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D E C I S I O N
of 8 March 1996

Case Number: T 0783/94 - 3.5.2

Application Number: 87112959.9

Publication Number: 0262455

IPC: H01B 3/22

Language of the proceedings: EN

Title of invention:
Electrical insulating oil composition

Applicant:
NIPPON PETROCHEMICALS COMPANY, LIMITED

Opponent:

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Headword:

-

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

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

Decisions cited:

-

Catchword:

-



Case Number: T 0783/94 - 3.5.2

D E C I S I O N
of the Technical Board of Appeal 3.5.2
of 8 March 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. 87 112 959.9 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. 87 112 959.9. 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: DE-A-2 718 905 and

D2: FR-A-2 395 576.

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. In reply to observations from the Board, the Appellant filed with the letter dated 21 February 1996 a new Claim 1 and amended pages 2 and 3 of the description.

IV. Claim 1 now reads:

"1. An electrical insulating oil, comprising a composition which consists of (a) m-ethylbiphenyl, (b) p-ethylbiphenyl, (c) m-isopropylbiphenyl and (d) p-isopropylbiphenyl, wherein the total amount of said ethylbiphenyls is not less than 40% by weight and the total quantity of solid phase in said composition at a temperature of -40°C is controlled to be not more than 45% by weight, the solid quantity of each component being obtained by calculating the equilibrium mole fraction X_i in the liquid phase of each component according to the following general equation of solid-liquid equilibrium:

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

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 ($\text{cal.mol}^{-1}.\text{K}^{-1}$)."

- 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:

Claim 1, filed with the letter dated 21 February 1996, Claims 2 to 6, filed with the grounds of appeal, received 13 September 1994;

Description:

Pages 1 and 2 and 5 to 23, as originally filed, Pages 3 and 4, filed 21 February 1996.

There are no drawings.

- 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 low viscosity at low temperature. This was achieved by providing a composition consisting of the four components (a) to (d) indicated in Claim 1, wherein the total amount of ethylbiphenyl is not less than 40% by weight. The equation in Claim 1 enabled the skilled person to adjust the relative ratios of the four 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 . The invention recognised that the calculation for mixtures of the four components (a) to (d) 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 is 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 claimed insulating oil composition according to 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 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 aforementioned equation was only applicable for two component liquid-solid systems exhibiting ideal behaviour so it was impossible to apply this equation for a composition comprising at least four defined components plus any undefined components in unspecified amounts (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_i 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

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

3.1.1 It is true that, strictly speaking, the above-mentioned equation applies only to ideal solutions (see D5, pages 128-130). 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 is clear from the description of the present application that it has been assumed that any insulating oil composition consisting of the four components (a) to (d) specified in Claim 1 can be regarded as an ideal solution (see page 5, lines 13 to 21 of the published application). The calculations are performed under the assumption that the activity coefficients of the components are equal to unity. It is stated that the results of the calculations coincide well with experimental results.

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 four components (a) to (d) have similar molecular sizes and chemical constitutions. They are formed by two non-condensed aromatic rings (biphenyl) 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) and justifying the adoption of a value 1 for the activity coefficients r_i of every component i (see description of the published application, page 5, lines 13 to 21).
- 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 3-19 and Figure 6.13 on page 147) and consider that the liquid phase exists substantially as a continuous phase (see description of the published application, page 6, lines 50-54).
- 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 intermixing of the components (a) to (d) recited 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 no 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. With four components, it is unavoidable that above a certain temperature $\sum X_i$ will exceed unity. It will be equal to 4 when all four 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 5, line 13 to page 6, line 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*

D2 corresponds to document US-A-4 266 264 cited in the description of the present application as filed. It discloses a capacitor with a dielectric spacer impregnated with a dielectric fluid comprising isopropylbiphenyl, of which at least 55% by weight is the meta isomer. D1 discloses a capacitor with a dielectric spacer impregnated with a dielectric fluid containing monoalkyl biphenyls in admixture with dialkyl biphenyls. The meta isomers are not specifically mentioned.

4.2 Claim 1 of the present application concerns an electrical insulating oil having particular properties. The claimed insulating oil can be defined by the following features:

- (A) it comprises a composition consisting of the four components:
 - (a) m-ethylbiphenyl,
 - (b) p-ethylbiphenyl,
 - (c) m-isopropylbiphenyl and
 - (d) p-isopropylbiphenyl;
- (B) the total amount of the ethylbiphenyls is not less than 40% by weight; and
- (C) the composition has the property that its solid mass proportion is not more than 45% by weight 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 feature (C) may be inferred.

4.3 The insulating oils known from D1 and D2 are not described as having any of the features (A) to (C). Therefore, the subject-matter of Claim 1 is new over the cited prior art.

5. *Inventive step*

5.1 Starting from the prior art according to D2, regarded as 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 and hydrogen gas adsorbing capacity.

5.2 D2 teaches that additional components may be mixed with isopropylbiphenyl: 3 to 6% by weight of di-isopropylbiphenyl, up to 1% of an antioxidant, up to 2% of hydrogen acceptor (e.g. anthraquinone) and up to 2% of an epoxy resin. D2 also discloses that a dielectric fluid containing m-isopropylbiphenyl would prevent the formation of a crystalline solid and so render capacitors suitable for operating temperatures below -40°C. However, although m-isopropyl biphenyl is mentioned in D2, there is no mention of any of the other components (a) (b) and (d) specified in the present application.

5.3 D1 discloses dielectric oils comprising mixtures of monoalkyl biphenyls and dialkyl biphenyls. The use of isopropylbiphenyls in combination with other compounds is described in D1, but there is no suggestion in D1

that the oil could consist only of monoalkyl derivatives (a) to (d) as specified in Claim 1 of the present application. Nor is there any hint at a proportion of at least 40% by weight of ethylbiphenyl. Moreover, the teaching of D1 gives the skilled person no incentive to choose particular isomers. D1 contains no instructions for selecting the proportions of the components so as to keep the proportion of solid phase below a particular percentage at a particular low temperature.

- 5.4 For these reasons, 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, which are properly dependent on Claim 1, also involves an inventive step.
6. In the opinion of the Board, a patent may be granted on the basis of the Appellant's request.

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:

The Chairman:

M. Kiehl

W. J. L. Wheeler

