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D E C I S I O N
of 9 November 2000

Case Number: T 0444/98 - 3.3.3

Application Number: 92921371.8

Publication Number: 0563354

IPC: C08G 63/80

Language of the proceedings: EN

Title of invention:

Process for the production of high molecular weight polyester resins

Applicant:

SINCO RICERCHE S.p.A.

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Amendments (main request) - added subject-matter (yes)"
"Novelty (first auxiliary request) - product-by-process claim (no)"
"Inventive step (second auxiliary request) - known process - improvement"

Decisions cited:

-

Catchword:

-



Case Number: T 0444/98 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 9 November 2000

Appellant: SINCO RICERCHE S.p.A.
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 3 December 1997
refusing European patent application
No. 92 921 371.8 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
A. Lindqvist

Summary of Facts and Submissions

I. This appeal, which was filed on 26 January 1998, lies against the decision of the Examining Division dated 3 December 1997, refusing European patent application No. 92 921 371.8 filed as PCT/EP92/02375 on 15 October 1992 in the name of M.& G. RICERCHE S.P.A. (now assigned to SINCO RICERCHE S.P.A.), and published under No. WO 93/08226 (EP-A-0 563 354). The appeal fee was paid in advance on 23 January 1998 and the Statement of Grounds of Appeal was filed on 8 April 1998.

II. The decision under appeal was based on Claims 1 to 13 filed with the submission dated 7 March 1996, independent Claims 1, 12 and 13 reading as follows:

"1. Process for the production of high molecular weight polyester resins starting from polyester resins with intrinsic viscosity lower than 0.57 dl/g comprising the following steps:

- 1) mixing in the melt a polyester resin having intrinsic viscosity lower than 0.57 dl/g with a polyaddition additive containing at least two groups capable of addition reactions with the terminal OH or COOH groups of the resin;
- 2) converting the melted mixture into solid particles and subsequently crystallizing the particles at temperatures higher than the Tg of the polyester resin and lower than its melting point;
- 3) heating the particles at temperatures higher than 150°C up to obtain the desired increase of the intrinsic viscosity."

"12. Reactive polyester resins having intrinsic viscosity equal or higher than 0.57 dl/g obtained by

mixing in the melt a polyester resin having intrinsic viscosity lower than 0.57 dl/g with a polyaddition additive having at least two groups capable of addition reactions with the terminal OH/COOH groups of the polyester resin."

"13. Polyethyleneterephthalate and copolyethyleneterephthalate containing up to 20% by mols of units deriving from isophthalic acid according to claim 12 obtained by mixing in the melt the corresponding polymers having intrinsic viscosity lower than 0.57 dl/g, with pyromellitic dianhydride."

The further Claims 2 to 11 were dependent on Claim 1.

III. The decision under appeal held that the subject-matter of Claims 12 and 13 was not novel

(i) under Article 54(3) EPC, with regard to the overlapping Contracting States, over the disclosure of EP-A-0 475 142 (hereinafter document D2), and

(ii) under Article 54(2) EPC over the products obtained according to D1 (EP-A-0 422 282).

This conclusion of lack of novelty was essentially based on the denial of the recognition of an improved upgrading kinetics of the claimed polyesters over those according to D1.

Furthermore, in the Examining Division's opinion, the subject-matter of Claims 1 to 11 lacked an inventive step over D1, because there was no convincing evidence that a decrease of the intrinsic viscosity of the

starting polyester resin used according to that document to below 0.57 dl/g led to any unexpected result or overcame a prejudice in the art.

- IV. In the appeal proceedings the Appellant - (i) together with the Statement of Grounds of Appeal, (ii) in response to the Rapporteur's communications of 5 May 2000 and 14 August 2000 and (iii) at the oral proceedings on 9 November 2000 - submitted several amended sets of claims.

The present decision relies on the set of 13 claims submitted at the oral proceedings as main request, on a set of 13 claims filed with the submission dated 6 October 2000 as first auxiliary request and on the set of 9 claims filed at the oral proceedings as second auxiliary request.

- (i) Claim 12 of the main request reads as follows:

"12. Reactive polyester resins having intrinsic viscosity lower than 0.55 dl/g obtainable by mixing in the melt a polyester resin having intrinsic viscosity lower than 0.55 dl/g with a polyaddition additive having at least two groups capable of addition reactions with the terminal OH/COOH groups of the polyester resin."

- (ii) Claim 12 of the first auxiliary request reads as follows:

"12. A polyester resin obtainable by melt-mixing a polyester resin with I.V. lower than 0.55 dl/g with a dianhydride of a tetracarboxylic acid, added in amount of from 0.1 to 1 wt%, wherein the

added dianhydride is present in a combined form."

(iii) Claim 1 of the second auxiliary request reads as follows:

"1. Process for the production of high molecular weight polyester resins starting from polyester resins with intrinsic viscosity lower than 0.57 dl/g comprising the following steps:

- 1) mixing in the melt a polyester resin having intrinsic viscosity lower than 0.57 dl/g with a polyaddition additive which is selected from the dianhydrides of tetracarboxylic acids;
- 2) converting the melted mixture into solid particles and subsequently crystallizing the particles at temperatures higher than the T_g of the polyester resin and lower than its melting point;
- 3) subsequently heating the crystallized particles at temperatures higher than 150°C and lower than the melting point of the resin to obtain the desired increase of the intrinsic viscosity."

V. In its written and oral submissions the Appellant argued as follows:

(i) The replacement in Claim 12 of the main request of the previous definition "Reactive polyester resins having intrinsic viscosity equal or higher than 0.57 dl/g" by the definition "Reactive polyester resins having intrinsic viscosity lower than 0.55 dl/g" was supported by the disclosure in the original application of a mixture of the

starting polyester and of the dianhydride prior to the melt blending operation.

- (ii) The polyester resins according to Claims 12 and 13 of the main request and of the first auxiliary request were novel and inventive over documents D1 and D2, because, unexpectedly, they exhibited an enhanced solid state polyaddition kinetics.

- (iii) The subject-matter of Claims 1 to 11 of all requests was unobvious over D1, because this document did not suggest, that the time for preparing a high intrinsic viscosity polyester could be reduced by lowering the intrinsic viscosity of the starting polyester; this effect being experimentally proved by the evidence contained in the Appellant's submission dated 8 April 1998 for homopolyethylene terephthalate (PET) (Tests A and B on page 5, lines 11 to 20 of said submission) as well as for copolyethylene terephthalate/isophthalate (COPET) (reworking of Example 3.9 of the application in suit on page 6, lines 10 to 16 of said submission).

- (iv) Nor was the subject-matter of Claims 1 to 11 suggested by document

D3: US-A-4 238 593,

because the evidence contained in the submission dated 6 October 2000 (cf. point 3-5 on page 2) showed that, despite the presence of "make-up terephthalic acid", the solid state polycondensation of the low intrinsic viscosity

polyesters used according to this document followed a slower kinetics.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of:

- main request: Claims 1 to 13 filed during oral proceedings;
- first auxiliary request: Claims 1 to 13 filed on 10 October 2000;
- second auxiliary request: Claims 1 to 9 filed during oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. *Admissibility under Article 123(2) EPC of the amendments in Claim 12*

The feature of Claim 12 "Reactive polyester resins having intrinsic viscosity lower than 0.55 dl/g" is not supported by the description as originally filed.

Steps 1 and 2 of Claim 1 as well as steps b) and c) on page 3, lines 6 to 14 of the application as filed refer to the melt-mixing of a polyester resin having an I.V. of lower than 0.57 dl/g (or lower than 0.55 dl/g: cf. page 4, lines 23 to 26 of the application) with a

polyaddition additive and to the subsequent conversion of the melted mixture into solid particles.

Pursuant to Examples 1 to 3 of the application this melt-processing step results in an increase of the I.V. of the starting polyester: according to Example 1 from 0.408 dl/g to 0.62 dl/g (± 0.07 dl/g), according to Example 2 from 0.408 dl/g to 0.58 dl/g (± 0.018 dl/g) and according to Example 3 from 0.52 dl/g to 0.61, 0.65 or 0.69 dl/g (dependent on the amount of PMDA) (cf. Example 1, page 8, line 26 to page 9, line 14; Example 2, page 9, line 31 to page 10, line 1; Example 3, page 10 lines 5 to 12, page 11, Table 1).

It is, thus, evident from the disclosure of the application in suit that the melt-preparation of a blend ("reactive polyester resin") of polyester and pyromellitic acid dianhydride (PMDA) cannot be performed without some increase of the I.V. of the starting polyester; consequently, the concept of a blend, whose I.V. is equal or lower than the upper I.V. limit of the starting polyester is not supported by this disclosure.

In the absence of any other disclosure in the application as filed concerning the possibility that the "reactive polyester resins" may have an intrinsic viscosity lower than 0.55 dl/g this feature lacks support.

3. Claim 12, therefore, contravenes the requirement of Article 123(2) EPC and is, thus, not admissible.
4. Since a request can only be considered as it stands, the entire main request must share the fate of Claim 12

and is, thus, not admissible.

First auxiliary request

5. *Admissibility under Article 123(2) EPC of the amendments in Claim 12*

While, in the Board's judgment, this claim is also deficient under Article 123(2) EPC, this issue needs not to be discussed, because - for the reasons to follow - its subject-matter is anyway not allowable under Article 54 EPC.

6. *Novelty of the subject-matter of Claim 12*

6.1 The subject-matter of Claim 12 of the first auxiliary request is anticipated by the disclosure of document D1.

6.1.1 According to its Claim 1 D1 relates to a process for the continuous production of high molecular weight polyester resin from polyester resin having a lower molecular weight, in which the resin is blended in a molten state with an additive adapted to accelerate the achievement of high viscosity, transformed into a granulate and then treated in a solid state polycondensation reactor, wherein said additive is a dianhydride of an aromatic tetracarboxylic acid, e.g. PMDA (Claims 2, 3).

The optimal concentration of PMDA with respect to the polyester is disclosed to be 0,1 to 1 % by weight (page 3, lines 2 to 3).

According to Example 1 (page 3, lines 19 to 45)

polyethylene terephthalate (PET) having an intrinsic viscosity (I.V.) of 0,57 dl/g was melt-mixed in an extruder with a mixture of 20% weight of PMDA in crystallized PET-powder (I.V. 0,58 dl/g). The extrudate was pelletized to chips of I.V. $0,6 \pm 0,02$ dl/g, which were subjected to solid state polycondensation for 5 hours at 202°C. The resulting PET had an I.V. of $1,16 \pm 0,022$ dl/g, indicating an upgrading kinetics of 0,108 dl/g per hour.

- 6.1.2 The only possible difference between the disclosure of D1 and the subject-matter of present Claim 12, thus, resides in the use, according to the latter, of a starting polyester having an I.V. of lower than 0.55 dl/g (whereas according to D1 the lowest I.V. disclosed is 0.57 dl/g).
- 6.1.3 However, this difference is not able to establish novelty, because the lower I.V. of the starting polyester does not manifest itself directly on the resulting polyester resin, nor can the presence of this feature be ascertained by analysis of the final product and/or any measurements carried out on it.
- 6.1.4 The latter observation also applies to the higher solid state polyaddition kinetics allegedly exhibited by the "inventive" products, because
- (i) this property is dependent on many variables, which are not defined in said claims, among which are *inter alia* the OH and COOH content of the starting polyester (cf. Fig. 1 and 2 of D2), the moisture content of the starting polyester (cf. page 2, lines 26 to 36 of D2) and the temperature of the solid state polyaddition (cf. application

in suit: Table 1, comparison of test series 3.1 vs. 3.3, 3.4 vs. 3.6 and 3.7 vs. 3.9) and because

- (ii) according to common general knowledge, the relationship between the I.V. of the starting polyester and the ensuing solid state polyaddition kinetics follows a continuous pattern and does not exhibit any discontinuity at an I.V. of 0.55 dl/g,
- (iii) with the consequence that any tentative inference of the I.V. of the starting polyester from the measured solid state polyaddition kinetics implies an unknown margin of error,
- (iv) rendering it impossible to conclude with any certainty, whether a starting polyester had an I.V. below or above a certain value, including the value of 0.55 dl/g according to present Claim 12.

6.1.5 That I.V. feature is not, therefore, capable of unambiguously distinguishing the matter for which protection is sought from matter beyond that I.V. limit, including such reactive polyester resins, which have been prepared from starting polyesters having an I.V. of at least 0.57 dl/g (Article 84 EPC, Rule 29 (1) EPC, Guidelines C III, 4.5a).

6.2 The same conclusion is arrived at with respect to the disclosure of document D2, which represents prior art under Article 54(3) EPC for the overlapping Contracting States.

6.2.1 Claim 1 of D2 relates to a PET-composition comprising a

reaction product of a PET resin, e.g. one having an intrinsic viscosity of 0.55 to 0.65 dl/g (Claim 6), and 0.05 to 5.0 wt% of a copolymer of maleic anhydride. According to Claim 10 the composition may furthermore comprise 0.05 to 0.5 wt% of PMDA.

The components of the composition are mixed in and formed by an extruder (page 3, lines 21 to 25 and lines 30 to 38 (Example)).

- 6.2.2 The only possible difference between the disclosure of D2 and the subject-matter of present Claim 12, thus, resides in the use, according to the latter, of a starting polyester having an I.V. of lower than 0.55 dl/g (whereas according to D2 the lowest I.V. disclosed is 0.55 dl/g).

For the reasons set out in points 6.1.3 to 6.1.5 supra this difference is not able to establish the novelty of the subject-matter of Claim 12 vis-à-vis D2.

- 6.3 Since Claim 12 thus contravenes the requirements of Article 54 EPC the first auxiliary request has to be rejected as a whole (cf. point 4 supra).

Second auxiliary request

7. *Admissibility under Article 123(2) EPC of the amendments in Claim 12*

- 7.1 Claim 1 differs from its version as originally filed

- (i) by the replacement of the statement "1) mixing in the melt a polyester resin ... with a polyaddition additive containing at least two

groups capable of addition of the resin;
reactions with the terminal OH or COOH groups of
the resin" by the statement "1) mixing in the
melt a polyester resin ... with a polyaddition
additive which is selected from the dianhydrides
of tetracarboxylic acids." and

- (ii) by the substitution of the statement "3)
subsequently heating the crystallized particles
..." for the original statement "3) heating the
particles".

Amendment (i) is based on original Claim 7, amendment
(ii) is based on original Claim 1 in conjunction with
the passage on page 8, lines 5 to 11 of the original
description.

7.2 Claims 2 to 9 are based on original Claims 2 to 4, 6
and 8 to 11.

7.3 The requirement of Article 123(2) EPC is, thus,
complied with.

8. *Novelty*

8.1 Document D1

The method of preparation of high I.V. polyester resins
disclosed in this document corresponds to that
according to present Claim 1 (cf. point 6.2 supra).
However, the lowest intrinsic viscosity of a starting
polyester mentioned in D1 is 0.57 dl/g (Example 1,
page 3, lines 19 to 23), whereas present Claim 1
requires that the intrinsic viscosity of the starting
polyester be **lower** than 0.57 dl/g.

8.2 Document D2

The PET-compositions according to this document are processed in an extruder; D2 does not disclose a solid state upgrading step (cf. point 6.2.1 supra).

8.3 Document D3

8.3.1 Claim 1 of this document relates to a method for the production of high molecular weight, high purity polyester, comprising the steps of

- (a) reacting a glycol and a dicarboxylic compound to form a polyester prepolymer having an intrinsic viscosity from about 0.40 dl/g to about 0.62 dl/g and having a carboxyl end group content from about 18% to about 40% total end groups,
- (b) polymerizing in a solid state in a static bed said polyester prepolymer so that a high molecular weight, high purity polycondensed polyester is formed having an intrinsic viscosity of at least 0.70 dl/g and an acetaldehyde impurity concentration less than about 3.0 parts per million.

According to Claim 8 the polyester prepolymer is formed by a two-stage reaction, said first stage reacting 1.0 mole of dicarboxylic acid and about 1.20 moles of glycol, and said second stage reacting make-up dicarboxylic acid, such that the overall glycol/dicarboxylic acid molar ratio is from about 1.02:1.0 to about 1.15:1.0.

The finished prepolymer is solidified and pelletized;

thereafter the particles are crystallized and dried at a temperature of from 140° to 180°C and finally fed into a solid state polymerization reactor having a temperature of from 200 to 240°C (column 6, lines 16 to 49).

8.3.2 The polyester preparation process according to present Claim 1 differs from that disclosed in D3 by the use of dianhydrides of tetracarboxylic acids instead of the "make-up" dicarboxylic acids, e.g. terephthalic acid, which may be used according to that document.

8.4 The subject-matter of Claim 1 is, thus, novel over the disclosure of documents D1, D2 and D3.

8.5 The same conclusion applies *a fortiori* to the subject-matter of the dependent Claims 2 to 9.

9. *Problem and solution*

9.1 Document D1 is considered to represent the closest state of the art, because the process disclosed therein for the production of high molecular weight polyester resins comprises the same sequence of steps (cf. point 6.1.1 supra).

9.2 The problem underlying the subject-matter of Claim 1 of the second auxiliary request vis-à-vis D1 is the provision of a process for the production of high molecular weight polyester resins, which is less time consuming and leads to a significant increase of the production capacity of the production plant (page 2, lines 4 to 18 of the original description).

9.3 According to Claim 1 the solution to this problem

resides in the use as starting material of a polyester resin having an intrinsic viscosity lower than 0.57 dl/g.

- 9.4 The available evidence demonstrates that by that feature the existing technical problem has effectively been solved.
- 9.4.1 This may be concluded, because (1) owing to the lower I.V. (and, thus, the lower degree of condensation) of the starting polyester it is self-evident that the time for preparing the "inventive" starting polyester must be lower than for the starting polyesters according to D1 having a higher I.V., and (2) because the comparative experiments submitted in the Statement of Grounds for Appeal demonstrate that also the solid state polyaddition time is shorter.
- 9.4.2 Tests A and B on page 5, lines 11 to 20 of the Statement of Grounds of Appeal compare the solid state polyaddition kinetics (12 hours at 170°C: cf. Example 1 of the original description) of reactive PET resins having been prepared by melt mixing starting polyesters having, respectively, an I.V. of 0.408 dl/g ("inventive" Test A) and of 0.60 dl/g ("comparative" Test B) with 0.4 % by weight of PMDA. The kinetics reported are 0.030 dl/g/h according to Test A and 0.016 dl/g/h according to Test B.
- 9.4.3 The reworking of Example 3.9 of the original application (cf. page 10, line 5 to page 11, Table 1 of the original description) set out on page 6, lines 10 to 16 of the Statement of Grounds of Appeal demonstrates that the same faster solid state polyaddition kinetics is also achieved with

polyethylene terephthalate copolyesters comprising 15 % by weight of units from isophthalic acid.

According to the "inventive" Example 3.9 the kinetics for a starting polyester having an I.V. of 0.52 dl/g is 0.059 dl/g/h (I.V. difference before and after the solid state polyaddition according to page 11, Table 1 of the application as filed: $1.4 - 0.69 = 0.71/12h = 0.059$]), while for the "comparative" reworking experiment using a starting polyester having an I.V. of 0.6 dl/g a solid state polyaddition kinetics of 0.045 dl/g/h was found.

9.4.4 It is, thus, established that the process for the production of high molecular weight homo- and copolyesters according to present Claim 1 exhibits a higher reaction rate than that according to D1, justifying thereby the conclusion drawn in point 9.4 supra.

10. *Obviousness*

This issue turns upon the question whether it was obvious or not to arrive at the solution of the existing technical problem as set out in point 9.2 supra by the measures taken according to Claim 1, i.e. by the lowering of the I.V. of the starting polyester to below 0.57 dl/g.

10.1 While D1 is concerned with the reduction of the solid state upgrading time, which it actually succeeds to do by the use of PMDA, this document is completely silent on the possibility of the use of starting polyesters having an I.V. lower than 0.57 dl/g. D1 cannot suggest, therefore, the solution of the existing technical

problem, i.e. the further reduction of the time for preparing high molecular weight polyester resins, by the reduction of the I.V. of the starting polyester to values below 0.57 dl/g (cf. point 9.4 supra).

- 10.2 D2 is not to be taken account of for the issue of obviousness, because it is to be considered only under Article 54(3) EPC.
- 10.3 The disclosure of document D3 encompasses the use of starting polyesters having an I.V. of 0.40 to 0.62 dl/g for the preparation of high molecular weight polyester resins (cf. Claim 1). However, according to this document the decisive parameter for a high reaction rate of the solid state upgrading step resides in the selection of a certain carboxyl end group content of from 18% to 40% (preferably 40 to 65 equivalents/ton) of the polyester prepolymer (column 8, line 63 to column 9, line 30). Since it is stated in column 9, lines 31 to 37 that the same correlation reaction "rate/COOH-content" exists within the I.V. range of from 0.35 to 0.62 dl/g, the skilled person had no reason to surmise that a sub-group of starting polyesters (prepolymers) within said I.V. range, i.e. those having an I.V. of below 0.55 dl/g would provide any benefit concerning the reaction rate, be it in the context of the polyester production method according to D3 itself or in the context of the method used according to D1.
- 10.4 Since, according to Figures 1 and 2 of D3, the reaction rate of a polyester having a COOH content of 10 equivalents/ton is much lower than that of polyesters having COOH contents 26 and 47 equivalents/ton, it is even more surprising that according to Tests A and B

(cf. point 9.4.2 supra) the solid state polyaddition kinetics of a starting polyester having an I.V. of 0.408 dl/g ("inventive" Test A), which has a COOH content of 7.8 equivalents/ton (cf. point 6-9, page 2 of the Appellant's submission dated 21 June 2000), is superior to the kinetics of a starting polyester having an I.V. of 0.60 dl/g ("comparative" Test B), which has a COOH content of 30 equivalents/ton. On the basis of D3 one would rather have expected the reverse effect.

10.5 The subject-matter of Claim 1 is, thus, not obvious over the disclosure of documents D1 and D3, alone or in combination; hence, this claim complies with the requirement of Article 56 EPC.

10.6 By virtue of their appendance to Claim 1 the same conclusion applies to Claims 2 to 9.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The main request is rejected as inadmissible.
3. The first auxiliary request is rejected.
4. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 9 submitted during oral proceedings as second auxiliary request, after any consequential amendment of the description.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin