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# DECISION
## of 6 March 2001

**Case Number:** T 0841/98 - 3.3.3

**Application Number:** 93120804.5

**Publication Number:** 0603889

**IPC:** C08G 63/81

**Language of the proceedings:** EN

**Title of invention:**
Process for the preparation of lactic acid polyesters

**Applicant:**
MITSUI CHEMICALS, INC.

**Opponent:**
-

**Headword:**
-

**Relevant legal provisions:**
EPC Art. 54, 56, 89

**Keyword:**
"Novelty - inherent disclosure (no)"
"Multiple priorities"
"Inventive step (yes)"

**Decisions cited:**
-

**Catchword:**
-

**Europäisches Patentamt**
Beschwerdekammern

**European Patent Office**
Boards of Appeal

**Office européen des brevets**
Chambres de recours

**Case Number:** T 0841/98 - 3.3.3

# DECISION
## of the Technical Board of Appeal 3.3.3
## of 6 March 2001

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|---|---|
| **Appellant:** | MITSUI CHEMICALS, INC.<br>2-5, Kasumigaseki 3-chome<br>Chiyoda-ku<br>Tokyo (JP) |
| **Representative:** | Strehl Schübel-Hopf & Partner<br>Maximilianstrasse 54<br>D-80538 München (DE) |
| **Decision under appeal:** | **Decision of the Examining Division of the European Patent Office posted 9 March 1998 refusing European patent application No. 93 120 804.5 pursuant to Article 97(1) EPC.** |

**Composition of the Board:**

**Chairman:** A. Däweritz
**Members:** C. Idez
J. De Preter

## Summary of Facts and Submissions

I. European patent application No. 93 120 804.5 filed on 23 December 1993 in the name of Mitsui Chemicals Inc., claiming priority of eight earlier patent applications in Japan and published under No. 0 603 889 on 29 June 1994, was refused by a decision of the Examining Division issued on 9 March 1998. The decision was based on the set of Claims 1 to 13 as originally filed, including independent Claims 1 and 13 which read as follows:

"1. A process for preparing a polyhydroxycarboxylic acid having an inherent viscosity of 0.3 dl/g or more by dehydrating polycondensation of lactic acid or of lactic acid and another hydroxycarboxylic acid as a raw material in an organic solvent in the presence or absence of a catalyst, wherein the content of one or more impurities selected from the group consisting of methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate in the raw material for the polycondensation is 0.3 % by mole or less in total, based on said lactic acid or lactic acid and another hydroxycarboxylic acid."

"13. A raw material for use in the preparation of a polyhydroxycarboxylic acid having an inherent viscosity of 0.3 dl/g or more, comprising lactic acid or lactic acid and another hydroxycarboxylic acid, wherein the content of one or more impurities selected from the group consisting of methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate is 0.3 % by mole or less in total, based on the amount of said lactic acid or of said lactic acid and the other hydroxycarboxylic acid."

Claims 2 to 12 related to preferred embodiments of the process according to Claim 1.

II. The Examining Division refused the application on the grounds that the subject-matter of Claims 1 to 13 lacked novelty over document D1 (EP-A-572 675) and that document D2 (EP-A-26 599) anticipated Claims 1 to 3, 5, 6 and 13. More specifically the decision held that D2 disclosed a copolymer of lactic acid and glycolic acid having an inherent viscosity of 0.08 to 0.30 dl/g which could be prepared at temperatures of from 100 to 250°C, optionally in the presence of a solvent and that, although D1 did not explicitly disclose the level of impurities in the starting hydroxycarboxylic acids, it could be deduced from the comparative Examples disclosed in the application and the inherent viscosity of the polyhydroxycarboxylic acids obtained in Example 3 of D1 that these impurities were below the level as required by Claim 1. Concerning the subject-matter of independent Claim 13, the decision stated that lack of impurities did not render a known product novel.

III. On 4 May 1998 a Notice of Appeal was lodged by the Appellant (Applicant) against this decision with simultaneous payment of the prescribed fees.

IV. In the Statement of Grounds of Appeal filed on 15 July 1998, the Appellant argued essentially as follows:

(i) D1 did not recognize the importance of the level of the specific impurities on the molecular weight of the polyhydroxycarboxylic acid obtained. The content of these impurities in the starting components was neither explicitly nor implicitly disclosed in D1.

(ii) The comparison between Example 3 of D1 and Example 1 of the present application showed that the conclusion drawn by the Examining Division concerning the level of impurities in the starting components of D1 had no basis since the polymer of Example 3 of D1 had been obtained under different conditions, in particular by applying a much longer reaction time than in Example 1 of the present application. Furthermore, the results of comparative Example 1 of the present application demonstrated that under the same reaction time as in Example 1 a polymer having a much lower molecular weight was obtained. In other words a high molecular weight could only be obtained in D1 if the reaction time was increased.

(iii) D1 did not teach to select in advance and to use raw materials having a specified level of impurities. Thus, the subject-matter of Claims 1 to 13 was novel over D1.

(iv) The subject-matter of these claims was not anticipated by document D2, since this document disclosed neither polymers with the requested inherent viscosity nor the level of impurities of the raw materials.

(v) The subject-matter of Claims 1 to 13 also involved an inventive step over D1 and D2, since there was no suggestion in these documents that by using raw materials containing less than a specified amount of impurities one could obtain a more efficient reaction as indicated by the high inherent viscosity and the high yields shown in the examples of the present application.

V. In an annex to the summons to oral proceedings dated 17 November 2000, the Appellant was informed about a number of essential questions to be discussed.

(i) The question arose whether the concentration of impurities as defined in Claim 1 under consideration had necessarily and inevitably been met in those examples of D1 which gave polymers having high molecular weights equivalent to inherent viscosities of at least 0.3 dl/g and whether the conclusion was correct which was drawn by the Examining Division on the basis of the low inherent viscosities and molecular weights obtained in the comparative examples in the present application that D1 anticipated the claimed process.

(ii) The raw material according to Claim 13 did not appear to be novel over lactic acid *per se*.

(iii) Having regard to the wording of Claim 1 and the fact that each of the eight earlier applications, the priority rights of which were claimed, referred to a maximum amount of only one of the impurities separately, the validity of the priority claims had to be considered. As a consequence thereof, D1 which was published in the priority interval was possibly to be taken into account as closest prior art for the assessment of inventive step.

(iv) The invention could then be regarded as a mere modification of the process of D1 (i.e. use of raw materials with a greater purity) and lacking therefore inventive step.

(v) An objection under Article 84 EPC was also raised against Claim 1 since the method for determining the inherent viscosity of the polyhydroxycarboxylic acid was not indicated therein.

VI. With its response filed on 2 February 2001, the Appellant submitted a new set of Claims 1 to 12 which differed from Claims 1 to 13 as originally filed by the indication in Claim 1 of the method for determining the inherent viscosity and by the deletion of independent Claim 13.

The Appellant argued essentially as follows:

(i) There was no basis for the conclusion that the raw material used in the Examples of D1 had necessarily and inevitably the required purity.

(ii) The feature of using pure starting components was not taught in D1. To the contrary the Examples of D1 clearly showed that the only required measure for obtaining the high molecular weight was the removal of water from the reaction mixture.

(iii) The control of the water content of the polymerisation system, which was an essential feature of the process of D1, was rather complicated, whilst the simple control of the purity of the raw materials was much easier to perform with better reliability and reproducibility.

(iv) Thus, Claims 1 to 12 met the requirements of Articles 54 and 56 EPC.

VII. At the Oral Proceedings held on 6 March 2001, the Appellant submitted a main and an auxiliary request comprising two versions of a new Claim 1 each followed by Claims 2 to 12 as submitted on 2 February 2001.

Claim 1 of the main request reads as follows:

"1. A process for preparing a polyhydroxycarboxylic acid having an inherent viscosity of 0.3 dl/g or more, measured at 20°C at a concentration of 0.1 g solid polymer/100 ml dichloromethane, by dehydrating polycondensation of lactic acid or of lactic acid and another hydroxycarboxylic acid as a raw material in an organic solvent in the presence or absence of a catalyst, while removing the water generated in the reaction by distillation of the solvent, dehydration of the solvent distilled off by treatment with a drying agent or by distillation and returning dehydrated solvent to the reaction system, wherein the content of one or more impurities selected from the group consisting of methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate in the raw material for the polycondensation is 0.3 % by mole or less in total, based on said lactic acid or lactic acid and another hydroxycarboxylic acid."

Claim 1 of the auxiliary request was further limited with respect to the water content of the solvent returned to the reaction mixture.

VIII. The Appellant requested that the decision of the Examining Division be set aside and that a patent be granted on the basis of sets of claims comprising Claim 1 according to the main request submitted in the Oral Proceedings or Claim 1 according to the auxiliary request submitted at the Oral Proceedings.

## Reasons for the Decision

1. The appeal is admissible.

*Main Request*

2. *Admissibility of amendments*

2.1 Claim 1 differs from Claim 1 as originally filed by

(a) the incorporation of the method for determining the inherent viscosity of the obtained polyhydroxycarboxylic acid, and

(b) the incorporation of the feature "while removing the water generated in the reaction by distillation of the solvent, dehydration of the solvent distilled off by treatment with a drying agent or by distillation and returning dehydrated solvent to the reaction system".

For these amendments, support can be found on page 8, lines 13 to 27 and page 9, lines 15 to 25 of the application as originally filed.

2.2 Hence, no objection arises under Article 123(2) EPC in respect of the amendments made, which are consequently admissible.

3. *Clarity*

An independent claim must define all the features essential to carry out the invention. According to the passage starting on page 8, line 13, the water generated during the dehydrating polycondensation must be removed from the reaction system, and inherent viscosity values depend on their measuring conditions (page 9, lines 15 to 25). In view of the amendments in Claim 1 defining these features, the Board is satisfied that Claims 1 to 12 meet the requirements of Article 84 EPC.

4. *Documents*

The two documents which have been considered in the examining procedure can be summarized as follows:

4.1 D1 discloses a process for the preparation of polyhydroxycarboxylic acid (referred to as PHA) comprising conducting dehydrating condensation of a hydroxycarboxylic acid or an oligomer thereof in a reaction mixture containing an organic solvent and in the substantial absence of water (Claim 1). The water generated by the reaction is removed by azeotropic distillation of the solvent from the reaction system, and the dehydrated solvent is returned thereto. The average molecular weight of the PHA obtained is in the range of 15 000 to 200 000, wherein an average molecular weight in the range of about 50 000 to 100 000 or more is said to correspond to an inherent viscosity of from 0.4 to 0.78 dl/g or more (measured at 20°C at a concentration of 0.1 g PHA in 100 ml dichloromethane). Example 3 refers to PHA having an average molecular weight of 100 000 and an inherent viscosity of 0.84. The reaction is preferably carried out at a temperature in the range of 80 to 200, more preferably 110°C to 170°C, in presence of solvents such as diphenyl ether or anisole. Tin catalysts may also be used in the polycondensation reaction. As indicated in D1, the molecular weight of the PHA obtained depends upon the kind and amount of solvent and catalyst used, reaction temperature, reaction time, water content of the solvent in the reaction system (Claims 1, 2, 4, 13 and 14; column 3, line 16 to column 5, line 39; column 5, line 48 to column 6, line 16; column 7, lines 9 to 37; column 8, lines 9 to 14; column 9, lines 27 to 38; Examples 1 to 29 and 31).

4.2 D2 refers to copolymers of lactic acid with glycolic acid, which comprise 60 to 95 percent by weight of lactic acid and 40 to 5 percent by weight of glycolic acid, said copolymers having an inherent viscosity of 0.08 to 0.30 (25 °C, polymer concentration in chloroform: 0.5 g/100 ml) and a molecular weight of 6000 to 35 000. These copolymers are prepared by condensation of the mixture of lactic acid and glycolic acid in the presence of a strong acid ion-exchange resin. Although reference is made to the optional presence of solvents, the reaction is preferably carried out in their absence (i.e. in the melt) and in such a manner that water formed during the polymerization is removed, e.g. by distillation (Claims 1, 9; page 6, lines 7 to 30; page 15, lines 4 to 17, Examples 1 to 7).

The polymers prepared in the examples have inherent viscosities of from 0.12 to 0.20. In the Table (page 20), two 90:10 copolymers are reported to have both an inherent viscosity of 0.20 dl/g and average molecular weights of 21 563 and 30 946, respectively.

## 5. *Novelty*

5.1 As indicated under point 4.1, D1 relates to a process for the manufacture of PHA by dehydrating condensation in the presence of a solvent. The document refers to a number of process features having a decisive influence on the average molecular weight of its products, i.e. kind and amount of solvent and catalyst, reaction temperature, reaction time and treating methods of azeotropically distilled solvent (column 7, lines 30 to 35), but makes no explicit reference to the impurity content of the hydroxycarboxylic acid used as starting components.

5.1.1 As pointed out by the Appellant during the oral proceedings, the characterising feature of the invention under consideration is the limitation of the maximum content of specific impurities in the reaction mixture (ethanol, methanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate) to a very small amount of at most 0.3% by mole, based on the hydroxycarboxylic acids used. D1 is however totally silent with respect to any impurities in the reaction mixture, let alone the presence of the specific compounds defined in Claim 1. Such a limitation is neither explicitly nor implicitly derivable from D1, even though most of its examples disclose the manufacture of PHA having average molecular weights of 50 000 or more, i.e. inherent viscosities of at least 0.40 dl/g (Examples 1 to 29 and 32), in which process the dehydrating polycondensation of lactic acid is carried out in the presence of a solvent, the water generated during the reaction is removed from the reaction system by azeotropic distillation and separation from the solvent, and the dehydrated solvent is returned to the reaction system.

5.1.2 The Board accepts the Appellant's argument that it cannot be concluded from the Examples of D1 that the starting components necessarily and inevitably meet the purity requirement of present Claim 1, because the process conditions used in these examples are not exactly the same as those used in the examples of the present application. In view of the various factors mentioned in D1 which affect the molecular weight of the PHA obtained, the influence of one factor, in particular the impurity content, can only be determined provided all the other parameters remain the same, as is demonstrated e.g. in Example 1 and comparative Example 1 of the present application. This requirement is not fulfilled when comparing examples from D1 directly with those in the application.

5.1.3 It follows that it cannot be deduced beyond any reasonable doubt, that the starting hydroxycarboxylic acids used in the Examples 1 to 29 and 31 of D1 would necessarily and inevitably have exhibited the level of purity as defined in present Claim 1 and, consequently, the novelty of the subject-matter of Claims 1 to 12 over D1 is acknowledged (Article 54 EPC).

5.2 The subject-matter of Claims 1 to 12 of the main request is novel over D2, since this document does neither disclose a process for the production of PHA having an inherent viscosity of at least 0.3 (as measured in the present application) by dehydrating polycondensation of lactic acid optionally together with another hydroxycarboxylic acid in a solvent comprising the return of the dehydrated solvent to the reaction mixture nor to limit the maximum presence of certain compounds in the reaction mixture as required by Claim 1.

5.3 Consequently, the subject-matter of Claims 1 to 12 meets the requirements of Article 54 EPC.

## 6. *Technical problem and its solution*

6.1 The patent application relates to a preparation process of polyhydroxycarboxylic acid.

6.2 Such processes are described in D1 and D2.

6.3 Since document D1 has been published on 8 December 1993, i.e. after the filing date (17 September 1993) of the last of eight Japanese patent applications, the priority of which is claimed in the present application, but before the filing date of the present application (23 December 1993), it is necessary to determine whether the priority claim is valid.

6.3.1 The present application claims priority of the following Japanese patent applications

P1: JP 346329/92, filed on 25 December 1992,

P2: JP 346330/92, filed on 25 December 1992,

P3: JP 77002/93, filed on 2 April 1993,

P4: JP 79219/93, filed on 6 April 1993,

P5: JP 82867/93, filed on 9 April 1993,

P6: JP 229796/93, filed on 16 September 1993,

P7: JP 231526/93, filed on 17 September 1993, and

P8: JP 231527/93, filed on 17 September 1993.

Each of these Japanese patent applications P1 to P8 requires that the amount of one specific impurity in the raw material made up of lactic acid, a mixture of lactic acid and another hydroxycarboxylic acid, or their oligomers does not exceed 0.3 mol % (i.e. P1: methanol, P2: methyl lactate, P3: ethanol, P4: acetic acid, P5: pyruvic acid, P6: fumaric acid, P7: ethyl lactate and P8: butyl lactate).

6.3.2 Present Claim 1 requires that one or more impurities selected from the group consisting of methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate be present in an amount of 0.3% by mole or less in total, based on the lactic acid or the mixture of lactic acid with another hydroxycarboxylic acid.

6.3.3 There is no disclosure in any one of P1 to P8 as to whether the total amount of the impurities selected from the group consisting of methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate should be 0.3% by mole or less in the lactic acid or in the mixture of lactic acid with another hydroxy carboxylic acid. Each of the documents P1 to P8 only limits the content of only one specific impurity but neither excludes the presence of one or more of the other specific impurities nor limits their amount.

6.3.4 The Appellant has emphasised in the oral proceedings that the application does not refer to impurities in general, but that it is essential for the claimed invention that the total amount of the specific impurities does not exceed 0.3% by mole. As shown above each of the priority documents P1 to P8 only refers to the presence of one specific impurity in the starting raw material, but they do not disclose this essential feature of the present invention. It follows that Claim 1 cannot have the benefit of the priorities claimed. The effective filing date is therefore the 23 December 1993. (Article 89 EPC).

6.3.5 Consequently, D1 has to be taken into account for the assessment of inventive step of Claims 1 to 12 , because D1 belongs to the state of the art according to Article 54(1)(2) EPC.

6.4 In fact, the Appellant itself considered D1 to represent the closest state of the art. D1 is the only document concerned with the manufacture of PHA of high molecular weight by dehydrating polycondensation of lactic acid or copolymer thereof with another hydroxycarboxylic acid in a solvent, while removing the water generated by the reaction by distillation of the solvent and returning the dehydrated solvent to the reaction system (cf. point 4.1).

As indicated in D1, the average molecular weight of the resulting polymer in the range of 15000 to 200000, as well as the product yield depend on the specific combinations of process features, including kind and amount of solvent and catalyst, reaction temperature, reaction time and treatment of the solvent distilled off (column 7, lines 30 to 37). This is clearly demonstrated by the examples of D1. Reference can e.g. be made to Examples 1 and 30 or 14 and 31, wherein the water contents have respectively been changed from 3 to 439 ppm and from 1 to 450 ppm. These examples show that in all cases the yield and the molecular weight depend heavily on the water content besides the other factors mentioned above.

6.5 Starting from D1 and as indicated in the description, the technical problem underlying the present application may be seen as providing an alternative process for producing high molecular weight PHA, which is easier to control and does not necessitate a strict monitoring of the water content of the reaction system.

6.6 According to the present application this problem is solved by preparation of PHA by dehydrating polycondensation of lactic acid or of lactic acid and another hydroxycarboxylic acid as a raw material in an organic solvent in the presence or absence of a catalyst, while removing the water generated in the reaction by distillation of the solvent, dehydration of the solvent distilled off by treatment with a drying agent or by distillation and returning dehydrated solvent to the reaction system, wherein the content of one or more specific impurities selected from methanol, ethanol, acetic acid, pyruvic acid, fumaric acid, methyl lactate, ethyl lactate and butyl lactate in the raw material for the polycondensation is 0.3 % by mole or less in total, based on said lactic acid or lactic acid and another hydroxycarboxylic acid.

The limitation of the content of impurities in the raw material for the polycondensation is simple and much easier to perform than a strict control of the water content of the reaction system which continuously changes during the polycondensation step.

6.7 In view of the present Examples and comparative Examples corresponding thereto (cf. in particular Example 1 and comparative Example 1 or Example 58 and comparative Example 10) which demonstrate, whilst being carried out under the same process conditions (temperature, reaction time, solvent..), that a small increase of the amount of specific impurities from 0.3 mol % or less to values slightly above this limit (e.g. 0.31 mol %) results in a significant decrease of the molecular weight of the obtained PHA and the yield, the Board is satisfied that the problem has effectively been solved.

## 7. *Obviousness*

It remains to be decided whether this solution was obvious to a person skilled in the art having regard to the cited prior art.

7.1 D1 discloses the influence of several process factors on the molecular weight of the obtained PHA and stresses the importance of the control of the amount of water in the reaction solvent (column 4, line 39 to column 5, line 5; column 7, lines 8 to 37), but D1 is totally silent on a possible influence of the amount of the specific impurities mentioned in Claim 1 of the present application on the molecular weight of the PHA obtained.

It follows that D1 by itself does not provide any incentive to solve the above technical problem in accordance with Claim 1.

7.2 D2 deals with a totally different problem, i.e. that of avoiding unpredictable degradation of lactic acid/glycolic acid copolymers due to the content of polymerisation catalyst as an impurity (page 2, lines 17 to 27). Hence, there is no indication either in D2 as to whether certain impurities and their content in the raw material may affect the molecular weight of PHA obtained by dehydrating polycondensation in a solvent.

Consequently, D2 does not provide any information either to solve the above technical problem nor any incentive to modify the teaching of D1 in such a way and to arrive at something falling within the terms of Claim 1.

7.3 Even in view of the general knowledge that the impurity content of the starting components should be as low as possible in polycondensation reactions in order to avoid side reactions, the critical effect of the total content of 0.3 mol % or less of the specific impurities (i.e. ethanol, methanol, acetic acid, pyruvic acid, maleic acid, methyl lactate, ethyl lactate and butyl lactate) of the starting raw materials on the molecular weight of the obtained PHA, as demonstrated by the experimental data in the application, could not have been foreseen.

8. It follows that the solution of the technical problem as defined in Claim 1 does not arise in an obvious way from the state of the art relied upon by the Examining Division. Consequently, the subject-matter of Claim 1 involves an inventive step within the meaning of Article 56 EPC.

Claims 2 to 12, which relate to preferred embodiments of the process according to Claim 1, are supported by the patentability of the main claim and thus also allowable.

*Auxiliary request*

9. Since the main request is allowable, it is not necessary for the Board further to consider the auxiliary request.

## Order

### For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of Claim 1 of the main request submitted at the oral proceedings and claims 2 to 12 submitted with the letter of 2 February 2001 and a description yet to be adapted.

The Registrar: The Chairman:

E. Görgmaier A. Däweritz