# New Synthetic Liquid Impregnants for Capacitors

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**An investigation has been made of the suitability of several new synthetic liquids having dielectric constants from approximately 7 to 23 for use as impregnants for fixed paper-dielectric capacitors. These included five partially fluorinated esters and two fluorosilicones. The characteristics of three chlorinated diphenyl commercial dielectric liquids were also determined. Studies were made of the variation of dielectric constant and dissipation factor with frequency from 60 cycles per second to 75 Mc. and with temperature from −70° to 200° C. Blends of esters were prepared to obtain optimum low temperature performance. Most of the new liquids exhibited electrical properties at low temperatures superior to those afforded by three chlorinated diphenyls. Pentachlorodiphenyl withstood exposure to elevated temperatures with less change in electrical characteristics than any of the liquids studied. The performance of the 250-cs. fluorosilicone was superior in every way to that of the other new materials. One partially fluorinated ester appears worthy of further study.**

THE SEARCH for improved liquid dielectrics has continued since the introduction of synthetic nonflammable organic compounds for this purpose in the 1930's (*3*). Interest has been accelerated in recent years by the growing need for high dielectric constant capacitor impregnants which can be used from −65° to 200° C. (*19*). To obtain better capacitor performance over a wider temperature range, silicone fluids and organic esters have been used (*6*). A partially fluorinated ester having a dielectric constant of about six has been found to given substantial improvement in low temperature properties over existing commercial synthetic liquids (*2*). As a result of this experience, additional compounds were prepared (*18*), and the electrical properties of those showing the greatest promise have been investigated. Parallel studies were made of two fluorosilicones and three commercially available dielectric liquids.

## LIQUIDS SELECTED FOR STUDY

A number of partially fluorinated esters synthesized by Faurote, Murphy, O'Rear, and others (*8, 12, 14*) were investigated. These included bis(1*H*,1*H*,5*H*-octafluoropentyl) phthalate and bis(1*H*,1*H*,5*H*-octafluoropentyl) 3-methylglutarate studied earlier (*16, 17*), and bis(1*H*,1*H*,3*H*-tetrafluoropropyl) diphenate, bis(1*H*,1*H*,5*H*-octafluoropentyl) diphenate, and tris(1*H*,1*H*,3*H*-tetrafluoropropyl) tricarballylate. A high dielectric constant partially fluorinated propyl ester, 1*H*,1*H*,3*H*-tetrafluoropropyl *o*-benzoyl benzoate, supplied by a commercial laboratory, was also studied.

To simplify the nomenclature of the partially fluorinated esters, the fluoroalkyl groups of these compounds (which have $-CF_2H$ terminal groups) are designated as $\psi'$ in this report after the system used previously (*8*). Thus the compound bis(1*H*,1*H*,3*H*-tetrafluoropropyl) diphenate is designated as bis($\psi'$-propyl) diphenate.

Two fluorosilicones with viscosities of 250 and 1000 cs. at 77° F., respectively (*4*), supplied as research samples by the Dow Corning Corp., Midland, Mich., were included in the investigation. Two nitrile silicone fluids having dielectric constants of about 7 and 17, respectively, and a partially fluorinated ester, octafluoropentyl carbonate with a dielectric constant of approximately seven were examined, but were too unstable to warrant further investigation. A sucrose derivative, cyanoethyl sucrose, had a dielectric constant of nearly 40, but the electrical loss characteristics seriously limited the potentiality of the substance as a capacitor impregnant.

To compare the new liquids with those presently recommended for use in the electrical industry (*10, 20*), studies were also made of trichlorodiphenyl, tetrachlorodiphenyl, and pentachlorodiphenyl obtained from the Monsanto Chemical Co., St. Louis, Mo., under the trade names of Aroclor 1242, Aroclor 1248, and Aroclor 1254, respectively (*9, 21*).

**Physical Properties.** The physical properties of those liquids which showed promise for use in capacitors are listed in Table I. Dielectric constants at 60 cycles per second (c.p.s.) and 20° C. range from 6.97 to 23.2 as compared with 5.06 to 5.92 for the chlorinated diphenyls. The new liquids have

Table I. Properties of Liquids Studied

| Compound | Dielectric Constant, 20° C.; 60 C.P.S. | Molecular Weight, Theory | Density 20° C., G./Ml. | Index of Refraction, $N_D^{20}$ | Viscosity, Cs. 68° F. | Viscosity, Cs. 210° F. | Boiling Point, ° C./Mm. Hg | Flash Point, ° F. | Pour Point, ° F. |
|---|---|---|---|---|---|---|---|---|---|
| Bis($\psi'$-propyl) 2,2′-diphenate | 9.87 | 470.31 | 1.4342 | 1.4891 | 1,190 | 8.47 | 168/1.3 | 425 | 0 |
| Bis($\psi'$-amyl) 2,2′-diphenate | 7.90 | 670.35 | 1.5622 | 1.4469 | 1,000$^{a}$ | 10.3 | 175/0.5 | 430 | −5 |
| Tris($\psi'$-propyl) tricarballylate | 10.7 | 518.26 | 1.5514 | 1.3810 | 283 | 5.90 | 154/0.7 | 410 | −30 |
| $\psi'$-Propyl *o*-benzoyl benzoate$^{b}$ | 23.2 | 340.23 | 1.354$^{c}$ | 1.5292 | 2,000$^{d}$ | 13.8 | 390$^{d}$ | 421 | 36 |
| Fluorosilicone, 250 cs.$^{b}$ | 6.97 | ... | 1.24$^{e}$ | ... | 300$^{a}$ | 24 | ... | 440 | −60$^{f}$ |
| Fluorosilicone, 1000 cs.$^{b}$ | 7.20 | ... | 1.28$^{e}$ | ... | 1,200$^{a}$ | 74 | ... | 530 | −40$^{f}$ |
| Pentachlorodiphenyl$^{b}$ | 5.06 | ... | 1.55 | 1.639–41 | 22,000$^{g}$ | 6$^{g}$ | 365–390/760 | None | 50 |
| Tetrachlorodiphenyl$^{b}$ | 5.46 | ... | 1.46 | 1.630–1 | 370$^{g}$ | 3$^{g}$ | 340–375/760 | 379–384 | 19 |
| Trichlorodiphenyl$^{b}$ | 5.92 | ... | 1.38 | 1.627–29 | 78$^{g}$ | 2.5$^{g}$ | 325–360/760 | 348–356 | 2 |

[a] Interpolated. [b] Physical properties from manufacturer. [c] Specific gravity 25° C./25° C. [d] Extrapolated. [e] Specific gravity at 77° F. [f] Freezing point. [g] Approximate.

lower viscosities than that of pentachlorodiphenyl and higher flash points than those of trichlorodiphenyl and tetrachlorodiphenyl. With the exception of that of the $\psi'$-propyl *o*-benzoyl benzoate, the pour or freezing points of the new materials are lower than those of the chlorodiphenyls.

**Purification.** The experimental liquids were available only in small research quantities which made it impractical to employ the more elaborate methods used commercially for purifying the chlorodiphenyls (*21*). All the liquids were passed through small percolation columns containing Florisil and alumina adsorbents. For certain studies of the bis($\psi'$-propyl)2,2'-diphenate, the bis($\psi'$-amyl),2'-diphenate, and of three chlorinated diphenyls, Linde-type 13X Molecular Sieves adsorbent was added to the Florisil and alumina. The percolation columns and receivers were purged with dry nitrogen immediately before use, and the system was protected against the entrance of moisture by drying tubes. The effectiveness of the purification process was determined by measuring the 60-c.p.s. dissipation factor of the percolates at 20° C. Purification was continued with new adsorbents on a batch basis until the dissipation factor of each substance reached a constant low value. Values of less than 0.0001 were achieved for the bis($\psi'$-amyl) diphenate and pentachlorodiphenyl. This is lower than that reported for the latter substance by another investigator (*20*). For the two fluorosilicones, the trichlorodiphenyl, and the tetrachlorodiphenyl, the 60-c.p.s. dissipation factors at 20° C. were less than 0.001. Although the method of purification was not considered the ultimate, it did permit comparison of the relative merits of the materials.

## METHODS USED IN THE MEASUREMENT OF ELECTRICAL PROPERTIES

The dielectric constants and dissipation factors of the liquids were obtained with a General Radio 716 Schering Bridge network together with the necessary amplifiers, oscillators, and detectors. The dielectric constant term referred to in this paper is the relative dielectric constant, a dimensionless number $\epsilon'$, the value of which is the ratio of the capacitance of a capacitor having a given medium between the plates to that of the same capacitor at the same voltage with a vacuum between the plates. An improved Elliot capacitance cell (*7*, *15*) was used to contain the liquid being measured. This cell has the distinct advantage over commercially available cells in that only about 0.3 ml. of liquid is required for filling. In view of the limited quantities available of some of the liquids, this feature was of considerable importance. However, the design of the cell is such that it is not possible to fill it and to make measurements while maintaining the sample under vacuum as is frequently done during the purification of the chlorodiphenyls (*21*). Attemps to operate the Elliott cell in a vacuum after filling were unsuccessful because of the introduction of bubbles in the sample between the closely spaced electrodes. During operation below room temperature, errors in measurement that could have arisen because of the deposition of moisture were prevented by enveloping the cell in a stream of dry nitrogen. Some degree of protection against the effect of air on the sample at elevated temperatures was afforded by the design of the cell. That portion of the sample being measured is surrounded by a considerable portion which lies in a labrynth outside the influence of the electrodes. It is likely that the air and moisture initially dissolved in the sample had more effect on the electrical properties of the materials at elevated temperatures than any further amounts taken up during the measurement.

Data were also obtained at 20° C. over the frequency range 100,000 c.p.s. to 75 Mc. with a calibrated Boonton 160-A Q-Meter and a two-terminal cell used in earlier work (*1*).

The volume resistivities of the several materials studied were obtained with the Elliott cell and a General Radio 544-B megohm bridge. A potential of 500 volts d.c. was applied for one minute.

## RESULTS

**Variation with Temperature of the Dielectric Constant and Dissipation Factor.** The dielectric constant–temperature and dissipation factor–temperature characteristics of the most promising liquids were determined over the temperature range −70° to 200° C. at frequencies of 60, 1000, 10,000, and 100,000 c.p.s. The behavior of bis($\psi'$-amyl)2,2'-diphenate under these conditions, typical of that of viscous liquids (*11*), is shown graphically in Figure 1. The 60-c.p.s. dielectric constant maximum, which occurs at −32° C., governs the temperature at which a capacitor impregnated with this material would undergo an abrupt change in capacitance (*5*). By comparison, tri-, tetra-, and pentachlorodiphenyl can be cooled to only about −20°, −10°, and 0° C., respectively, without undergoing maxima in the 60-c.p.s. dielectric constant (*20*). Because of this limitation, trichlorobenzene has sometimes been added to these materials to obtain capacitor impregnants with improved low temperature characteristics (*20*). This has the disadvantage that trichlorobenzene is a relatively volatile substance as compared with the chlorodiphenyls.

The dissipation factor maxima shown in Figure 1 indicate the approximate temperatures at which heating would occur for four frequencies in an operating capacitor impregnated with bis($\psi'$-amyl) 2,2'-diphenate. Since the maximum dissipation factor of a liquid always occurs at a temperature a little below that at which the maximum in dielectric constant occurs, heating of the capacitor could move the impregnant up into the peak dielectric constant region, all

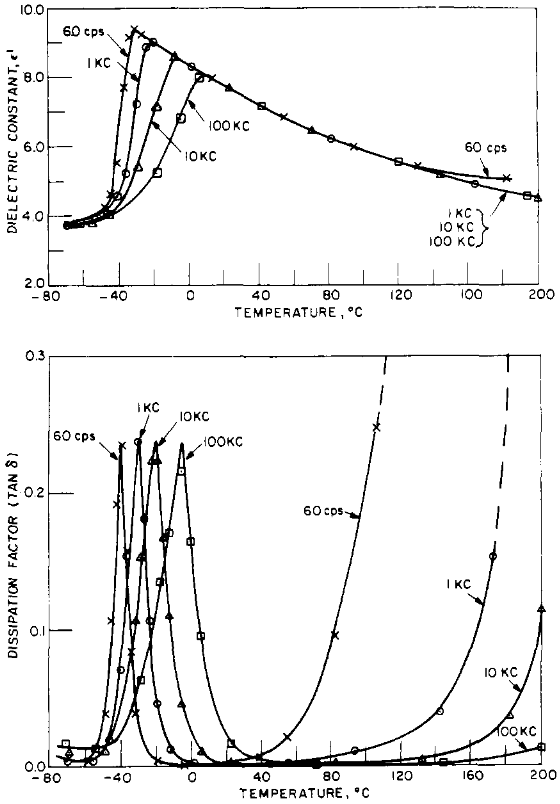

Figure 1. Variation with temperature of the dielectric constant and dissipation factor of bis($\psi'$-amyl)2,2'-diphenate

of which results in an unstable condition. However, a capacitor impregnated with bis($\psi'$-amyl)2,2'-diphenate could be operated at 60 c.p.s. down to about −30° C. without undergoing a sudden change in dissipation factor.

The variation with temperature of the 60-c.p.s. dielectric constants of most of the liquids studied is shown graphically in Figure 2. The dielectric constants are considerably higher at any given temperature than those of the chlorodiphenyls studied (*20*) and all maxima occur at lower temperatures. Most outstanding are the two fluorosilicones which can be cooled approximately 40° C. lower than trichlorodiphenyl (*20*), and 60° C. lower than pentachlorodiphenyl before abrupt changes in dielectric constant and dissipation factor occur. Capacitors impregnated with any of the new liquids in Figure 2 would have superior low temperature characteristics and higher energy-storage–to-volume ratios than similar capacitors impregnated with the chlorodiphenyls recommended for this purpose.

A study was made of the possibility of blending two liquids to obtain improved low temperature characteristics and still maintain a reasonably high dielectric constant. The two partially fluorinated esters studied earlier (*16, 17*) were included as blending agents: bis($\psi'$-amyl)phthalate, chosen because of its high dielectric constant (8.6 at 60 c.p.s. and 20° C.) and bis($\psi'$-amyl)3-methylglutarate, selected because the maximum 60-c.p.s. dielectric constant occurs well below −70° C. (*17*).

Three blends were prepared consisting in each case of 40% of the constituent having the better low temperature characteristics. The results are shown in Table II. Well-defined maxima were obtained and they fell approximately midway between those of the parent compounds. The data indicate that it is possible to obtain improved low temperature properties by using proper blending techniques.

Attempts to blend the two fluorosilicones with the aromatic esters were unsuccessful because of the insolubility of the fluorinated compounds in these esters. On the other hand, the tris($\psi$-propyl)tricarballylate is miscible with the fluorosilicones. However, similar blends made with these compounds would not result in dielectric constant maxima at temperatures any lower than that obtained with the bis($\psi'$-amyl)3-methylglutarate because the 60-c.p.s. dielectric constant maxima of both fluorosilicones occur at temperatures higher than that of the 3-methylglutarate ester.

**Variation with Frequency of the Dielectric Constant and Dissipation Factor.** Although the 60-c.p.s. electrical characteristics of the new dielectric liquids are of primary interest, capacitors impregnated with these materials would be exposed in use to much higher frequencies as well. For the proper design of electronic equipment, it is important to know the frequency at which a capacitor may undergo severe changes in energy-storage ability and in dissipation factor. Since this involves the frequency characteristics of the impregnant, the dielectric constants and dissipation factors of seven liquids were determined at 20° C. over the frequency range 60 c.p.s. to 75 Mc.

The data for the 250-cs. fluorosilicone are shown graphically in Figure 3 and follow the typical pattern (*13*). A summation of all the data is given in Table III. The dielectric constants and dissipation factors of most of the new liquids remained virtually unchanged over a wide range in frequency. In contrast, the dielectric constants of the pentachlorodiphenyl and the benzoate ester began to de-

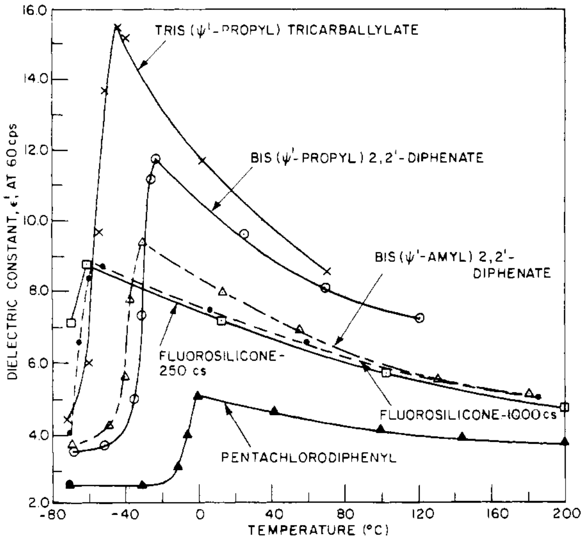

Figure 2. Variation with temperature of the dielectric constant of the most promising liquids

Table II. Effect of Blending on the Maximum Dielectric Constants and Temperatures at which Maxima Occur in the 60-C.P.S. Dielectric Constant and Dissipation Factor

| Material | Max. Dielectric Constant at ° C. | Temperature at Max. Dissipation Factor, ° C. |
|---|---|---|
| Bis($\psi'$-amyl)3-methylglutarate | at < −70 | < −70 |
| Tris($\psi'$-propyl)tricarballylate | 15.5 at −44 | −56 |
| 40–60% blend | 14.3 at −58 | −70 |
| Bis($\psi'$-amyl)phthalate | 12.2 at −48 | −67 |
| Bis($\psi'$-propyl)2,2'-diphenate | 11.7 at −24 | −32 |
| 40–60% blend | 12.1 at −35 | −44 |
| Tris($\psi'$-propyl)tricarballylate | 15.5 at −44 | −56 |
| Bis($\psi'$-propyl)2,2'-diphenate | 11.7 at −24 | −32 |
| 40–60% blend | 12.9 at −34 | −42 |

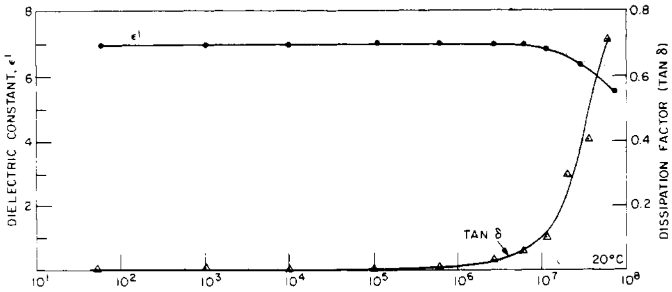

Figure 3. Variation with frequency of the dielectric constant and dissipation factor at 20° C. of 250-cs. fluorosilicone

Table III. Dielectric Constant–Frequency and Dissipation Factor–Frequency Characteristics of the Materials Studied at 20° C.

| Material | Dielectric Constant Is[a] Independent of Frequency from 60 C.P.S. to, C.P.S. | Max. Dissipation Factor Occurs at, C.P.S. |
|---|---|---|
| Fluorosilicone, 250 cs. | $6 \times 10^6$ | $>6 \times 10^7$ |
| Fluorosilicone, 1000 cs. | $2 \times 10^6$ | $4 \times 10^7$ |
| Tris($\psi'$-propyl)tricarballylate | $1 \times 10^6$ | $3 \times 10^7$[b] |
| Bis($\psi'$-propyl)2,2'-diphenate | $3 \times 10^5$ | $4 \times 10^6$[b] |
| Bis($\psi'$-amyl)2,2'-diphenate | $2 \times 10^5$ | $2 \times 10^8$ |
| $\psi'$-Propyl *o*-benzoyl benzoate | $1 \times 10^4$ | $1 \times 10^6$[b] |
| Pentachlorodiphenyl | $1 \times 10^4$ | $5 \times 10^5$ |

[a] Within limits of experimental error. [b] Approximate.

crease at about 0.01 Mc. Since minimum change in capacitance over a wide range in frequency is desired, a manufacturer would find several of the new liquids (the 250-cs. fluorosilicone in particular) to be attractive candidates as capacitor impregnants from this standpoint.

**Stability at Elevated Temperatures.** In addition to the requirement that a good capacitor be useful over a substantial range in temperature and frequency, it should possess long life. If the capacitor has been properly manufactured, life is governed by the ability of the impregnant to withstand elevated temperatures under electrical stress for prolonged periods without undergoing serious deterioration. The effects on the electrical properties of exposing the new dielectric liquids to 100° C. for one hour in the Elliott cell are detailed in Tables IV and V.

The magnitude of the dissipation factors of the chlorodiphenyls at 100° C. at the beginning of the exposure period was greater than that normally expected of these materials (*10*, *20*). This was probably a result of the somewhat limited purification procedure used for all the dielectric liquids studied. However, the magnitude of the initial 100° C. volume resistivities of the tetrachlorodiphenyl, the pentachlorodiphenyl, and the fluorosilicones were above that required for use in capacitors (*21*). The pentachlorodiphenyl withstood exposure to 100° C. with less alteration of electrical characteristics than any of the experimental liquids or the other two chlorinated diphenyls. The bis($\psi'$-amyl)2,2'-diphenate and the two fluorosilicones stood up well enough to be of interest in view of their superior low temperature properties. The three propyl esters were much less promising; their stability was not sufficiently high to justify consideration for high temperature applications in spite of their high dielectric constants. This indicates that the propyl alcohol group used to enhance the magnitude of the dielectric constant severely reduces the ability of the esters to resist deterioration at elevated temperatures. The increased dissipation factor and decreased resistance shown by all the liquids at 100° C. result both from increased

Table IV. Effect of Exposure at 100° C. on the Dissipation Factor

| | Dissipation Factor × $10^4$ | | | | | |
|---|---|---|---|---|---|---|
| | 60 C.P.S. | | | | 1000 C.P.S. | |
| | Measured at 20° C. | | Measured at 100° C. | | Measured at 100° C. | |
| Material | Initial | After 1 hr. at 100° C. | Initial | After 1 hr. at 100° C. | Initial | After 1 hr. at 100° C. |
| Bis($\psi'$-propyl)2,2'-diphenate | 13 | 108 | 6,162 | 10,310 | 410 | 681 |
| Bis($\psi'$-amyl)2,2'-diphenate | <1 | 17 | 804 | 1,270 | 49 | 78 |
| Tris($\psi'$-propyl)tricarballylate | 401 | 3,516[a] | [b] | [a] | 6,100 | [a] |
| $\psi'$-Propyl *o*-benzoyl benzoate | 20 | 145 | [b] | [b] | 6,893 | 11,250 |
| Fluorosilicone, 250 cs. | 9 | 228 | 481 | 1,473 | 29 | 90 |
| Fluorosilicone, 1000 cs. | 8 | ... | 1,047 | 1,693 | 68 | 75 |
| Pentachlorodiphenyl | <1 | 1 | 246 | 324 | 16 | 21 |
| Tetrachlorodiphenyl | 2 | 11 | 887 | 2,772 | 54 | 174 |
| Trichlorodiphenyl | 6 | 108 | 776 | 2,275 | 81 | 143 |

[a] Compound was heated to 100° C. and immediately allowed to cool. [b] Dissipation factor at 60 c.p.s. was beyond range of capacitance bridge.

Table V. Effect of Exposure at 100° C. on the Volume-Resistivity

| | Volume-Resistivity × $10^{-11}$ (Ohm-Cm.)[a] | | | |
|---|---|---|---|---|
| | Measured at 20° C. | | Measured at 100° C. | |
| Material | Initial | After 1 hr. at 100° C. | Initial | After 1 hr. at 100° C. |
| Bis($\psi'$-propyl)2,2'-diphenate | 110 | 21 | 0.76 | 0.50 |
| Bis($\psi'$-amyl)2,2'-diphenate | 420 | 150 | 9 | 5.7 |
| Tris($\psi'$-propyl)tricarballylate | 2.5 | 0.93[b] | 0.065 | [b] |
| $\psi'$-Propyl *o*-benzoyl benzoate | 13 | 1.8 | 0.015 | 0.015 |
| Fluorosilicone, 250 cs. | 480 | 94 | 37 | 22 |
| Fluorosilicone, 1000 cs. | 110 | ... | 16 | 9.3 |
| Pentachlorodiphenyl | >930 | >930 | 30 | 24 |
| Tetrachlorodiphenyl | 330 | 110 | 12 | 6.9 |
| Trichlorodiphenyl | 740 | 51 | 7.5 | 5.4 |

[a] 500 volts d.c. for one minute. [b] Compound was heated to 100° C. and immediately allowed to cool.

Table VI. Effect of Exposure at 100° C. on the Dissipation Factor

| | Dissipation Factor × $10^4$ | | | | | |
|---|---|---|---|---|---|---|
| | 60 C.P.S. | | | | 1000 C.P.S. | |
| | Measured at 20° C. | | Measured at 100° C. | | Measured at 100° C. | |
| Material | Initial | After 24 hrs. at 100° C. | Initial | After 24 hrs. at 100° C. | Initial | After 24 hrs. at 100° C. |
| Bis($\psi'$-amyl)2,2'-diphenate | <1 | 77[a] | 1,626 | 8,027 | 295 | 517 |
| Fluorosilicone, 250 cs. | 10 | 452 | 873 | 5,236 | 49 | 323 |
| Fluorosilicone, 1000 cs. | 4 | 621[b] | 2,328 | 13,370 | 162 | 866 |
| Pentachlorodiphenyl | <1 | 3[c] | 291 | 1,179[d] | 23 | 77 |

[a] After 26 hours at 100° C. [b] After 30 hours at 100° C. [c] After 45 hours at 100° C. [d] 1301 after 45 hours at 100° C.

ionization of the liquid or of impurities in it, and from the lower viscosity of the hot liquid. The resistivity at 100° C. of the 250-cs. fluorosilicone after one hour of exposure approached that of the pentachlorodiphenyl. The final measurements at 20° C. provide some indication of the irreversible increase in ion content as a result of exposure to the higher temperature. These data indicate that pentachlorodiphenyl undergoes very little irreversible change during one hour at 100° C., and that the bis(ψ′-amyl)2,2′-diphenate is the most stable of the experimental liquids studied. The high resistivity of the ester at 20° C. after exposure may reflect some influence of the viscosity of the material at this temperature.

Results of exposing the new dielectric liquids to 100° C. for a short time indicated the desirability of studying the effects of exposure for a longer period. The effect of heating the bis(ψ′-amyl)2,2′-diphenate, the fluorosilicones, and pentachlorodiphenyl for 24 hours at 100° C. are summarized in Tables VI and VII. The further deterioration of the electrical qualities of the liquids after a longer exposure to elevated temperatures is apparent. This results in part from the formation of new ionizable compounds by progressive decomposition of the liquids. However, the pentachlorodiphenyl is again seen to be less altered by the prolonged exposure than the experimental liquids, though the resistivity at 100° C. is somewhat lower than that attained by the 250-cs. fluorosilicone. The dissipation factor and resistivity data indicate the lower viscosity fluorosilicone to be superior to the other new liquids at 100° C. However, data obtained at 20° C. after cooling the heated samples again indicate that the ester has undergone the least irreversible change of the experimental liquids.

Since capacitors are frequently required to operate at temperatures considerably higher than 100° C., the four liquids studied for 24 hours were subjected to an exposure of one hour at 150° C. The effect on the electrical characteristics by this treatment are summarized in Tables VIII and IX. The pentachlorodiphenyl is again seen to withstand exposure to 150° C. better than the experimental liquids, though the resistivity characteristics at 150° C. after one hour closely parallel those of the 250-cs. fluorosilicone. The characteristics of the latter compound at the test temperature were superior to those of the other experimental liquids. After exposure and cooling, the dissipation factor of the ester indicated it to be more stable than the other two liquids, as had been observed in the two previous experiments. From the standpoint of change in resistivity, the 250-cs. fluorosilicone appeared to be the most stable.

## CONCLUSIONS

The low temperature electrical properties of five new dielectric liquids are superior to those of tri-, tetra-, and pentachlorodiphenyl. The two fluorosilicones are the most outstanding; they can be cooled to approximately −60° C. before undergoing severe changes in dielectric constant.

Improved low temperature properties of the aromatic fluoroesters can be obtained by suitable blending with aliphatic fluoroesters, but at the expense of high temperature stability. If the fluorosilicones could be rendered compatible with aromatic fluoroesters by use of a suitable cosolvent or by minor changes in structures, it would be possible to prepare a stable material of higher dielectric constant than the fluorosilicone which would be useful at both high and low temperatures.

Table VII. Effect of Exposure at 100° C. on the Volume-Resistivity

| | Volume-Resistivity × $10^{-11}$ (Ohm-Cm.)[a] | | | |
|---|---|---|---|---|
| | Measured at 20° C. | | Measured at 100° C. | |
| Material | Initial | After 24 hrs. at 100° C. | Initial | After 24 hrs. at 100° C. |
| Bis(ψ′-amyl)2,2′-diphenate | 740 | 19 | 4.1 | 0.54 |
| Fluorosilicone, 250 cs. | 320 | 16 | 42 | 9.7 |
| Fluorosilicone, 1000 cs. | 120 | 11[b] | 8.4 | 2.5[b] |
| Pentachlorodiphenyl | >930 | 860[c] | 16 | 5.7[d] |

[a] 500 volts d.c. for one minute. [b] After 30 hours at 100° C. [c] After 45 hours at 100° C. [d] 5.2 after 45 hours at 100° C.

Table VIII. Effect of Exposure at 150° C. on the Dissipation Factor

| | Dissipation Factor × $10^4$ | | | | | |
|---|---|---|---|---|---|---|
| | Measured at 20° C. | | Measured at 150° C. | | Measured at 150° C. | |
| Material | Initial | After 1 hr. at 150° C. | Initial | After 1 hr. at 150° C. | Initial | After 1 hr. at 150° C. |
| Bis(ψ′-amyl)2,2′-diphenate | 7 | 112 | 6,678 | 17,810 | 446 | 1,203 |
| Fluorosilicone, 250 cs. | 9 | 164 | 2,437 | 3,042 | 153 | 188 |
| Fluorosilicone, 1000 cs. | 8 | 316[a] | 7,048[a] | [a] | 535 | [a] |
| Pentachlorodiphenyl | <1 | 4 | 769 | 1,963 | 53 | 125 |

[a] Material was held at 100° C. for one hour, raised to 150° C. and immediately allowed to cool.

Table IX. Effect of Exposure at 150° C. on the Volume-Resistivity

| | Volume-Resistivity × $10^{-11}$ (Ohm-Cm.)[a] | | | |
|---|---|---|---|---|
| | Measured at 20° C. | | Measured at 150° C. | |
| Material | Initial | After 1 hr. at 150° C. | Initial | After 1 hr. at 150° C. |
| Bis(ψ′-amyl)2,2′-diphenate | 190 | 13 | ... | 0.075 |
| Fluorosilicone, 250 cs. | 100 | 26 | 2.3 | 2.0 |
| Fluorosilicone, 1000 cs. | 110 | 18[b] | 1.6[b] | [b] |
| Pentachlorodiphenyl | >930 | >930 | ... | 1.6 |

[a] 500 volts d.c. for one minute. [b] Material was held at 100° C. for one hour, raised to 150° C., and immediately allowed to cool.

The dielectric constants and dissipation factors of all five new liquids are independent of frequency over a very wide range in contrast with pentachlorodiphenyl. The 250-cs. fluorosilicone is the most outstanding; frequency has little effect on the magnitude of the dielectric constant below 6 Mc.

Studies of the electrical properties at elevated temperatures show that those compounds containing propyl groups are not sufficiently stable to justify consideration for high temperature applications. Pentachlorodiphenyl withstands exposure to elevated temperatures with less change in electrical characteristics than any of the new liquids studied. The electrical properties of the 250-cs. fluorosilicone at elevated temperatures are in general superior to those of the other new liquids under the same conditions.

Capacitors impregnated with the bis($\psi'$-amyl)2,2′-diphenate or one of the fluorosilicones would be capable of storing more electrical energy for a given size and would give better performance at low temperatures than those impregnated with the chlorodiphenyls recommended for this purpose. Further studies are needed to determine the performance characteristics of such capacitors under all operating conditions and to learn whether improvements might be obtained through the use of stabilizing additives.

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# Solubility Measurements Using a Sealed Ampoule

## Dynamic *vs.* Static Method

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**The solubility of several organic solids in a volatile solvent using a sealed ampoule was determined using different rates of heating. Even the slowest rate of heating gave a significant overshoot of the true solubility temperature, and use of the static method of holding the temperature constant for at least three hours was recommended.**

TWO METHODS, each utilizing a sealed ampoule, are in common use for determining the solubility of a solid in a volatile solvent. In the dynamic method (*3*), a sealed glass ampoule containing a weighed amount of solute and solvent is shaken in a liquid bath while the temperature is raised at a constant rate. The bath temperature at which the last portion of solid phase disappears is taken to be the solubility temperature.

In the static method (*1*), the ampoule is shaken in a constant temperature bath for at least three hours. If the last portion of solid has not dissolved, the temperature is raised 0.1° C. and held there for another three hours (smaller temperature increments such as 0.05° can be used if greater accuracy is desired). This process is repeated until the last portion of solid disappears. The temperature at which this occurs is taken to be the solubility temperature. This paper reports the results on several ampoules using both methods and shows the desirability of using the static method for accurate work.

## EXPERIMENTAL

**Chemicals.** Baker's reagent grade benzene (3 liters) was purified by agitation with C.P. sulfuric acid until the acid no

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