LIQUID DIELECTRICS

ACKNOWLEDGMENT

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Although solid insulators play an all-important role in the electrical industry—that of isolating conductors from one another and from ground while supporting them physically—liquid insulators are equally important in many electrical products. Transformers, circuit breakers, capacitors, and cables are some of the more common devices that utilize liquid dielectrics. In the transformer industry alone, insulating oil by the tens of millions of gallons is used annually.

Through a better understanding of how liquid dielectrics behave in electric fields, engineers have the opportunity to further increase the operating efficiency of their products. For the development of liquids with better electrical properties will yield transformers of greater power capacity per unit volume, capacitors of greater electric capacity per unit volume, and so on.

What electrical properties determine the performance of a liquid dielectric? Primarily they are: 1) ability to resist breakdown under electrical stress, 2) the electric capacitance per unit of volume determined by the dielectric constant, and 3) loss factor—the energy loss per unit volume per cycle. These properties depend on the liquid's operating temperature and frequency as well as on the structure of its constituent molecules.

Let's look first at electrical phenomena in liquids at low voltages, then proceed to the less understood phenomenon of electrical breakdown at much higher voltages. (We'll confine our attention to electrical properties alone—not the physical ones such as heat transfer and mechanical forces.)

DIELECTRIC CONSTANT

In deciding what liquids may be classed as insulators, you can arbitrarily select those having resistivities greater than a certain value, say, 10-million ohm-centimeters. (An ohm-centimeter is the resistance between opposite faces of one cubic centimeter of the liquid.) On this basis, for example, you would class water as a liquid insulator, although its resistivity is a million times smaller than other commonly used insulating liquids.

Actually, the application dictates the

permissible lower limit of resistivity. Thus water, with a high dielectric constant, can be a perfectly respectable dielectric in some applications and an extremely poor one in others.

What do we mean by a dielectric constant? Perhaps the most familiar property of liquid dielectrics is the capacitance increase observed when an air capacitor is immersed in a liquid. In fact, this ratio of the liquid- to air-filled capacitance is often used to define the dielectric constant of the liquid in question. Explained in this way, its magnitude becomes a property of the liquid, independent of the measuring voltage or geometry of the test capacitor. Extremes of such ratios for liquid dielectrics range from 1.05 for liquid helium to 78 for water; most commonly used liquids have dielectric constants between 2 and 5. On the other hand, the dielectric constants of all gases are very nearly unity. And so, gases do not offer the larger capacitances per unit volume that liquids do.

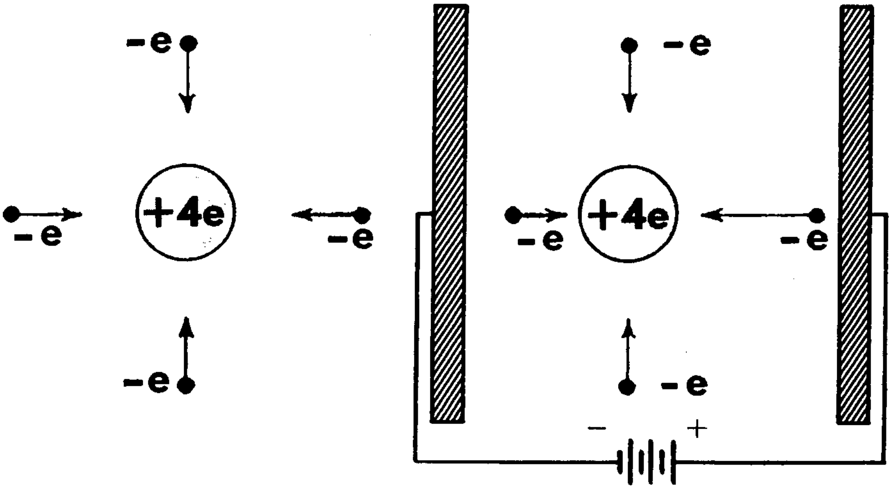


Fig. 1. INDUCED DIPOLES are formed in a neutral atom (*left*) when placed in an electric field (*right*). Arrows indicate the magnitude and direction of polarization.

POLARIZATION

The dielectric constant varies with the temperature and molecular structrue of a liquid and with the frequency of the applied voltage wave. To understand the reason for this variation and why liquids differ broadly in their dielectric properties, you need to become acquainted with a phenomenon called *poralization,* common to all liquid dielectrics.

When a unit positive charge $+q$ and a unit negative charge $—q$ coincide in space, their electric fields cancel one another and their net charge is zero. If they are displaced a distance

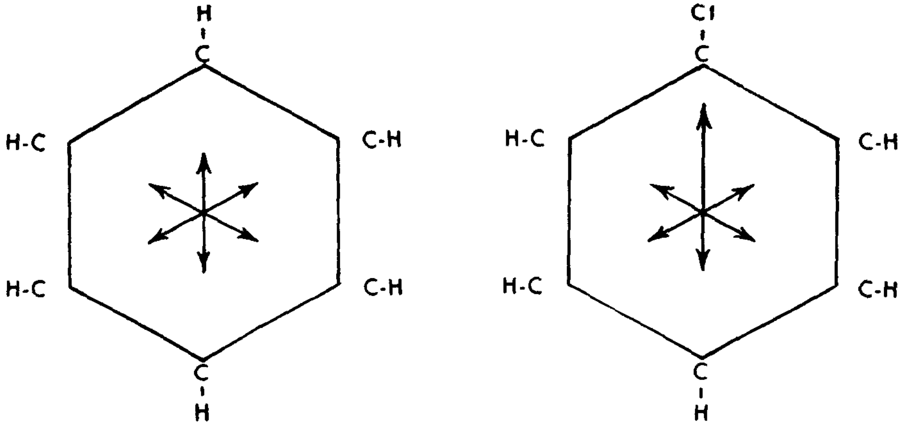


Fig. 2. ELECTRICAL BALANCE of a neutral benzene molecule with six dipoles (*left*) is upset when an atom of hydrogen is interchanged with an atom of chlorine.

l apart, a resultant field arises. Such a pair of charges now constitutes a dipole, and has a moment μ equal to the product of their displacement and magnitude of either charge, or lq.

For example, an atom (Fig. 1, left) with four electrons symmetrically located about a nucleus of four protons is the equivalent of four dipoles. The moment of each pair, electron and proton, is represented by the length and direction of the arrows. You can see in this instance that all the individual dipoles balance each other and that the atom as a whole is electrically neutral. But place the same atom in an electric field between the plates of a capacitor and see what happens: Now the individual dipoles no longer mutually cancel each other (Fig. 1, right) because the electrons are attracted to the positive plate. Conversely, the positive nucleus is slightly displaced toward the negative plate. The resultant dipoles are called induced dipoles because they exist only by virtue of the externally applied field.

You may remember from your studies of chemistry that different kinds of atoms have different amounts of attraction for electrons, called *valence*. Thus, when a molecule is formed from its constituent atoms, it may not be completely in electrical balance. As an example, take first the molecular structure of benzene (Fig. 2, left), a common dielectric liquid. Here the individual dipoles arising from the different electron affinities of carbon and hydrogen are in complete electrical balance. If, however, a chlorine atom is substituted for one of the hydrogens (Fig. 2, right) there is no longer complete neutralization of the individual dipoles, and the molecule is out of electrical balance. This kind of molecule has a *permanent* dipole moment because it doesn't owe its existence to any external electric field.

The general term polarization refers to the movement of electric charges within a molecule in response to an applied field. You may further classify it according to the kind of body that carries the charge—electronic, atomic, or dipole. Electronic polarization, explained in connection with the atom, involves the relative displacement of electrons with respect to their atomic nucleus. Atomic polarization, on the other hand, is caused by shifts in the equilibrium positions of atoms within the molecule with respect to each other. Dipole polarization (not illustrated) arises when the randomly oriented permanent dipoles of a molecule are oriented by an applied electric field.

At a given frequency and temperature, the value of the dielectric constant will depend on the number of different kinds of polarization that can form. Although all liquids will have electronic and atomic polarizations that contribute to their dielectric constants, the amount of dipole polarization will vary greatly according to the magnitude of the dipole moment. If the molecule is electrically symmetrical, it will be zero. The dielectric constant of a liquid may therefore be large or small, according to the arrangement of atoms within its molecules.

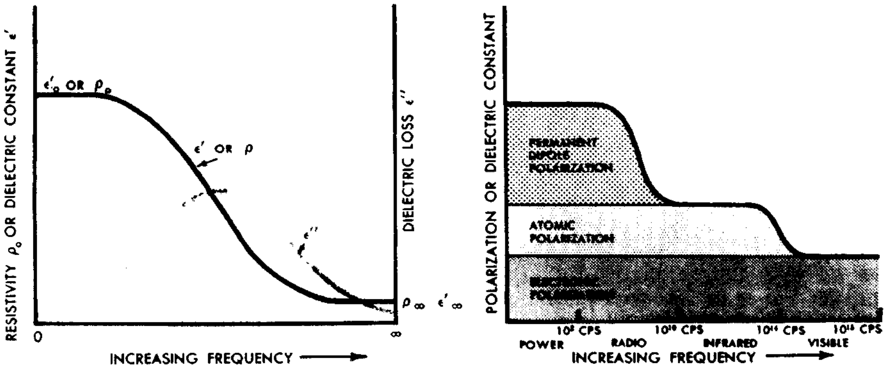


Fig. 3. FREQUENCY DEPENDENCE of the dielectric constant, resistivity, and dielectric loss (*left*) is depicted for one type of polarization. Superimposing curves, shown in heavy black, for the different polarizations result in the composite picture.

FREQUENCY DEPENDENCE - - -

If you use a low frequency or d-c voltage to measure the dielectric constant of a liquid having permanent dipoles, all the polarizations will be complete, and you'll obtain the highest possible value of dielectric constant. If, however, you increase the frequency of the applied a-c voltage, the direction of the electric field may be reversed before polarization has time to form. This leads to a decrease of dielectric constant from some steady-state value ϵ'_0 (Fig. 3, left) at low frequencies to another steady-state value ϵ'_∞ at some higher frequency.

The size and weight of the various charge carriers give a clue to the time required for the different types of polarization to form. Electronic polarization (Fig. 3, right), involving the extremely light and small electron, persists from power frequencies through the visible frequencies. Atomic polarization, requiring the movement of the heavier atom, usually does not persist beyond the infrared range. Dipole polarization needs even longer times because it requires the movement of entire molecules, and usually it contributes to the dielectric constant only at power and radio frequencies.

One by one the permanent dipole

and atomic polarization cease to contribute to the total dielectric constant as the frequency is increased from d-c or low-power frequencies to the higher visible frequencies. For this reason water has a dielectric constant of 78 at power frequencies (60 cps) and only 2 at visible frequencies (10^{14} through 10^{15} cps).

The heat generated in a liquid dielectric under stress is determined by the combined effect of: 1) a frequency-independent resistivity measured with d-c voltage and 2) a frequency-dependent resistivity that contributes when a-c voltage is applied. Determined principally by the number of free ions in the liquid, the d-c resistivity can be reduced to an extremely small value in the case of organic fluids. For example, carefully purified water has a d-c resistivity of about 10^7 ohm-centimeters. Thus it represents a highly "lossy" liquid dielectric. On the other hand, a hydrocarbon oil—a liquid such as petroleum containing only carbon and hydrogen—may have a d-c resistivity of 10^{15} ohm-centimeters, making it almost a perfect insulator.

Referring back to Fig. 3 (left), you can see how the measured resistivity starts at its d-c value ρ_0 and decreases to some limiting value ρ_∞. The loss factor ϵ'' is a measure of the amount of heat generated per cycle as a result of this resistivity.

- - - AND TEMERATURE EFFECTS

Why does the dielectric constant change with temperature? Remember that its value at any particular temperature is determined by the combined contributions of the different kinds of polarization. In the instance of electronic and atomic polarization, the influence of temperature is slight because these polarizations are affected only as the density of the liquid changes. Hence, they become slightly smaller as the liquid's temperature is raised, because the number of contributing atoms per unit volume becomes smaller. Conversely, they become slightly larger as the temperature is lowered.

A much stronger temperature dependence is experienced in dipole polarization: the entire molecule rotates instead of just its charges being displaced. Because a rise in temperature generally lowers viscosity, the molecules experience less molecular friction as they rotate among their neighbors. They undergo an increased motion as the temperature rises. This action promotes a random orientation of the dipoles that opposes the ordering influence of the applied electric field. And so, the net effect of temperature increase on dipole polarization is a lowering of the measured dielectric constant.

(Because the dielectric "constant" of a liquid may depend strongly on frequency and temperature, you might conclude that the name isn't too descriptive. This is true. However it has gained rather universal acceptance, although various other terms are occasionally found in literature. One that is sometimes used is specific inductive capacity, and another is dielectric coefficient.

HIGH-VOLTAGE PHENOMENA

When liquid dielectrics are utilized as insulating material in high-voltage equipment, they are important because of a property differing entirely from that associated with the polarization of molecules. This difference arises from their ability to withstand high electrical stresses for long periods of time. For these purposes, engineers usually prefer liquids that are thermally stable and nonflammable.

If the voltage applied to any dielectric insulator placed between two metal electrodes is increased sufficiently, an electric spark will pass through it. When this occurs the insulator has broken down electrically. And the heat accompanying such a breakdown spark usually results in the accumulation of a large quantity of decomposition products along its path. Because such products weaken the dielectric even more, an initial breakdown often renders a piece of equipment permanently inoperative.

When designing high-voltage equipment using liquid insulation, the engineer wants to know what he can expect of a liquid exposed to high electric fields. Unfortunately, this isn't always predictable. Any property that deals with the violent destruction of matter is often complicated and difficult to reproduce. As a result, the phenomenon of electrical breakdown in liquids is not well understood as yet. Surprisingly enough, the great influence of extraneous factors not associated with the liquid itself makes it difficult to correlate breakdown with any known molecular properties. The engineer, then, is forced to use abnormally large safety factors when designing his equipment.

WHAT IS ELECTRICAL BREAKDOWN?

The appearance of an electric spark is certainly evidence of electrical breakdown; but it doesn't tell you why breakdown occurred. Extremely large currents in sparks do, however, suggest this: Electrical breakdown involves the failure of the insulator to retain its initially high electrical resistance. Hence, a general definition of a liquid's breakdown voltage is that amount of voltage required to convert it rapidly from an insulator to a conductor.

How does such a conversion take place? And what does the magnitude of the voltage have to do with electrical breakdown? Perhaps the most widely accepted theory of explaining this phenomenon deals with the electron avalanche.

Consider a system (Fig. 4) composed of a liquid dielectric placed between two parallel-plate metal electrodes. Choose the system so that the electric field will be homogeneous throughout. When you apply a voltage to the electrodes, free electrons emitted by the cathode—or already present in the liquid itself—are accelerated toward the anode. This acceleration will continue until the rate at which electrons lose energy by collision with molecules just equals the rate at which they gain energy from the electric field. Under these conditions the electric current will remain small. If, however, you in-

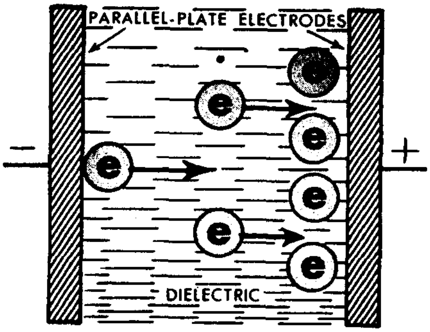


Fig. 4. ELECTRON AVALANCHE begins when a free electron gains enough energy to knock another electron from a molecule.

crease the voltage so that each free electron gains sufficient energy to knock another electron from a molecule of the liquid—called *ionization*—the number of current carriers, and the current, is multiplied by two.

To take it a step further, suppose you increase the voltage until each starting electron can multiply in this manner a number of times as it crosses the gap. Since each new electron produced by collision is also free to undergo the same process, the result is an electron avalanche. Mathematically it works like this: If a single starting electron makes n ionizing collisions while crossing the gap, then $2\ n$ electrons will reach the anode. When n reaches some critical value, the electric current through the liquid will be high enough to lead to a runaway process culminating in the spark. Thus, you can define electric strength as the voltage that will just cause breakdown divided by the distance between electrodes.

THEORY AND PRACTICE

From what has been said you might expect the electric strength of a liquid to be controlled by its ability to resist the acceleration of electrons to those energies causing ionization. This would be true only under ideal conditions. However, as mentioned earlier, many extraneous factors often overshadow the properties of the liquid itself. Usually they prevent measurement of the liquid's maximum electric strength.

Of these complications, one of the most serious is the shape of electrodes. Seldom do you find smooth, flat electrodes in actual equipment. Irregularities on electrode surfaces—such as sharp edges or points—intensify the electric field in their immediate vicinity. Such a situation often gives rise to localized electric fields far more intense than the uniform field throughout the liquid. And if their intensity is great enough, electron avalanches can be sufficiently large to cause breakdown. You would thus measure an apparent electric strength far below that realized in the uniform field.

Often, however, electrode irregularities give rise to electron avalanches of limited magnitude—too small to cause complete breakdown directly. This is so because the field strength throughout most of the liquid is low, and localized avalanches are unable to proceed through the entire gap before they are snuffed out. (The occurrence of ionization leads nonetheless to a gradual decomposition of the liquid and in time may cause its failure. Engineers commonly call pre-breakdown avalanches of this kind *corona*—observed electrically as small bursts of current.)

Closely associated with the effect of electrode shape are small particles on the electrode surfaces or suspended in the liquid. Having a dielectric constant higher than the surrounding medium, these particles will tend to move into the regions of highest electric field. Their effect is to create intense localized fields and cause premature breakdown.

But perhaps the most difficult external factor to control in the laboratory is the electrode material itself. Here's why: The electric fields involved in the breakdown of liquids are of the order of one-million volts per centimeter. At these voltages, most metals emit electrons profusely by a process known as *field emission,* or *cold emission.* The electric field at which this emission becomes extensive has a high sensitivity not only to the kind of electrode metal but also to the nature of its surface. For example, if a highly polished electrode is partially covered with an oxide film, its emission characteristics may vary a considerable amount from one part of its surface to another.

Again, if electrons are emitted by this mechanism at a faster rate than they can be swept to the anode by the electric field, there results a large electronic space charge near the cathode. This gives rise to an intense field in the vicinity of the anode. And such behavior invariably results in low values of the apparent electric strength of the liquid.

THERMAL BREAKDOWN

Although the distortion caused by intense localized fields may lower the apparent electric strength of a liquid, it doesn't alter the general mechanism by which disruption of the dielectric occurs. For example, another complication that may not be considered external doesn't depend on field distortion at all. If ionic impurities are present in the liquid, the application of an electric stress may cause a large enough current to flow even without ionization to cause a definite increase in temperature. The same thing can happen if the liquid itself possesses a relatively low electrical resistance at low fields.

This rise in temperature is usually observed in liquids that require long-time exposure to high voltage. If it becomes high enough, the liquid may decompose thermally. More often, however, bubbles of once-dissolved gases —or vaporized liquid—that are ejected at the elevated temperatures cause premature failure. This phenomenon is called *thermal breakdown.*

LABORATORY TRICKS

You might infer from what has been said that any measurement of the electric strength of a liquid has little or no meaning in terms of the liquid's structure and composition. This isn't necessarily true. For if you make a concentrated effort to either fix or remove extraneous factors, it's possible to observe significant differences in the electric strengths of liquid dielectrics. You can do this by carefully purifying and filtering the liquids before testing and by carefully selecting and preparing electrodes.

Hemispherical electrodes are usually used to test liquid dielectrics in the laboratory for a number of reasons. For one, they are relatively easy to fabricate and polish. For another, if their radii or curvature are large compared to the gap spacing between them, a small region in the center of the gap will have a configuration approaching that of two parallel-plane electrodes. This electrode curvature, reducing the electric field in all other parts of the gap, limits breakdown to the small center region of the electrodes where the field is nearly uniform. And by using rectangular voltage pulses of approximately one microsecond's duration, the danger of interference from thermal breakdown is easily removed. (When pulses of such short duration are applied singly to the breakdown cell, there's not enough time for harmful temperature changes to take place.)

With these techniques it's possible to measure the electric strengths of liquid hydrocarbons within an accuracy of

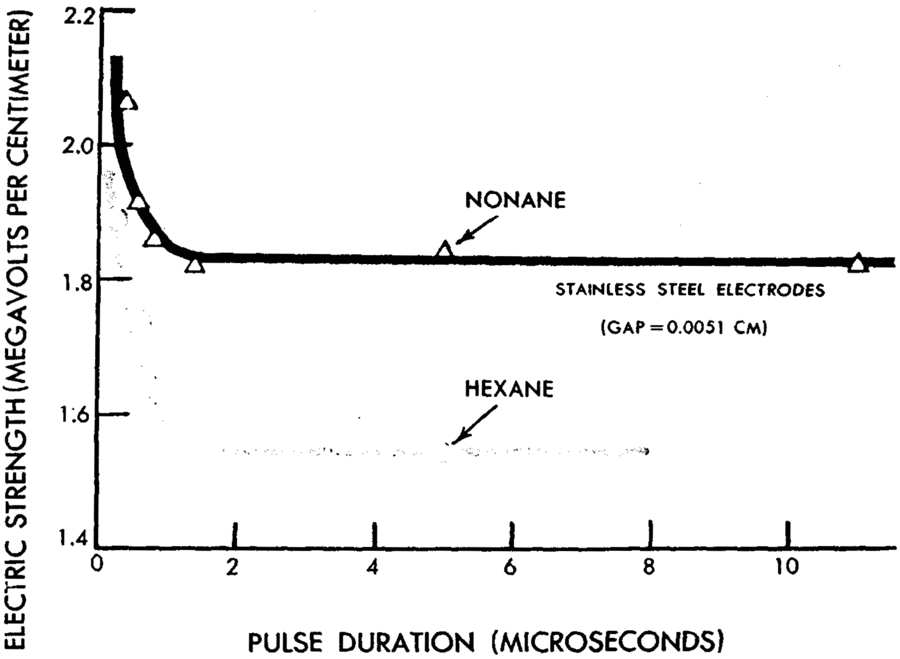


Fig. 5. *INVERSE RELATIONSHIP* of pulse duration to liquid-hydrocarbon strength ends beyond one microsecond. Beyond 10 microseconds, thermal effects complicate matters.

four per cent or less. From such measurements (Fig. 5) the electric strength is found to be independent of pulse duration for times greater than one *microsecond*. Thus the time-independent value of electric strength is the same value you would measure under long-time voltage application—if you could prevent thermal effects. (The latter begin to interfere when pulse duration reaches a value greater than 10 micro-seconds.)

Field emission effects, on the other hand, can only be fixed but not eliminated. And so, the time-independent value of electric strength represents a combined property of the liquid and the electrode material.

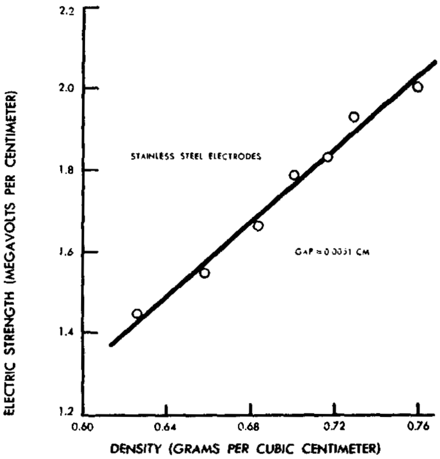


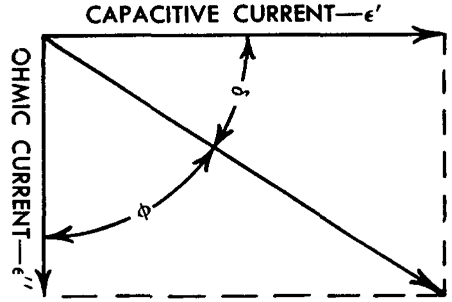
Fig. 6. ELECTRIC STRENGTH of a liquid hydrocarbon, stressed by high-voltage pulses of short duration, is a function of the liquid's density and molecular structure.

HOW THE LOW-VOLTAGE GOODNESS OF A LIQUID DIELECTRIC IS DETERMINED

That most liquid dielectrics are not perfect insulators has been experimentally proved. But they do have a measurable power loss that's manifest as heat. This fact is taken into account by the equation

$$\epsilon^* = \epsilon' - j\epsilon''$$

where ϵ^* is a liquid's complex dielectric constant, having direction as well as magnitude. Its real component ϵ' is the absolute value of the dielectric constant and gives rise to the current out of phase with the applied voltage. The imaginary component ϵ'', called the loss factor, is a measure of the in-phase current or the heat generated per cycle of applied voltage; j equals $\sqrt{-1}$. This is shown graphically—



A quantity expressing the merit of a liquid dielectric is the loss tangent, defined by the relationship, *tan* $\delta = \epsilon''/\epsilon'$. *As you can see from the diagram, the* complement of the angle δ is ϕ; and the cos ϕ is defined as the dielectric's power factor. For low-loss liquids, δ is almost zero, making ϕ nearly 90 degrees; therefore, tan δ=cos ϕ. And so, the loss tangent equals the power factor when tan δ is less than 0.1.

Liquids having a power factor less than about 0.05 at their operating frequency are usually considered satisfactory for commercial use. To give you an idea of magnitudes, the Table lists values for some commonly used liquid dielectrics, including water.

DIELECTRIC PROPERTIES OF SOME LIQUIDS
AT 25 C AND 60 CPS

Liquid	ϵ'	ϵ''	Loss Tangent (Tan δ)	Power Factor (Cos ϕ)
Pyranol†	5.3	0.005	0.001	0.001
Hydrocarbon oil	2.2	0.002	0.001	0.001
Dibutyl sebacate	4.5	0.040	0.010	0.010
Water	78.0	78,000	1000	1.000

†Reg. Trade-mark of General Electric Company

By applying the pulse technique to liquid hydrocarbons of different molecular weight (Fig. 6), it is found that electric strength increases in a linear fashion with the density of the liquid.

THE SUMMING UP

In a general way, some of the more important electrical properties of liquid dielectrics have been presented. And though these properties were divided into two distinct groups—low and high voltage—it doesn't necessarily mean that there is any line sharply dividing dielectrics in usage. For in many applications you'll find liquids with good characteristics at both high and low voltages.

The relationship of a liquid's electric strength to its molecular properties is obviously of value to the engineer in search of a liquid dielectric to fill his specific need. But unfortunately, theories that explain such correlations —or predict the electrical behavior of liquids in high dielectric fields—are presently qualitative at best. Perhaps more information of the type discussed will, in the future, contribute to a better understanding of liquid dielectrics.