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D E C I S I O N
of 16 April 1996

Case Number: T 0967/92 - 3.3.1

Application Number: 86308812.6

Publication Number: 0225738

IPC: C07C 67/60

Language of the proceedings: EN

Title of invention:

Method of recovering useful components at least containing dimethyl terephthalate from high-boiling byproducts occurring in the production of dimethyl terephthalate

Patentee:

Teijin Petrochemical Industries Ltd.

Opponent:

HOECHST Aktiengesellschaft Zentrale Patentabteilung

Headword:

Dimethyl terephthalate/TEIJIN

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step (yes) - unobvious improvement"

Decisions cited:

-

Catchword:

-



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Case Number: T 0967/92 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 16 April 1996

Appellant:
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Respondent:
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 23 September 1992
rejecting the opposition filed against European
patent No. 0 225 738 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: A. J. Nuss
Members: R. K. Spangenberg
S. C. Perryman

Summary of Facts and Submissions

- I. The appeal lies against the decision of the Opposition Division of the EPO rejecting an opposition against European patent No. 0 225 738. This patent was granted in response to European patent application No. 86 308 812.6. The patent specification contained 11 claims. Claim 1 read as follows:

"1. A method of recovering useful components at least containing dimethyl terephthalate from high-boiling byproducts occurring in the production of dimethyl terephthalate, which comprises oxidizing p-xylene and/or methyl p-toluate with a molecular oxygen-containing gas in the presence of a heavy metal oxidation catalyst, subjecting the resulting oxidation product to esterification with methanol, separating dimethyl terephthalate and esters having lower boiling points than dimethyl terephthalate from the esterification reaction product by distillation, and thereafter treating the distillation residue containing byproducts having higher boiling points than dimethyl terephthalate with methanol to recover at least dimethyl terephthalate from the treated product, characterised in that the treatment of the distillation residue with methanol is carried out at a temperature of 110 to 240°C under a pressure sufficient to maintain methanol in the liquid phase."

Claims 2 to 11 depend on Claim 1 and relate to specific embodiments thereof. In the decision under appeal among others the following document was considered:

DD-A-29314 (D1)

II. The Opposition Division held that the subject-matter of the patent in suit was novel and involved an inventive step. In particular, it was held that D1 concerned only the preparation of dimethyl terephthalate by oxidation of p-xylene, followed by the esterification of the oxidation product with methanol in liquid phase and its separation by distillation, but did not disclose the treatment of the **distillation residue** obtained in this process. In the decision under appeal it was further held that the citations referred to in the introduction of the patent in suit, in particular US-A-4 092 481, corresponding to FR-A-2 273 794 (cited in the search report and referred to hereinafter as D6), represented the closest prior art, disclosing processes corresponding to the preamble of claim 1.

In the light of this prior art the technical problem underlying the opposed patent was found to be the **increase** of the recovery of the useful components such as dimethyl terephthalate (DMT), methyl p-toluate (MPT) and methyl p-hydroxymethylbenzoate (MHB) in the distillation residue. This problem was held to be actually solved if the methanol treatment was carried out with methanol in liquid instead of vapour phase as in the prior art, having regard to the comparison tests filed during the examination proceedings. Although the Opposition Division admitted that D1 might have suggested that the esterification of terephthalic acid in a distillation residue with methanol could be made in liquid as well as in vapour phase, this citation was not found to suggest this measure as a solution of the problem underlying the opposed patent.

- III. Oral proceedings took place on 16 April 1996.
- IV. During the oral proceedings as well as in the statement of grounds of appeal the Appellant (the Opponent) submitted that, in addition to the disclosure of D1, the common general knowledge represented by

D5 L.F.Fieser/M.Fieser, Lehrbuch der organischen Chemie, Verlag Chemie GmbH, Weinheim/Bergstr. (1957), page 139.

should be taken into account when assessing the inventive step of the process according to Claim 1. In his opinion, in respect of D1 and D5 the claimed process was no more than the result of a routine modification of the process described in D6 with a view to improving the yield of DMT, i.e. with a view to solving the technical problem defined by the Opposition Division.

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

- V. The Respondent (the patent proprietor) requested that the appeal be dismissed.

In support of his request, he submitted that the Appellant's line of argument was developed with the benefit of hindsight. In particular, he submitted that the distillation residue obtained as a result of carrying out the distillation step according to Claim 1 of the patent in suit was not a simple mixture of an acid, an alcohol and an ester such as was discussed in D5, but was rather a complex mixture of high-boiling materials not containing any terephthalic acid, as could be seen from analytical data submitted by him in the letter dated 18 February 1992, as well as from D6. For this reason, he argued, neither D1 alone nor in

combination with the common general knowledge represented by D5 could suggest a solution to the technical problem underlying the patent in suit.

- VI. At the end of the oral proceedings, the decision of the Board to dismiss the appeal was announced.

Reasons for the Decision

1. The appeal is admissible.
2. The Board is satisfied that the process according to Claims 1 to 11 is novel with respect to the cited state of the art. Since novelty was no longer disputed by the Appellant at the oral proceedings, there is no need to give reasons for this finding.
3. The only issue that remains to be decided in these appeal proceedings is that of inventive step.
 - 3.1. In the Board's judgment, the decision under appeal correctly stated that D6, corresponding to US-A-4 092 481 (see the patent in suit, page 2, lines 31 to 40), represents the closest state of the art. This was accepted by the Appellant during the oral proceedings. This document discloses a method of producing dimethyl terephthalate and intermediate products of the dimethyl terephthalate manufacture from the high-boiling byproducts obtained during the air oxidation of p-xylene and methyl p-toluate in the liquid phase, subsequent esterification of the resulting acids and separation of the so-produced esters, which comprises treating the high-boiling byproducts with methanol at an elevated temperature and then separating the resultant product by distillation with the addition

of a carrier such as methanol (see e.g. Claim 1). The document further states that the treatment of the byproducts with methanol at an elevated temperature is carried out at a temperature of 180 to 370°C and a pressure of 0 to 60 atms gauge (see e.g. Claim 2). In Example 1, giving the best yields of DMT, the treatment with methanol is carried out at a temperature of 250°C and under a pressure of 24 atms gauge. The Appellant did not dispute the Respondent's submission that under these conditions no liquid methanol could have been present.

- 3.2. In respect of this state of the art, the patent in suit sets out to solve the technical problem of providing reaction conditions which allow to obtain, from the said high-boiling distillation residue, DMT and other useful products, such as MPT and MHB, in higher yields than were obtained according to Example 1 of D6 (see the description of the patent in suit, page 2, lines 41 to 56).

As solution of this technical problem, the patent in suit essentially proposes to carry out the treatment of the distillation residue with methanol at a temperature of 110 to 240°C under a pressure sufficient to maintain methanol in the liquid phase (see Claim 1 in point I above).

It follows from the data contained in Tables 1 and 2 of the patent in suit that this technical problem is thereby solved. This was not disputed by the Appellant.

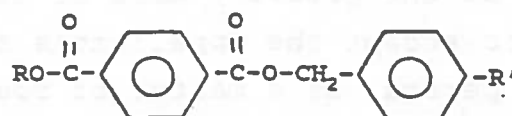
- 3.3. Since D6 (see point 3.1 above) does not contain any suggestion to carry out the methanol treatment under such conditions that liquid methanol is present in the treating system, nor that thereby higher yields of useful products can be obtained, it remains to be examined whether this document in combination with D1

and/or the common general knowledge represented by D5 would have suggested to a person skilled in the art that the particular reaction conditions set out in the present Claim 1 would solve this technical problem.

3.3.1. D1 describes in Example 3 a process for manufacturing DMT comprising that DMT is separated predominantly by crystallisation, and not by distillation. It is a mother liquor remaining after separation of DMT crystals which is subsequently treated by distillation in order to recover methanol for recirculation to the esterification reaction. The bottoms liquid 21 from distillation column 20 can then subsequently be treated to remove residual methanol and water and the residue of that distillation, which is not the same as the distillation residue to be treated in accordance with the present Claim 1, is then recycled to the oxidation reactor 1 (see the top of column 6 and Figure 2). At the end of Example 3 there is a discussion of the possibility of distilling the methanol-containing residue. The fraction boiling below 250°C is recycled to the oxidation reactor 1 and DMT is recovered from the fraction boiling from 250 to 300°C by crystallisation. D1 also generally proposes to recover the desired ester by distillation (column 3, lines 13 to 16). However, there is no reference to any use being made of higher boiling fractions. The technical problem addressed in D1 is the improvement of the esterification of the reaction products obtained by the oxidation of aromatic compounds such as p-xylene. According to column 3, lines 32 to 61 the objectives of the process of D1 were to avoid having to separate out the acids prior to esterification, and to avoid the use of esterification catalysts which were difficult to remove prior to the recycling of incompletely oxidised products.

3.3.2. D5 refers to the well-known and undisputed fact that the esterification of an acid with an alcohol is an equilibrium reaction and that the equilibrium can be shifted, according to the law of mass action, by the application of an excess of either the acid or the alcohol.

3.4. During the oral proceedings, the Appellant submitted that the distillation residue obtained according to D6, which was to be treated according to the process of the patent in suit, was practically identical with the oxidation product esterified in the process of D1, because it contained substantial amounts of free terephthalic acid (TA). This submission, which was not supported by any kind of evidence, was based on the observation that the distillation residue treated in Example 1 of the patent in suit contained 15% by weight of DMT, whereas the product obtained after the methanol treatment of 100 g of the said residue contained 32.6 g of DMT. The Board, however, does not find this submission convincing, since it is not only contrary to the analytical data of the distillation residue treated with methanol in Example 1 of D6, which do not mention TA, and since, more importantly, it does not take into account the experimental evidence submitted by the Respondent during the opposition proceedings and, again, in the statement of grounds of appeal. The Respondent has shown by this evidence that the said distillation residue is not a simple mixture of an acid, an alcohol and an ester such as is discussed in D5, but is a complex mixture of high-boiling materials comprising benzyl benzoate products of the following type:



R = H, CH₃, aromatic benzyl group
R' = -CH₃, -COOH, -COOCH₃, -COOR

In the Board's judgment, it is immediately apparent that the reaction of the above products with methanol is capable of yielding DMT, so that the yields of DMT obtained in Example 1 of the patent in suit do not support the Appellant's allegation that the distillation residue treated in this example had to contain free TA.

- 3.5. It is also clear to the Board that, according to the law of mass action, the treatment of the above products with an excess of methanol, be it liquid or gaseous, would yield DMT and other useful esters, as was already shown by D6.

However, neither D1 nor D5 relate to the problem of finding optimal reaction conditions for the transformation of the above compounds **contained in the distillation residue** in useful products such as DMT. In particular, the undisputed common general knowledge that such a transformation would require the presence of an excess of methanol would not have suggested to the skilled person to modify the process described in D6 in the way set out in the patent in suit, i.e. to use relatively low reaction temperatures and to apply a pressure sufficient to keep the methanol in the liquid stage, with a view to a further improvement of the recovery of the said useful products. In this respect, the Board observes that the law of mass action as well as the content of D5 were both common general knowledge already at the priority date of D6. The Board is thus unable to accept the Appellant's submission that a skilled person, as a matter of routine, would have applied for this purpose the reaction conditions applied in D1 to a quite different starting material.

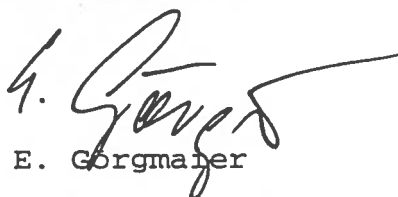
4. Therefore, the Board holds that the process according to Claims 1 to 11 of the patent in suit involves an inventive step and that the patent can be maintained as granted.

Order

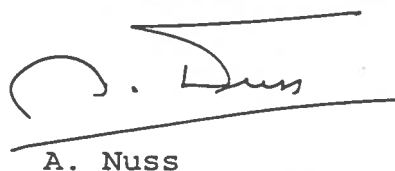
For these reasons it is decided that:

The appeal is dismissed.

The Registrar:


E. Gorgmaier

The Chairman:


A. Nuss

