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(54) **Liquid dielectric composition.**

(57) A liquid dielectric composition obtained as a result of a process which comprises reacting benzene with ethylene in the presence of an alkylation catalyst to obtain an alkylation product containing largely unreacted benzene, ethylbenzene, polyethylbenzenes, 1,1-diphenylethane and heavier products, separating benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane from said alkylation product and thereafter recovering from said heavier products a fraction having a boiling point in the temperature range of about 275° to about 420°C., preferably about 280° to about 400°C., as said liquid dielectric composition.

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Liquid Dielectric Composition

The invention defined herein relates to a liquid dielectric composition obtained as a result of a process which comprises reacting benzene with ethylene in the presence of an alkylation catalyst to obtain an alkylation product containing largely unreacted benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane and heavier products, separating benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane from said alkylation product and thereafter recovering from said heavier products a fraction having a boiling point in the temperature range of about 275° to about 420°C., preferably about 280° to about 400°C., (including any portion thereof) as said liquid dielectric composition.

Polychlorinated biphenyls have been extensively employed commercially in the electrical industry over a long period of time as liquid insulating fluids, but because of environmental and toxicological problems associated therewith, substitutes therefor are required.

We have found that a liquid dielectric composition can be obtained from a process which comprises reacting benzene with

ethylene in the presence of an alkylation catalyst to obtain an alkylation product containing largely unreacted benzene, ethylbenzene, polyethylbenzenes, 1,1-diphenylethane and heavier products separating benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane from said alkylation product and thereafter recovering from said heavier products a fraction having a boiling point in the temperature range of about 275° to about 420°C., preferably about 280° to about 400°C., as said liquid dielectric composition.

In the alkylation of benzene with ethylene an alkylation product is obtained containing largely unreacted benzene, ethylbenzene, polyethylbenzenes and a higher-boiling product. From said alkylation product the unreacted benzene, ethylbenzene and polyethylbenzenes are recovered and said higher-boiling product is said to have only fuel value. In U.S. Patent No. 4,011,274, dated March 8, 1977, Masaaki et al recover from said higher-boiling product 1,1-diphenylethane and state that the resulting residue is still available as fuels. In the present application we have found, unexpectedly, that from said resulting residue we can obtain a fraction or fractions useful as liquid dielectric compositions.

Briefly, the process employed in obtaining the new liquid dielectric compositions defined and claimed herein comprises reacting benzene with ethylene in the presence of an alkylation catalyst to obtain an alkylation product containing largely unreacted benzene, ethylbenzene, polyethylbenzenes, 1,1-diphenylethane and heavier, still higher-boiling, products, separating benzene,

ethylbenzene, polyethylbenzenes and 1,1-diphenylethane from said alkylation product and thereafter recovering from said heavier products a fraction having a boiling point in the temerature range of about 275° to about 420°C., preferably about 280° to about 400°C., as said liquid dielectric composition.

The alkylation of benzene with ethylene that can be employed to obtain the new liquid dielectric compositions claimed herein can be any of the processes known in the art for producing a product containing ethylbenzene, for example, either liquid phase alkylation or vapour phase alkylation. The molar ratios of benzene to ethylene employed can be, for example, in the range of about 25:1 to about 2:1, preferably about 10:1 to about 3:1. In the liquid phase reaction, for example, the benzene and ethylene, together with an alkylation catalyst, for example a Friedel Crafts catalyst, such as aluminium chloride or aluminium bromide or some other organo-aluminium halide; Lewis acids, such as promoted $ZnCl_2$, $FeCl_3$ and BF_3; and Bronsted acids, including sulfuric acid, sulfonic acid and p-toluenesulfonic acid, hydrofluoric acid, etc., in an amount corresponding to about 0.002 to about 0.050 parts, preferably about 0.005 to about 0.030 parts, relative to ethylbenzene produced, are reacted in a temperature range of about 20° to about 175°C., preferably about 90° to about 150°C., and a pressure in the range of about atmospheric to about 250 pounds per square inch gauge (about atmospheric to about 17.6 kilograms per square centimeter), preferably about seven to about 200 pounds per square inch gauge (about 0.5 to about 14 kilograms per square

centimeter), for about ten minutes to about 10 hours, preferably for about 20 minutes to about three hours. In the vapour phase, for example, the reactants can be passed over a suitable alkylation catalyst bed containing alkylation catalysts, such as phosphoric acid on kieselguhr, silica or alumina, aluminium silicates, etc., at a convenient hourly space velocity in a temperature range of about 250° to about 450°C., preferably about 300° to about 400°C., and a pressure of about 400 to about 1200 pounds per square inch gauge (about 28 to about 85 kilograms per square centimeter), preferably about 600 to about 1000 pounds per square inch gauge (about 42 to about 70 kilograms per square centimeter).

As a result of such reactions, an alkylation product is obtained containing unreacted benzene, the desired ethylbenzene, polyethylbenzenes, such as diethylbenzens and triethylbenzene, 1,1-diphenylethane and higher-boiling products.

The alkylation product can be treated in any conventional manner to remove any alkylation catalyst present therein. For example, when aluminium chloride is used as catalyst, the alkylation product can be sent to a settler wherein the aluminium chloride complex is removed and recycled to the reaction zone and the remaining product can then be water washed and neutralized.

The resulting alkylation product is then distilled at atmospheric pressure or under vacuum to recover unreacted benzene (B.P. 80°C.), ethylbenzene (B.P. 136°C.), polyethylbenzenes (B.P. 176°-250°C.) and 1,1-diphenylethane (B.P. 270°C.).

The heavier product remaining after removal of benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane, as described above, is a dark, viscous, high-boiling material from which the novel liquid dielectric compositions defined and claimed herein are obtained. To obtain the claimed novel liquid dielectric composition, the said heavier product is simply subjected to distillation and those portions recovered having a boiling point at atmospheric pressure (14.7 pounds per square inch gauge or 760 millimeters of mercury) in the temperature range of about 275° to about 420°C., preferably about 280° to about 400°C., constitute the desired and novel liquid dielectric composition. The remaining heavier material or residue is a black asphalt-like material solid at ambient temperature believed, in part, to be polynuclear structure having fuel value only.

A number of liquid dielectric compositions were prepared from the residue, or heavier products, obtained as a result of the production of ethylbenzene. This residue was obtained as follows: Benzene and ethylene in a molar ratio of 9:1 were contacted in the liquid phase, while stirring, in a reactor at a temperature of 130°C. and a pressure of 70 pounds per square inch gauge (4.9 kilograms per square centimeter) in the presence of $AlCl_3$ catalyst over a period of one hour, which was sufficient to convert all of the ethylene, The $AlCl_3$ complex catalyst was prepared by dissolving $AlCl_3$ in a polyethylbenzene cut from a previous run so that after the addition the composition of the catalyst complex was as follows:

31.3 weight per cent $AlCl_3$, 7.0 weight per cent benzene, 19.3 weight per cent ethylbenzene, 29.8 weight per cent polyalkylated benzenes, 3.4 weight per cent 1,1-diphenylethane and 9.0 weight per cent higher-boiling components. The amount of $AlCl_3$ present in the catalyst mixture amounted to 0.0034 parts by weight per one part by weight of ethylbenzene produced. Also present in the catalyst was ethyl chloride promoter in an amount corresponding 0.0034 parts by weight per one part by weight of ethylbenzene produced to maintain a high catalyst efficiency. Analysis of the alkylation product showed the presence of 49.0 weight per cent benzene, 32.9 weight per cent ethylbenzene, 17.5 weight per cent of polyalkylated benzenes (6.0 weight per cent diethylbenzene, 2.7 weight per cent triethylbenzenes, 2.1 weight per cent tetraethylbenzenes and 6.7 weight per cent other alkylbenzenes), 0.1 weight per cent 1,1-diphenylethane and 0.4 weight per cent residue. The alkylation product was subjected to distillation to recover unreacted benzene, ethylbenzene, polyalkylated benzenes and 1,1-diphenylethane, and the benzene and polyalkylated benzenes were recycled to the reaction zone. The residue remaining was a dark, viscous, high-boiling material, and was produced in an amount corresponding to 0.012 parts for each part of ethylbenzene produced. By using aged aluminium chloride complex the amount of high-boiling residue formed can be increased substantially.

The residue obtained above was subjected to distillations at atmospheric pressure and cuts, or combination of cuts, that

were recovered were subjected to tests (ASTM-D924) at 25°C to determine their power factors and dielectric constants. In the first series of tests a sample was arbitrarily fractionated into two cuts, one boiling between 280° and 300°C. and a second boiling between 300° and 405°C. When these cuts were subjected to the above tests the following results were obtained:

TABLE I

	Dielectric Strength kV.	Power Factor, Per Cent
Cut No. 1	50+	0.032
Cut No. 2	50+	0.005

Another sample of the residue defined above was fractionated into two cuts: a first boiling between 301° and 307°C. (Cut No. 3) and a second boiling between 319° and 399°C. (Cut No. 4). These cuts were similarly tested and found to have excellent dielectric strength.

TABLE II

	Dielectric Strength kV	Power Factor, Per Cent
Cut No. 3	50+	Not determined
Cut No. 4	50+	Not determined

Still another sample of the residue defined above was fractionated into two cuts: a first boiling between 286° and 303°C. (Cut No. 5) and a second boiling between 303° and 400°C. (Cut No. 6). These cuts were also treated as above with the following results:

TABLE III

	Dielectric Strength kV.	Power Factor, Per Cent
Cut No. 5	50+	0.14
Cut No. 6	50+	0.01

To show that combinations of the above cuts will give similarly good results, a composition was prepared using equal amounts of some of Cuts Nos. 5 and 6. When this composition was tested, the following results were obtained:

TABLE IV

	Dielectric Strength kV.	Power Factor, Per Cent
Cuts Nos. 5 and 6	50+	0.02

The above values clearly show that the compositions defined and claimed herein are useful as liquid dielectric compositions, particularly for use in capacitors.

It is understood that the present compositions can be further treated, if desired, for example, to further improve their properties for a particular purpose, for example, to improve their flash point, interfacial tension, pour point, viscosity, oxidation stability, corrosion resistance, etc.

Obviously, many modifications and variations of the invention, as hereinabove set forth, can be made without departing from the spirit and scope thereof, and therefore only such limitations should be imposed as are indicated in the appended claims.

Claims.

1. A liquid dielectric composition obtained as a result of a process which comprises reacting benzene with ethylene in the presence of an alkylation catalyst to obtain an alkylation product containing largely unreacted benzene, ethylbenzene, polyethylbenzenes, 1,1-diphenylethane and heavier products, separating benzene, ethylbenzene, polyethylbenzenes and 1,1-diphenylethane from said alkylation product and thereafter recovering from said heavier products a fraction having a boiling point in the temperature range of about 275° to about 420°C. as said liquid dielectric composition.

2. The composition of claim 1 wherein said fraction has a boiling point in the range of about 280° to about 400°C.

3. The composition of claim 1 wherein said catalyst is $AlCl_3$.

4. The composition of claim 1 wherein said benzene and said ethylene are reacted in the presence of $AlCl_3$ in a temperature range of about 20° to about 175°C.

5. The composition of claim 1 wherein said benzene and said ethylene are reacted in the presence of $AlCl_3$ in a temperature range of about 90° to about 150°C.

European Patent Office

EUROPEAN SEARCH REPORT

Application number

EP 78 30 0086

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim
A	BE - A - 504 293 (K.BRINKMANN et al.) * Example * --	1
	US - A - 2 385 187 (F.H.BLANDING) * Page 4 * ----	1,3-5

CLASSIFICATION OF THE APPLICATION (Int. Cl.³)

H 01 B 3/22
C 07 C 3/56

TECHNICAL FIELDS SEARCHED (Int.Cl.³)

H 01 B 3/22
H 01 B 3/20
C 07 C 3/52
C 07 C 3/56
C 07 C 3/54

CATEGORY OF CITED DOCUMENTS

X: particularly relevant
A: technological background
O: non-written disclosure
P: intermediate document
T: theory or principle underlying the invention
E: conflicting application
D: document cited in the application
L: citation for other reasons

&: member of the same patent family, corresponding document

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
The Hague	18-10-1978	VAN GEYT

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