

HIGH FREQUENCY BREAKDOWN OF DIPOLAR DIELECTRIC LIQUIDS

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Introduction

In the previous report,¹⁾ the author indicated that we can not always hold good the theory that the high frequency electric breakdown in liquid is attributed to the thermal breakdown originated in the dielectric loss in H. F. field. Because, there is such a strange phenomenon that the dipolar liquid as nitrobenzene presents a peak value of breakdown voltage at the frequency near 2 MC. The author reported this peak to be presented when slight impurity especially moisture is absorbed in the medium. In this paper, some results of successive measurement and the author's point of view for the phenomenon will be stated. And the author is expecting many discussions to be given.

Experiments

1. Influence of a kind of medium

Fig. 1 shows the frequency characteristics of H. F. breakdown voltage of pure nitrobenzene, pure water and a little wet nitrobenzene with the needle-plane electrodes. Neither the former two substances show irregular characteristics in the measured region, but the mixture of them shows a very evident peak at 2 MC. It is also evident by Fig. 2 that the frequency of the peak is not changed by the shape of electrodes.

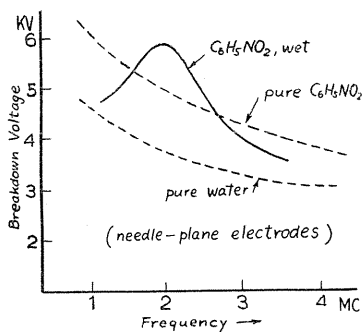


FIG. 1. Breakdown characteristics of nitrobenzene and pure water.

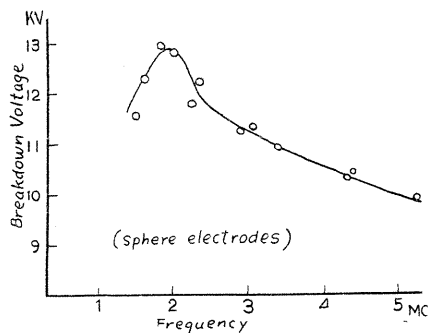


Fig. 2. Breakdown characteristics of nitrobenzene (gas absorbed).

Fig. 3 shows the frequency characteristics of other dipolar liquid. Not only the strong dipolar liquid as nitro benzene or *iso*-propylalcohol but also the relatively weak dipolar liquid as toluene shows irregular characteristics at 2 MC. This results indicate that the medium molecule is not concerned in the cause of the irregularity.

2. Influence of a kind of absorbed molecule

It was also reported that the frequency characteristics of dipolar liquid do not present any kind of irregularity when the absorbed molecules are non-dipolar, and merely, as was expected, the breakdown voltage descends considerably by impurities over whole range of frequency.

Fig. 4 shows the frequency characteristics of nitrobenzene in which several kinds of dipolar molecules are absorbed. As seen in Fig. 4, alcohol or toluene shifts the frequency of irregularity from that by water. This shows that the frequency of peak voltage is principally decided by the mixed molecule. However, the frequency difference is unexpectedly little considering that the characteristics of mixed molecules fairly differ from each other. From this fact, it may be thought that the irregularity of alcohol-absorbed nitrobenzene is not owing to the alcohol molecule itself but to the water molecule which is absorbed in alcohol. But this point will be once more discussed afterward.

3. Temperature characteristics of breakdown voltage

The frequency region under consideration is believed to be that of thermal breakdown through the so-called dielectric heating. Therefore, the non-dipolar liquid which does not present any peak in his frequency characteristics of breakdown voltage, or entirely pure dipolar liquid has such characteristics that go down in inverse proportion to the square root of f . The breakdown condition is given when the equilibrium temperature between the electric heating and cooling becomes higher than the "breakdown temperature" of the medium. With the relatively simple liquids the "breakdown temperature" is suggested to be identical to the boiling temperature, but with the polymerized substances it can be lower than the boiling point.

Fig. 5 shows the temperature characteristics of entirely degassed nitrobenzene and toluene. The temperature is controlled by a oil- or glyceline-bath and a sensitive thermoregulator. Both curves are nearly flat up to the temperature lower

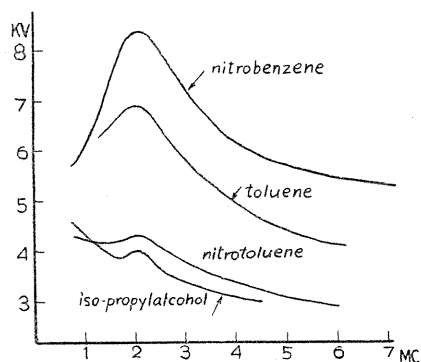


FIG. 3. Breakdown characteristics of dipolar liquids (moisture absorbed).

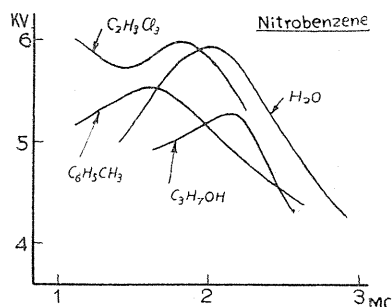


FIG. 4. Breakdown characteristics of nitrobenzene (absorbed several kind of molecules).

than the boiling point by about 20°C, and then they go down rapidly with temperature.

Fig. 6 shows curves of Xylene both degassed and not degassed, measured on 1.5 and 3.3 MC. It is noticeable that the degassed curve of 1.5 MC. is higher than the not-degassed curve, while the former of 3.3 MC. is lower than the latter. It can be understood by Fig. 7, in which the above used two frequencies are shown to exist on the both side of the peak of the frequency characteristics. Besides, the not-degassed curve of 1.5 MC. suddenly goes down at 100°C, i.e. the boiling point of moisture. This is a positive proof for moisture to make irregular rise in the breakdown voltage of dipolar liquid.

4. Temperature characteristics when two kinds of molecule are absorbed

It is suggested in the Experiment 2, that, according to the phenomenon that the frequency difference between the peak positions of water-absorbed and alcohol-absorbed nitrobenzene is unexpectedly little, the cause of irregular rise of breakdown voltage does not exist in alcohol molecule but in water molecule inconsiderably absorbed in alcohol. In fact, almost all dipolar liquids attract strongly water molecules. But this suggestion is disproved by Fig. 8. There is a curve of nitrobenzene which absorbs both of water and *iso*-propylalcohol, and the curve, nearly flat from 0°C to 70°C, suddenly goes down at first at 80°C, the boiling point of alcohol, and further once more at 100°C, the boiling point of water. Evidently the two steps show the irregularities by alcohol and water molecules respectively. In addition, the height of each steps are respectively equal to the height of irregular rise when nitrobenzene have absorbed

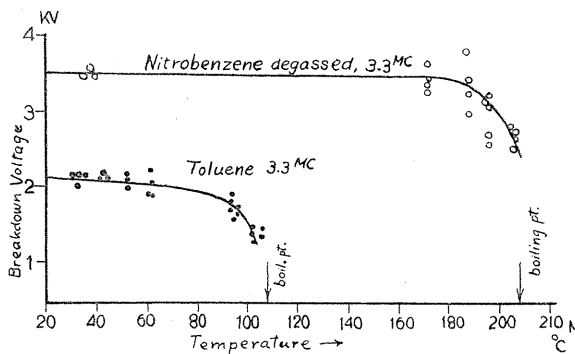


FIG. 5. Temperature dependence of H. F. breakdown voltage.

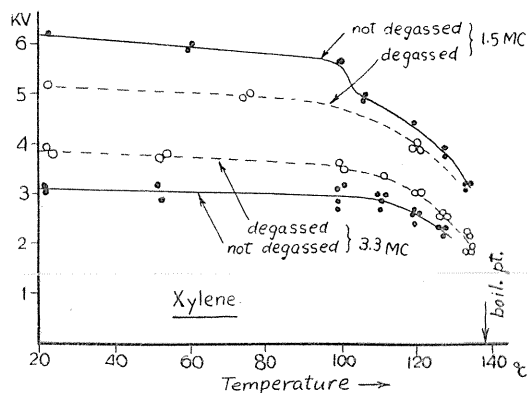


FIG. 6. Temperature dependence of breakdown voltage of Xylene.

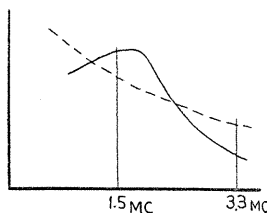


FIG. 7. Frequency characteristics of H. F. breakdown voltage of Xylene.

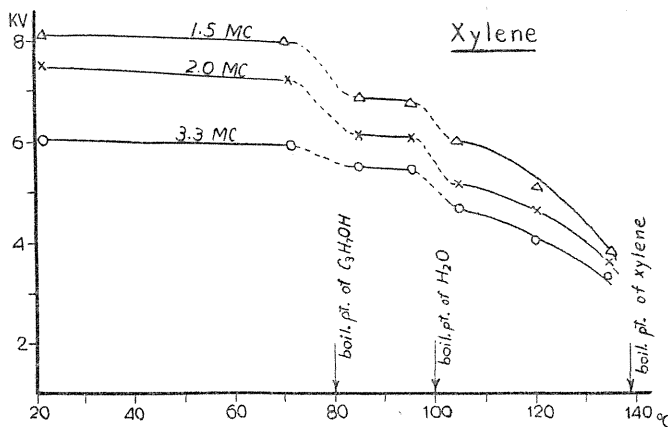


FIG. 8. Temperature dependence of breakdown of Xylene, absorbed C₃H₇OH and H₂O mixed.

alcohol or water independently; i.e. the different kinds of molecules which are absorbed together in a medium act independently for each other to make irregular rise of breakdown voltage.

Therefore, in Fig. 4 it should be considered that the peak of the characteristics of nitrobenzene-alcohol system is made certainly by the alcohol molecules.

Theoretical Consideration

1. On the thermal breakdown

In the electrodes arrangement shown in Fig. 9, the temperature distribution along the direction of electrode separation is given as follows:

$$\lambda \frac{d^2\theta}{dx^2} = -Q, \text{ or } \frac{d^2\theta}{dx^2} = -\frac{Q}{\lambda} \quad (1)$$

where, Q is the rate of heat generation, λ is the rate of heat conduction, and x is the coordinate along the axis.

When Q and λ are constant, solution of this equation is simple:

$$\left. \begin{array}{l} \text{at the centre, } x=0: \quad \theta_c = \theta_0 + Ql^2/8\lambda \\ \text{at } x=x: \quad \theta = \theta_c - Qx^2/2\lambda \end{array} \right\} \quad (2)$$

here, θ_0 is the temperature of electrodes.

Heat generation by the dielectric loss is:

$$Q = AE^2\omega\varepsilon \tan \delta \quad (3)$$

and

$$\theta_c - \theta_0 = (l^2/8\lambda)AE^2\omega\varepsilon \tan \delta \quad (4)$$

In this case the breakdown voltage V_d must be in inverse proportion to the square root of $\theta_d - \theta_0$, θ_d is the "breakdown temperature".

But generally, since ε and $\tan \delta$ of dielectrics are the function of temperature, distribution of the electric field becomes a function of temperature, and then Q

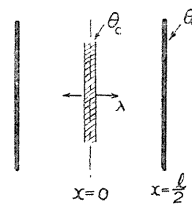


FIG. 9

does not become constant. So the equation of temperature distribution is as follows:

$$\frac{d^2\theta}{dx^2} = f(\theta) \tag{5}$$

This can be solved when the form of $f(\theta)$ is given,²⁾ but it will be enough to know that when $f(\theta)$ is increasing function with θ the distribution of temperature is steeper than parabolic and the breakdown voltage—temperature relation is as curve (b) in Fig. 10, and when decreasing function, the relation as curve (c).

Now, the curves in Fig. 5 and Fig. 6 can be rewritten as in Fig. 11 with respect to the coordinate of $\theta_d - \theta_0$. These curves in Fig. 11 have a tendency similar to the curve (b) in Fig. 10. Therefore, $f(\theta)$ in Equation (6) must be an increasing function with the temperature.

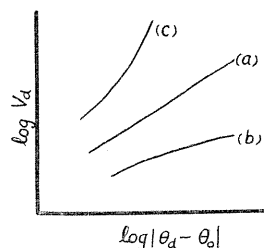


FIG. 10

2. Abnormal dispersion of breakdown voltage

Let us name the irregular rise of breakdown voltage when a dipolar liquid absorbs dipolar molecules “abnormal dispersion.” This kind of dispersion occurs on the frequency range which is widely different from the frequency of the ordinary dispersion, but it seems that a treatment similar to that of the ordinary dispersion can be applied.

Since in dipolar liquid molecules attract each other by the Coulomb’s force and make a sort of net of static force over the whole volume, a particle put in the medium is polarized and is moving under the restriction of surrounding medium. Then the particle will oscillate when the external field makes the net of molecules of the medium oscillate. The counter force for the oscillation is the viscosity of the medium.

In the case that the particle is very small one as a molecule, resistance of viscosity gives such an equation of motion that angular velocity of the particle is in proportion to the moment of force (angular mobility is nearly constant). But

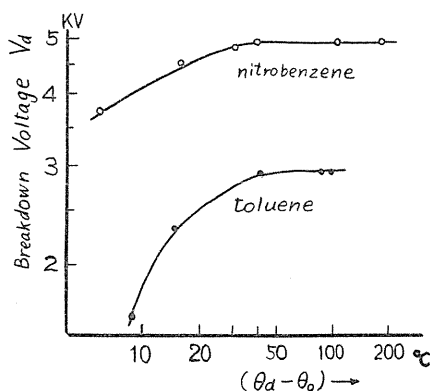


FIG. 11. Characteristics of H. F. breakdown voltage with respect to $(\theta_d - \theta_0)$.

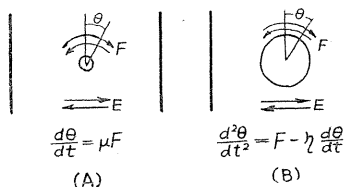


FIG. 12. Models of absorbed particle.

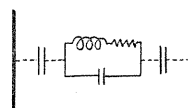


FIG. 13. Equivalent circuit for the Model of Fig. 12 (B).

in the case that the particle is more or less large and can be considered to be a solid sphere, the equation of angular motion is written in the form shown in Fig. 11 (B), and the equivalent circuit is represented by Fig. 12.

When the frequency of external field becomes nearly equal to the resonance frequency of L - C circuit, electric energy from the external field is given effectively to the particle. Therefore, if the particle is a substance that should not easily be broken down thermally for the large heat capacity or its high breakdown temperature, it is easily explained that the electric field in the surrounding medium becomes lower and the breakdown voltage of a little impure medium becomes higher than that of pure one.

For the resonance condition of the particle, the equation same as of the ordinary dielectric dispersion can be applied. The relaxation time is

$$\tau = 4 \pi \eta a^3 / kT \quad (6)$$

here, η is the coefficient of viscosity, and a the radius of the particle. The maximum consumption of energy occurs at $\omega\tau = 1$, and then, the frequency of peak in the characteristics of breakdown is kept on:

$$f = kT / 8 \pi^2 \eta a^3 \quad (7)$$

If we use for the value of $\eta = 1 \times 10^{-2}$, the value of a to make f equal to 2×10^6 is near 30 \AA . Even when the value of η between nitrobenzene and the absorbed particles may be somewhat greater than that of the medium itself, the value is about ten times greater than the radius of ordinary molecule. Therefore, in case of the above mentioned mechanism the particle is not a single molecule of absorbed impurity but a "clustered" particle. In this case the cause of force which makes "cluster" at the centre of it will be an unorganic ion which is included probably in the medium, although the medium is purified to the highest grade in the laboratory.

According to the results shown in Fig. 8, water molecules and alcohol molecules are believed to make up independent *clustered* particle respectively. And, since the radius of *cluster* sphere is decided mainly by the character of a central ion, the frequency difference of irregular dispersion with respect to the kind of absorbed molecule will be unexpectedly little.

The existence of the "cluster" particle which has the above suggested radius has not proved as yet in the physical or chemical field. But the hydrated unorganic ions in electrolytic solution may have radii near 6 \AA , and it may be able to suggest that in water-insoluble medium the sphere of hydration may become larger than that in water solution.

Literatures

- 1) M. Ueda: Memoirs of the Faculty of Eng., Nagoya Univ., Vol. 8, No. 1, May, p. 95 (1956).
- 2) T. Masaki: Researches of the Electrotechnical Laboratory, No. 481 (1948) (in Japanese).