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D E C I S I O N
of 15 May 1996

Case Number: T 0786/94 - 3.5.2

Application Number: 87112960.7

Publication Number: 0262456

IPC: H01B 3/22

Language of the proceedings: EN

Title of invention:
New 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:
-

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Case Number: T 0786/94 - 3.5.2

D E C I S I O N
of the Technical Board of Appeal 3.5.2
of 15 May 1996

Appellant: NIPPON PETROCHEMICALS COMPANY, LIMITED
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Chiyoda-ku
Tokyo (JP)

Representative: Strehl Schübel-Hopf Groening & Partner
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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. 87 112 960.7 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. 87 112 960.7. The reason given for the refusal was that the application contravened Articles 83 and 84 EPC and 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-3 127 905,
D2: EP-A-0 136 230 and
D3: 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 12 September 1994 the Applicant filed a set of amended 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

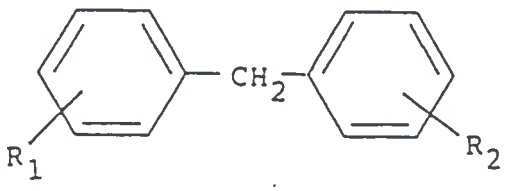
$$X_i = \exp\left[\frac{\Delta H_i^f}{R}\left(\frac{1}{T_i^f} - \frac{1}{T}\right)\right]$$

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. The Appellant filed with the letter dated 26 March 1996 a new set of amended Claims 1 to 6 and amended pages 3, 3a, and 11 of the description. He further filed with the letter dated 3 May 1996 an amended first part of Claim 1 and a new page 11 of the description adapted thereto.

IV. Claim 1 now reads:

"1. An electrical insulating oil comprising a composition having improved low temperature characteristics which composition consists of a mixture of 40% by weight to 90% by weight of benzyltoluene and as the remainder one or more members selected from alkyl substituted diphenylmethanes having 15 to 17 carbon atoms which are represented by the following general formula:



wherein R_1 and R_2 are hydrogen or C_1 to C_4 alkyl groups and the total number of carbon atoms in R_1 and R_2 is 2 to 4, provided that not both R_1 and R_2 are methyl groups; and the proportion of the total quantity of solid phase calculated according to the following general solid-liquid equilibrium equation is 45 % by weight or less relative to the total quantity of said composition at -40°C:

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

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

Claim 2 is dependent on Claim 1. 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: First part of Claim 1, filed with the letter dated 3 May 1996 and second part of Claim 1 as filed with the letter dated 26 March 1996,
Claims 2 to 6, filed with the letter dated 26 March 1996.

Description: Pages 1, 2, 4 to 10 and 12 to 39, as originally filed,
Pages 3, 3a, filed with the letter dated 26 March 1996,
Page 11, filed with the letter dated 3 May 1996.

Drawings: Figures 1 to 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 electrical properties, not to keep the solid content of the insulating oil as low as possible as alleged by the Examining Division. The invention provided a rule, specified in Claim 1, to enable the skilled person to choose the necessary components in order to obtain an insulating oil composition which fulfilled the requirements. Applicant had found that the calculated value of solid phase is an effective means for selecting the necessary components and their amounts from the groups defined in Claim 1. 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. 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). This 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 declared that the reasons for which the involvement of an inventive step was denied were not clearly stated in the contested decision, and presented arguments showing that the prior art according to D1, D2 and D3 did not disclose or render obvious the claimed subject-matter.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

Claim 1 essentially corresponds to Claim 1 filed with the grounds of appeal, with the first amendment that the word "new" has been cancelled and the expression "except that R_1 and R_2 are simultaneously methyl groups" has been replaced by "provided that not both R_1 and R_2 are methyl groups" and the second amendment that the expression "the total number of carbon atoms in R_1 and R_2 is 1 to 4" has been replaced by "the total number of carbon atoms in R_1 and R_2 is 2 to 4". The first amendment has been carried out in order to improve clarity. The second amendment stems from the obvious fact that, since the two aromatic rings and the CH_2 group of the substituted diphenylmethane according to the formula in Claim 1 provide for 13 carbon atoms, R_1 and R_2 have to supply at least 2 carbon atoms in order to comply with the requirement "15 to 17 carbon atoms" specified in Claim 1. This amendment is supported by the description of the application as originally filed. Claim 2 corresponds to Claim 2 filed with the grounds of appeal, with the exception that it has been adapted to the amended Claim 1. Claims 3 to 6 correspond to Claims 4 to 7 as originally filed, respectively. The description has

been amended to adapt it to the amended Claim 1 and include an acknowledgement of the prior art disclosed in D1. In the opinion of the Board, 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 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 which may contain unspecified further components in unspecified proportions. 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.

(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 multicomponent 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.

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. As a consequence, the originally filed application did not contain sufficient information to carry out the invention and thus contravened Article 83 EPC.

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. Since it is stated in the description of the application in suit that the calculations have been performed under the assumption that the activity coefficients of the components were equal to unity, it has to be concluded that it has been assumed that the insulating oil composition consisting of a mixture of benzyltoluene and one or more components selected from alkyl substituted diphenylmethanes as specified in Claim 1 could be regarded as an ideal solution (see page 7, lines 6 to 34 of the published application). 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:

- All the components embraced by Claim 1 have similar molecular sizes and chemical constitutions. They are formed by two non-condensed aromatic rings (benzyltoluene, diphenyl) and aliphatic substituents (alkyl groups) 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 r_i from the solid-equilibrium equation

indicated in Claim 1 and used for the calculations 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^f_i 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 multicomponent 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.

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^f_i. 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 ΣX_i to be equal to 1,

and, in general, it will not be. For any composition made of n members as specified in Claim 1, it is unavoidable that, above a certain temperature, Σx_i will exceed unity. It will be equal to n when all n components are completely dissolved (i.e. 100% mole fraction dissolved, for each component). Thus the fact that Σx_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 7, lines 34 to 51 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.

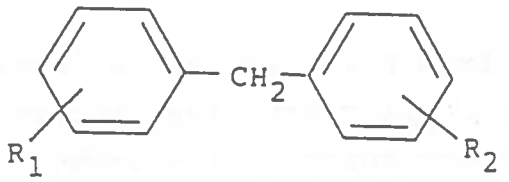
4. *Novelty*

D1 discloses dielectric oil compositions comprising methyl-substituted diphenylmethanes according to the formula shown on page 4 of this document. It is considered that D1 discloses the closest prior art.

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

(A) it is made of a mixture of benzyltoluene and alkyl-substituted diphenylmethanes,

(B) the diphenylmethanes are chosen to have 15 to 17 carbon atoms and are represented by the general formula



wherein R_1 and R_2 are hydrogen or C_1 to C_4 alkyl groups and the total number of carbon atoms in R_1 and R_2 is 2 to 4, provided that not both R_1 and R_2 are methyl groups,

(C) the mixture must comprise 40 to 90% by weight of benzyltoluene,

(D) the composition has the property that its solid phase 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 (D) may be inferred.

4.2 D1 does not suggest that benzyltoluene could be added to the methyl-substituted diphenyl methanes and is silent as to the proportion of the solid phase of the composition. 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 D1, 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, a low viscosity and improved hydrogen gas adsorbing capacity.

5.2 As already pointed out, D1 does not suggest that benzyltoluene could be added to methyl-substituted diphenyl methanes and is silent as to the proportion of the solid phase of the composition. Since the compositions cited in D1 have been tested at temperatures not lower than -25°C (see table on page 12) this document does not inform the skilled person that the solid mass of the composition at -40°C might not exceed 45% by weight.

It is concluded that the insulating oil known from D1 lacks the features (A) to (D) of the claimed insulating oil.

5.3 D2 corresponds to document US-A-4 523 044, cited in the description. It concerns the use of mixtures of polyarylalkane oligomers as components of electrical insulating oil. The description of D2 mentions some disadvantages of benzyl and dibenzyltoluenes are known (see page 1, line 34 to page 2, line 15). Moreover, although the oligomer mixtures known from D2 might comprise a proportion of methyl diphenylmethanes, it is apparent from this document (see for example Claims 1 to 3) that an important proportion of higher molecular components with more than 17 carbon atoms has to be present in the mixtures. This is contrary to the restrictive conditions required by feature (B) for the choice of the substituted diphenylmethanes used in the claimed insulating oil. Thus the teaching of D2 leads the skilled person away from features (A) and (B) of Claim 1, namely the use as an insulating oil of a mixture of benzyltoluene and alkyl-substituted diphenylmethanes, wherein the diphenylmethanes are chosen to have 15 to 17 carbon atoms.

In addition, D2 does not disclose the proportion of the solid phase at low temperatures. D2 reveals that the oligomer mixtures have been tested at temperatures not lower than -20°C or -30°C (see examples 1 and 2), so that it is not possible to conclude that the solid mass at -40°C might not exceed 45 % by weight. Therefore, feature (D) is not disclosed in D2.

5.4 D3 deals with a capacitor filled with a dielectric liquid comprising a mixture of at least two bicyclic aromatic compounds having alkyl groups of 2 to 4 carbon atoms. The use of benzyltoluene in admixture with alkyl

substituted diphenylmethanes is not suggested. The teaching of D3 does not render obvious features (A) and (C) of Claim 1 of the present application. Moreover, instructions for selecting the proportions of the components in such a manner that the solid phase at low temperature could be kept below a determined threshold are not hinted at. Hence, feature (D) of Claim 1 of the present application is not known from D3.

5.5 Thus, the Board concludes that documents D1, D2 and D3, considered alone or in combination, neither disclose nor suggest an oil composition according to the features (A) to (D) of Claim 1.

5.6 For assessing the merit of the claimed subject-matter it must be borne in mind that the skilled person would regard the melting points of benzyltoluenes and of alkyl diphenylmethanes as being generally too high and widely distributed in a broad range for using these compounds as an insulating oil having good low temperature properties. It is the merit of the invention to have recognised that a mixture of these compounds showing excellent properties at low temperature could be indeed obtained on the basis of a calculation using the equation defined in Claim 1, under the requirement that the solid phase mass does not exceed 45% by weight at -40°C. A surprisingly low amount of solid phase in a mixture comprising benzyltoluene and alkyl substituted diphenylmethanes at a temperature as low as -40°C could not be achieved or expected with the knowledge of the art according to D1, D2 and D3.

The Board observes 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 multicomponent 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 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. Kiehl

The Chairman

W. J. L. Wheeler