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File Number: T 507/89 - 3.3.3

Application No.: 81 303 826.2

Publication No.: 0 046 670

Title of invention: Process for the preparation of polybutylene terephthalate

Classification: C08G 63/22

DECISION  
of 31 January 1991

Applicant:

Proprietor of the patent: Celanese Corporation

Opponent: Akzo N.V.

Headword:

EPC Articles 54, 56, 104

Keyword: "Novelty (yes)"  
"Inventive step (yes) - Hindsight analysis - Time factor"  
"Apportionment of costs (no)"

Headnote



Case Number : T 507/89 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 31 January 1991

Appellant :  
(Opponent)

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Decision under appeal :

Decision of Opposition Division of the European  
Patent Office dated 8 June 1989 rejecting the  
opposition filed against European patent  
No. 0 046 670 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : F. Antony  
Members : C. Gérardin  
M. Aúz Castro

## Summary of Facts and Submissions

- I. The mention of the grant of the patent No. 46 670 in respect of European patent application No. 81 303 826.2 filed on 21 August 1981 and claiming priority of 25 August 1980 from an earlier application in the United States, was published on 8 May 1985 on the basis of 13 claims.

Claim 1 read as follows:

"A process of preparing polybutylene terephthalate in which terephthalic acid and 1,4-butanediol are reacted in a first stage at elevated temperature and in the presence of polyesterification catalyst and, after a major part of the terephthalic acid has been consumed, but before the conversion of substantially all the terephthalic acid has been reached in the first stage, the reaction products of the aforesaid first stage are further reacted in a second stage at a temperature which is higher than that of the first stage to distill 1,4-butanediol in the presence of polycondensation catalyst to provide polybutylene terephthalate characterised in that the temperature of the first stage reaction is up to 215°C."

- II. On 22 January 1986 the Opponent filed a notice of opposition against the grant of the patent on the grounds that the requirements of Article 100(a) EPC were not met and that the patent specification 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). While the latter objection was not substantiated, the former was based on the following documents

(1) DE-A-2 338 824 = (1a) FR-A-2 215 437

- (2) Journal of Polymer Science (Polymer Chemistry Edition), April 1981, Volume 19, No. 4, 1021 to 1032
- (3) US-A-4 014 858
- (4) NL-A-7 112 807 = (4a) GB-A-1 363 446
- (5) JP-A-121 697/77 (Derwent abstract)

and on the fact that the experimental results in the patent in suit did not demonstrate the criticality of the features of the process Claim 1.

III. By an interlocutory decision dated 8 June 1989 the Opposition Division maintained the patent in amended form, the amendments consisting in specifying, first, that 1,4-butanediol and terephthalic are reacted in a molar ratio of 1.2:1 to 2:1 and, secondly, that the second stage starts after at least 60 weight percent of the terephthalic acid has been consumed.

In that decision, it was first stated that document (2) was published after the publication date of the patent in suit and could not consequently be taken into consideration. The distinguishing feature of the process according to the patent in suit with respect to document (1a), regarded as the closest state of the art, was that in the former the first stage was operated at up to 215°C, whilst according to that citation the esterification reaction was carried out at 220 to 280°C. Since no skilled operator would use temperatures higher than necessary, it could be fairly concluded that the teaching of document (1a) led away from using temperatures of 215°C or less. The experimental data in the patent in suit provided evidence that, when operating at temperatures of 215°C or less in the first stage, considerably less tetrahydrofuran (THF) was formed than when operating at 220°C. Since the other documents relied upon by the Opponent did not give

the skilled man any incentive to adopt such a low temperature range, an inventive step was acknowledged.

- IV. The Opponent (Appellant) thereafter filed a notice of appeal against that decision on 27 July 1989 and paid the prescribed fee at the same time. In the statement of grounds of appeal filed on 4 October 1989 the Appellant reiterated first that the conclusions which could be drawn from the experimental results in the patent in suit were not as clear as contended by the Patentee (Respondent). In this respect, reference was made to document (2), which showed that, even at a temperature of 210°C, there were conditions leading to an unduly high THF production. Further, the temperature range of up to 215°C for the first stage was suggested by documents (3), (4) and (5); the combination thereof with the general teaching of document (1a) could not consequently be regarded as inventive.

Oral proceedings were requested in case the Board should consider the maintenance of the patent in any form.

- V. The Respondent did not file any counterstatement of appeal, nor did he appear at the oral proceedings which were held on 31 January 1991, to which he had been duly summoned.

During these oral proceedings, although the Chairman had underlined in his introduction that document (2) was not a prior art document within the meaning of Article 54(2) EPC, the Appellant nevertheless relied on that citation to interpret document (5) as a novelty destroying disclosure, as well as to demonstrate the alleged lack of relevance of the experimental data in the patent in suit.

Further, the Appellant put forward that, because of the absence of any comments on the Respondent's side, he had incurred higher costs and that consequently an apportionment of costs in his favour seemed appropriate.

- VI. The Appellant requested that the decision under appeal be set aside, the patent be revoked and costs be apportioned in his favour.

There was no formal request from the Respondent's side.

#### Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.
2. The first question which arises is whether document (2), an article written by the two inventors incorporating the subject-matter of the patent in suit into a broader teaching, forms part of the state of the art within the meaning of Article 54 EPC.

According to Article 54(2) EPC the state of the art shall be held to comprise everything made available to the public before the date of filing of the European patent application.

That citation was published in April 1981, that is eight months after the priority date of the patent in suit (25 August 1980). It, therefore, does not belong to the state of the art and is not novelty destroying; nor can it be used as a reference to interpret the teaching of prior art documents to support any substantive objection.

3. The current wording of the claims does not give rise to any objections under Article 123 EPC.

With regard to Claim 1 originally filed, the present Claim 1 differs in substance by the indication of the ratio of butanediol to terephthalic acid, i.e. from 1.2:1 to 2:1, the incorporation of the amount of terephthalic acid, i.e. at least 60 weight percent, which has to be consumed in the first stage, and by the introduction of the upper limit of 215°C for the temperature of the first stage reaction. These three features, which are disclosed respectively in Claims 6, 12 and 7 of the application as originally filed, have a counterpart in respectively Claims 3, 12 and 1 of the patent as granted. The other amendments, especially the shifting of features from the characterising part into the preamble of the claim, are basically of formal nature and do not affect the scope of the protection sought.

Further, Claim 2 corresponds to Claim 2 of the patent as granted, which in turn derives from the combination of the ranges of temperature mentioned in Claim 7 and 8 of the application as originally filed. Claims 3 to 9, 11 and 12 correspond respectively to Claims 4 to 10, 12 and 13 of the patent as granted and to Claims 9, 2, 3, 4, 5, 10, 11, 12 and 13 of the application as originally filed, with their numbers and appendancies adjusted. Finally, present Claim 10 corresponds to Claim 11 as granted, which is in turn supported by lines 6 and 7 of page 4 of the original application.

4. The patent in suit concerns a process for the preparation of butylene terephthalate. A similar process for obtaining such a polymer is disclosed in document (3), which the Board regards as a state of the art closer than document (1a) considered by the Opposition Division, since the

latter document deals only with the preparation of polyesters in general, as will appear hereinafter. The process described in document (3) comprises reacting, in the first stage, (i) 1,4-butanediol or a mixture thereof with a polyhydroxyl compound, wherein 1,4-butanediol provides from 75 to 100 equivalent percent of the total hydroxy groups, and (ii) terephthalic acid or a mixture thereof with a polycarboxylic acid, wherein terephthalic acid provides from 75 to 100 equivalent percent of the total carboxyl groups, in a concentration of 1.1 to 4 hydroxyl equivalents per carboxyl equivalent, at a temperature of 180 to 250°C, and in the presence of a tetravalent tin catalyst compound having one organo-to-tin linkage (Claim 1). This particular tin catalyst serves to achieve a rapid first stage polyesterification of terephthalic acid with 1,4-butanediol at temperatures comprised between 180 and 250°C to form a clear polyester melt as well as to limit the loss of 1,4-butanediol by undesirable formation of THF to economically acceptable levels (column 2, lines 35 to 44; column 4, lines 11 to 31). Throughout that document it is emphasised that the first stage reaction has to be completed, i.e. the reaction mixture must become clear, before the second stage is begun (column 3, lines 40 to 44; column 5, lines 59 and 60; column 7, lines 43 to 45). After substantially all of the water of esterification has been removed, the polyester can be condensed to high molecular weight using conventional means with or without further addition of catalyst, typically at reduced pressure at 240°C for 2 hours (column 5, lines 3 to 30; column 7, Example XXXV). The experimental results in Tables I and II show that, by using butyl stannic anhydride as catalyst, the number of moles of THF formed per mole of 1,4-butanediol charged to the reactor is commonly 0.05 and can even be as low as 0.025 (Examples XV and XX). Although such figures can be regarded as satisfactory as far as the



overall yield of the reaction is concerned, they can only be obtained by using specific tetravalent tin catalysts.

In the light of this restrictive condition of the prior art, the technical problem underlying the patent in suit can thus be seen in providing a more flexible process which, without increasing the rate of conversion of 1,4-butanediol to THF, would not be tied down to the use of specific first stage esterification catalysts.

According to the patent in suit this problem is solved by conducting the first stage esterification reaction at a temperature of up to 215°C until at least 60 weight percent, but not all of the terephthalic acid has been consumed, and only then commencing the second stage polycondensation reaction at a higher temperature.

5. The Board cannot follow the Appellant's argument that the experimental data in the patent in suit did not demonstrate that the operative features specified in Claim 1 would necessarily lead to the desired result and, thereby, provide a general solution to the above-defined problem.

The experiments reported in Table I show the effect of the temperature on the production of THF during the first stage esterification/oligomerisation of 1,4-butanediol and terephthalic acid. For the three temperatures of 190, 200 and 210°C, the rate of conversion of 1,4-butanediol to THF can be regarded as acceptable, even when the first stage reaction is carried out to completion, as in the third run; by contrast, at 220°C the amount of THF produced would be considerably higher. These results, thus, illustrate the Respondent's argument, according to which between 210 and 220°C there is a marked change in the

production rate of THF (statement filed on 28 November 1986 in opposition procedure, point 18).

Similarly, the experiments reported in Table III show the effect on the production of THF of commencing the second stage polycondensation before reaching the clearing point. Example 1 demonstrates that keeping the temperature at 210°C until the completion of esterification, i.e. the clearing point, which occurs after 75 minutes, causes 4% of the butanediol to be converted to THF. Examples 4, 7 and 9 show that there is no increase at all or, in any case, a still acceptable increase in the amount of THF formed when, before the completion of the first stage reaction, the temperature is raised to 235°C and a vacuum applied. In the Board's view, this clearly illustrates that the combination of operative features as specified in Claim 1, i.e. the conditions regarding the first and second stage temperatures as well as the starting point of the second stage reaction, provides effectively a solution to the above-defined technical problem.

The fact that the above data confirm well-known trends already mentioned in document (3) (column 4, lines 27 to 31) about the influence of the reaction temperature on the duration of the full esterification and on the rate of conversion of 1,4-butanediol to THF, does not affect the validity of these data or the conclusions which may be drawn therefrom, as the Appellant argued in the statement of grounds of appeal (page 2, paragraphs 5 and 6). The purpose of these data is not to illustrate the influence of temperature on the course of the esterification reaction, but to demonstrate the existence of a critical temperature.

6. The Appellant argued for the first time during oral proceedings that the solution claimed in the patent in

suit was not novel with regard to the teaching of document (5).

That citation describes the production of polyester mouldings from 81.34 kg terephthalic acid, 1.66 kg isophthalic acid and 72 kg 1,4-butanediol, which corresponds to a molar ratio diol:diacid of 1.6, together with 42 g tetra-n-butyltitanate as catalyst. The first stage reaction is carried out between 160 and 210°C under ordinary pressure for 3.5 hours while distillate is continuously distilled out; after further addition of catalyst, the temperature is raised to 250°C and polycondensation is performed under 0.3 mm Hg for three hours. On the basis of the identity of the aforementioned compositional and operative features with the requirements specified in Claim 1 of the patent in suit, the Appellant has concluded that the condition of only partial reaction at the end of the first stage must implicitly be met in that prior art as well.

This argumentation cannot be accepted by the Board. First, the Appellant, who being the Opponent has the onus of proof, (see Decision T 219/83 "Zeolites/BASF" of 26 November 1985 published in OJ EPO 1986, 211, point 12, paragraphs 4 and 5), has not supported his contention by an appropriate comparative test carried out under the same experimental conditions showing that in the process according to document (5) the second stage is indeed begun before the completion of the first stage. Secondly, none of the documents relied upon by the Appellant could be of any help in interpreting document (5) in the Appellant's desired sense. For these reasons, the Board regards the Appellant's novelty objection as an unsupported allegation.

Since the issue of novelty has not been raised with regard to the other cited documents and since the Board after examination of these citations has come to the conclusion that the claimed subject-matter is not described in any one of them, it can be concluded that the solution claimed in the patent in suit is novel within the meaning of Article 54 EPC.

7. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the cited documents.

7.1 As noted above in point 4, document (3) is very specific regarding the type of catalyst to be used. This appears quite clearly from Table I, wherein the amount of THF formed is compared when using as catalysts the typical butyl stannic anhydride (uneven Examples I to XV) and the conventional tetrabutyl titanate (even Examples II to XVI), the other compositional features, in particular the quantity of the respective catalyst and the butanediol:terephthalic acid ratio, being the same; the last two Examples, XVII and XVIII, wherein respectively butyl tin oxide and octyl tin oxide are used as catalysts, are further comparative examples related to the reference Example XI. The experimental results show that in all cases significantly less THF is formed when butyl stannic anhydride is used, in practice only between 26% (Examples IX/X) and 83% (Examples V/VI). It is self-evident that any solution to the above-defined problem must at least compensate the advantages provided by such specific tin catalysts which will be lost.

The general remarks in document (3) about the various factors which may influence the formation of THF do not give the skilled man any incentive to adopt a solution along the line claimed in the patent in suit. In that

citation, it is first said that a higher butanediol:terephthalic acid ratio tends to shorten the first stage reaction and thereby reduce the undesirable formation of THF (column 1, line 65 to column 2, line 5); further, it is indicated that the lower the reaction temperature, the longer it takes to obtain full esterification, while the higher the reaction temperature, the more butanediol is converted to THF (column 4, lines 27 to 31). These conclusions would thus suggest to increase the butanediol:terephthalic acid ratio and/or reduce the temperature of esterification. In the above first case, however, this would result in both (i) a larger reactor to produce an equivalent amount of polybutylene terephthalate and (ii) a larger butanediol recycle system; this would make the direct method of production of polybutylene terephthalate economically unattractive compared with the traditional transesterification route using dimethyl terephthalate (column 1, lines 35 to 42; column 2, lines 6 to 19). This would lead to the practical conclusion that, for a reduced conversion of butanediol to THF, one should operate the first stage reaction at lower temperature until formation of a clear polyester melt, which would be unattractive for kinetic reasons.

- 7.2 The feature of only partial esterification reaction is known from document (1a). According to its broad disclosure, which deals with the preparation of polyesters in general by direct esterification of a dicarboxylic acid and a diol, only partial esterification is performed in the first stage, following which polycondensation is carried out at high temperature (Claim 1). More specifically, the first stage is normally carried out up to 70 to 95% of completion at a temperature between 220 and 280°C at superatmospheric pressure (Claims 3 and 4), optionally in the presence of special additives in order

to limit the undesirable formation of ether by-products (page 3, lines 1 to 15). This conversion is followed by an intermediate second step, in which the unreacted diol as well as any by-products are eliminated at reduced pressure at a temperature comprised between 150 and 220°C (Claims 2 and 5). In a third stage, final polycondensation is carried out at a temperature comprised between 240 and 300°C at a still lower pressure (page 4, lines 10 to 19).

More specifically, as far as the esterification stage is concerned, the temperatures actually used in the examples of document (1a) vary between 230°C (Examples 5, 7, 9 of Table I and 19 on page 9) and 241°C (Example 21 of Table III). According to Example 19, which is specifically directed to the preparation of polybutylene terephthalate, the partial esterification reaction is carried out at 230°C. In the Board's view, such temperatures must be regarded as an essential feature of that prior art process. As mentioned in the introduction of document (1a), the teaching disclosed therein aims at providing a general process of preparation of polyesters by the direct esterification route, wherein some of the typical problems associated with that embodiment, in particular the formation of undesirable by-products in higher amounts than during transesterification, can be overcome to a large extent (page 1, lines 24 to 34). That aim is achieved by carrying out an only partial esterification, which has the further economic advantage of shortening the whole process (page 2, lines 3 and 4). Since no skilled worker would use temperatures higher than necessary, the fact that the temperature is kept high during that stage in spite of the well-known advantage of lower temperatures in terms of by-products formed, can only mean that such a high temperature is necessary when esterification is not carried out to completion; there is thus no reason to

depart from that teaching by formally combining the features of document (1a) with those of document (3).

7.3 Document (4) describes a process for the preparation of polyesters from dicarboxylic acid or esters thereof and 1,4-butanediol involving temperatures lower than 200°C during the first stage. According to Claim 1 of that citation, the process comprises, first, condensing 1,4-butanediol with the acid or ester reactant in a molar ratio of from 1.05:1 to 1.25:1 at a temperature below 200°C until a major proportion of non-reacted diol and volatile condensation products have distilled off, then, subjecting the resulting condensate to further condensation in a vacuum at a temperature of from 250 to 310°C. In the description, it is emphasised that the condition not to increase the temperature of esterification beyond 200°C until the excess of free butanediol has been substantially removed, which obviously indicates a fully completed reaction, is in fact essential to obtain polyesters not contaminated with decomposition products (page 4, lines 9 to 16).

Although document (4) deals primarily with the transesterification process starting from dialkyl terephthalates, as illustrated in all the examples, it is clear that the direct esterification route is encompassed as well within that teaching; the main change occurring according to that alternative concerns the catalyst, which should no longer promote transesterification reactions and is, therefore, selected among titanium compounds (page 5, lines 15 to 25). This means that the essential teaching of that citation is to carry out the esterification reaction to completion at a temperature lower than 200°C in order to limit the formation of by-products.

7.4 As noted above in point 6, document (5) describes the preparation of polybutylene terephthalate by using the direct esterification route. According to the only example of that abstract, whilst the range of temperatures suitable to carry out the esterification stage, i.e. 160 to 210°C, and the temperature chosen for the polycondensation stage, i.e. 250°C, correspond to those according to the claimed subject-matter, nothing is said about the degree of completion of the first stage reaction. In the Board's view, a specific interpretation of that operative feature, in particular in the sense suggested by the Appellant, can only be made by hindsight. Nor is it obvious to relate that undisclosed feature to the flexibility of the process according to the above-defined technical problem.

7.5 It follows that the skilled man, when attempting to find an alternative method to the catalyst solution disclosed in document (3), is faced with teachings whose main features are in fact mutually exclusive. Whereas document (1a) recommends to carry out only a limited esterification reaction at high temperature, document (4) suggests to complete the esterification reaction, but at a lower temperature. Although the claimed subject-matter might, at first sight, appear as a mere combination of those teachings, i.e. as the combination of partial esterification according to document (1a) and of low temperature according to document (4), closer examination shows that these features are in fact incompatible.

As already noted in the analysis of document (1a) above, the authors of that citation were well aware of the negative influence of high temperature on the amount of by-products formed; this means that partial esterification reaction and high temperature must be regarded as a whole and that, consequently, the concept of reaction not



carried out to completion should not be isolated from its technical context. Further, it is essential to appreciate that the low temperature required according to the teaching of document (4) concerns a process wherein the butanediol:terephthalic acid ratio, i.e. a parameter which has a considerable bearing on the production of THF, is significantly lower (1.1:1 according to Example 1) than in document (1a) (1.5:1 according to Example 19) and in document (3) (between 1.5:1 and 4:1 according to the examples in Tables I and II); that value of 1.1:1 corresponds in fact to the lower limit of the suitable ranges according to these two documents. In this respect, thus, the process disclosed in document (4) differs substantially from the other teachings. It follows that, for the skilled man, there could be no incentive to combine a specific feature from document (4) with the other disclosures and that the combination of low temperature according to that document with partial esterification reaction according to document (1a) can only be made by hindsight analysis.

Another element to consider is the time factor, i.e. the time which elapsed between the date of publication of documents (1a) and (4), respectively 1974 and 1972, and the date of priority of the patent in suit (25 August 1980). In the Board's view, such a number of years in a technical field of commercial importance to which considerable attention is directed, is evidence that the claimed solution, in particular the existence of a critical temperature at 215°C, was not obvious and speaks in favour of its inventiveness.

7.6. In conclusion, for the various reasons given above, the subject-matter of Claim 1 involves an inventive step.

8. Claim 1 being allowable, the same applies to the dependent Claims 2 to 12, which represent preferred embodiments of the subject-matter of Claim 1 and whose patentability is supported by that of the main claim.
  
9. A further question to be considered is whether the fact that the Respondent did not file any counterstatement, nor appear at the oral proceedings, could justify an apportionment of costs in the Appellant's favour.

Article 104(1) EPC states the principle that each party to the proceedings shall meet the costs he has incurred and that a different apportionment of costs can only be ordered for reasons of equity.

First, the Board notes that the appeal file does not contain any statement by the Appellant other than the statement of grounds of appeal and that during oral proceedings the Appellant did not submit any experimental test report; this means that the costs actually incurred by the Appellant did not exceed the costs normally related with the filing of an appeal. Further, the Board observes that oral proceedings were requested by the Appellant himself at so early a stage as the statement of grounds of appeal, thus without even waiting for possible arguments from the other party or comments from the Board, which could have made these oral proceedings superfluous.

All costs incurred in appeal proceedings were thus caused by the Appellant. The Respondent remained completely passive and did not contribute to the costs in any respect. Therefore the question of equity does not arise and there is no reason to deviate from the principle stated above.

Order

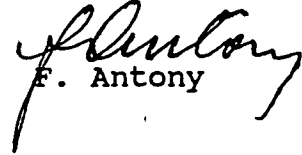
For these reasons, it is decided that:

1. The appeal is dismissed.
2. The request for apportionment of costs is rejected.

The Registrar:

  
E. Görgmaier

The Chairman:

  
F. Antony

CG 4/6/91  
HC 4/6/91