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

Case Number: T 0785/94 - 3.5.2

Application Number: 87112957.3

Publication Number: 0262454

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:

"Sufficient disclosure of the invention in the description
(yes)"

"Clarity of the independent claim (yes)"

"Inventive step (yes)"

Decisions cited:

-

Catchword:

-



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Boards of Appeal

Chambres de recours

Case Number: T 0785/94 - 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: NIPPON PETROCHEMICALS COMPANY, LIMITED
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Decision under appeal: Decision of the Examining Division of the European
Patent Office posted 18 April 1994 refusing
European patent application No. 87 112 957.3
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. 87 112 957.3. 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-4 266 264,

D3: US-A-4 493 943,

D4: DE-A-2 718 905.

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 submitted with the letter dated 29 August 1994 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

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

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 16 February 1996 an amended first page of Claim 1, containing the beginning of the claim up to the second occurrence of the expression "pure substance", and amended pages 11, 22, 23, 24, 24a and 25 of the description.

IV. Claim 1 now reads:

"1. An electrical insulating oil, comprising a composition having good low temperature characteristics which composition consists of at least 4 members selected from the group consisting of the following 7 components:

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

wherein the proportion of the total quantity of solid phase at a temperature of -40°C of said composition is controlled to be not more than 45% by weight and the proportion of the total quantity of solid phase obtained by calculating the equilibrium mole fraction X_i of each of the components 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 of said 7 components 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 ($\text{cal.mol}^{-1}.\text{K}^{-1}$)."

Claims 2 to 6 are dependent on Claim 1.

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

Claims: Claim 1, first page (which goes up to the second occurrence of the expression "pure substance"), filed with the letter dated 16 February 1996; and Claim 1, second part and Claims 2 to 6, as filed with the grounds of appeal on 29 August 1994.

Description: Pages: 1 to 10, 12 to 21 and 26 to 46 as originally filed, and
Pages: 11, 22, 23, 24, 24a and 25 as filed with the letter dated 16 February 1996.

Drawings: Figures: 1, 2, 3-A, 3-B, 3-C and 4 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. This was achieved by providing a composition consisting of at least four components taken from the list of the seven components (a) to (g) indicated in Claim 1, in accordance with a selection rule. The equation in Claim 1 enabled the skilled person to adjust the relative ratios of the at least 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 at least four of the seven components (a) to (g) 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, D3 and D4 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 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).

Amendments in the description consist of editorial corrections and adaptation to the claims. 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 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 is clear from the description of the present application that it has been assumed that any insulating oil composition consisting of at least four of the seven components defined in claim 1 could be regarded as an ideal solution (see in particular page 6, line 43 to page 7, line 5 and page 10, lines 12 to 17 of the published application).

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 seven components (a) to (g) have similar molecular sizes and chemical constitutions. They are formed by two non-condensed aromatic rings (biphenyl or benzyltoluene) 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 7, lines 3 to 5 and page 10, lines 16 and 17).
- 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 description of the published application, page 6, lines 43 to 45 and page 10, lines 12 to 15; see also D5, page 129, lines 3 to 19 and Figure 6.13 on page 147).
- 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** the 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 (g) recited in Claim 1, if several of 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 number of the components (from four to seven) present. 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 (the minimum required by Claim 1), 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 10 of published application, lines 7 to 34). 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*

4.1 D3 discloses insulating oils containing diarylalkanes in admixture with an aromatic olefine such as 1,1 - diphenylethylene (see Table 1, Examples 11 to 13 and 18) and which are reported to exhibit superior electrical insulating properties (see column 10, lines 31 to 40) even at temperatures lower than -50°C (see Table 2, Examples 11 and 13). It is considered that D3 represents the closest prior art document.

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 a choice made from the following components:

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

(B) at least four of these components are selected; and

(C) the composition has the property that its solid mass proportion is 45 % by weight or less 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 D3 are not described as having any of the features (A) to (C). Therefore, the subject-matter of Claim 1 is new over the prior art disclosed in this document.

5. *Inventive step*

5.1 Starting from the prior art according to D3, 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 D3 refers only to mixtures of components which differ from those listed in Claim 1 of the application, apart from component (g). According to D3, page 17, capacitors filled with this mixture have been tested at 30°C, but there is no disclosure of their behaviour at low temperatures of the order of -40°C. There is no suggestion in D3 that by choosing a determined number of certain components, their proportions could be determined to keep the proportion of solid phase below a particular percentage at a particular low temperature.

- 5.3 D1 describes a capacitor with a dielectric spacer impregnated with a dielectric fluid comprising isopropyl biphenyl, of which at least 55% by weight is the m-isomer. Other components may be added, such as 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. D1 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, the components in this known dielectric fluid are entirely different from the components (a) to (g) specified in the present application.
- 5.4 D4 describes a capacitor with a dielectric spacer impregnated with a dielectric fluid comprising a mixture of at least two bicyclic aromatic compounds, including biphenyl compounds, having alkyl groups of 2 to 4 carbon atoms. The mixing of at least four of the components (a) to (g) specified in the present application is not disclosed. There is no suggestion in D4 that by choosing a determined number of certain components, their proportions could be determined to keep the proportion of solid phase below a particular percentage at a particular low temperature.
- 5.5 The Board observes that documents D1, D3 and D4, considered alone or in combination, neither disclose nor even remotely suggest any one of the aforementioned features (A), (B) and (C) of Claim 1. The selection of at least four components among (a) to (g) is not obvious in view of the teaching of these documents.

- 5.6 It 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.
- 5.7 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 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. Kiehl

The Chairman:


W. J. L. Wheeler

