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D E C I S I O N
of 16 April 1997

Case Number: T 0989/93 - 3.3.1

Application Number: 85306447.5

Publication Number: 0176281

IPC: C09K 11/06

Language of the proceedings: EN

Title of invention:
New scintillation media and methods of detecting beta ray emissions

Patentee:
Fisher Scientific U.K. Limited

Opponent:
Packard Instrument B.V.

Headword:
Scintillation Media/FISHER SCIENTIFIC

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - yes"
"Common general knowledge (no); burden of proof"
"Analogous substitution (no)"
"Obvious to try (no)"

Decisions cited:
T 0219/83, T 0795/93

Catchword:
In the absence of relevant common general knowledge no conclusions are possible on the basis of the known properties of one group of chemical compounds (here: benzene derivatives) regarding the properties of a different group of chemical compounds (here: naphthalene derivatives; see point 9.1).

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0989/93 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 16 April 1997

Appellant: (Opponent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 21 September 1993 rejecting the opposition filed against European patent No. 0 176 281 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: R. K. Spangenberg
Members: P. Krasa
W. Moser

Summary of Facts and Submissions

I. This appeal lies from the Opposition Division's decision rejecting an opposition against the European patent No. 0 176 281, Claim 1 of which read:

"A scintillation medium comprising at least one diisopropylnaphthalene, wherein said at least one diisopropylnaphthalene is liquid at a temperature of 5 °C."

II. The ground of opposition was that the subject-matter of the patent was not inventive. The opposition was based, inter alia, on the documents

(1) US-A-3 444 089

(2) Carter, Christophorou, "Organic Scintillators with 2-Ethyl Naphthalene as a Solvent" J. Chem. Phys. 46(5) 1967, 1883 to 1890

(3) Koike, "Liquid scintillators based on normal alkanes and intermediate solvents" Nuclear Instruments and Methods 109, 1973, 269 to 274

(5) Buck, Swank, "Use of Isopropylbiphenyl as Solvent in Liquid Scintillators" Rev. Sci. Instr. 29, 1958, 252

(6) Birks, "The Theory and Practice of Scintillation Counting", Pergamon Press, 1964, 277 to 279

(8) Addison, "PCB replacements in dielectric fluids", Environ. Sci. Technol., 17(10) 1983, 486A to 494A.

(9) Kureha Chemical Industry Co., Ltd. "Solvent for carbonless copying paper KMC-113".

In the course of the opposition proceedings, the Respondent (patent proprietor) submitted further documents, among them

(F3) ter Wiel, Hegge, "Advances in Scintillation Cocktails" in Ross, Noakes, Spaulding (eds.), "Liquid Scintillation Counting and Organic Scintillators", Lewis Publishers Inc., Michigan, 1991, 51 to 67

III. The Opposition Division decided that the subject-matter of Claims 1 to 9 of the patent in suit involved an inventive step. They found in particular that document (1) disclosed the most relevant prior art and taught an improved organic scintillation composition comprising a particular solvent such as dimethylnaphthalene (DMN) or 2-ethylnaphthalene (2-EN).

In relation to this state of the art, the Opposition Division defined the technical problem underlying the patent in suit as to provide scintillation media with an improved performance in respect to flash point, vapour pressure, permeation through polyethylene or other plastic material, resistance to quenching, toxicity, odour, formation of coloured compounds in the presence of basic material, and compatibility with polymethylmethacrylate glasses.

The Opposition Division found that this technical problem was solved by the subject-matter of Claim 1 of the patent in suit and that this subject-matter was not rendered obvious by any of the citations.

IV. The Appellant (opponent) introduced further documents into the appeal proceedings, among them:

(10) Birks, "The Theory and Practice of Scintillation Counting", Pergamon Press, 1964, 625 to 628 and

(11) Horrocks, "Applications of Liquid Scintillation Counting", Academic Press, New York and London, 1974, 35 to 46.

He submitted in essence that

- the properties of diisopropylnaphthalene (DIPN), which were important for its use in a liquid scintillation counting (LSC) medium, were known from documents (9) or (8) which should be chosen as starting point for defining the technical problem to be solved;

- it had been obvious for a skilled person to suggest the application of DIPN as a LSC solvent in view of its known properties;

- DIPN was not commercially available when document (1) was published, in contrast to DMN and 2-EN, and that the filing of the patent in suit only about 5 or 6 years after the commercial introduction of DIPN in Europe was an indication of obviousness;

- if the technical problem were defined in relation to documents (1) and (2), these citations suggested to use excimer-forming solvents like DMN and 2-EN in LSC mixtures, so that it was obvious for the skilled person to try other alkylnaphthalenes and, thus, also the higher homologue DIPN as a component in LSC mixtures, so much the more as document (3) mentioned alkyl naphthalenes together with monoisopropylbiphenyl (IPBP) as intermediate solvents for LSC mixtures, the latter compound having been suggested as a LSC solvent also in documents (5) and (6);

- document (9), in combination with document (1), rendered the use of DIPN as a solvent in LSC obvious for a skilled person;

- document (11) disclosed for 2-EN a quantum yield which was superior to that of 1-methylnaphthalene (1-MN) rendering plausible to a skilled person that further improvements were to be expected for a propyl naphthalene solvent and, applying experiences gained with alkyl substituted benzene derivatives, in particular for DIPN;

- most of the Respondent's comparative tests were not apt to demonstrate a superior performance of DIPN as compared with 2-EN under realistic working conditions. If DIPN, according to the available data, were slightly better than 2-EN in certain respects, it was not better or even inferior in other respects, such minor (and practically irrelevant) improvements were probably due to differences in the impurities and not the solvent as such. Moreover, according to the Appellant, such improvements were to be expected by the skilled person in view of the state of the art.

V. At the oral proceedings, which took place on 16 April 1997 before the Board, the Respondent submitted amended Claims 1 to 8 and pages 2 to 5 of the description, with amendments on pages 2 and 3. Amended Claim 1 reads:

"A liquid scintillation medium comprising a fluor and at least one diisopropylnaphthalene, wherein said at least one diisopropylnaphthalene is liquid at a temperature of 5 °C."

The Respondent argued in essence that

- a skilled person wishing to improve scintillation media would not have considered documents (8) or (9) which were concerned with the replacement of PCB in a dielectric fluid and with carbonless copying paper, respectively;

- DIPN was not a true homologue of 2-EN;

- the preference given in documents (1) and (2) to 2-EN as compared with DMN would have dissuaded a person skilled in the art to use di-substituted naphthalenes in scintillation media;

- the comparative data, which were obtained according to standard methods accepted in the art, demonstrated various improved properties of DIPN under realistic working conditions, which improvements were not to be expected by a skilled person.

VI. The Appellant requested that the decision under appeal be set aside and the patent be revoked. As a further request, the Appellant requested to be given the opportunity to show that document (9) belongs to the state of the art, if necessary.

The Respondent requested that the decision under appeal be set aside and that the patent be maintained with claims 1 to 8 and the description, pages 2 to 5, submitted during oral proceedings.

VII. At the end of the oral proceedings the chairman announced the decision of the Board to allow the Respondent's request.

Reasons for the Decision

1. The appeal is admissible

Amendments

2. The Board is satisfied that the amended claims and the amended description comply with the requirements of Article 123. The Appellant, on inquiry, did not raise any objections in this respect. Therefore, it is not necessary to comment on this issue in detail.

Novelty

3. The Board is also satisfied that the subject-matter of the claims as amended is not disclosed in any of the citations and is, therefore, novel. This not being contested either, no detailed reasoning needs to be given.

Inventive step

4. According to the patent in suit (see page 2, lines 10 to 23), known scintillation solvents, such as toluene, the xylenes, cumene, ethylbenzene, etc., suffer from several disadvantages, e.g. such as

- a low flash point,
- a high vapour pressure,
- high rates of permeation through polyethylene or other plastic material,
- easy quenching,
- high toxicity, and/or
- formation of coloured compounds in the presence of basic material, and/or
- their expensiveness.

According to the patent in suit, the drawbacks of the solvents belonging to the state of the art were overcome by providing liquid scintillation media comprising at least one DIPN, which is liquid at a temperature of 5 °C (see page 2, line 3 and lines 24 to 27, in combination with page 3, lines 14 to 18 of the patent as granted). Further, it is stated that the claimed liquid scintillation media do not attack polymethylmethacrylate glasses (page 3, lines 16 to 17 of the patent as granted).

5. Liquid scintillation media which, like the claimed ones, contain alkyl naphthalenes, are known from a number of documents, in particular from citations (1), (2) and (3).

5.1 Document (1) aims at liquid scintillation media having improved radiation detection efficiency due to a reduction of the effects of quenching materials present in the scintillation medium (column 1, lines 58 to 65). It discloses that excimer-forming solvents, such as 1,6-DMN or 2-EN, enhance the energy transfer from solvent to solute and show the desired reduced quenching (page 2, lines 7 to 40). While 1,6-DMN and 2-EN are only disclosed as examples for such excimer-forming solvents (column 2, lines 24 to 33) no other ones are specified in document (1). 2-EN is preferred because of its superior detection efficiency (column 2, lines 33 to 40). Document (1) relates to the same technical field and the same - or a very similar - technical problem as does the patent in suit (document (1) column 1, lines 58 to 65). It discloses state of the art which is more relevant in respect of the subject-matter claimed in the patent in suit than the state of the art (i.e. the solvents) discussed in the latter.

5.2 Documents (2) and (3) also relate to the use of DMN and 2-EN in LSC solvents, but do not provide technical information coming closer to the claimed subject-matter.

6. Therefore, the Board takes document (1) as the starting point for evaluating inventive step (regarding the Appellant's approach starting from citation (9), see No. 12, below).

7. The Appellant has conceded that DIPN, while exhibiting about the same counting efficiency, colour quench resistance, colour formation in alkaline media, and photoluminescence decay as 2-EN, is superior ("somewhat better") to 2-EN in respect to

- chemical quench resistance against carbon tetrachloride as quenching agent,
- resistance to light induced yellowing,
- rates of permeation through polyethylene or other plastic material, and
- plastic glass compatibility

but alleged that these improvements were either exaggerated by the Respondent or of no practical relevance and, moreover, probably due to differences in the impurities of the compositions used in the respective experiments and not to the compounds used as solvents per se.

7.1 There is no evidence on file supporting the Appellant's submission that all beneficial effects of the claimed LSC medium are indeed caused by differences in the impurities and not by the respective solvents as such. At the oral proceedings, the Appellant, who had the

burden of proof in this respect (see T 0219/83, Reasons for the Decision Nr.12, OJ 1986, 211), confirmed on inquiry that no such evidence was available to him. Under these circumstances, the Board cannot take into account this allegation when determining the technical problem which has been solved by the claimed compositions.

7.2 Further, the Board cannot accept the Appellant's assertion that the superior chemical quench resistance against carbon tetrachloride of DIPN as compared with 2-EN is of no relevance and outweighed by the DIPN's poorer performance against ethanol and acetone as quenching agents. Carbon tetrachloride is accepted as a standard quenching agent for testing the quench characteristics of LSC solvents (see e.g. document (12), page 11, second sentence of the first complete paragraph). Document (F3), which does not belong to the state of the art but which can be used as an expert opinion, deals with "Advances in Scintillation Cocktails". The superiority of the high flashpoint solvent DIPN as compared with conventional LSC solvents is demonstrated, inter alia, by comparison of the respective quench characteristics with carbon tetrachloride (document (F3), page 55, table 3). The Board finds document (F3) to be sufficient evidence that, when evaluating LSC solvents, those skilled in the art of liquid scintillation relied, inter alia, on quenching tests with carbon tetrachloride. Therefore, in the Board's judgement, the quenching tests with carbon tetrachloride cannot be depreciated or even be disregarded for the sole reason that DIPN is inferior to 2-EN in other quenching tests with, e.g. ethanol or acetone.

7.3 The Appellant's more general argument that the above improvements were, in practice, compensated by disadvantages in other aspects, relates, in the Board's judgement, rather to the issue of technical progress which, however, is not a requirement of the EPC. Since the Appellant did not argue that these disadvantages would have prevented the intended use of the claimed compositions, this argument is also not to be considered when assessing the technical problem which has been solved.

7.4 It follows that the technical problem underlying the patent in suit can be defined as providing further liquid scintillation media with about the same counting efficiency as 2-EN and, inter alia, an improved chemical quench resistance. In view of the available data, the Appellant's concessions, and the above considerations, the Board is satisfied that this existing problem is not only technically meaningful but also solved by the subject-matter of Claim 1.

8. The Appellant further argued that a person skilled in the art would have known from common general knowledge that DIPN was an excimer-forming solvent and, thus, was useful as a LSC solvent, according to document (1) (see above no. 6). According to the Appellant, it would have been obvious for the skilled person to use DIPN in LSC cocktails in particular since its properties were known from documents (9) or (8).

This argument must fail for the following reasons:

8.1 The Respondent contested that there existed common general knowledge, according to which all alkylated naphthalenes are excimer-forming compounds. Since the Appellant did not submit supporting evidence for his allegation the Board cannot accept that such common general knowledge existed (see T 0219/83, Reasons for the Decision Nr.12, OJ EPO 1986, 211).

8.2 Regarding the known properties of DIPN, which, according to the Appellant, were useful for LSC, the following is to be noted:

8.2.1 Document (9) is a customer print by Kureha Chemical Industry Co., Ltd. dealing with DIPN (designated as KMC-113) and its use as a solvent for carbonless copying paper. No publication date can be found on the copy which had been submitted by the Appellant. On inquiry at the oral proceedings, the Appellant maintained that the brochure had been published "around 1980", i.e. at the time when DIPN became commercially available. Already in the notice of opposition, the Appellant had submitted that DIPN became commercially available "... early in the 1980s ..." (page 6, first line 4 of the Facts and Arguments). Prior to the oral proceedings, this was never objected to by the Respondent, who only then questioned whether, in view of the lacking publication date, document (9) was indeed relevant to the patent in suit.

8.2.2 The Appellant's submission that DIPN became available on large scale in the early 1980's is corroborated by document (8), published 1983. Table 1 on page 487A discloses that a product manufactured by "Kreha (sic!) Co. (Japan)" and designated KMC-A is DIPN and is used, inter alia, in pressure sensitive copying paper. Relying on a reference (20), it is further stated in table 2 on page 489A that DIPN was manufactured in quantities of several thousand metric tons per year.

The said reference (20) in turn was published in 1977 (document (8), page 494A, middle of the left hand column). Therefore, the Board finds that DIPN was available in commercial quantities in the early 1980's. Further, taking into account the similarities of the product designations KMC-A and KMC-113 and of the fields of application, as well as the chemical identity of the products concerned, the Board concludes that "Kreha" is an obvious misprint for Kureha.

8.2.3 It is common practice of a manufacturer to inform his customers on the properties and possible uses of a product, e.g. by publishing and distributing pamphlets and customer prints. Thus, the Board concludes that it is more likely than not that Kureha's product leaflet on KMC-113, i.e. on DIPN, was available to the public prior to 19 September 1984, the priority date of the patent in suit. Therefore, on the balance of probabilities, the Board holds that the subject-matter disclosed in document (9) is state of the art within the meaning of Article 54(2) EPC.

8.2.4 Document (9) discloses that DIPN has, inter alia, a low volatility, a low permeability through micro-capsule walls, a boiling point of 300 °C, a pour point of -42 °C, and further that it is odourless, colourless, and practically non toxic (chapter 1, part 2, in particular lines 8 and 9, and tables 1 and 3).

The Board accepts the Appellant's submission that all these are valuable properties of a LSC solvent. However, there is no indication at all in document (9) that DIPN could be used in LSC, let alone that the counting efficiency of 2-EN would be matched by that of DIPN. Document (8), concerning replacement fluids for polychlorinated biphenyls (PCB) and in addition mentioning "electrical uses" for DIPN, is also completely silent on LSC.

9. Further, the Appellant submitted that document (11) disclosed an increased quantum yield for 2-EN (0.26) as compared with 1-methylnaphthalene (0.21; see document (11) page 43, table III-6) and that it was known from table III-1 on page 38 of document (11) that increasing the number of methyl substituents in benzene derivatives resulted in a corresponding increase of their relative scintillation yield: toluene = 100, p-Xylene = 110, and 1,2,4-Trimethylbenzene = 112. The Appellant concluded therefrom that it had been obvious for a skilled person not only to increase the number of alkyl substituents in naphthalene derivatives but also to increase their chain length and, thus, to try DIPN as a liquid scintillation solvent taking into account not only the known valuable properties of this compound (see above no. 6.4), but also that it is a homologue of 2-EN.

These arguments are not convincing. Rather, they are the result of an interpretation of the content of document (1) with the benefit of knowing the disclosure of the patent in suit, and are thus not suitable for demonstrating the obviousness of the claimed subject-matter.

9.1 Thus, quite different conclusions than those drawn by the Appellant from document (11) result from the data contained in the table on page 626 of document (10). This table gives the following values of the scintillator light output in small samples (relative pulse height) for various solvents (amounts and nature of primary and secondary solute being identical):

toluene = 102
m-xylene = 101
p-xylene = 106
1,2,4-trimethylbenzene = 108
1,3,5-trimethylbenzene = 67

Whereas the figures of the series: toluene - p-xylene - 1,2,4-trimethylbenzene seemingly support the Appellant's conclusion that increasing (methyl) substitution improves the performance of a compound as a LSC solvent, a conclusion to the contrary could be drawn from the respective figures of the series: toluene - m-xylene - 1,3,5-trimethylbenzene. In any case, the above figures demonstrate that for benzene derivatives the position of the substituents is at least of the same importance as their number. Therefore, a skilled person relying on these experimental data could not have reasonably expected that an increase of the number of substituents would regularly promote the scintillation efficiency of benzene derivatives. But even if a general rule could have been established relating the degree of substitution of benzene derivatives to their respective scintillation efficiency - which is not the case as demonstrated above - the Board is not aware of any common general knowledge, and the Respondent contested the existence of such knowledge, on the basis of which a skilled person could have applied such a hypothetical rule for **benzene derivatives** to **naphthalene derivatives**. On inquiry, the Appellant could not give any reason why a skilled person should have applied information on benzene derivatives to naphthalene derivatives in this particular technical field.

9.2 Further, even if a skilled person had disregarded the different positions of the alkyl substituent in 2-EN and in 1-methylnaphthalene and additionally had contemplated an elongation of the ethyl group, this would have required him to select between a propyl naphthalene or an isopropyl naphthalene without any guidance available which alternative to select. The Board cannot accept the Appellant's argument that reference (3) contained an incentive for a person

skilled in the art to select the isopropyl naphthalene. It is true that this document disclosed the use of **mono**isopropyl**biphenyl** (IPBP) as an intermediate solvent for LSC. The dependency of a particular property (i.e. the relative pulse height) of a LSC cocktail on the concentration of IPBP is compared with such dependency on varying concentrations of 1-methylnaphthalene and of 1,6-DMN. However, document (3) is silent on the possible effects of introducing one, let alone two, isopropyl groups into the naphthalene system. Moreover, the figures 2, 3, and 4 on page 271 of document (3) show that the relative pulse height of IPBP is inferior to that of 1-methylnaphthalene and of 1,6-DMN at all concentrations. Therefore, the Board concludes that there was no incentive in document (3) for a skilled person to turn to DIPN as a LSC solvent with the reasonable expectation that this solvent would solve the existing technical problem. Documents (5) and (6) also disclose the use of IPBP as a LSC solvent, but add nothing relevant to the information which a skilled person could gain from document (3).

9.3 In respect to the Appellant's allegation that DIPN and 2-EN, are homologues, which was contested by the Respondent, the following is to be stated:

A homologue is a compound which differs from a given organic compound only by the number of carbon atoms of a carbon atom chain, or, in other words, by the number of $-CH_2-$groups, i.e. by the chain length. A DIPN, on the contrary, does not result from a simple elongation of the ethyl group of 2-EN. This compound contains only one CH_3-substructure in the side-chain whereas each side-chain of a DIPN comprises two CH_3-groups. Moreover,

any DIPN has necessarily a substitution pattern of the aromatic nucleus different from that of 2-EN. Therefore, in the Board's judgement, a DIPN is not a homologue of (or better, does not belong to the same homologous series as) 2-EN.

Therefore, the Board is not prepared to accept the Appellant's argument that the claimed solution of the existing technical problem does not involve an inventive step, since it makes use of a compound which is "homologuous" to 2-EN.

10. Document (2) is a scientific publication. Apart from corroborating the results disclosed in citation (1), it discloses 1,2-DMN as a further LSC solvent (document (2), e.g. page 1885, left hand column, lines 8 to 10). This citation, therefore, even when considered in combination with documents (3), (5), (6), (8), (9), (10), and (11) contains no incentive for the skilled person to use DIPN as a LSC solvent for substantially the same reasons which have been set out in points 8 and 9 above.

11. From these considerations it also follows that the subject-matter claimed in the patent in suit would not have been obvious in view of the discussed state of the art, even if it had to be considered as a mere alternative to 2-EN without showing any improvement as compared with the latter.

12. The Appellant also suggested that document (9) would have been the appropriate starting point to decide on inventive step. The Board cannot share this view, since document (9) relates to carbonless copying paper, a technical field completely different from LSC. According to the established jurisprudence of the Boards of Appeal a proper application of the so-called "problem-solution-approach" requires the assessment of

a document as the starting point which discloses state of the art as close as possible to the subject-matter of the respective patent (see T 0795/93, nos. 5.1.3 to 5.1.5; not published in the OJ EPO). Therefore, a document serving as the starting point for evaluating the inventive merits of an invention should relate to the same or a similar technical problem or, at least, to the same or a closely related technical field as the patent in suit, which criteria are not fulfilled by document (9).

A document belonging to a different technical field is much less likely to provide any sound reason why a person skilled in the art **would** (and not only could or might) have arrived at the subject-matter claimed.

Nevertheless, the Board finds it appropriate to answer the Appellant's submission.

In the present case, the only conceivable technical problem, which could be formulated in view of document (9), would be to find a further application for DIPN in view of its properties as disclosed therein (see no. 8.2.4, above). This was also suggested by the Appellant during oral proceedings. The Appellant further stated that all the properties of DIPN disclosed in citation (9) were valuable properties for LSC solvents (see no. 8.2.4, above) and concluded that, therefore, a skilled person would have considered the use of DIPN as a LSC solvent.

The Board does not concur with this conclusion, since document (9) is completely silent about the scintillation efficiency of DIPN. In the absence of information about this essential property, however, a skilled person would not, in the Board's judgement, have reasonably expected that DIPN was useful as a LSC

solvent. No other conclusion could be reached when taking into account the additional information available from documents (1) to (3), (5), (6), (8), (10) and (11) (see points 8 to 10 above), since these documents do not provide a clear and unambiguous basis for assuming that DIPN would be sufficiently effective as a LSC solvent, as explained in point 9, above.

13. The Board therefore holds that the subject-matter of Claim 1 involves an inventive step in the sense of Article 56 EPC.

Dependent Claims 2 to 7 relate to particular embodiments of Claim 1 and derive their patentability from that of Claim 1, as does Claim 8, relating to a method of detecting β-ray emissions using a scintillation medium comprising DIPN as specified.

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 maintain the patent with the Claims 1 to 8 and the description pages 2 to 5, submitted during oral proceedings.

The Registrar

E. Görgmaier

The Chairman

R. Spangenberg