

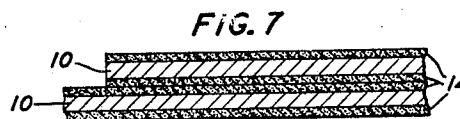
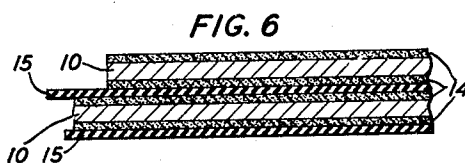
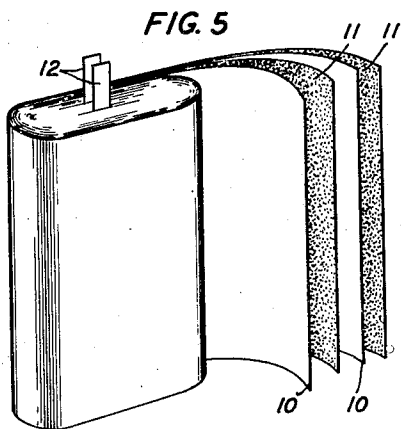
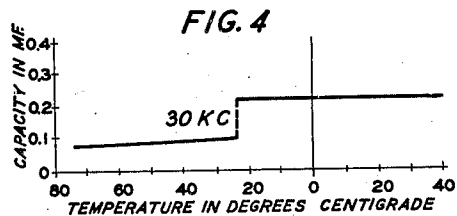
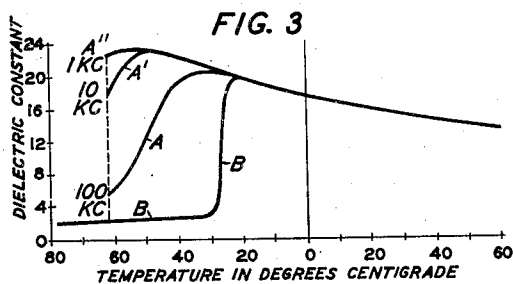
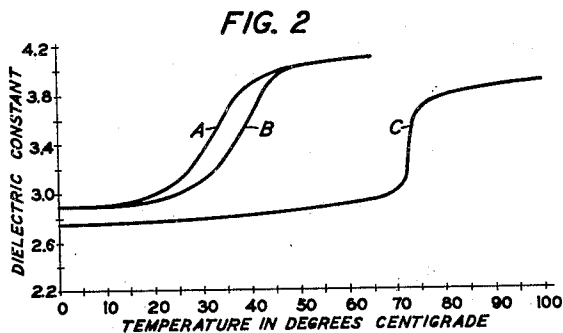
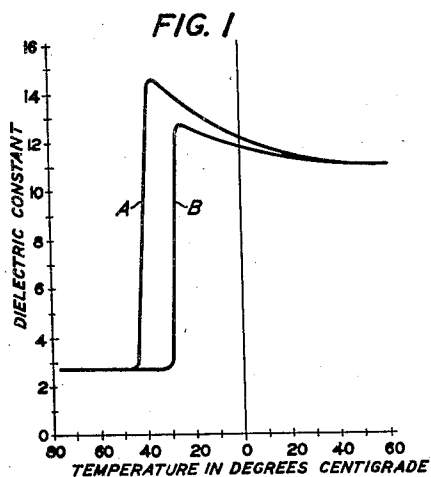
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A. H. WHITE ET AL

2,126,363

DIELECTRIC MATERIAL

Filed March 28, 1935



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DIELECTRIC MATERIAL

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This invention relates to dielectric materials for use in electrical devices, and more particularly to such materials for use in capacitors, wave guides and the like which are adapted to the use of either a liquid or a solid material, either alone or in combination with a porous non-conducting material such as paper.

An object of this invention is to provide dielectric materials of high dielectric constant for use in electrical apparatus such as capacitors, where it is desired to secure a maximum of capacity consistent with other desirable electrical and physical characteristics.

Another object of this invention is to secure dielectric materials which, under the conditions of use, will be solids and which will, either alone or in conjunction with another dielectric such as paper, provide high capacity in electrical apparatus, and which at the same time will have the physical properties, such as melting points, compressibility, viscosity, and penetrative properties, which make them readily applicable to such apparatus.

Another object of this invention is to secure dielectric materials which are soluble in common organic liquids, so as to be suitable for use in solutions, mixtures or pastes, and which are also compatible with other organic solids, so as to permit their use in mixtures having more desirable melting points, fluidities, and penetrabilities than the materials themselves.

Another object of this invention is to provide dielectric materials having a high dielectric constant which is retained at high frequencies.

Another object of this invention is to provide dielectric materials of a water-proof or non-hygroscopic character, and which do not undergo electrolytic changes under the conditions of use in electrical apparatus.

Another object of this invention is to provide a dielectric material which changes its dielectric constant sharply at a certain temperature or within a certain range of temperatures, without undergoing liquefaction.

Another object of this invention is to provide a dielectric material, which when used in conjunction with paper will yield a capacitor having a low temperature coefficient of capacitance.

To accomplish these objects and in accordance with one feature of the invention, a group of dielectric materials is provided which may be defined as polar derivatives of polymethylene cyclic carbon compounds, the dielectric constants of which in the crystalline state are as high or higher than in the liquid state.

In accordance with another feature of the invention one or more of these materials is combined with an organic material such as an insulating wax or a hydrocarbon oil to alter the physical properties of the dielectric without materially affecting its electrical properties.

In accordance with still another feature of the invention these materials may be used either alone or in combination with paper or other porous material as the non-conducting sheets of a capacitor.

It is well known that many materials have a high dielectric constant in the molten or liquid state, but that on passing into the crystalline solid form at the melting point, the dielectric constant of these materials decreases to a low value of about 3. Many other materials have low values of dielectric constant in the liquid state which increase slightly, in proportion to the increase in density on solidifying, but still have low values of the dielectric constant, that is, about 3. The dielectric constant of these classes of materials in the solid state is approximately equal to the square of their refractive index. We have discovered that the following materials, d camphor (natural gum camphor), d-1-camphor (synthetic camphor), camphoric anhydride, borneol, isoborneol, bornyl chloride (pinene hydrochloride), cyclohexanol, chlorocyclohexane and cyclohexene to which this invention relates, have dielectric constants in the crystalline solid state as high as, or higher than, in the liquid state, and also higher than the square of the refractive index for visible light for the respective materials. Also, these materials are applicable as dielectrics in electrical apparatus, where they possess the desirable physical characteristics of solids and yet retain the high dielectric constants usually found only in liquids. There are two kinds of solids, that is, crystalline solids which show a regular arrangement of atomic or molecular building units in an X-ray photograph and glasses or amorphous materials which show a random arrangement of atomic or molecular units. Just as the dielectric behavior of the materials of this invention differs from that of others which solidify to crystalline solids, their behavior also differs from those materials which solidify to glasses, or amorphous, or non-crystalline solids, in a way which will be described later.

We have also discovered that the foregoing substances to which this invention relates have the characteristic that with increasing temperature, the solid crystalline substances undergo at

some temperature below the melting point, a transition from a solid crystalline state of low dielectric constant to a solid crystalline state of high dielectric constant, without undergoing liquefaction. With decreasing temperature, the transition is from a state of high to a state of low dielectric constant. At this transition point, a discontinuous change of density has been observed in the materials to which this invention relates, the change being from a higher density below the transition point to a lower density above the transition point or vice versa; this change being observed on approaching the transition temperature from either direction. An evolution of heat on passing through the transition point in the direction of decreasing temperature has also been observed in these materials, and a corresponding absorption of heat on passing through the transition with increasing temperature. It has also been found that the specific heat follows the same general behavior as the dielectric constant, having a higher value above the transition point than below it.

A practically important feature of these materials is their high dielectric constant in the solid state at temperatures above a transition point. Some of the properties serve only to describe and identify the materials as belonging to the class to which this invention relates.

In the following table are shown the transition temperatures, dielectric constants and densities above and below the transition temperature of certain of the materials to which this invention relates:

Compound	Approximate transition temperature (rising temp.)	Dielectric constant above transition	Dielectric constant below transition	Density below transition	Density above transition
d Camphor.....	-30°	12.5	2.7	1.088	1.008
d-l Camphor.....	-65°	11.5	2.9	1.066	1.026
Camphoric anhydride.....	+135°	24.5	2.5	-----	-----
Borneol.....	+72°	3.9	2.3	0.991	0.987
Isoborneol.....	+25°	4.1	2.9	1.017	1.008
Bornyl chloride.....	-115°	7.0	2.5	1.14	1.10
Cyclohexanol.....	-28°	20.3	2.5	-----	-----
Chlorocyclohexane.....	-55°	10.8	2.9	-----	-----

Substances capable of undergoing such a transition with respect to dielectric properties will be referred to hereafter as transition dielectrics and the temperature at which such transitions occur will be referred to as transition temperatures. The transition from one solid form to the other which these materials show, does not in all cases take place sharply at one temperature, but may occur over a range of temperatures. Also, the temperature at which the transition takes place with decreasing temperature may be different from that with increasing temperature. This behavior is hereafter referred to as a hysteresis of the transition.

The invention both as to its further objects and features will be more clearly understood from the following detailed description taken together with the accompanying drawing in which:

Fig. 1 shows the dielectric constant and temperature hysteresis of transition of d camphor;

Fig. 2 similarly shows the dielectric constant and temperature hysteresis of isoborneol and borneol;

Fig. 3 shows the dielectric constant and temperature hysteresis of cyclohexanol at different frequencies;

Fig. 4 shows the change in capacity at various temperatures for a capacitor, the non-conducting

sheets of which are made of paper impregnated with cyclohexanol;

Fig. 5 shows an embodiment of the invention in which applicant's material is employed as an impregnating material for the porous non-conducting sheets of the capacitor;

Fig. 6 shows in cross-section a modified capacitor structure in which the dielectric material of this invention is applied directly to the conducting sheets of the capacitor and used in combination with sheets of insulating material; and

Fig. 7 shows in cross-section a structure similar to that of Fig. 6 in which the insulating sheets are eliminated.

Referring to the drawing, Fig. 1, shows a temperature hysteresis curve of the transition for d camphor. For decreasing temperatures, shown on curve A, the transition takes place at -37° C. while for increasing temperatures, as shown on curve B, transition takes place at -30° C. Similar hysteresis curves are shown on Fig. 2 in which curve A shows the hysteresis in the transition for isoborneol with a descending temperature and curve B shows the similar transition for ascending temperatures. With this material it will be noted that the hysteresis is spread out over a much wider range of temperatures than was the case for d camphor shown in Fig. 1. Curve C of Fig. 2 is a corresponding curve for borneol in which it will be noted that there is no hysteresis of the transition, the values of dielectric constant for ascending and descending temperatures falling on the same curve in this case. For the materials illustrated by Figs. 1

and 2, the dielectric constant curves shown are the same for all frequencies from 60 to 10,000,000 cycles per second. In some of the transition dielectrics a dependence of the dielectric constant upon frequency has been observed. For example, with d-l camphor, the change in capacity is independent of frequency for temperatures above -90° C. but below that point it has a dielectric constant when measured at 100 kilocycles, somewhat below that when employing a frequency of 1 kilocycle. Bornyl chloride behaves similarly. The dielectric constant above the transition and during a part of its course through the transition is independent of frequency, but at lower temperatures depends somewhat upon frequency. In these cases the frequency behavior is the same, irrespective of whether the transition is approached with ascending or descending temperatures.

A still different type of dielectric behavior in the transition dielectrics is illustrated by Fig. 3 for cyclohexanol. Starting at low temperatures with the dielectric in the low temperature crystalline form, the dielectric constant is independent of frequency and increases sharply at the transition point to its high value along curve B, still essentially independent of frequency. Starting from higher temperatures, however, the

transition does not take place at the same temperature but shows a hysteresis and one which is dependent upon frequency in the manner shown. Curve A for 100 kilocycles begins to decrease at a higher temperature than curve A' for 10 kilocycles. Curve A'' for 1 kilocycle decreases at still lower temperatures. At the temperature indicated by the dotted line the super-cooled material recrystallized to the low temperature form. The purity of the material has been found to be one factor, though not necessarily the only one, which may cause this observed supercooling and dependence of dielectric constant upon frequency. Knowledge of the fact and temperature of transition, and of the hysteresis which may be present, permits one to bring about solidification under conditions so as to insure the presence of either form as desired.

Examination of the accepted chemical formulae for the transition dielectrics shows them to be polar derivatives of polymethylene cyclic hydrocarbons. They may be mono-cyclic as in cyclohexanol or polycyclic as in borneol. They may consist of rings of five like atoms as in the polycyclic terpene derivatives or of six like atoms as in cyclohexanol. The substances exhibiting the dielectric transition phenomena contain at least one substituent group of polar type so situated as to lead to its classification as a polar compound. This polar group may be carbonyl as in camphor, hydroxyl as in borneol, isoborneol or cyclohexanol, or chlorine as in bornyl chloride or chlorocyclohexane. Or they may contain two polar groups as in the anhydride structure of camphoric anhydride. Or the polar group may be an oxime as in camphor oxime. Or polarity may be contributed by partial unsaturation as in cyclohexene.

While the previous examples describe polar derivatives of polymethylene hydrocarbons which contain only carboxyl, hydroxyl, chlorine or oxime groups or which have an anhydride structure or which are partially unsaturated other derivatives may be used as well. For example, the methyl, ethyl or other alkyl substituted derivatives or the nitro, or nitrile, or methoxy, or carboxy, or ester derivatives of polymethylene cyclic hydrocarbons may be used. We do not exclude other substituted groups, such as bromine or iodine, but prefer the substituted groups or atoms of lower atomic weight.

The magnitude of the dielectric constant which these transition dielectrics have in the solid state is related to the polarity of the substituent group and the arrangement of these polar groups in the cyclic structure. For the highest values of dielectric constant, the preferred substituent groups are nitro (NO₂), nitrile (CN), carboxyl (CO), chlorine (Cl) and hydroxyl (OH). The preferred arrangement of these groups in the case of compounds containing more than one substituent group is an asymmetrical one. For example, ortho-dichloro cyclohexane is preferred to paradichlorocyclohexane.

The temperature at which the transition occurs is related to the intermolecular forces acting in the solid and to the chemical nature of the substituent group, although in a complicated manner. For a low transition temperature, the preferred case is a solid of low intermolecular forces or internal fields.

The transition dielectrics to which this invention relates are, therefore, described and dis-

tinguished by the fact that they are polar derivatives of polymethylene cyclic carbon compounds.

The following substances which resemble one or more of the substances to which this invention relates in some feature of chemical constitution do not exhibit the above described transition. Hexane is a saturated hydrocarbon but is not a transition dielectric, being non-polar and non-cyclic. Chlorobenzene is a cyclic, polar substituted hydrocarbon, but is not a transition dielectric, being an aromatic, or aryl cyclic rather than polymethylene compound. Benzoic anhydride is a cyclic polar substituted compound having an anhydride structure, but it is not a transition dielectric, being an aromatic cyclic compound. In this way we distinguish transition dielectrics from all other classes of dielectrics in that they are polar derivatives of polymethylene cyclic hydrocarbons.

In Fig. 5 the invention is shown as embodied in a capacitor unit of the rolled type, which consists of a pair of conducting sheets 10, 10 alternating with sheets 11, 11 of porous non-conducting material such as paper or textile fabric, these sheets being impregnated with a dielectric material containing a polar derivative of polymethylene cyclic carbon compounds. The non-conducting sheets 11, 11 may be impregnated before their assembly in alternate position with the conducting plates and then wound, but preferably the non-conducting and conducting sheets are wound into a unit and then impregnated with insulating material. Terminal members 12, 12 are provided as shown to permit connecting the respective conducting sheets in an electrical circuit.

In the modified construction as shown in cross-section in Fig. 6 the dielectric material of this invention is applied directly as coatings 14, 14 to the conducting sheets 10, 10. Insulating sheets 15, 15 are positioned as shown and the assembled sheets wound into unit form as shown on Fig. 5. The structure of Fig. 7 differs from that of Fig. 6 only in that the insulating sheets 15, 15 are dispensed with. The dielectric materials of this invention are also applicable to the use in the stacked type of capacitor in which the conducting plates instead of being rolled are formed into a stack in the well-known manner.

The paper insulated capacitor of the type shown in Fig. 5 and designed for operation at low voltage, 500 volts or less, having cubical dimensions of 13.3 cm³ has, when impregnated according to standard practice with chlorinated naphthalene, a commercial dielectric, a capacity of 1.0 microfarad. When such a paper insulated capacity unit is similarly impregnated with cyclohexanol, its capacity is increased to approximately 1.4 microfarads, an increase of approximately 40%. If no increase of dielectric capacity is desired, the size of the capacitor may be reduced when impregnated with the materials to which this invention relates. For example, a one-microfarad capacitor of the type illustrated in Fig. 5 when impregnated with chlorinated naphthalene or chlorinated diphenyl has a cubical volume of 13.3 cm³. When impregnated with cyclohexanol the one-microfarad capacitor of corresponding dielectric thickness need have a cubic volume of only 9.5 cm³.

Due to the fact that the temperature coefficient of dielectric constant of cyclohexanol has the opposite sign from that of paper over a considerable range of temperatures, a paper insulated capacitor of the type disclosed in Fig. 5

when impregnated with cyclohexanol may have zero or a very small temperature coefficient of capacitance over a wide temperature range. This is clearly illustrated in Fig. 4 of attached drawing which shows that over the temperature range from -20°C. to $+40^{\circ}\text{C.}$ the capacity measured at a frequency of 30,000 cycles changes only to a very slight degree. A paper insulated condenser impregnated with cyclohexanol was found to change its capacity at the transition point by approximately 100%. In other cases using these transition dielectrics for impregnating paper condensers similar changes of 25% or more take place. Dielectric constants of the solids previously listed in tabular form range from 4 to 24 as shown and the transition points range from -115°C. to $+135^{\circ}\text{C.}$ Therefore, capacitors may be prepared from combinations of these or other transition dielectrics with paper, for example, having a wide range of capacities and a great variety of temperature characteristics.

Heretofore, it has been common practice to use as separator in capacitors intended for impregnation a relatively dense paper, because the dielectric constant of the cellulosic material constituting the paper is higher than that of the available dielectric materials suitable for impregnation. With these new dielectric materials the dielectric constant of the paper is no longer the limiting factor but higher capacities can now be obtained by the use of more porous paper and hence a larger percentage of impregnating material.

The use of dielectric materials undergoing such a transition is not limited to the pure forms of the materials. In suspensions, mixtures or pastes, where there is some transition dielectric present as a solid phase, the transition is observed.

Some of the transition dielectrics described herein melt at temperatures too high to permit their use alone for impregnating condensers according to the usual procedure. Such transition dielectrics may, however, be used in solutions or mixtures with other dielectric materials such as mineral oil, rosin oil, chlorinated diphenyl, halowax or superlawax, in such a manner as to retain the advantage due to their high dielectric constant. In such mixtures the dielectric constants represent roughly a mean between the dielectric constants of the two components approximately in proportion to the molar fraction of each.

A paper insulated capacitor of the type disclosed in Fig. 5, when impregnated with a solution of chlorinated diphenyl containing 40% by weight of d camphor, had a capacity approximately 25% higher than a similar capacitor similarly impregnated with the chlorinated diphenyl alone. This 40% solution of d camphor in chlorinated diphenyl was not saturated with respect to camphor at room temperature. At 80°C. , a more suitable temperature for impregnation, the d camphor is soluble to the extent of 60% and good impregnation results. A 60% solution of d camphor in chlorinated diphenyl is supersaturated with respect to d camphor at room temperature and a mush or paste results on cooling. Capacitors impregnated with the 60% solution had about 30% higher capacity than those impregnated with chlorinated diphenyl alone. This mixture is essentially non-hydroscopic and non-water soluble. It does not un-

dergo electrolytic changes under conditions of measurement.

These transition dielectrics when intimately mixed with certain organic materials, by melting the two to a homogeneous solution and then solidifying, may show either of two types of dielectric behavior. In one of these, to be designated as solid mixtures of type A, the transition point of the mixture is the same as that of the pure transition material and the value of the dielectric constant is roughly proportional to the mol fraction and dielectric constant of the transition dielectric. Mixtures of d camphor with phthalic anhydride (having 9, 21 and 37 mol. per cent phthalic anhydride) are examples of this type. In the other of these, to be designated as solid mixtures of type B, the transition point is displaced by the presence of the second solid. Mixtures of d camphor with camphoric anhydride containing 25, 50 and 75 mol. per cent of d camphor show a progressive lowering of the transition point of camphoric anhydride. In this way, mixtures may be prepared having the transition point at any temperatures desired. For satisfactory operation it is desirable that these mixtures be liquid above 100°C. and solidify at approximately 50°C.

To obtain mixtures of transition solids with other solids having the dielectric behavior of type A solid mixtures, the preferred choice of the second component is one which forms simple eutectic mixtures with the transition solid.

We have made a theoretical study of the matter in order to establish a sound basis for defining the class of materials to which this invention relates. According to our study of the matter, the theoretical interpretation of the phenomenon of transition is as follows. The dielectric constant of a material may be expressed in terms of its dielectric polarizability or the ability of the electric charges to be displaced by an electric field. There are many equations for this relationship but the preferred one is

$$p = 3 \left(\frac{K-1}{K+2} \right)$$

where p is the polarizability and K the dielectric constant. The polarizability may be mathematically defined by the equation

$$p = \sum_{j=1}^K N_j \frac{e^2}{f_j} + \sum_{j=K+1}^m \frac{N_j e^2}{(f_j - m_j w^2 + i r_j w)} + \sum_{n=m+1}^{\infty} \frac{N_n e^2}{(f_n + i r_n w)}$$

The purpose of this equation is to show that p , the polarizability, and hence K , the dielectric constant, may be defined in terms of f_j , a constant depending upon the restoring force which opposes the displacement of any charge, for example, of the j th type, m_j , a constant depending upon the mass of the charged particle, of the j th type, for example, and r_j , a constant depending upon the frictional resistance to the displacement of the charged particle, of the j th type, for example.

These are also the factors which determine the vibrations or rotations of charges or groups of charges of which the material is composed. The motions of the charges within a dielectric may, for the purpose of illustration, be regarded as separable into vibrations or rotations of the electrons or atoms, or groups of atoms, or molecules or groups of molecules, which may be regarded as the units of which the dielectric is

composed. Each of these vibrations or rotations has a characteristic frequency and amplitude and it is the summation of the effect of an electric field upon these vibrations and rotations which determines the dielectric behavior of any material.

The dielectric constant of a material is affected by all of the factors which affect the vibrations or rotations of the atoms or molecules, or groups of atoms or molecules. These factors include temperature, pressure, voltage, frequency and factors, such as internal field, which determine the state of aggregation of the material. On this basis, reasonable explanations of the effects of these factors upon dielectric behavior have been developed and described in the literature.

The high dielectric constant of certain substances in the liquid state is due largely to the fact that their molecules or aggregates of molecules which act as the unit possess permanent electric dipoles and are, therefore, classed as polar dielectric materials. A molecule is said to possess a permanent electric dipole when it is electrically asymmetrical, that is, when the center of gravity of the positive charges is separated from the center of gravity of the negative charges by a finite distance. A molecule containing a permanent electric dipole will tend to orient itself in an electric field analogously to the well-known behavior of a permanent magnet orienting itself in a magnetic field.

Among organic materials of the sort to which this invention relates, there is but little difference in dielectric constant, apart from that due to the permanent dipole. For example, benzene, hexane, naphthalene, paraffin, cyclohexane, carbon tetrachloride, and other symmetrical or non-polar materials all have dielectric constants in the liquid state between two and three, these being considered low values. Substitution of polar groups in these materials increases the dielectric constant by an amount which depends upon the polar group. For example, liquid nitrobenzene has a dielectric constant of 34 as compared to 2.3 for benzene. Liquid chlorobenzene has a dielectric constant of 5.5, the chlorine being less polar than the nitro group. These materials are distinguished from transition dielectrics in that their dielectric constants decrease to a value between 2 and 3 at their freezing points. The position of substitution determines the dielectric constant. For example, ortho-dichlorobenzene has a dielectric constant approximately 10, while paradichlorobenzene, which differs in that the substituted groups are symmetrically located, has a dielectric constant of only 2.5 in the liquid state.

The concept of dielectric behavior which has been outlined is the basis for expecting that the dielectric materials of this invention, being polar, should have high dielectric constants in the liquid state, where the polar molecules are free to orient themselves. The only further requirement to account for the high dielectric constants in the solid state is to show that molecules or parts of molecules are also able to orient or rotate in the solid state, at least in one direction, and within certain limits. Recent theoretical and experimental study has produced a basis for believing that rotation is possible in the solid state.

The materials to which this invention relates belong to a class having a transition within a range of temperatures at which some rearrangement of the forces holding the molecules to-

gether takes place, such that the intermolecular forces are weakened to an extent that at higher temperatures they are able to take up, from whatever source, enough energy to cause them to rotate.

It is an essential part of our invention that we have discovered that the preferred class of dielectric materials to provide high dielectric constants in the solid state is the polymethylene cyclic polar substituted hydrocarbons. Although the theory of dielectric behavior outlined above very completely describes our experimental observations, we do not limit our invention to this theoretical interpretation nor confine ourselves to such substances to which this interpretation may ultimately be found to apply.

What is claimed is:

1. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic carbon compound which in the solid state undergoes a transition in dielectric properties at a particular temperature.

2. A dielectric material for electrical apparatus comprising as a major constituent a substance capable of undergoing a transition in the solid state, at a particular temperature, from a state of high dielectric constant to a state of lower dielectric constant.

3. A dielectric material for electrical apparatus comprising a substance capable of undergoing a transition in the solid state, within a range of temperatures, passing from a state of high dielectric constant to a state of lower dielectric constant with decreasing temperature and from a state of low dielectric constant to a state of higher dielectric constant with increasing temperature.

4. A dielectric material for electrical apparatus comprising as a major constituent comprising a polar derivative of a polymethylene cyclic hydrocarbon having a dielectric constant in the solid state, above a transition, greater than four, which value is substantially independent of frequency between sixty cycles and ten million cycles per second.

5. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic hydrocarbon having a dielectric constant in the solid state above a transition greater than the square of the optical refractive index of the material for visible light.

6. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic hydrocarbon having a dielectric constant in the solid state, greater than four, which value increases with decreasing temperature in the range of temperatures between the melting point and the transition point.

7. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic hydrocarbon which is essentially non-hygroscopic and non-water soluble.

8. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic hydrocarbon which is substantially free from electrolytic changes under the conditions of use in electrical apparatus.

9. A dielectric material for electrical apparatus comprising a mixture of a polar derivative of a polymethylene cyclic hydrocarbon which in the solid state undergoes a transition in dielectric

properties at a particular temperature with other dielectrics, said mixtures being molten above 100° C. and solid below 50° C.

5 10. A dielectric material for electrical apparatus comprising a mixture of a polar derivative of a polymethylene cyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature with other dielectrics, said mixture being a homogeneous solution at 100° C. and a suspension or paste of crystalline transition dielectric in a liquid phase composed mainly of the second component below 50° C.

15 11. A dielectric material for electrical apparatus comprising a mixture of a polar derivative of a polymethylene cyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature with a non-polar dielectric, said polymethylene cyclic hydrocarbon comprising a major portion of said mixture.

20 12. A dielectric material for electrical apparatus comprising a polar derivative of a polymethylene monocyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature.

25 13. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene bicyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature.

30 14. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a cyclic terpene which in the solid state undergoes a transition in dielectric properties at a particular temperature.

35 15. A dielectric material for electrical apparatus comprising a polar derivative of a polymethylene monocyclic hydrocarbon having a dielectric constant in the solid state above a transition greater than the square of the optical refractive index of the material for visible light.

40 16. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene bicyclic hydrocarbon having a dielectric constant in the solid state above a transition greater than the square of the optical refractive index of the material for visible light.

50 17. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a cyclic terpene having a dielectric constant in the solid state above a transition

greater than the square of the optical refractive index of the material for visible light.

18. A dielectric material for electrical apparatus comprising a polar derivative of a polymethylene monocyclic hydrocarbon having a dielectric constant in the solid state greater than four, the value of the dielectric constant increasing with decreasing temperature in the range of temperatures between the melting point of said derivative and the transition point thereof.

10 19. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene bicyclic hydrocarbon having a dielectric constant in the solid state greater than four, the value of the dielectric constant increasing with decreasing temperature in the range of temperatures between the melting point of said derivative and the transition point thereof.

20 20. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a cyclic terpene having a dielectric constant in the solid state greater than four, the value of the dielectric constant increasing with decreasing temperature in the range of temperatures between the melting point of said derivative and the transition point thereof.

25 21. A dielectric material for electrical apparatus comprising as a major constituent a polar derivative of a polymethylene cyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature, said derivative having one polar substituent.

30 22. A dielectric material for electrical apparatus comprising a polar derivative of a polymethylene cyclic carbon compound which in the solid state undergoes a transition in dielectric properties at a particular temperature, said derivative having two polar substituents.

35 23. A dielectric material for electrical apparatus comprising as a major constituent an asymmetric polar derivative of a polymethylene cyclic hydrocarbon which in the solid state undergoes a transition in dielectric properties at a particular temperature.

40 24. A dielectric material for electrical apparatus comprising iso-borneol.

45 25. A dielectric material for electrical apparatus comprising camphoric anhydride.

50 26. A dielectric material for electrical apparatus comprising cyclohexanol.

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