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D E C I S I O N
of 4 December 2002

Case Number: T 1060/99 - 3.3.1

Application Number: 93810519.4

Publication Number: 0581737

IPC: C07C 7/20

Language of the proceedings: EN

Title of invention:

Inhibiting polymerization of vinyl aromatic monomers

Patentee:

ONDEO NALCO ENERGY SERVICES, L.P.

Opponent:

UNIROYAL CHEMICAL CO INC

Headword:

Polymerisation Inhibitors/ONDEO NALCO ENERGY SERVICES

Relevant legal provisions:

EPC Art. 56

Keyword:

"Main request, first to fourth auxiliary request: inventive step (no) - obvious combination of two types of known polymerisation inhibitors - no improvement in view of the closest state of the art"

Decisions cited:

T 0164/83, T 0254/86

Catchword:

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Case Number: T 1060/99 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 4 December 2002

Appellant: UNIROYAL CHEMICAL CO INC
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Representative: Harrison, David Christopher
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 14 October 1999
rejecting the opposition filed against European
patent No. 0 581 737 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: A. J. Nuss
Members: P. F. Ranguis
J. P. B. Seitz

Summary of Facts and Submissions

I. The appeal lies from the decision of the Opposition Division to reject the opposition filed against the European patent No. 0 581 737 (European patent application No. 93 810 519.4) pursuant to the provisions of Article 102(2) EPC.

II. The patent was granted with twenty three claims, independent Claim 1 reading:

"1. A composition which comprises
(a) a vinyl aromatic compound, and
(b) an effective inhibiting amount, sufficient to prevent premature polymerization during distillation or purification of said vinyl aromatic compound, of a mixture of
(i) 5 to 95% by weight, based on the total weight of components (i) and (ii), of a stable hindered nitroxyl compound, and
(ii) 95 to 5% by weight, based on the total weight of components (i) and (ii), of an aromatic nitro compound."

III. The opposition sought revocation of the patent in suit on the grounds that its subject matter did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Article 100(b) EPC) and did not involve an inventive step (Article 100(a) EPC). In support of lack of inventive step the following documents were cited:

- (1) JP-A-1 165534 (translation into English)
- (2) US-A-4 469 558
- (3) Lanzhou Daxue Xuebao, Ziran Kexueban, 1987, 23(3),

pp. 138-140 (translation into English)

(4) Experimental report from the Opponent

The Patentee (Respondent) submitted document

(5) WO-A-96/16921 (not prior art)

One week after the time limit set for submissions under Rule 71(a) EPC, the Opponent submitted the following documents:

(6) DE-A-21 49 670

(7) US-A-3 988 212

(8) JP-A-63316745 (translation into English)

IV. In its decision, the Opposition Division found that in view of the patent as a whole the person skilled in the art had enough information to carry out the invention. Regarding inventive step, the Opposition Division disregarded, first, the newly cited documents as late-filed and no more relevant than the documents cited with the statement of grounds of opposition, i.e. documents (1) to (4). On the substance, the Opposition Division, starting from document (2) as the closest state of the art, acknowledged the increase of activity of the claimed mixture over the isolated components and defined the technical problem to be solved as providing polymerisation inhibitors exhibiting improved activity. It, furthermore, held that it would not have been obvious for the person skilled in the art wanting to solve the above defined technical problem to arrive at the claimed solution in view of the prior art cited, in particular documents (1) and (2).

V. Oral proceedings took place on 4 December 2002. In

addition to its request that the appeal be dismissed, the Respondent (Proprietor of the patent) filed four sets of claims as first to fourth auxiliary request.

Claim 1 of the first auxiliary request read as follows:

"1. Use of a composition in a distillation or purification process of a vinyl aromatic compound, the composition comprising

- (a) a vinyl aromatic compound, and
- (b) an effective inhibiting amount, sufficient to prevent premature polymerization during the distillation or purification process of said vinyl aromatic compound, of a mixture of
 - (i) 5 to 95% by weight, based on the total weight of components (i) and (ii), of a stable hindered nitroxyl compound, and
 - (ii) 95 to 5% by weight, based on the total weight of components (i) and (ii), of an aromatic nitro compound."

Claim 1 of the second auxiliary request was in substance the same as Claim 11 of the patent as granted and read as follows:

"1. A process for inhibiting the premature polymerization of a vinyl aromatic compound during distillation or purification of said vinyl aromatic compound which comprises incorporating therein an effective inhibiting amount, sufficient to prevent premature polymerization during distillation or purification of said vinyl aromatic compound, of a mixture of

- (i) 5 to 95 % by weight, based on the total weight of components (i) and (ii), of a stable hindered

nitroxyl compound, and
(ii) 95 to 5% by weight, based on the total weight of components (i) and (ii), of an aromatic nitro compound."

Claim 1 of the third auxiliary request read as follows:

- "1. A composition which comprises
- (a) a vinyl aromatic compound, and
 - (b) an effective inhibiting amount, sufficient to prevent premature polymerization during distillation or purification of said vinyl aromatic compound, of a mixture of:
 - (i) 5 to 95 % by weight, based on the total weight of components (i) and (ii), of a stable hindered nitroxyl compound, selected from the group consisting of:
bis(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) sebacate,
1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl benzoate,
2,4,6-tris-[N-butyl-N-(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl)]-s-triazine,
4,4'-ethylenebis-(1-oxyl-2,2,6,6-tetramethylpiperazin-3-one),
1-oxyl-2,2,6,6-tetramethylpiperidin-4-ol,
1-oxyl-2,2,6,6-tetramethylpiperidin-4-one, and
1-oxyl-2,2,6,6-tetramethylpiperidine
and
 - (ii) 95 to 5% by weight, based on the total weight of components (i) and (ii), of an aromatic nitro compound, selected from:
2,6-dinitro-4-methylphenol,
1,3-dinitrobenzene,
2,4-dinitro-6-methylphenol,

2-nitro-4-methylphenol,
2,4-dinitrophenol, and
2,4,6-trinitrophenol (picric acid)."

Claim 1 of the fourth auxiliary request differed from Claim 1 of the third auxiliary request in that 2,6-dinitro-4-methylphenol was deleted as compound (ii).

VI. At the oral proceedings before the Board, the Appellant (Opponent) withdrew his ground of opposition based on Article 100(b) EPC. Regarding inventive step (Article 100(a) EPC), he relinquished reliance on documents (6) to (8) and other documents filed with the statement of grounds of appeal. He only relied on documents (1), (2) and (4) and argued as follows:

Document (1) should be considered the closest state of the art since it disclosed piperidine-1-oxyls within the definition of compounds (i) of the claimed invention as polymerisation inhibitors to be used when styrenes are recovered by distillation at high temperatures, namely the same objective as the patent in suit. Furthermore, document (1) disclosed that known polymerisation inhibitors such as aromatic nitro compounds within the definition of compounds (ii) of the claimed invention were insufficient in their polymerization inhibiting effectiveness and were to be replaced by piperidine-1-oxyls. By contrast, document (2), earlier state of the art than document (1), was not the closest prior art since it disclosed 2,6-dinitro-p-cresol as polymerisation inhibitor, the effectiveness of which was lower than the piperidine-1-oxyls as taught by document (1). Piperidine-1-oxyls of document (1) achieved an improvement vis-à-vis the nitrophenols and, for this reason, document (1) was to

be considered the starting point.

The only missing link between the opposed patent and the disclosure of document (1) was that document (1) did not disclose the mixture of both types of polymerization inhibitors, i.e. (i) and (ii). However, such a combination was obvious in the absence of any unexpected result. Indeed, contrary to the opinion of the Patentee, a synergistic effect could not be acknowledged within the whole claimed area. In particular, it was to be noted that the invention as disclosed in the patent in suit stated that the composition of polymerisation inhibitors could be used with or without the presence of oxygen. However, the experiments submitted as document (4) showed that in the presence of oxygen those mixtures exhibited only an additional effect, namely an expected result. It was to be concluded that the claimed subject-matter could only be viewed as an alternative to the disclosure of document (1). Under those conditions, it would have been obvious for the person skilled in the art to combine two well-known polymerization inhibitors and obtain the result to be expected. The same applied to the subject-matter of the four auxiliary requests, let alone the fact that the subject-matter of Claim 1 of the third and fourth auxiliary requests extended beyond the content of the application as filed.

VII. The Respondent's submissions in the written proceedings and during oral proceedings may be summarised as follows:

It was immaterial whether document (1) or document (2) was considered as the starting point, since one document disclosed the polymerisation inhibitor (i),

while the other disclosed the compound (ii). It was nevertheless the case that none of those two documents suggested combining (i) and (ii) in order to obtain a synergistic, and therefore, unexpected effect as set out in the patent in suit.

In that context, the experiments submitted by the Appellant (document (4)) were not a fair comparison since distillation of styrene or other vinyl aromatic compounds was carried out on an industrial scale in the absence or in the presence of a very small amount of oxygen. Those experiments were, therefore, to be rejected.

It followed that the Appellant's submissions amounted to unsubstantiated allegations and were not to be taken into account by the Board. Under those circumstances, the Appellant had submitted nothing relevant to substantiate the alleged non-synergistic effect and had, therefore, not discharged the onus upon him. In the absence of evidence to the contrary, the synergistic effect was to be acknowledged. Should any doubt remain in that respect, the auxiliary requests relating to the use of a composition in a distillation process of a vinyl aromatic compound (first auxiliary request) or to a process for inhibiting the premature polymerization of a vinyl aromatic compound during distillation or purification (second auxiliary request) made it clear that the claimed invention involved a very small amount of oxygen.

If the Board could not acknowledge a synergistic effect, it was nevertheless the case that neither document (1) nor document (2) suggested combining the polymerization inhibitors (i) and (ii).

VIII. The Appellant requested that the decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed or that the patent be maintained on the basis of one of the four auxiliary requests filed during the present oral proceedings.

IX. At the end of the oral proceedings, the decision of the Board was announced orally.

Reasons for the decision

1. The appeal is admissible.

Main request

2. *Article 56 EPC - Inventive step*

2.1 The patent in suit as reflected by Claim 1 as granted (cf. point II above) relates to a composition of two types of polymerization inhibitors, namely (i) and (ii), for reducing premature polymerization of readily polymerizable vinyl aromatic compounds during monomer manufacturing processes, in particular distillation or purification (cf. page 2, lines 3 to 24). The objective to be achieved in the most general form, as indicated in the patent in suit, consists in offering a composition enabling the distillation and purification of vinyl aromatic compounds in manufacturing plants to operate more safely at an increased production rate compared to prior art processes because of its greater effectiveness with or without oxygen. This permits higher distillation temperatures with minimal polymer

formation with accompanying economic and environmental advantages (cf. page 5, lines 17 to 20). Examples Nos. 1 to 5 relate to experiments involving bis(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) sebacate **(i)** and 2,6-dinitro-4-methylphenol **(ii)** as polymerization inhibitors of styrene, alone in the same amount, or in mixture with different proportions but still in the same amount as the compounds alone. Those experiments confirm that at the same total concentration, greater monomer stabilization efficacy is achieved by using the combination of both bis(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) sebacate and 2,6-dinitro-4-methylphenol as polymerization inhibitors than by using either component **(i)** or **(ii)**.

2.2 In accordance with the "problem-solution approach" consistently applied by the Boards of Appeal to assess inventive step on an objective basis, it is necessary to establish the closest state of the art being the starting point, to determine in the light thereof the technical problem which the invention addresses and solves, to verify whether the problem is solved by substantially all the embodiments encompassed within the scope of the claimed subject-matter and to examine the obviousness of the claimed solution to this problem in view of the state of the art. In this context, the Boards of Appeal have developed certain criteria in order to identify the closest state of the art being the starting point. One such criterion is that the "closest prior art" is normally a prior art document disclosing subject-matter aiming at the same objective as the claimed invention and having the most relevant technical features in common.

2.3 In that context, the Board observes that the Opposition

Division started from document (2) and that there has been a debate in the appeal proceedings between the parties to determine which of document, (1) or (2), was to be elected as the closest state of the art.

- 2.4 Document (1) relates to polymerization inhibitors composed of piperidine-1-oxyls, i.e. a type of compounds within the definition of compounds (i) of the claimed subject-matter, which are used when styrenes are recovered by distillation at high temperatures (cf. page 2, point 3). The quantity of piperidine-1-oxyls to be used should be 0.002wt% or more, generally between 0.005 to 0.5wt% with respect to the styrene (cf. page 3), which corresponds to the prescribed amount of polymerization inhibitors in the patent in suit (1 ppm to 2,000 ppm, page 4, line 57). Examples Nos. 1 to 4 describe the valuable effect for inhibiting the polymerization of styrene of four different piperidine-1-oxyls in amount of 200 ppm. In addition, this documents provides comparative examples with "publicly-known" polymerization inhibitors such as dinitrophenol and 2,6-dinitro-p-cresol which show that the piperidine-1-oxyls reveal a higher inhibiting activity than the dinitrophenol or 2,6-dinitro-p-cresol.

This document (1) aims at the same objective as the claimed invention and has one of the two relevant technical features in common, i.e. compound (i).

- 2.5. Document (2) relates to a process for the distillation of readily polymerizable vinyl aromatic compounds using as polymerization inhibitor 2,6-dinitro-p-cresol, namely one of the compound encompassed within the definition of compounds (ii) of the claimed invention.

In addition, the amount of inhibitor to be used is similar to that of the patent in suit, i.e. 50 to 3000 ppm (cf. col 3, lines 56 to 60).

This document (2) aims at the same objective as the claimed invention and has one of the two relevant technical features in common, i.e. compound (ii).

2.6 However, the Board concurs with the Appellant that the piperidine-1-oxyls disclosed in document (1) represent an improvement vis-à-vis 2,6-dinitro-p-cresol disclosed in document (2) as confirmed by the examples set out in document (1) (cf. point 2.4 above). Furthermore, document (1) is later than document (2) by five years. It follows, under those circumstances that the most promising springboard towards the invention which was available to the skilled person (cf. T 254/86, OJ EPO 1989, 115, point 15 of the reasons) is document (1) which thus qualifies as the closest state of the art.

2.7 In the next step, the technical problem which the invention addresses in the light of the closest state of the art is to be determined.

The Appellant argued that the technical problem to be solved could not be viewed in the provision of a synergistic mixture since the experiments provided in document (4) showed that in the presence of oxygen no synergistic effect could be obtained while the patent in suit encompassed the use of the inhibitor composition in the presence of oxygen (cf. point 2.1 above). The synergistic effect was, therefore, not shown for all the claimed embodiments.

However, if comparative tests are provided to

demonstrate that the technical problem as formulated in the patent in suit is not solved, a basic requirement to be met is that those tests reflect the teaching of the closest state of the art and do not go beyond that teaching. In that context, the disclosure of document (1) relates to distillation of styrene at high temperature. It is set out in that respect that the quantity of dissolved oxygen in the distillation tower during styrene manufacturing is frequently extremely small (cf. page 2, first paragraph of "Conventional Technology). Furthermore, all the examples disclosed in document (4) are made in the presence of pure nitrogen (cf. page 4, Example No. 1). The experiments provided with document (4) are, therefore, not a fair comparison with the closest state of the art and must be disregarded.

The Respondent argued that the examples provided in the patent in suit showed that the mixtures of compounds (i) and (ii) were unexpectedly good in view of the efficiency of the individual compounds (i) and (ii). This was, therefore, a reliable indicator of an inventive step and under those circumstances, the invention necessarily solved a technical problem. However, as noted by the Appellant, the merit of what is presented as an invention is not to be assessed per se but as opposed to the prior art. In that sense, a technical effect resulting from a combination of compounds, plays no role until it is acknowledged as non-obvious **having regard** to the state of the art. Therefore, it is only in comparison with the closest state of the art that the technical problem must be determined and, on this basis, the inventive step to be assessed.

In that context, document (1) discloses that no polymerization of styrene occurs when styrene is heated at 110°C during one hour in presence of 200 ppm of three piperidin-1-oxyls, respectively 1-oxyl-2,2,6,6-tetramethylpiperidine (example No.1), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one (example No. 2), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl benzoate (example No. 4). Those three compounds are clearly within the definition of (i) of Claim 1 and explicitly mentioned in Claim 7. Furthermore, the experimental conditions are also within the teaching of the patent in suit, i.e. 5 to 1,000 ppm of polymerization inhibitor (cf. page 4, lines 58-59), 50°C to 150°C (cf. page 5, line 1) and in the examples of the patent in suit, the time of the treatment is similar (45 minutes).

Reverting now to the experiments described in the patent in suit, the Board can recognise no improved effect in view of the disclosure of document (1). It is true that in the patent in suit the examples Nos. 1 to 5 demonstrate a synergistic effect between bis(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) sebacate and 2,6-dinitro-4-methylphenol. However, bis(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) sebacate, although being within the scope of the claimed subject-matter, does not represent a comparison vis-à-vis the nitroxyl compounds disclosed in document (1) which are also within the scope of the claimed subject-matter and explicitly mentioned in sub-claim 7. If the Patentee chooses to give evidence by comparative tests, these must be carried out in respect of the relevant closest state of the art (cf. T 164/83 OJ EPO 1987, 149, point 6 of the reasons). For these reasons, examples Nos. 1 to 5 are not a fair comparison and are to be disregarded. Moreover, example No. 11 relating to a

mixture of 1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl benzoate and 1,3-dinitrobenzene yields a polystyrene content of 2.2 % by weight, example No. 15 relating to a mixture of 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one and 2,4-dinitrophenol yields a polystyrene content of 0.5% by weight and example No. 16 relating to a mixture of 1-oxyl-2,2,6,6-tetramethylpiperidine and 2,4,6-trinitrophenol yields a polystyrene content of 0.3% by weight. All those results are less good than that of document (1) and do not show any improvement.

It must be concluded that neither the Appellant nor the Respondent have been able to discharge the burden of proof which were upon them, with the consequence that their unsubstantiated allegations are not to be taken into account by the Board.

- 2.8 In the Board's judgment, having regard to the fact that there is no evidence for an improvement for all the claimed subject matter, the technical problem underlying the patent in suit in the light of the closest state of the art can only be seen in the provision of an alternative composition of polymerization inhibitors for vinyl aromatic compounds.
- 2.9 In view of the technical information present in the patent in suit, in particular in the examples, the Board is satisfied that the problem as defined in point 2.8 has been solved.
- 2.10 The remaining question is, thus, whether the prior art as a whole would have suggested to a person skilled in the art solving the technical problem indicated above in the claimed way.

2.11 Since document (1), on the one hand discloses piperidin-1-oxyls as polymerization inhibitors for styrene (a vinyl aromatic compound) and since it also discloses the activity of publicly-known polymerization inhibitors such as dinitrophenol and 2,6-dinitro-p-cresol (2,6-dinitro-4-methylphenol) for styrene which was not contested by the Appellant, the presumption prevails, in the absence of evidence to the contrary, that it would have been obvious for the person skilled in the art, faced with the technical problem defined in point 2.8 above, to combine both type of compounds thereby arriving without inventive ingenuity at the composition of Claim 1, which is the solution proposed by the patent in suit. The Board observes, in that respect, that the patent in suit wherein piperidin-1-oxyls and 2,6-dinitro-p-cresol are acknowledged as known polymerization inhibitors for vinyl aromatic compounds (cf. page 2, lines 25 to 36 of the patent in suit) does not provide any indication which would have deterred the person skilled in the art from envisaging the claimed mixture to achieve the desired inhibiting effect. Nor was anything relevant submitted in the appeal proceedings.

2.12 Since the Board can only decide on a request as a whole, the patent in suit cannot be maintained in the form as granted and this request must be rejected for lack of inventive step.

First and second auxiliary request

3. *Article 56 EPC - Inventive step*

The inventive step issue raised by Claim 1 of the first and second auxiliary request is not changed compared

with that of Claim 1 of the main request. Indeed, document (1) also relates to the use of piperidine-1-oxyls (and dinitrophenol or 2,6-dinitro-p-cresol as comparative examples) as polymerization inhibitors in the recovery of styrenes by distillation. Therefore, the same considerations as mentioned in point 2 above apply and the subject-matter of Claim 1 of the first and second auxiliary requests does not involve an inventive step and those requests must also fail.

Third auxiliary request

4. *Article 56 EPC - Inventive step*

The inventive step issue raised by Claim 1 of the third auxiliary request is not changed compared with that of Claim 1 of the main request. Indeed, document (1) also relates to the use of 1-oxyl-2,2,6,6-tetramethylpiperidine (example No. 1), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one (example No. 2), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl benzoate (example No. 4) (and dinitrophenol or 2,6-dinitro-p-cresol as comparative examples) as polymerization inhibitors in the recovery of styrenes by distillation. The presumption prevails, in the absence of evidence to the contrary, that it would have been obvious for the person skilled in the art, faced with the technical problem defined in point 2.8 above, to combine both type of compounds thereby arriving without inventive ingenuity at the composition of Claim 1 which is the solution proposed by the patent in suit in the form of the third auxiliary request. The subject-matter of Claim 1 of the third auxiliary requests does not involve an inventive step and, therefore, this request must also fail.

Fourth auxiliary request

4. *Article 56 EPC - Inventive step*

The inventive step issue raised by Claim 1 of the fourth auxiliary request is not changed compared with that of Claim 1 of the main request. Indeed, document (1) also relates to the use of 1-oxyl-2,2,6,6-tetramethylpiperidine (example No. 1), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one (example No. 2), 1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl benzoate (example No. 4) (and dinitrophenol as comparative example) as polymerization inhibitors in the recovery of styrenes by distillation. The presumption prevails, in the absence of evidence to the contrary, that it would have been obvious for the person skilled in the art, faced with the technical problem defined in point 2.8 above, to combine both types of compounds, thereby arriving without inventive ingenuity at the composition of Claim 1 which is the solution proposed by the patent in suit in the form of the fourth auxiliary request. The subject-matter of Claim 1 of the fourth auxiliary request does not involve an inventive step and, therefore, this request must also fail.

Since none of the requests put forward comply with the requirements of the EPC, the patent is revoked.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:

The Chairman:

N. Maslin

A. Nuss