecules of solvent and the strong dipole of water molecules. But it is rather strange that the frequencies of peaks are almost in neighbour of 2 MC independent of kinds of solvent.

[Notes]: In above written all experiments the frequency peak are all 2 MC, and this makes us doubt whether any parts of used equipment has a particular resonant frequency. So the author measured them for several years by changing oscillator, measuring circuit, measuring vessels and changing testing peoples. But the curve was not changed. Besides, the fact that by the same equipment itself transformer oil and hexane show no irregularity proves the peak of 2 MC not to be made by the equipment.

What does the frequency, 2 MC, mean? The well known dielectric dispersion of liquid dielectrics occurs at above 1,000 MC. The frequency of dispersion is very different by its order, but we may consider a mechanism that resembles to the phenomenon, i.e., when the diameter of oscillating particles becomes tenth of molecular diameter, the frequency of dispersion becomes lower by 10^3 , to be 10^6 c/s. These particles may be large collective molecules.

If the diameter of the collective molecules is not decided by the solvent, but by water, the frequency of peak with most solvent including water will be nearly equal. Fig. 33 shows the results of the experiment in which pure nitro-

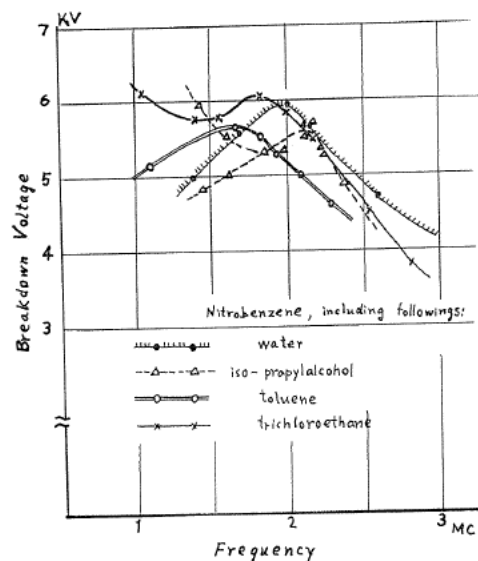


FIG. 33. H. F. Breakdown voltage of nitrobenzene including several kinds of dipolar molecules.

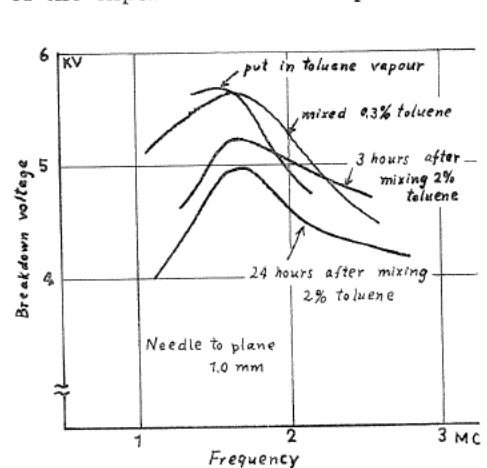


FIG. 34. Characteristics of nitrobenzene, includes different amount of toluene.

benzene as solvent is used, and as solute water, isopropylalcohol, toluene and trichloroethane were selected. Clearly all the curves have some irregularities between 1 to 3 MC, and the locations of peak are more less different each other. With water the peak exists at 2 MC, with isopropylalcohol it exists at 2.2 MC and with trichloroethane and toluene at 1.85 and 1.7 MC respectively. Further, here is some question that the solubilities of above mentioned molecules in nitrobenzene are different each other, and then difference of solubility occurs difference of peak frequency. In the next figure, Fig. 34, four grades of concentration of toluene in nitrobenzene are measured. This figure shows that the value of breakdown strength varies by the concentration of toluene, but the location of peak is not varied.

At last, one question remains. In this experiment used solute molecules are dipolar molecules, and generally dipolar liquids have strong affinity with water and it is difficult to remove water. Then, when we solve trichloroethane into nitrobenzene, trace of water absorbed in trichloroethane may affect on nitrobenzene and it shows the irregularity near 2 MC. This question will be solved in the next section.

5. Effects of absorbed gasses on temperature dependence

Fig. 35 shows temperature dependence of high frequency breakdown of xylene. Used frequencies are 1.5 and 3.3 MC, the former is near and before the peak and the latter is fairly higher than the peak. In this figure, the effects of

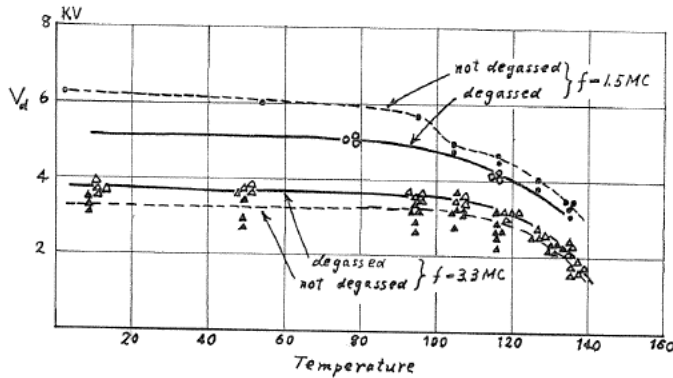


FIG. 35. Effect of degassing on the temperature characteristics of H. F. breakdown of xylene.

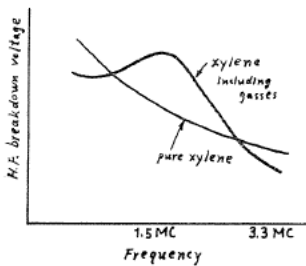


FIG. 36. Characteristic curves of xylene.

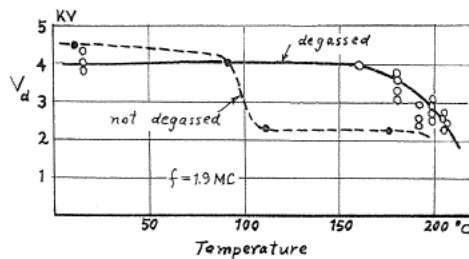


FIG. 37. Effect of degassing on the temperature characteristics of nitrobenzene.

absorbed gas are entirely inverse for 1.5 and 3.3 MC. At 1.5 MC it makes the value of strength higher and at 3.3 MC it makes lower. This facts are made clear by Fig. 36. Besides, at the temperature range higher than 100°C the effect becomes very little for both frequencies. It shows that the principal role of irregularity is water which has a boiling point of 100°C.

An experiment with nitrobenzene has been done, and the results by 1.9 MC is shown in Fig. 37. Curve of impure nitrobenzene goes rapidly down at 100°C. At 100°C only water is removed and remained impurities make the value lower than the pure medium.

In Fig. 38 curves of temperature dependence of xylene including both of propylalcohol and water are shown. These curves have rapid decreasing point at 80°C and 100°C, boiling point of alcohol and water respectively. By this figure we can say that the molecules of iso-propylalcohol and water make irregularities independently. At the last paragraph of the former section one question remained whether water molecules absorbed in trichloroethane make irregularity in nitrobenzene. But now we must consider that water molecules and trichloroethane molecules make irregularities independently each other in nitrobenzene.

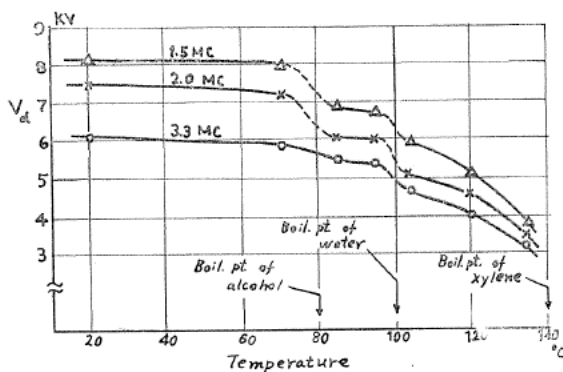


FIG. 38. Temperature characteristics of xylene including both of water and propylalcohol.

Now, here presents a difficult problem why the dipolar molecules absorbed in dipolar liquid make the breakdown strength of the liquid higher than pure one.

§ 2.7. Measurement with silicone oil

1. Physical characters of silicone oils

All over the wide range of polymerization, presented by its viscosity from 10 cs. to 500,000 cs., ϵ , $\tan \delta$ are nearly constant and specific gravities are nearly equal to 1. These characters are also seen in series of saturated hydrocarbons, from pentane (C_5H_{12}) to hexadecane ($C_{16}H_{34}$). Next table shows the physical characters of silicone oils. In these characteristic factors ϵ and $\tan \delta$ do not depend on temperature, but viscosities vary fairly by temperature and the variation is nearly exponential as seen in Fig. 39.

TABLE 5. Physical characters of silicone oils

Viscosity (25°C)	Density (25°C)	Dielect. constant	Thermal conduct (25°C)	Dielect. loss factor	Degree of polymeriz. (Mol. Wgt.)
10 cs	0.940	2.65	0.000317	$< 0.4 \times 10^{-4}$	1200
20	0.950	2.68			1900
50	0.955	2.72			3700
100	0.968	2.74	0.000368	< 0.4 "	6700
500	0.972	2.75			19000
1000	0.973	2.76	0.000381	0.6 "	26400
12500	0.973	2.82			5.62×10^5
30000	0.973	2.77	0.000370	< 0.4 "	2.24×10^5

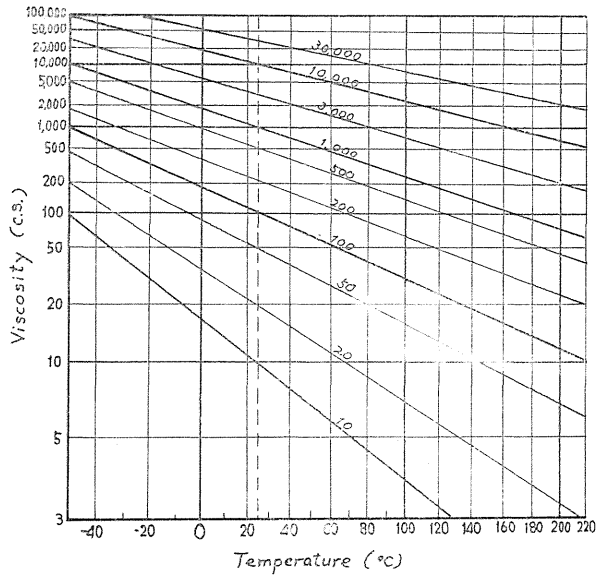


FIG. 39. Viscosity variation of silicone oils.

2. Frequency characteristics of breakdown strength

Already in Fig. 15 frequency characteristics of silicone oil with uniform field were shown. Here in Fig. 40 that with needle-plane electrodes are shown in logarithmic scale. From both figures we can see the following interesting facts:

- (i) Curve of 20 cs is nearly parallel in log. scale to the line of $V_d \propto 1/\sqrt{f}$.
- (ii) Curves of 100 cs and 800 cs are nearly parallel to the line of $V_d \propto 1/\sqrt{f}$ in the lower frequency

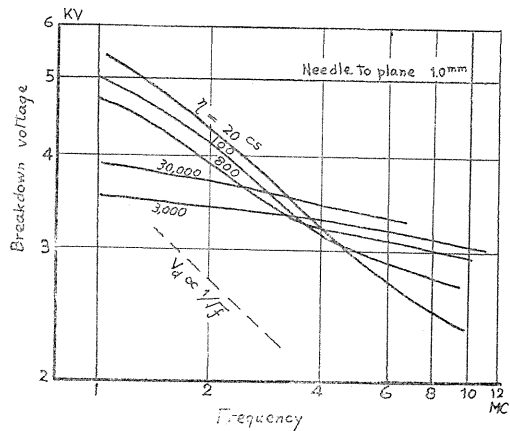


FIG. 40. H. F. Breakdown of silicone oils.

range, but they become flat at higher frequency range.

(iii) Curves of 3,000 cs and 30,000 cs are flat in whole measured frequency range. They are very different from the relation of $V_d \propto 1/\sqrt{f}$.

(iv) In lower frequency range, (strictly to say in the range that curves are parallel to the line $V_d \propto 1/\sqrt{f}$, the higher the viscosity, the lower is the breakdown strength. On the contrary in the higher frequency range (i.e. in the range of flat characteristics), the higher the viscosity, the higher is the strength.

From these facts, we can assume that the curves of 3,000 cs and 30,000 cs have turning point in the range lower than 1 MC and at left side of it they are parallel to $V_d \propto 1/\sqrt{f}$. And the curve of 20 cs has a turning point at the range higher than 10 MC and in the right side of it it has a flat characteristic. Then the fact that at 1 MC the sequence of strength is as 20 cs > 100 cs > 800 cs > 30,000 cs > 3,000 cs, in spite of that at 8 MC 30,000 cs > 3,000 cs > 800 cs > 100 cs > 20 cs, is very reasonable. If we make a figure of wide frequency range according the above mentioned assumption, it will be as Fig. 41.

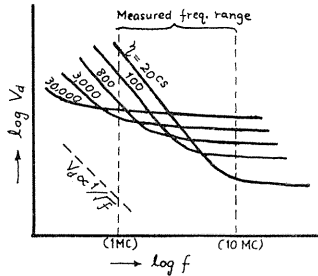


FIG. 41. Suggested frequency characteristics of H. F. breakdown of silicone oils.

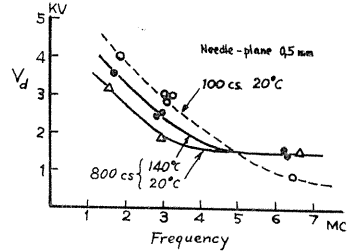


FIG. 42. Variation of freq. characteristic of 800 cs silicone oil by temperature.

3. Temperature dependence of silicone oil

Fig. 42 shows the frequency characteristics with the parameter of temperature. From Fig. 39 we can see that the viscosity of 800 cs (20°C) becomes 150 cs at 140°C, then the curve in Fig. 42 of 140°C must move to be near the curve of 100 cs (20°C). But in Fig. 42 the curve of 140°C moves to neighbour of dotted line of 100 cs (20°C) at the range lower than 4 MC, and in the range higher than 4 MC the result is very different. The curve runs on almost same position of 800 cs (20°C). If the breakdown strength under high frequency field is decided by thermal breakdown as Schlegelmilch's theory, the curve must move uniformly upward or downward. And if the characteristics as shown in Fig. 41 are decided only by their viscosities, the curve of 800 cs (20°C) in Fig. 42 must become near the curve of 20 cs (20°C). This results of the experiment deny the above consideration. The curve goes upward in the lower frequency range in which the law of $V_d \propto 1/\sqrt{f}$ consists and becomes to resemble to it of lower viscosity one but does not move in the higher frequency range in which the curve is flat. In the range where V_d is independent of frequency, V_d is also independent of temperature.

Fig. 43 shows the temperature dependence of 800 cs (20°C) silicone oil with parameters of frequency 1.4 and 6.5 MC. From the above mentioned consideration

1.4 MC belongs to the lower frequency range in which $V_d \propto 1/\sqrt{f}$ exists, and 6.5 MC belongs to higher frequency range in which the temperature dependence is very little. In the figure the curve of 1.4 MC at first increases with increasing temperature and then decreases after passing a peak value. On the other hand the curve of 6.5 MC goes nearly flat.

4. On variation of viscosities and breakdown

Generally we can say that the higher molecules have higher boiling point and are more difficult to be decayed than lower molecules (or polymers). Then when the mixed solution of two kinds of hydrocarbon, for example, is heated, the liquid of smaller molecules will at first become vapor. We can say the situation will be same under high frequency electric field. On silicone oil, contrary, the situation is rather different. Silicone oils higher than 5 cs have not "boiling temperature" and when they are strongly heated, vaporation does not occur, organic bases are separated and at last silica is isolated. Energy will be at first given to chop Si-C bonds and then Si-O bonds. It seems to be one of the reason why the curve of temperature dependence as in Fig. 43, does not go rapidly down as toluene or xylene, does go down slowly.

Here, a mixed solution of two different grades of silicone polymers is applied by high frequency electric field. If there occurs pure thermal breakdown by dielectric heating, both polymers are broken down in same probability and there will be not variation of mixing ratio. But if the larger molecules are easily broken, viscosity will become small. Ordinarily commercial silicone oils have a some distribution of grade of polymerization. So several kinds of commercial silicone oil, described by brands of viscosities, are tested by viscosity measurement after following treatments: (A) leaved for a long time, (B) heated and (C) broken down by high frequency voltage for many times. Next table shows that results. Decrease of viscosities by (A) treatment is not so large, so they seem to include very little evaporative components. By (B) treatment they also don't vary. But by (C) treatment viscosities increase, especially the oils of 1,500 and 3,000 cs are remarkable.

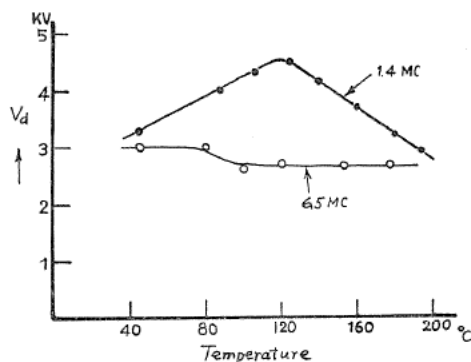


FIG. 43. Temperature characteristics of 800 cs silicone oil.

TABLE 6. Variation of viscosity of silicone oils (presented in centi-strokes)

Name of medium	New oil	Heated up to 100°C	Heated up to 135°C	Leave for long time	Repeated H. F. discharge
100	83.5			(2 years) 73	94
500	602	620	592	(2 months) 615	615
800	990				1125
1500	1571	1630	1552		3352
3000	3718				4350

To explain the increase of viscosities there may be two methods of consideration, one is the cross-linking by the after-discharge current, and the other is that the lower grade molecules are broken and the higher components remain. By the former, viscosities of all kinds of oil must increase uniformly. This table seems to make us consider the latter. The fundamental data for the latter, i.e. the smaller molecule is easier to be broken than the larger molecule, is already shown in the range over 4 MC in Fig. 40. For the fact that the molecules of the least diameter are at first broken down in the mixed solution of several kinds of molecules of different diameters, the criterion of breakdown must be decided by the molecular size. The author assumes that the energy of oscillation of charged particles trapped in intermolecular space becomes great enough. The reason why 100 cs and 500 cs show very little increase of viscosities can be explained in Fig. 40 and Fig. 41. In these figures, the characteristic curves of less viscous oils are almost near to the line of $V_d \propto 1/\sqrt{f}$, i.e. heat has large effect on breakdown. Heat is given for all molecules uniformly and the breakdown of all kinds of molecules of different diameters are broken in same probability. Hence, the difference of curve of frequency characteristic of breakdown strength shows existence of the different mechanisms of breakdown.

Chapter III. Discussion—Correction of the Theory of High Frequency Breakdown of Liquids

§ 3.1. Regions of frequency characteristics

The author considers the three regions in frequency characteristics as in Fig. 44. The dotted line is well known to be "thermal breakdown" character, and the full line is written by the author. In the region (B) the curve is near to the dotted line and may be considered to be thermal region. The region (A)

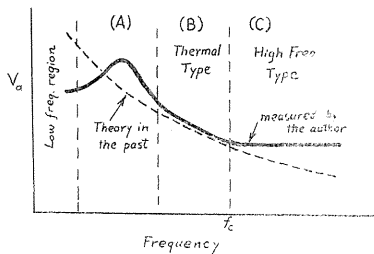


FIG. 44. Division of regions.

is also same to the dotted line for some kinds of liquids but especially for dipolar liquids very different. The region (C) can be seen in more or less viscous liquid, and there the breakdown voltage is independent of frequency and temperature. The boundary frequency between (B) and (C) is higher as the viscosity is higher, but is not decided only by the viscosity. The author call the frequency the boundary frequency f_c .

§ 3.2. The region (B); thermal region

In the region (B) the breakdown strength goes down monotonously with increasing frequency. By logarithmic co-ordinates the characteristic curve is near to the straight line of $V_d \propto 1/\sqrt{f}$. In these region there is no question for thermal phenomenon by the dielectric loss to be principal role of breakdown. But the author notices that the heat is principal role before the breakdown but is not at starting of breakdown. The true leading phenomenon of breakdown is not now determined.

§ 3.3. Region (C); high frequency type

1. Existence of the region (C)

As in Fig. 15 and 40, silicone oil of 3,000 cs and 30,000 cs, and in Fig. 14 phenylethanolamine have frequency independent breakdown strength at more or less higher frequency range. This region seems not to be thermal. Quite different mechanism from the thermal mechanism must be assumed.

For detailed discussion Fig. 45 is shown.

In this figure written the neighbour of the boundary frequency f_c in logarithmic scale.

In the left side of f_c the line has inclination of $1/2$, and in the right side of f_c it goes almost flat. If we extrapolate the line of (B) region into the right side, it goes under the real breakdown curve (dotted line). In other words, in the region (C) the medium is not broken down by the field, which is high enough for the thermal criterion. The substantial constants as ϵ , $\tan \delta$ or viscosity must not be varied by such a little change of frequency.

According to the thermal breakdown theory the medium must reach the "breakdown temperature" on the full line in (B) and the extrapolated dotted line in (C).

But in (C) the medium is not broken down. Then the "breakdown temperature" is not the enough condition, but the necessary condition, and there must exist another condition to make breakdown.

If we extrapolate the full line in (C) into the region (B), it goes under the full line of (B). This says that in (B) the another necessary condition were already fulfilled at the field on the full line.

2. New mechanism of h. f. breakdown

As already explained, the characteristic curve in the region (C) is independent of frequency and temperature. The new mechanism, now we must consider, consist of a factor which does not depend on frequency and temperature but on the molecular size. This factor may be assumed to be a molecular diameter or dimension of inter-molecular space.

Now, a charged particle, its mobility is b , has following velocity under an electric field of $E = E_m \cdot \cos \omega t$:

$$v = Eb = E_m b \cos \omega t \quad (22)$$

The maximum energy is equal to $1/2 M(E_m b)^2$, and the maximum amplitude is

$$x_m = 2 E_m b / \omega \quad (23)$$

The value of b for silicone oil is not well known, but referring the value of paraffines by Adamzewsky⁽⁹⁾ we put it about 2×10^{-4} cm²/V sec. E_m is given from Fig. 15 to be 1.8×10^5 V/cm and put $f = 5 \times 10^6$, then we get $x_m \approx 230$ Å.

Now the next table shows the molecular dimension of silicone oil by the author's calculation, by the internal viscosity of Barry⁽¹¹⁾. From the table we see the silicone oil over 500 cs (25°C) has mean diameter larger than 200 Å. Intermolecular space may be same as the molecular dimension.

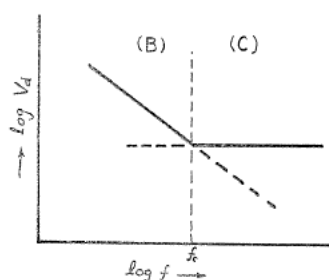


FIG. 45. Neighbour of f_c .

TABLE 7. Dimensions of silicone molecules

Viscosity at 25°C (cs)	Internal viscosity (η)	Molecul. weight (Osm)*	$(\frac{r_0^2}{M})^{1/2}$	Mean radius r_0 (Å)	Diameter \bar{D}_0 (Å)
20	0.040	1900	0.688×10^{-5}	30	60
100	0.069	6200	0.747 "	59	118
500	0.130	18300	0.772 "	104.3	208
800	0.150	24500	0.771 "	120	240
1500	0.180	35800	0.775 "	146	292
3000	0.220	40000	0.806 "	161	322
30000	0.360	82000	0.795 "	227	454

* (Osm): by the method of osmotic pressure.

In silicone oil of lower viscosity (<100 cs), charged particles collide with silicone molecules during the oscillating motion by the field, and the energy of the particles, given to the molecules, is converted to heat. But in one of higher viscosity, charged particles are trapped in the intermolecular space. Here is one question why the author makes the value of mobility b to be same order to the value in hydrocarbons. It is known by Eying's method that the unit volume of viscous flow of silicone oils is the volume including six atoms of Si, i.e. the segment of silicone oil is same as the hexamer. Then the value of mobility will be not so differ from the value above assumed.

3. Breakdown strength and dimension of molecules

Generally the breakdown strength of liquid has been known to become great when the molecular dimension becomes large. Fig. 46 is written on the abscissa of molecular diameter and the parameter of frequency from the Fig. 40. The scales are logarithmic. At the frequency over 5 MC the lines are nearly linear, i.e. $V_d \propto \bar{D}_0^n$. At lower frequencies the curves have this relation only in the range that the molecular diameter is great enough. Lewis⁽²⁾ measured the breakdown strength of silicone oils of the least polymers, from dimer to pentamer. The author's calculation on the length of molecules of dimer, trimer, tetramer and pentamer, suggested to be not configured, is shown in the next table. This values and the Lewis's breakdown strength are plotted in Fig. 46 by the dotted line. It is interesting that the dotted line has the inclination near $V_d \propto \bar{D}_0^n$ of the author's full lines.

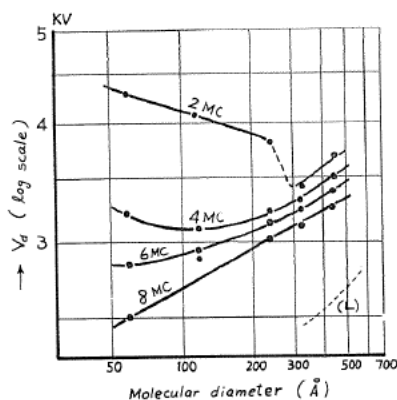


FIG. 46. H. F. Breakdown strength and molecular diameter.

TABLE 8. Length of Molecules

Polymer	Dimer	Trimer	Tetramer	Pentamer
Molecular diameter	7 (Å)	10	13.5	17

§ 3.6. High frequency breakdown voltage of dipolar liquids

1. Motion of dipolar molecules in high frequency electric field

From the theory of dielectric dispersion, under the high frequency field, $E = E_0 e^{j\omega t}$, the molecules of dipolar moment μ get the torque force of

$$M = -\mu E_0 e^{j\omega t} \sin \theta \tag{24}$$

here, θ is the angle between the dipolar moment and the direction of electric field. The factor of internal friction (viscosity) to be ζ , the distribution function F must be as follows:

$$F = A \left[1 + \frac{1}{1+j\omega\tau} \cdot \frac{\mu E}{\kappa T} \cos \theta \right] \tag{25}$$

here, τ is the relaxation time,

$$\tau = \zeta / 2 \kappa T \tag{26}$$

And the mean dipolar moment is:

$$\bar{m} = \frac{\mu^2}{3 \kappa T} \cdot \frac{E}{1+j\omega\tau} = \frac{\mu^2}{3 \kappa T} \cdot \frac{E_0 e^{j\omega t}}{1+j\omega\tau} \tag{27}$$

Fig. 47 shows ϵ and $\tan \delta$ in function of frequency from the result of Eq. (27). The frequency f_d , at which exists the relation $2\pi f_d \cdot \tau = 1$, is called frequency of dielectric dispersion. When the molecules are nearly spheres of radius a , ζ is as following from Stokes's law.

$$\zeta = 8\pi\eta a^3 \quad (\eta: \text{viscosity}) \tag{28}$$

$$\text{then} \quad \tau = 4\pi\eta a^3 / kT \tag{29}$$

Ordinary liquid as water or nitrobenzene has η of the order of 10^{-2} , and the radius is nearly $2 \sim 3.5 \text{ \AA}$, then f_d is $3,000 \sim 400 \text{ MC}$.

If we suggest the mechanism, resembling to the dielectric dispersion phenomenon, the diameter of molecules corresponding to the value of f_d of 2 MC must be the order of $10 \sim 30 \text{ \AA}$.

2. Model of dipolar liquid absorbing some dipolar molecules

The absorbed molecules may be not entirely diffused in the medium, but make some collective molecules and attract some medium molecules, also dipolar, around the collective group. This great collective molecules will be also dipolar and the dimension is calculated from the value of η to be radius $r = 20 \text{ \AA}$.

This large collective molecules resemble to a solid sphere in the medium and oscillate rotary under high frequency field. In Fig. 48, (A) shows the motion of small particle under the alternating field. His angular velocity is proportional to the torque μ . But (B) shows the motion of large sphere. His angular ac-

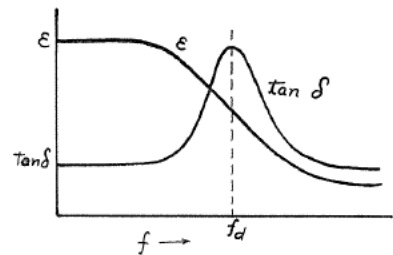


FIG. 47. Dielectric dispersion.

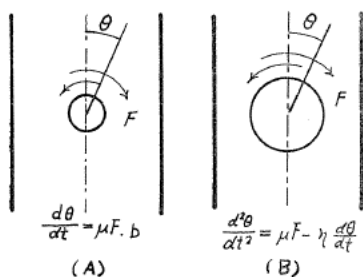


FIG. 48. Model of revolution of molecules.

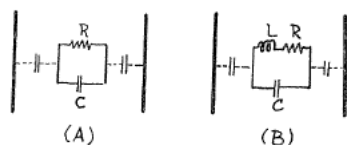


FIG. 49. Equivalent circuit.

celeration is proportional to the torque and the viscosity is the attenuation term of the equation of motion. These equivalent circuit are shown in Fig. 49. (A) and (B) show small and large particle respectively. The condition of $\omega\tau=1$ is presented by the resonance state of LCR circuit in figure (B). And then, the current in LCR tank circuit increases but the partial voltage of either capacitive sides, representing the medium itself, decreases. Therefore, the field strength impressed on the medium decreases, and if the particle is not broken down by the increased loss in the tank circuit, the total breakdown voltage must be higher.

Dimension and character of the collective particles are proper to the absorbed molecules in a medium. But it is very strange that the dimensions of collective particles made by water, alcohol or nitrobenene, in spite of these molecular character are very different each

other, seem to be not so different each other. On the other hand the non-dipolar molecules absorbed in some media do not make such collective molecules or they do not act the role to absorb energy even if they make such collective particles.

Conclusion and Acknowledgement

The breakdown phenomena of liquids under high frequency field have been studied in detail and two important points have been made clear. One of them, there is a frequency range decided by molecular diameter of the medium in which the breakdown strength depends little on frequency and temperature. In this range the breakdown seems to occur not by the thermal criterion but by the energy of charged particles trapped in intermolecular spaces.

The other point concerns to dipolar liquids. Dipolar impurity molecules absorbed in dipolar medium make large collective molecules. These collective molecules are suggested to have diameters of the order of 30 \AA . This is rather interesting. For example, it is well known that water molecules make some associated molecules consisting from two or three molecules. Between the dimension of above mentioned collected molecules and the dimension of bulk water there is some range of dimension in which physical or chemical measuring methods are difficult to be used. Now the author does not know whether the experiment in this paper can be or not the first step of research such a problem, but he believes that successive research must be continued.

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References

- 1) A. Nikuradse: Das Flüßige Dielektrikum.
- 2) for example Toriyama; Electric Insulation (J.).
- 3) M. Ueda: Report of Meeting, Kansai Div., IEE of Japan (1957).
- 4) W. O. Schumann: Z. f. Phys. **76**, 707 (1932).
- 5) Baker and Boltz: Phys. Rev. **51**, 275 (1937).
- 6) W. LePage and L. Dubridge: Phys. Rev. **58**, 61 (1940).
- 7) H. Edler and O. Zeier: Z. f. Phys. **84**, 356 (1933).
- 8) *ibid.*
- 9) R. W. Dornte: Ind. Eng. Chem. **32**, 1529 (1940).
- 10) L. Onsager: J. of Chem. Phys. **5**, 599 (1934).
- 11) M. Wien: Ann. d. Phys. **83**, 327 (1927).
- 12) K. H. Reiss: *ibid* **28**, 325 (1937).
- 13) H. J. Plumley: Phys. Rev. **59** (1941).
- 14) Y. Toriyama and U. Shinohara: J. of IEE of Japan **56**, 164 (1938).
- 15) K. W. Wagner: Arch. f. Elektrot. **2**, 371 (1914).
- 16) R. Becker: *ibid* **30**, 340 (1936).
- 17) A. Güntherschultze: cf. (2).
- 18) L. Inge and A. Walther: Z. f. Phys. **9**, 279 (1930).
- 19) F. M. Klark: J. Franklin Inst. **215**, 39 (1933).
- 20) F. Koppelman: E.T.Z. **46**, 1413 (1931).
- 21) Y. Toriyama: p. 153 (2).
- 22) Hirano: Tech. Rep. of IEE of Japan **4**, 193 (1943).
- 23) W. D. Edwards: Canad. J. of Phys. **29**, 310 (1951).
- 24) B. Salvage: PIEE part IV Monog. II (1951).
- 25) Bragg, Sharbaugh and Crowe: J. App. Phys. **28**, 382 (1954).
- 26) D. W. Goodwin and K. A. Macfadyen: P. Phys. Soc. B **66**, 85 (1953).
- 27) J. L. Makasiejewski and H. Tropper: PIEE II **101**, 183 (1954).
- 28) W. B. Green: J. App. Phys. **26**, 1257 (1955), **27**, 920 (1956).
- 29) T. Suita and C. Yamanaka: J. of IEE of Japan **73**, 589 (1953).
- 30) V. S. Komellkov: Contes Renbus d l'Akademie des Science de l'URSS Moscow **47**, 265 (1945).
- 31) S. Nagao and Y. Toriyama: Report of Meeting. Tohoku Div., IEE of Japan, no. 30, B-7 (1957).
- 32) R. Naehner: Arch. f. Elektrot. **21**, 169 (1929).
- 33) J. Sorge: *ibid* **13**, 139 (1924).
- 34) K. Dräger: *ibid* **13**, 366 (1924).
- 35) W. Schlegelmilch: Phys. Z. **34**, 497 (1933).
- 36) Hirano and Nemoto: Report of Meeting, Tokyo Div. IEE of Japan No. 1-17 (1944).
- 37) T. Masaki: J. of IEE of Japan **64**, 744 (1943).
- 38) M. Ueda and M. Ito: J. of IEE of Japan **79**, 3 (1959), Report of Annual Meeting, IEE of Japan No. 95 (1957).
- 39) M. Ueda and K. Nakano: *ibid* No. 96 (1958).
- 40) M. J. Adamczewski: Ann. d. Phys. Paris **8**, 309 (1935).
- 41) A. J. Barry: J. App. phys. **17**, 1020 (1946).
- 42) T. J. Lewis: PIEE II A **104**, 493 (1957).
- 43) Phenomena written in this paper had been reported partially as follows:
 - i) On the breakdown of silicones: J. of the IEE of Japan, p. 3, vol. **79** (1959); Electro-technical J. of Japan, p. 56, vol. **5** (1959); Memoirs of the Fac. of Eng., Nagoya Univ., p. 180, No. 1, vol. **9** (1957).
 - ii) On the breakdown of dipolar liquids: Memoirs of the Fac. of Eng., Nagoya Univ., p. 95, No. 1, vol. **8** (1956), *ibid*, p. 174, No. 1, vol. **9** (1957), A synthetic report on the dipolar liquids has been send to the Journal of the Applied Physics on January 1965.