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**Datasheet for the decision
of 2 April 2008**

Case Number: T 0550/04 - 3.3.01

Application Number: 94304782.9

Publication Number: 0632106

IPC: C09B 48/00

Language of the proceedings: EN

Title of invention:

Process for the production of 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester, and process for the production of quinacridone from said ester as intermediate

Patentee:

TOYO INK MANUFACTURING CO., LTD.

Opponent:

Ciba Specialty Chemicals Holding Inc.

Headword:

Production of quinacridones/TOYO INK

Relevant legal provisions:

EPC Art. 56, 114(2)

EPC R. 103, 124(1)

RPBA Art. 13(3)

Relevant legal provisions (EPC 1973):

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Keyword:

"Admissibility of a late filed test report (no)"

"Inventive step (yes) - determination of the closest prior art - no-obvious solution of the technical problem"

"Request to record matter in the minutes of the oral proceedings (refused)"

"Reimbursement of the appeal fee (no)"

Decisions cited:

T 0928/98, T 0263/05

Catchword:

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Case Number: T 0550/04 - 3.3.01

D E C I S I O N
of the Technical Board of Appeal 3.3.01
of 2 April 2008

Appellant:
(Opponent)

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Representative:

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Respondent:
(Patent Proprietor)

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Decision under appeal:

Interlocutory decision of the Opposition
Division of the European Patent Office posted
8 March 2004 concerning maintenance of European
patent No. 0632106 in amended form.

Composition of the Board:

Chairman: P. Ranguis
Members: J. Jonk
C. Brandt

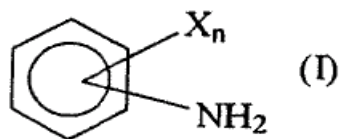
Summary of Facts and Submissions

I. The Appellant (Opponent) lodged an appeal against the interlocutory decision of the Opposition Division, which found that the European patent No. 0 632 106 (European patent application No. 94 304 782.9) in the form as amended during the opposition proceedings according to the set of claims 1 to 12 filed on 17 February 2004 met the requirements of the EPC.

II. Independent claim 1 of this set of claims read as follows:

"A process for the production of a quinacridone, which process comprises:

a) preparing a 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester by a condensation reaction between 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester) and an aromatic amino compound of the formula (I) in the presence, as a catalyst, of hydrochloric acid or sulfuric acid in an amount of 0.04 to 1.10 mol per mol of the 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester) and in the present, as a solvent, of a lower alcohol having 1 to 4 carbon atoms, in an oxygen-free atmosphere at a reaction temperature between 80°C and 130°C, wherein the condensation reaction takes place for from 3 to 5 hours,



wherein X is -F, -Cl, -Br, -I, -OH, -NO₂, -CF₃, an alkyl group having 1 to 4 carbon atoms, an alkoxy group having 1 to 4 carbon atoms, a substituted alkoxy group having 1 to 4 carbon atoms, a phenyl group, a cyclohexyl group, a phenoxy group, -COOH, a -COOC₁₋₄ alkyl group, -SO₃H, a phenylamino group, a benzamino group, -N(CH₃)₂, -SO₂NH₂, -SO₂N(CH₃)₂, a pyridino group, -CONH₂ or CON(CH₃)₂, and n is 0 or an integer of 1 to 4, provided that a hydrogen atom is present at at least one ortho-position relative to the -NH₂, the amount of the aromatic compound of formula (I) being from 2.0 to 4.0 per mol of the 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester);

b) heating the 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester thus obtained in an organic solvent to a temperature between 250°C and 350°C in an oxygen-free atmosphere, thereby proceeding with an intra-molecular-elimination reaction to convert the 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester to 6,13-dihydroquinacridone; and

c) oxidizing the 6,13-dihydroquinacridone."

Independent claim 7 of this set of claims related to another process for the production of quinacridone, but it comprised the same condensation step a) as defined in present claim 1.

III. The opposition was filed against the patent as a whole, and based on the ground of lack of inventive step as indicated in Article 100(a) EPC. It was supported by several documents including:

- (1) DE-A-3 605 976,
- (2) DE-A-2 222 177,
- (4) Hans Liebermann, Liebigs Annalen der Chemie 404 (1914), 272-277, 300-301,
- (6) US-A-3 317 539, and
- (9) GB-A-0 975 466.

IV. The Opposition Division held that the subject-matter of claim 1 of the patent in suit as granted lacked inventive step, but that the subject-matter of the claims filed during the oral proceedings on 17 February 2004 was found to meet the requirements of the EPC. Concerning the required inventive step the Opposition Division considered that the yield and purity of the reaction products, as well as the shortness of the reaction time achieved according to the claimed process were surprising in the light of document (1) as the closest prior art and that the claimed subject-matter was not obvious in view of document (2) or document (6).

V. Oral proceedings before the Board were held on 2 April 2008.

VI. The Appellant argued that the subject-matter of claim 1 of the set of claims filed on 17 February 2004 forming the basis for the decision of the Opposition Division lacked inventive step, since there was no unexpected effect over the cited prior art as had been shown by

the test results submitted on 15 December 2003 and 14 March 2008, and because the claimed process was obvious to the skilled person in the light of the cited documents, whereby each of the documents (1), (2), (6) or (9) could be considered as the closest prior art.

He also argued that the refusal of the opposition division to consider the relevance of the late filed document (9) constituted a substantial procedural violation rendering it equitable to reimburse the appeal fee.

VII. The Respondent (Patentee) argued that, compared to the closest prior art as represented by document (1) or document (2), the process of present claims 1 and 7 of the patent in suit provided the desired products of the condensation step a) in higher yields and purity as had been shown by the examples of the patent in suit and the test results submitted on 13 March 2000 and 27 March 2003. The cited prior art as a whole did not provide any incentive to the skilled person that these advantages could be achieved by the process as now claimed.

VIII. The Appellant (Opponent) requested:

- that the decision under appeal be set aside,
- that the patent be revoked,
- that in the minutes of the oral proceedings be recorded that the feature "oxygen-free atmosphere" in claims 1 and 7 as indicated under a) meant that the atmosphere was completely free from oxygen, and

- the reimbursement of the appeal fee.

The Respondent (Patentee) requested:

- that the appeal be dismissed or alternatively that the patent be maintained on the basis of one of the two sets of claims filed as First and Second Auxiliary Request with the response dated 14 March 2008, and
- that the test report submitted by facsimile dated 14 March 2008 be not admitted into the proceedings.

IX. At the conclusion of the oral proceedings the Board's decision was pronounced.

Reasons for the Decision

1. The appeal is admissible.
2. *Admissibility of the late filed test report (Article 114(2) EPC and Article 13(3) RPBA)*
 - 2.1 The present appeal procedure is governed by the Rules of Procedure of the Boards of Appeal of the EPO (RPBA) as amended on 25 October 2007 (see OJ EPO, 11/2007, 536). According to Article 24 of these Rules they entered into force upon entry into force of the revised text of the European Patent Convention, i.e. on 13 December 2007 in accordance with Article 8 of the Revision Act.

According to Article 13(3) of these RPBA amendments sought to be made after oral proceedings have been

arranged shall not be admitted if they raise issues which the Board or the other party or parties cannot reasonably be expected to deal with without adjournment of the oral proceedings.

- 2.2 In the present case the Appellant submitted a test report by facsimile on 14 March 2008, i.e. about 14 days before the oral proceeding before the Board, and as indicated in said facsimile he also sent a copy of it to the Respondent.

Furthermore, the Board found in assessing the admissibility of the test report:

(i) that the tests did not relate to the process of present claim 1, since they dealt with the preparation of a mixture of 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester by a condensation reaction between a 1,4-cyclohexadione-2,5-di(carboxylic acid alkyl ester) and a mixture of aromatic amino compounds corresponding to formula (I) of present claim 1 indicated above under point II, so that the question had to be answered whether the conclusions of the Appellant based on the test report would be valid for the present process of the patent in suit;

(ii) that the test results did not contain values for the yields rendering it impossible to make a comparison with the yields of the process of the patent in suit; and

(iii) that in view of the short time before the oral proceedings before the Board it would not be possible

for the Respondent to deal with the test results and the Appellant's submissions in this respect without an adjournment of the oral proceedings.

2.3 Under these circumstances and in line with the RPBA, the Board decided not to admit the test report into the proceedings (Article 114(2) EPC).

3. *Inventive step*

3.1 For deciding whether or not a claimed invention meets this criterion, the Boards of Appeal consistently apply the problem and solution approach, which involves essentially identifying the closest prior art, determining in the light thereof the technical problem which the claimed invention addresses and successfully solves, and examining whether or not the claimed solution to this problem is obvious for the skilled person in view of the state of the art. This approach ensures assessing inventive step on an objective basis.

In this context, the Boards of Appeal have developed certain criteria that should be adhered to in order to identify the closest state of the art to be treated as 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 objectives as the claimed invention and having the most relevant technical features in common.

3.2 Object of the patent in suit was the provision of a process for the production of a quinacridone and in particular a method for preparing an intermediate 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl

ester as indicated in step a) of present claim 1 in a high yield and a high purity and in a short period of time (see page 2, first paragraph, page 4, paragraph 0019, and examples 1 to 8).

According to said step a) a 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester) is subjected to a condensation reaction with an aromatic amino compound of the formula (I) using the following reaction conditions:

- the presence of hydrochloric acid or sulphuric acid as a catalyst;
- an amount of the catalyst of 0.04 to 1.10 mol per mol of the 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester);
- a lower alcohol having 1 to 4 carbon atoms as a solvent;
- an oxygen-free atmosphere;
- a reaction temperature between 80°C and 130°C; and
- a reaction time of from 3 to 5 hours.

3.3 Having regard to the objectives indicated in the patent in suit with respect to the process of step a) and in view of the relevant technical features of said process, a selection among the documents cited in the proceedings must be made as to which is to be considered as the closest prior art. The Appellant submitted that document (1), (2) or (9) represented the closest prior art, whereas the Respondent considered that document (1) or, preferably, document (2) was the closest state of the art.

3.4 Document (1), which was published about 6 years before the priority date of the patent in suit, relates to a

process for the production of an optionally substituted quinacridone compound involving two reaction steps, whereby in the first step an optionally substituted 2,5-di(arylamino)-3,6-dihydroterphthalic acid dialkyl ester is converted to a 6,13-dihydroquinacridone in the presence of a dimethyldiphenyl ether isomer mixture as a solvent and the resulting product is oxidised in a customary manner (see claims 1 and 8).

3.4.1 The optionally substituted 2,5-di(arylamino)-3,6-dihydroterphthalic acid dialkyl esters used as starting compounds in the process of document (1), which substantially correspond to the desired products of the reaction step a) of the process of present claim 1 of the patent in suit, can be prepared by a condensation reaction of aniline or derivatives thereof with dialkyl succinylsuccinate in the presence of a solvent and an acid catalyst at an elevated temperature and if appropriate under pressure (see page 4, lines 3 to 8).

Suitable acid catalysts are, for example, hydrochloric acid, sulphuric acid, acetic acid and p-toluenesulphonic acid (see page 4, line 7).

Suitable solvents are, for example, methanol, ethanol, toluene, xylene or the dimethyl diphenyl ether isomer mixture as applied in the subsequent conversion step (see page 4, lines 8 and 9).

Furthermore, the preparation of the 2,5-di(arylamino)-3,6-dihydroterphthalic acid dialkyl ester compounds as disclosed in this prior art document (1) is exemplified by examples 1, 1a, 1b and 3, whereby:

- according to example 1 a mixture of:

- 600 ml **methanol**,
- 34,3 ml (0.566 mol) **glacial acetic acid**,
- 95,9 ml (1,03 mol) **aniline** and
- 100 g of dimethyl succinylsuccinate

is heated to 100°C in a 1.2 l VA autoclave in the course of 30 minutes and is kept **at 100°C to 105°C** for **6 hours**. The desired 2,5-dianilino-3,6-dihydroterphthalic acid dimethyl ester is recovered in a yield of 96.5% with a purity of 96 to 98%;

- according to example 1a the aniline is replaced by **p-chloro- or m-chloroaniline** and following the procedure as described in example 1 2,5-di(p-chloroanilino)-3,6-dihydroterphthalic acid dimethyl ester and 2,5-di(m-chlorophenylamino)-3,6-dihydroterphthalic acid dimethyl ester are obtained in an amount of 96.7% and 95.5%, respectively;

according to example 1b the aniline is replaced by **p-methylaniline** and following the procedure as described in example 1 2,5-di(p-methylanilino)-3,6-dihydroterphthalic acid dimethyl ester is obtained in an amount of 97.3%; and

- according to example 3 a reaction mixture of:

- 50 g of dimethyl succinylsuccinate (DMSS),
- 250 ml of **a dimethyldiphenyl ether isomer mixture** as a solvent,
- 150 ml of **aniline** and

- 0.5 ml of 30% strength **hydrochloric acid** as a catalyst

is first stirred at 20°C-30°C for about 30 minutes and is then warmed to 105°C-110°C in the course of 1 hour with thorough stirring and under a **vacuum of 70 mmHg**, after which the mixture is then kept at **105°C-110°C** for **3 hours**. Subsequently a suspension consisting of 2,5-dianilino-3,6-dihydroterphthalic acid dimethyl ester and the dimethyldiphenyl ether isomer mixture is recovered, which is, as such, applied for the preparation of pure 6,13-dihydroquinacridone in a yield of 87,8%.

3.4.2 Furthermore, having regard to the technical information of the examples of document (1) as a whole the Board concludes that the process of example 1 represents the most preferred embodiment, since:

- the same process of example 1 is also used for preparing the 2,5-di(p-chloroanilino)-, 2,5-di(m-chloroanilino)- and 2,5-ditoluidino-3,6-dihydroterephthalic acid dialkyl esters (see examples 1a and 1b);

- example 1 is the only example making use of a reaction medium corresponding to that of the patent in suit and providing information of both yield and purity of the resulting product;

- the yield of 6,13-dihydroquinacridone obtained by combining examples 1 and 2, which includes the isolation, purification and use of the intermediate product of example 1 is higher than that of example 3, namely 96,5% (yield of example 1) x 94,1% (yield of

example 2) = 90,8% compared to 87,8% (yield of example 3); and

- all the other examples of document (1) relating to the preparation of further intermediate compounds and quinacridone end products include the use of the process of example 2 and, as indicated in this example, also the use of example 1.

3.4.3 Thus, in view of these considerations, document (1) discloses a process for preparing a group of the optionally substituted 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl esters and a corresponding group of quinacridones broadly overlapping those of the patent in suit, whereby it indicates most of the reaction conditions, which can be applied in step a) of the process of present claim 1 of the patent in suit.

3.5 Document (2), which was published about 20 years before the priority date of the patent in suit, relates to a particular compound, namely **2,9-dicarboxyquinacridone**, having improved pigment properties compared to the then prior art quinacridone compounds and methods for its preparation (see page 1 to page 2, formula III). Moreover, it discloses several methods for its preparation, whereby a preferred method involves the preparation of a 2,5-di(4-carboxyanilino)-3,6-dihydroterephthalic acid dialkyl ester as an intermediate compound (see pages 2 and 3), i.e. a compound obtainable according to step a) of the process of the patent in suit.

3.5.1 According to the sole example in document (2) with respect to the preparation of such an intermediate **2,5-**

di(4-carboxyanilino)-3,6-dihydroterephthalic acid dialkyl ester, namely example 2(A), a mixture having the following components:

- 2.56 g (0.01 mol) diethyl succinylsuccinate
- 100 ml absolute **ethanol**
- 4.1 g (0.03 mole) **p-amino benzoic acid** and
- 0.5 ml concentrated **hydrochloric acid**

was heated at **reflux temperature** under a **nitrogen atmosphere** for **3 hours**. The reaction product had a melting point of 347 to 350°C and was obtained in a yield of 91.1%.

3.5.2 Although the process of this example only differs from the reaction conditions indicated for process of step a) of the patent in suit by the use of a reflux temperature of about 78°C, i.e. a reaction temperature outside the claimed range of 80 to 130°C, the Board does not consider document (2) as the closest prior art in view of document (1), since the main object of document (1) was the provision of an improved process for the preparation of an overlapping group of compounds including the preparation of corresponding intermediate compounds, whereas the object of document (2) essentially concerns the provision of a single new product having improved properties, whereby the method of its preparation was apparently not of particular significance.

3.6 Document (6), which was published about 26 years before the priority date of the present patent, relates to a novel, beta polymorphic **2,9-dimethoxyquinacridone** having very useful pigment properties and a process for

preparing it (see column 1, lines 8 to 31, and column 2, lines 54 to 71). It discloses that one of many known methods of preparing said compound involves cyclisation of a 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester, followed by oxidation of the resulting dihydroquinacridone (see column 1, lines 60 to 68, and example 1).

3.6.1 According to the sole example 1(a) indicating the preparation of the intermediate **2,5-di(4-methoxyanilino)-3,6-dihydroterephthalic acid diethyl ester**, i.e. a product obtainable according to step a) of the process of the patent in suit, a mixture having the following components:

- 256 parts (1 mol) diethyl succinylsuccinate
- 4000 parts of denaturated **ethanol**
- 369 parts (3 mols) **p-methoxyaniline** and
- 35 parts of concentrated **hydrochloric acid**

was heated **under reflux**, i.e. at a temperature outside the claimed range of 80 to 130°C, and under an **inert nitrogen atmosphere** for **3 hours**. After isolation and purification the reaction product had a melting point of 190 to 191°C and was obtained in a yield of 98.0%.

3.6.2 Thus, as in case of document (2), the object to be achieved by document (6), therefore essentially relates to the provision a single new product having improved properties, so that document (6) cannot be considered as the closest prior art for the same reasons as indicated under point 3.5.2 above.

3.7 Document (9), which was published about 30 years before the priority date of the present patent, relates to a process for the production of **2,5-di(arylamino)-terephthalic acids**, which is characterised by a simultaneous oxidation and saponification method, in which 2,5-di(arylamino)-3,6-dihydroterephthalic acid esters as obtainable according to step a) of the process of the patent in suit are used as starting compounds (see page 1, lines 56 to 68). It also discloses that as a simplification of this process, it has been found that it is not necessary to use as starting materials 2,5-di(arylamino)-3,6-dihydroterephthalic acid esters in the form of the isolated and purified substances, since they may directly be employed in the simultaneous oxidation and saponification method in the form of reaction mixtures which are obtained by reacting 1 mol of a succinylosuccinic acid ester (i.e. a 1,4-cyclohexanedione-2,5-di(carboxylic acid ester)), in an alcoholic solution with 2 mols of an aryl amine (see page 2, lines 9 to 18).

3.7.1 According to example 4 in this document, being the sole example of said simplified process involving the *in situ* preparation of an intermediate 2,5-diarylamino-3,6-dihydroterephthalic acid diethyl ester, a reaction mixture of:

- 25 parts of succinylosuccinic acid diethyl ester,
- 26 part **p-chloroaniline**,
- 195 parts **n-butanol** in the presence and
- 0.6 parts **hydrochloric acid**

was heated at a temperature of **90-100°C** for **1 hour**, whereby the intermediate 2,5-di(p-chloroanilino)-3,6-dihydroterephthalic acid diethyl ester was formed. Subsequently, 145 parts 22% by weight sodium hydroxide solution and 3 parts anthrachinone-1-sulphonic acid were added to the reaction solution. Whilst introducing air, the solution was then heated for 4 to 5 hours at 80 to 90°C. After working up of the resulting mixture the desired 2,5-di(p-chloroanilino)-terephthalic acid was obtained in a yield of 86% of the theory.

- 3.7.2 Therefore, document (9) substantially relates to the preparation of different compounds, namely 2,5-di(arylamino)-3,6-terephthalic acids instead of 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl esters. Moreover, it does not disclose the preparation of a 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl ester in an isolated and purified form at all.
- 3.7.3 Under these circumstances, and having regard to the objectives of the patent in suit (see also point 3.2 above, first paragraph), document (9) cannot represent the closest prior art either.
- 3.8 In view of the considerations above with respect to the documents suggested as candidates for being treated as the closest prior art, the Board thus concludes that document (1) is the correct choice.
- 3.9 Having regard to the Respondent's submissions the technical problem underlying the patent in suit in the light of document (1) can be seen in the provision of a process for producing quinacridone compounds in which intermediate 2,5-di(arylamino)-3,6-dihydroterephthalic

acid dialkyl esters are prepared in improved yields and purity for a short period of time (see also paragraph [0019] of the patent in suit).

3.10 The patent in suit suggests as the solution of this problem, a process for producing quinacridone compounds in accordance with present claim 1, whereby in the light of the preferred embodiment of the process of document (1), i.e. the process of example 1 (see point 3.4.2 above), the preparation of the intermediate 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl esters as indicated in step a) is essentially characterised by the use of:

- hydrochloric acid or sulphuric acid instead of acetic acid as a catalyst,
- a catalyst to 1,4-cyclohexadinone-2,5-di(carboxylic acid alkyl ester) mol ratio of 0,04 to 1.10,
- an oxygen-free atmosphere, and
- a reaction time of from 3 to 5 hours.

3.11 With respect to the question whether or not the technical problem as defined above (point 3.9) has indeed been solved the Respondent referred to the examples in the patent in suit and his test reports submitted on 13 March 2000 and 27 March 2003.

3.11.1 According to **examples 1, 3 and 4 of the patent in suit** 2,5-dianilino-, 2,5-di(p-chloroanilino) and 2,5-di(p-toluidino)-3,6-dihydroterephthalic acid dimethyl ester were obtained in a yields and purities of 99.3% yield and 99.5% purity, 98.7% yield and 99.5% purity, and 98.9% yield 99.6% purity, respectively, whereas

according to **examples 1, 1a and 1b of document (1)**, i.e. under the reaction conditions indicated under point 3.4.1 above, in particular by using **glacial acetic acid as a catalyst** and probably without an oxygen-free reaction atmosphere, these compounds were obtained in a yields and purities of 96.5% yield and 96-98% purity, 96.7% yield (no purity value indicated) and 97.3% yield (no purity value indicated), respectively.

- 3.11.2 According to **Experimental Annex 2 of the test report submitted on 27 March 2003** the process of example 1 of the patent in suit for preparing 2,5-dianilino-3,6-dihydroterephthalic acid dimethyl ester had been repeated, except that glacial acetic acid was used instead of hydrochloric acid as a catalyst. The yield and the purity by using **glacial acetic acid** were diminished to 92.2% and 97.6%, respectively.
- 3.11.3 According to the **comparative examples 1, 3 and 4** of the patent in suit making use of a reaction temperature of **65°C instead of a reaction temperature of 100°C**, the yields and purities of the obtained 2,5-dianilino-, 2,5-di(p-chloroanilino)- and 2,5-di(p-toluidino)-3,6-dihydroterephthalic acid dimethyl esters were diminished to 95.8% yield and 94.3% purity, 94.4% yield and 95.1% purity and 94.1% yield and 94.6% purity, respectively.
- 3.11.4 According to the **test report submitted on 13 March 2000** the preparation of 2,5-dianilino-3,6-dihydroterephthalic acid dimethyl ester was carried out according to the patent in suit in ethanol at a **reaction temperature of 84°C** (example A) and for comparison purposes by the same process, but at a

- reaction temperature of 78°C** (reflux temperature) (example B). The yield and purity achieved in example A were 99.3% and 99.5%, respectively, whereas the yield and purity achieved in example B were 96.3% and 94.5%, respectively.
- 3.11.5 According to **Experimental Annex 1 of the test report submitted on 27 March 2003** 2,5-dianilino-3,6-dihydroterephthalic acid dimethyl ester was prepared according to example 1 of the patent in suit in ethanol at a **reaction temperature of 80°C** (example B) and also by applying the same process, but at a **reaction temperature of 78°C** (reflux temperature) (example A). The yield and purity achieved in example B were 98.0% and 99.2%, respectively, whereas the yield and purity achieved in example A were 96.1% and 96.7%, respectively.
- 3.11.6 According to **Experimental Annex 3 of the test report submitted on 27 March 2003** the process for preparing 2,5-dianilino-3,6-dihydroterephthalic acid dimethyl ester of example 1 of the patent in suit had been repeated, except that an inert atmosphere was not used. The yield and the purity by **not using an inert atmosphere** were diminished to 78.2% and 96.3%, respectively.
- 3.12 In view of this technical information consistently showing small improvements of about 1 to 2% of yields and purities compared to document (1), which improvements are significant in view of the fact that the process of the patent in suit is concerned with a large scale industrial process, the Board finds it

plausible that the technical problem as defined above has been solved.

Moreover, the board notes that it has been shown by the experimental evidence that the reaction temperature, the catalyst and the oxygen-free atmosphere as specified in step a) of present claim 1 are essential features of the claimed invention.

3.13 In this context, the Appellant disputed that the technical problem had been solved within the whole scope of present claim 1, since no improvement of yield and purity could be established by performing the claimed process at a reaction temperature of 80°C instead of 78°C as had been shown in a test report submitted on 15 December 2003 and because a number of examples in the patent in suit did not provide information about the yields and in some cases showed a lower purity of the respective products.

3.13.1 However, the experimental evidence provided by the Respondent with respect to the effect of the use of reaction temperatures below the lower limit of the claimed range of 80°C to 130°, as indicated under points 3.11.3 to 3.11.5 above, credibly showed a decrease of yields and purities at lower temperatures, whereas the lack of any effect as shown in the test report of the Appellant might, as submitted by the Respondent, result from an insufficient oxygen-free atmosphere. Furthermore, it is true that the examples 19 to 28 of the patent in suit relate to the preparation of further 2,5-dianilino-3,6-dihydroterephthalic acid dialkyl esters being substituted at the 2,5-dianilino groups without giving

information about the yields and in some cases showing relative low purity, e.g. 94.2% in case of 2,5-di(2,3,4,5-tetrachloroanilino)-3,6-dihydroterephthalic acid dimethyl ester and 94.8% in case of 2,5-di(p-dimethylaminocarbonylanilino)-3,6-dihydroterephthalic acid dimethyl ester. However, such results depend on the particular substituents and the Appellant did not provide any evidence showing that compared to the closest prior art process no improvement could be achieved. Therefore, the submissions of the Appellant, who carries under these circumstances the burden of proof for what he alleges or contests, cannot be accepted by the Board in the absence of convincing evidence.

- 3.14 The question now is whether the solution of the technical problem as defined above by the process of present claim 1 would have been obvious to the skilled person in view of the cited prior art.
- 3.15 As indicated under points 3.4.1 and 3.4.2 above, document (1) discloses a method for preparing optionally substituted 2,5-di(arylamino)-3,6-dihydroterephthalic acid dialkyl esters by performing a condensation reaction of aniline or derivatives thereof with dialkyl succinylsuccinate in the presence of a solvent and an acid catalyst at an elevated temperature and if appropriate under pressure, whereby according to the preferred process indicated in example 1 methanol was used as a solvent and a reaction temperature of 100 to 105°C was applied. However, having regard to the fact that the document does not provide any hint to the skilled person to the claimed solution of the present technical problem by combining these reaction

conditions with the use of hydrochloric acid or sulphuric acid as a catalyst, a oxygen-free reaction atmosphere and a reaction time of 3 to 5 hours, it cannot render the claimed subject-matter obvious by itself.

3.15.1 The Respondent disputed the existence of an inventive step by arguing that it was known from document (1) and in particular from example 3 that the reaction could be carried out by using hydrochloric acid as a catalyst, a reaction atmosphere having a reduced oxygen content (70 mm Hg vacuum) and a reaction time of 3 hours. However, apart from the fact that a different solvent outside the scope of claim 1 of the patent in suit had been used, the skilled person would not have any reason to amend the reaction conditions of example 1, since according to example 3 a significant lower yield of the desired 2,5-dianilino-3,6-dihydroterphthalic acid dimethyl ester was obtained (see also under points 3.4.1 and 3.4.2 above).

3.15.2 It is true, that documents (2) and (6) disclose the preparation of particularly substituted 2,5-dianilino-3,6-dihydroterphthalic acid dialkyl esters and that the exemplified preparation methods only differed from the process of step a) of claim 1 of the patent in suit in that a reaction temperature of 78°C instead of at least 80°C was applied (see also under points 5.3 to 3.6.1 above). However, starting from the disclosure of the closest prior art document (1) and in particular from the preferred embodiment represented by example 1, the skilled person would have had no reason to amend the reaction conditions of example 1 of document (1) by replacing acetic acid as a catalyst and by applying an

oxygen-free atmosphere in order to achieve improved yields and purities, since according to the technical teaching of document (1) acetic acid, hydrochloric acid and sulphuric acid are equally applicable and no particular significance had been given to the use of a reduced pressure of 70 mmHg as applied in example 3. In fact, documents (2) and (6) do not attach any significance to the use of a nitrogen atmosphere too. Therefore, these document do not provide a pointer to the solution of the technical problem underlying the patent in suit as defined above either.

3.16 The Appellant also argued that the use of an oxygen-free atmosphere as applied according to the patent in suit was obvious to the skilled person in view of document (9) and (4).

3.16.1 However, in view of the fact that document (9) was published about 30 years before the priority date of the present patent and because this document does not relate to the technical problem underlying the patent in suit (see also under points 3.7 to 3.7.2 above), the skilled person would not have any reason to take this document into consideration for its solution.

3.16.2 Furthermore, the same conclusion can be drawn with respect to document (4), which was published about 50 years before the priority date of the patent in suit and relates to 1,4-cyclohexanedione-2,5-di(carboxylic acid alkyl ester) and their reactions with ammonia and primary **aliphatic** amino compounds. It is true, that it has been noted in this document that **aromatic** amino compounds condensate with a 1,4-cyclohexanedione-2,5-di(carboxylic acid alkyl ester) at boiling temperature

in an open reactor in glacial acetic acid or a mixture of glacial acetic acid and a small amount of ethanol, but that by applying **a mixture of glacial acetic acid and much ethanol** in the presence of **sufficient air** often p-diarylamino terephthalic acid esters are formed (see page 276, line 8 from below, to page 277, line 10). Therefore, the skilled person would rather conclude from this teaching that the condensation reaction, while avoiding the forming of said by-product, should be carried out by using glacial acetic acid or a mixture of glacial acetic acid and a small amount of ethanol as a solvent system without the need of the presence of an oxygen-free reaction atmosphere, i.e. by applying a preparation method leading away from the process of the patent in suit.

3.16.3 In any case, documents (9) and (4), alone, in combination or in combination with the other cited documents do not provide any incentive to the skilled person to the claimed solution of the present technical problem, which involves a combination of all the reaction conditions specified in step a) of present claim 1.

3.17 The Board notes that in view of the teaching of the cited documents and for the reasons as submitted by the Appellant, the skilled person **could** have performed the preparation of the optionally substituted 2,5-di(arylamino)-3,6-dihydroterphthalic acid dialkyl esters under reaction conditions as specified in present claim 1 of the patent in suit. However, according to the consistent case law of the Boards of Appeal for determining lack of inventive step, it is necessary to show that considering the teaching of the

prior art as a whole, without using hindsight based on the knowledge of the claimed invention, the skilled person **would** have arrived at the claimed solution of **the technical problem to be solved**. However, as indicated above, the skilled person, when trying to solve the technical problem underlying the patent in suit, would not have expected that a process such as the one now claimed would solve the present technical problem with a reasonable chance of success.

3.18 In conclusion the Board finds that the subject-matter of the claims as maintained by the first instance involves an inventive step in the sense of Article 56 EPC.

4. *Auxiliary requests*

4.1 In the light of the above findings, it is not necessary to consider the Respondent's auxiliary requests.

5. *Request to record matter in the minutes*

5.1 During the oral proceedings before the Board the Appellant requested to record in the minutes of the proceedings that the expression "oxygen-free atmosphere" in present claim 1 had the meaning of "completely oxygen-free atmosphere".

5.2 However, according to the jurisprudence of the Boards of Appeal (see T 928/98 of 8 November 2000 and T 263/05 of 28 June 2007, point 8 to 8.11 (to be published in OJ EPO)) it is not the function of the minutes to record statements which a party considers will be of use to it in any subsequent proceedings in national courts, for

example in infringement proceedings as to the extent of protection conferred by the patent in suit. This is because such statements are not "relevant" to the decision which the Board has to take, within the meaning of Rule 124(1) EPC. Such matters are within the exclusive jurisdiction of the national courts.

5.3 In the present case, the reasons for applying an oxygen-free atmosphere and a method to achieve it are indicated in the patent in suit (see page 5, line 57 to page 6, line 12, and example 1). Moreover, the statement in question would not have an impact on the definition of the subject-matter of the patent for the questions the Board had to decide in these proceedings.

5.4 It follows that the desired statement is not a proper subject-matter of the minutes. Consequently, the request is refused.

6. *Reimbursement of the appeal fee*

6.1 According to Rule 103 EPC, reimbursement of the appeal fee shall be ordered where the Board of Appeal deems an appeal to be allowable and if such reimbursement is equitable by reason of a substantial procedural violation.

6.2 In the present case, the Appellant has not been successful on appeal to the extent requested. Thus, already for this reason the reimbursement of the appeal fee has to be refused.

Order

For these reasons it is decided that:

1. The appeal is dismissed.
2. The request for reimbursement of the appeal fee is refused.

The Registrar:

The Chairman:

M. Schalow

P. Ranguis