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D E C I S I O N
of 30 April 1996

Case Number: T 0784/94 - 3.5.2

Application Number: 87112958.1

Publication Number: 0259798

IPC: H01B 3/22

Language of the proceedings: EN

Title of invention:
Electrical insulating oil composition

Applicant:
NIPPON PETROCHEMICALS COMPANY, LIMITED

Opponent:

-

Headword:

-

Relevant legal provisions:
EPC Art. 56, 83, 84

Keyword:
"Sufficiency of disclosure (yes)"
"Clarity (yes)"
"Inventive step (yes)"

Decisions cited:

-

Catchword:

-



Case Number: T 0784/94 - 3.5.2

D E C I S I O N
of the Technical Board of Appeal 3.5.2
of 30 April 1996

Appellant:

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Representative:

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Decision under appeal:

Decision of the Examining Division of the European Patent Office posted 4 May 1994 refusing European patent application No. 87 112 958.1 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: W. J. L. Wheeler
Members: M. R. J. Villemin
M. Lewenton

Summary of Facts and Submissions

I. The Appellant filed an appeal against the decision of the Examining Division to refuse the European patent application No. 87 112 958.1. The reason given for the refusal was that the application contravened Articles 83 and 84 EPC. Moreover, the subject-matter of Claims 1 to 6 did not involve an inventive step within the meaning of Article 56 EPC, having regard to the following prior art:

D1: US-A-4 493 943 and

D2: DE-A-3 127 905.

In addition to the above-mentioned documents, the Board has taken into account the following document:

D5: Book "Physical Chemistry" by Walter J. Moore, Prentice-Hall, third edition, 1960, chapter 6, pages 118 to 159.

Chapter 6 of the second edition of this book is cited in the description of the present application.

II. With the grounds of appeal the Applicant filed new Claims 1 to 6 and a document (pages 1 to 17) explaining by way of examples the method of calculation of the solid mass using the liquid-solid equilibrium equation

$$X_1 = \exp \left[\frac{\Delta H_1^f}{R} \left(\frac{1}{T_1^f} - \frac{1}{T} \right) \right]$$

mentioned in Claim 1. With the letter dated 7 November 1994, the Appellant submitted a declaration by Professor Dr Ulrich K. Deiters concerning the applicability of the above equation for determining the proportion of solid mass.

III. The Appellant filed with the letter dated 26 March 1996 amended Claims 1 to 6 and amended pages 3, 3a, 18, 19, 19a and 20 of the description.

IV. Claim 1 now reads:

"1. An electrical insulating oil comprising a composition which consists of 45% by weight or more of at least 2 members selected from the group consisting of the following 6 components (a) to (f) and at least 10% by weight up to 55% by weight of non-condensed bicyclic aromatic hydrocarbons having not more than 17 carbon atoms other than the said 6 components:

- (a) m-ethylbiphenyl,
- (b) p-ethylbiphenyl,
- (c) o-benzyltoluene,
- (d) m-benzyltoluene,
- (e) p-benzyltoluene, and
- (f) 1,1-diphenylethane

wherein when the temperature of said composition is -40°C ,

(I) the total quantity of solid phase obtained by calculating the equilibrium mole fraction X_i in the liquid phase of each of the components according to the following solid-liquid equilibrium equation is 45% by weight or less relative to the total quantity of said composition, and

(II) in the liquid phase composition calculated by the following solid-liquid equilibrium equation, the total

quantity of said compounds (a) to (f) is 40% by weight or more and the viscosity of this liquid phase is 500 cSt or lower,

$$X_i = \exp \left[\frac{\Delta H_i^f}{R} \left(\frac{1}{T_i^f} - \frac{1}{T} \right) \right]$$

wherein X_i is the equilibrium mole fraction of a component i in the liquid phase of said composition, ΔH_i^f is the heat of fusion (cal.mol^{-1}) of said component i as a pure substance, T_i^f is the melting point (K) of said component i as a pure substance, T is the temperature (K) of the system, and R is the gas constant ($\text{cal.mol}^{-1}.\text{K}^{-1}$).

Claim 2 is dependent on Claim 1. Further dependent Claims 3 to 6 concern an oil-filled electrical capacitor impregnated with the insulating oil according to Claim 1 or 2.

V. The Appellant requested that the decision of the Examining Division be set aside and that a patent be granted with the following documents:

Claims: 1 to 6, filed with the letter dated 26 March 1996,

Description:

Pages 1, 2, 4 to 17 and 21 to 40 as originally filed, Pages 3, 3a, 18, 19, 19a and 20, filed 26 March 1996.

Drawings: Figures 1 to 5 as originally filed.

VI. The Appellant's arguments may be briefly summarised as follows:

The decision of the Examining Division seemed to be based on a misinterpretation of the problem underlying the claimed invention. The purpose of the invention was to provide an insulating oil composition having excellent low temperature characteristics and hydrogen gas adsorbing capacity as well as other excellent electrical characteristics, in particular a high breakdown voltage. This was achieved by providing a composition consisting of at least two members selected from the six components (a) to (f) indicated in Claim 1, wherein the amount of said selected components is 45% by weight or more, and at least 10% by weight up to 55% by weight of non-condensed bicyclic aromatic hydrocarbon having not more than 17 carbon atoms other than the above indicated six components. The equation in Claim 1 enabled the skilled person to adjust the relative ratios of the components in order to obtain an oil composition showing a solid phase proportion of not more than 45% by weight at a temperature of -40°C , and the invention taught that the total quantity of said components (a) to (f) in the liquid phase composition, again calculated by the solid-liquid equilibrium equation, had to be 40% by weight or more at -40°C and the value of the viscosity of the liquid phase 500 cSt or lower. The invention recognised that the calculation for mixtures of the components recited in Claim 1 may be carried out using the given equation which was established for an ideal system (see Chapter 6 of D5). The equation could be used to calculate the quantity of solid crystallized out for each component, and therefore the total, in a basically simple way described in the application. In practice and on the basis of this description, the skilled person, who was considered to be a team of experts including a mathematician, was able to calculate the solid content

at -40°C for different compositions. Therefore, it was considered that the disclosure of the present invention was sufficient to comply with Article 83 EPC and the subject-matter of Claim 1 was clear as required by Article 84 EPC.

With regard to inventive step, the Appellant presented arguments showing that the prior art according to D1 and D2 did not suggest or render obvious the subject-matter of Claims 1 to 6.

Reasons for the Decision

1. The appeal is admissible.
2. *Amendments*

Claim 1 essentially corresponds to original Claim 1, except that it is worded in the one-part form and contains some editorial amendments which have been carried out in order to improve clarity. Claims 2 to 6 correspond to original Claims 2 to 6, with the exception that Claim 2 has been adapted to the amended Claim 1. In addition to some obvious grammatical corrections, the description has been amended to adapt it to the new wording of Claim 1 and to include an acknowledgement of the prior art disclosed in D1. In the opinion of the Board, the wording of Claim 1 in the one-part form is appropriate (see paragraph 4.3 below) and the present form of the application does not infringe Article 123(2) EPC.

3. *Articles 83 and 84 EPC*

It appears that the major criticism put forward by the Examining Division in the decision of rejection is that the equation mentioned in Claim 1 was not applicable for calculating the solid content of the claimed insulating oil and violated principles of physical chemistry. The Examining Division alleged in particular that:

- (a) according to the rules of physical chemistry as developed in D5, the afore-mentioned equation was only applicable for two component liquid-solid systems exhibiting ideal behaviour and which do not form mixed crystals. Such a relation only existed for a solid/liquid equilibrium of one single component A in one (entirely liquid) solvent B. Since these properties depended on the chemical structure of the ingredients, it was impossible for these prerequisites to be met for a composition of 45% by weight of the components, which were not defined but could be freely selected. It was inconceivable that the non-ideal components, not only in admixture but also when employed as additives, were able to overcome basic principles of physical chemistry (see point 4.2.1. of the decision under appeal).

- (b) the application of the equation in most cases resulted in mole fractions the sum of which was > 1 , and it was not justified to expect a skilled person to apply the equation, contrary to the rules of physical chemistry, to a non-ideal multi-component system and to carry out the calculation of X_1 in such a way as if the other components of the mixture were not present (see point 4.2.2. of the decision under appeal).

The Examining Division concluded that the instructions given in the application as originally filed would never have enabled an expert in physical chemistry, let alone a person skilled in the art of insulating oils, to obtain the envisaged insulating oils.

- 3.1 For the following reasons, the objections raised by the Examining Division appear to stem from a wrong interpretation of the purpose of the solid-liquid equilibrium equation

$$X_1 = \exp \left[\frac{\Delta H_1^f}{R} \left(\frac{1}{T_1^f} - \frac{1}{T} \right) \right]$$

- 3.1.1 It is true that, strictly speaking, the above-mentioned equation applies only to ideal solutions (see D5, pages 128 to 130). However, it appears that the Applicant hypothesized that the study of phenomena involved in changes of states (i.e. depression of freezing point and solubility) in a solution which does not really constitute an ideal system, would be facilitated when it could be admitted that the composition shows properties close to the colligative properties which characterize ideal solutions. It appears from the description of the present application that it has been assumed that any insulating oil composition consisting of at least two members selected from the components (a) to (f) and non-condensed bicyclic aromatic hydrocarbons specified in Claim 1 can be regarded as an ideal solution (see particularly page 10, lines 4 to 8 of the published application). It further appears that the calculations have been performed under the assumption that the activity coefficients of the components were equal to unity. The validity of these assumptions is discussed below.

3.1.2 According to the Appellant's opinion, supported by the declaration by Professor Dr Ulrich K. Deiters, it was reasonable to admit that the various possible compositions specified in Claim 1 met the conditions required for ideal solutions justifying the use of the equation

$$X_i = \exp \left[\frac{\Delta H_i^f}{R} \left(\frac{1}{T_i^f} - \frac{1}{T} \right) \right]$$

For the following reasons, the Board shares the views of the Appellant:

- The six components (a) to (f) and the other aromatic hydrocarbons mentioned in Claim 1 have similar molecular sizes and chemical constitutions. They are formed by two non-condensed aromatic rings (biphenyl, benzyltoluene, other non-condensed bicyclic aromatic hydrocarbons having not more than 17 carbon atoms) and aliphatic substituents with no functional groups. This meets the conditions required for approaching ideality as indicated in D5 (page 130, lines 9 and 10). Thus, the omission of the activity coefficients from the solid-equilibrium equation as recited in the present application is justified by the reasonable assumption that the value of the activity coefficient for every component *i* is close to 1.

- All the components have different molecular shapes preventing the formation of mixed crystals of different components. Thus it is reasonable to assume immiscibility in the solid phase (see D5,

page 129, lines 3 to 19 and Figure 6.13 on page 147) and consider that the liquid phase exists substantially as a continuous phase.

- The enthalpy and entropy of melting of every component may be taken as constant over the temperature range of interest (between melting points and -40°C according to Claim 1).

Summarising, it may reasonably be admitted that all the various possible compositions specified in Claim 1 at least approximately meet the conditions required for ideal solutions.

- 3.1.3. The properties of a solution are said to be colligative when they depend only on the number of solute particles present, not on their identities. Although it is not strictly a colligative property, the solubility of a solute *i* in a solvent may be estimated by the use of the solid-liquid equilibrium equation:

$$X_i = \exp \left[\frac{\Delta H_i^f}{R} \left(\frac{1}{T_i^f} - \frac{1}{T} \right) \right]$$

when it can be assumed that the solute and solvent constitute an ideal system. To establish this equation (see D5, pages 128 to 130) it is inferred that, when the solute is left in contact with the solvent, it dissolves until the solution is saturated. Saturation implies equilibrium, and the above-mentioned equation rests on the key statement that, at equilibrium, the chemical potential of the component *i* as pure solid solute and the chemical potential of the same component *i* in solution are equal. It is crucial to observe that, under the reasonable assumption that the compositions

specified in Claim 1 may be regarded as ideal solutions, the calculation of the equilibrium mole fraction X_i of a component i at a temperature T with the solid-liquid equilibrium equation involves **only** thermodynamic parameters ΔH_i^f and T_i^f related to this component i and is **therefore fully independent of the nature of the solvent constituted by the other components**. This is clearly pointed out in D5, page 130, lines 8 to 10: "This is the mole fraction of naphthalene in **ideal solution, whatever the solvent may be**" (emphasis added).

3.1.4 Therefore, given that the conditions for colligative properties are met for any combination of the components in the manner as specified in Claim 1, if these components are intermixed, they form an ideal solution. Consequently, any one of them can be regarded as a solute in solution in a solvent constituted by the others, independently of their kinds. It follows that in the multi-component composition specified in Claim 1, it is reasonable to carry out the calculation of the X_i 's in such a way as if the other components of the composition were not present.

3.1.5 It is concluded that, contrary to the opinion of the Examining Division, it is admissible to use the equation

$$X_i = \exp \left[\frac{\Delta H_i^f}{R} \left(\frac{1}{T_i^f} - \frac{1}{T} \right) \right]$$

for calculating the mole fraction of any component i in the liquid phase of the compositions specified in Claim 1, whatever the proportions of the components might be. This remains true even if the application of the equation leads to calculated amounts which do not conform exactly to those actually occurring in practice.

3.2 For the following reasons, the objection raised by the Examining Division, that the application of the equation of solid-liquid equilibrium recited in Claim 1 in most cases results in mole fractions, the sum of which is > 1 , appears to originate from a misunderstanding of the very aim of this equation.

3.2.1 If a component i is a solute in a solvent, both forming an ideal solution, this equation permits calculating the value of the molar fraction of this solute-component i which is dissolved in the liquid phase in equilibrium with the pure solute-component i . In other words, X_i is nothing more than the expression of the solubility of i in a solvent, expressed in mole fraction, **whatever the solvent may be, provided the conditions for ideal solutions are met**. It is clear from the equation that the solubility X_i of i decreases exponentially as the temperature T is lowered from the melting point T_i^f . The (temperature variable) solubility X_i of a component i should not be confused with the (constant) mole fraction of that component in the composition of the insulating oil. There is no reason at all for $\sum X_i$ to be equal to 1, and, in general, it will not be. For any composition made of n members ($n \geq 2$) selected from the list (a) to (f) and m hydrocarbons specified in Claim 1, it is unavoidable that above a certain temperature $\sum X_i$ will exceed unity. It will be equal to $n+m$ when all $n+m$ components are completely dissolved (i.e. 100% mole fraction dissolved, for each component). Thus the fact that $\sum X_i$ may exceed 1 does not violate the basic principles of physical chemistry. The present application is, therefore, not unclear, or otherwise deficient, in this respect.

3.3 The manner in which the equation given in Claim 1 can be used to calculate the proportion of the solid phase is outlined in the description (see page 10, lines 4 to 22

and page 11, lines 1 to 6 of published application). As explained in the Appellant's submissions, the calculations involve only routine mathematics. They may be rendered less laborious by the use of a computer. If this is beyond an average physical chemist (and the Board is not saying it is), he can be expected to obtain the help of a mathematician. It is well established practice that the notional "person skilled in the art" may, if appropriate, comprise a team.

3.4 Summarising, the Board accepts that the disclosure in the present application is sufficient to enable a skilled person to make insulating oils having the composition and properties specified in Claim 1. This Claim 1 is clear and properly supported by the description. It is concluded that the present application meets the requirements of Articles 83 and 84 EPC.

4. *Novelty*

D1 discloses electrical insulating oil containing diarylalkanes in admixture with an aromatic mono- and/or diolefins (see e.g. table 1) having two condensed or non-condensed aromatic hydrocarbons in arbitrary ratios (see D1, column 10, lines 41 and 42). It is considered that D1 discloses the closest prior art.

4.2 Claim 1 of the present application concerns an electrical insulating oil which can be defined by the following features (A) to (F):

(A) it comprises a composition consisting of at least two members selected from the following six components:

- (a) m-ethylbiphenyl,
- (b) p-ethylbiphenyl,

- (c) o-benzyltoluene,
 - (d) m-benzyltoluene,
 - (e) p-benzyltoluene, and
 - (f) 1,1-diphenylethane;
- (B) the amount of components (a) to (f) not less than 45% by weight;
- (C) the composition further comprises at least 10% by weight up to 55% by weight of non-condensed bicyclic aromatic hydrocarbons having not more than 17 carbon atoms other than components (a) to (f);
- (D) the composition has the property that its solid mass proportion is not more than 45% by weight at -40°C;
- (E) the composition has the further property that, in its liquid phase, the total quantity of compounds (a) to (f) is not less than 40% by weight at -40°C;
- (F) the viscosity of this liquid phase is not less than 500 cSt at -40°C.

The additional information that the proportion of the total quantity of solid phase is obtained by calculating the equilibrium mole fraction X_i of each of the components according to the equation of solid-liquid equilibrium given in Claim 1, is hardly a further limiting feature. It is rather merely a brief indication of a method by which the presence of features (D) and (E) may be inferred.

- 4.3 D1 is silent as to the specific features listed in Claim 1, in particular the selection of at least two members from the compounds (a) to (f) mixed with non-condensed bicyclic aromatic hydrocarbons having not more

than 17 carbon atoms, and the proportions of solid phase at low temperature. Therefore, the subject-matter of Claim 1 is new over the prior art disclosed in document D1.

5. *Inventive step*

5.1 D2 is cited in the description of the present application (see published application page 9, lines 13 to 16). It discloses a dielectric mixture comprising methyl-substituted diphenyl-methanes (see example 1 and Claim 1 in D2).

Starting from D1, regarded as the document describing the closest prior art, the problem underlying the invention according to the present application is to provide an insulating oil comprising a composition of several components the proportions of which are controlled to confer on the oil excellent low temperature electrical characteristics, a low viscosity and improved hydrogen gas adsorbing capacity.

5.2 D1 concerns an electrical insulating oil to be used for impregnating electrical appliances such as oil-filled capacitors. This oil comprises

- at least one member of diarylalkanes and
- at least one member selected from the group of mono-and/or diolefins having two condensed or non-condensed aromatic nuclei, the examples of which include hydrocarbons.

Although the names of some components indicated in Claim 1 of the present application would fall within the list shown in columns 2 and 3 of D1, the teaching of this document gives the skilled person no reason to

select the particular compounds (a) to (f) and non-condensed bicyclic hydrocarbons having not more than 17 carbon atoms. Moreover, D1 is totally silent about the proportions according to features (B), (C), (D) and (E), and gives no indication of the value of the viscosity of the liquid phase at a temperature as low as -40°C . D1 reveals that capacitors have been tested at a temperature of $+30^{\circ}\text{C}$, but gives no indication of their electrical properties at low temperatures of the order of -40°C .

Aromatic olefins (alkenes) or diolefins are used in D1. The presence of reactive functional groups in the form of one or two double bonds renders these hydrocarbons more reactive than alkanes by permitting additions across the double bond. The resulting increased reactivity makes it unreasonable to assume that the components of the oils disclosed in D1 might be considered as constituting ideal solutions as discussed in section 3.1.2 above. Olefins or diolefins do not belong to the series of components mentioned in Claim 1 of the present application and the published description clearly points out that compounds having double bonds are not appropriate because they show polymerisation activity due to these double bonds (page 4, lines 22 to 24).

It is concluded that the teaching of D1 cannot help solve the problem underlying the present application in the manner defined by the features in Claim 1.

- 5.3 The problem to be solved according to document D2 is to achieve improved breakdown voltage properties of capacitors filled with dielectric oil compositions comprising methyl-substituted diphenyl methane according to the formula shown on page 4 of this document.

D2 does not suggest, even remotely, the specific features recited in Claim 1: in particular the selection of at least two members from the compounds (a) to (f) mixed with non-condensed bicyclic aromatic hydrocarbons having not more than 17 carbon atoms, or the idea of selecting the proportions of these components so as to keep the proportion of solid mass below a particular percentage at a particular low temperature. Moreover, since the compositions cited in D2 have not been tested at temperatures lower than -25°C (see table on page 12), it is not possible to conclude that the solid mass of the composition at -40°C might not exceed 45% by weight.

5.4 The inventors of the claimed oil composition have conducted detailed experiments which led to the conclusion that a composition comprising at least two bicyclic aromatic hydrocarbons having 14 carbon atoms, in particular components (a) to (f), in an amount not less than 45% by weight had excellent hydrogen gas absorbing capacity and voltage withstanding characteristics, but that it was advantageous to add from 10 to 55% by weight of further bicyclic aromatic hydrocarbons having not more than 17 atom carbons in order to reduce the viscosity of the composition. Documents D1 and D2 do not render it obvious that such a synergistic effect could be attained by mixing the components as specified in Claim 1.

5.5 It also appears that before the priority date of the present application, it was common belief that the equation mentioned in Claim 1 could not be applied in the study of a multi-component system, such as a composition of the kind specified in Claim 1, because it did not take into account the interaction between the different components. The present application recognises that, for particular choices of components, this

equation can be applied for calculating the equilibrium mole fractions of the components in the liquid phase, and hence the total quantity of solid phase.

- 5.6 For the reasons developed above, the Board concludes that the subject-matter of Claim 1 involves an inventive step within the meaning of Article 56 EPC. The subject-matter of Claims 2 to 6 also involves an inventive step.
6. In the opinion of the Board, a patent may be granted in the form requested by the Appellant.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to grant a patent according to the Appellant's request (see section V above).

The Registrar:


M. Riehl

The Chairman:


W. J. L. Wheeler

