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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Process for the production of high molecular weight polyester

Klassifikation / Classification / Classement : C08G 63/26

ENTSCHEIDUNG / DECISION

vom / of / du 8 August 1989

Anmelder / Applicant / Demandeur : The Goodyear Tire & Rubber Co.

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step"

Leitsatz / Headnote / Sommaire

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Case Number : T 168/87 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal
of 8 August 1989

Appellant : The Goodyear Tire & Rubber Company
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Decision under appeal : Decision of Examining Division 012 of the European Patent Office dated 5 December 1986 refusing European patent application No. 83 630 012.9 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : K. Jahn
Members : C. Gérardin
G.D. Paterson

Summary of Facts and Submissions

I. The European patent application No. 83 630 012.9 filed on 18 January 1983, claiming priority of 2 February 1982 from the earlier application US 345 172 and published under the publication No. 85 643, was rejected by a decision of the Examining Division dated 5 December 1986.

This decision was based on a set of three claims filed on 27 June 1985 of which Claim 1 reads as follows:

"A method for the continuous solid state polymerization of a prepolymer of a crystalline linear polyester having fiber-forming properties which is composed of by at least 85% by weight terephthalic acid or an ester-forming derivative thereof and a dihydric alcohol in a moving bed reaction zone wherein said crystalline polyester is counter-currently contacted within the reaction zone with a heated inert gas characterized in that said polyester has a moisture content of between 0.005% and 0.01% and in that there is a temperature gradient within the reaction zone which increases the temperature of the polyester in the reaction zone from an initial temperature between 170°C and 180°C to a final temperature between 200°C and 225°C."

II. The ground for this decision was non-compliance with the requirements of Article 56 EPC with regard to the teaching of following documents:

(1) US-A-4 271 287
(2) US-A-4 161 578

which both concern a process for the continuous production of polyesters in which crystallisation of granulated polymer is followed by a solid state polymerisation

reaction. It was generally objected that the only advantage with regard to the stage-wise process disclosed in document (1) was of economical nature and that the combination of features as claimed by the Appellant (Applicant) was obvious in view of the prior art.

More specifically, it was stated that the moisture content of the polyester was known from document (2) and that merely transposing the stage at which humidity was removed from the crystallisation stage, as taught in this document, to the final step as presently claimed, could not be regarded as an inventive feature.

III. A notice of appeal was lodged against this decision on 6 February 1987 with payment of the prescribed fee. The arguments presented in the Statement of Grounds of Appeal filed on 11 April 1987 can be summarised as follows:

The combination of drying, preheating and solid state polymerising a polyester prepolymer in a single moving bed reaction zone overcomes a prejudice, since it was generally admitted that a moisture content higher than 0.005% of the prepolymer caused depolymerisation and/or degradation of the polyester. Further, the temperature gradient mentioned in document (1) is not the zone wherein the solid state polymerisation takes place, but the crystalliser. Last, the prior art documents failed to appreciate the criticality of the initial temperature range in the reaction zone.

IV. The Appellant requests the impugned decision to be set aside and the grant of a patent on the basis of the set of claims filed on 27 June 1985.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. The wording of the claims does not give rise to objections under Article 123(2) EPC.

With regard to the original version, the present formulation of Claim 1 differs basically by the identification of the polyester, the indication of the moisture content and the definition of the initial temperature in the reaction zone by means of a range. More specifically, the fiber-forming properties of the polyester and the starting compounds for the polycondensation reaction, i.e. terephthalic acid or an ester-forming derivative thereof and a glycol, are disclosed on page 1, lines 9 to 14 of the application. The range for the moisture content of the prepolymer is defined from the preferred value of this parameter as upper limit and the conventional value thereof as lower limit, which are both mentioned on page 9, lines 13 to 18. As to the upper limit of 180°C of the initial temperature in the reaction zone, it is to be found on page 15, lines 14 to 19 in connection with a temperature gradient in the reaction zone. Regarding the other amendments, such as the formulation as method claim instead of process claim, they are merely of editorial nature without impact on the scope of the claim.

As far as the characterising features of Claims 2 and 3 are concerned, i.e. the specific polyester and the initial intrinsic viscosity thereof, they derive from original Claim 3.

3. The application in suit relates to a process for the continuous production of polyesters from a crystalline

prepolymer based on terephthalate sub-units. The conventional method to prepare such polymers consists in a step-wise process, as illustrated in document (1); according to the general teaching of Claim 1 thereof, this process comprises

(A) first preheating an amorphous polyethylene terephthalate under forced motion to a temperature between 180 and 200°C without substantial increase in the intrinsic viscosity and while maintaining the crystallinity below 40%;

(B) at least partially crystallising the preheated polymer by feeding it into a fixed bed crystalliser at a temperature between 180 and 200°C, maintaining it in said bed for a time sufficient to raise the crystallisation up to 40 to 60% while maintaining the intrinsic viscosity of the polymer below 0.75, and thereafter removing the latter at a temperature between 195 and 215°C; and

(C) carrying out the polycondensation reaction under forced motion at a temperature between 220 and 245°C until the intrinsic viscosity reaches the value of 0.7 up to 1.4,

steps (B) and (C) being conducted under a non-reactive gas atmosphere.

Although this process leads to polyesters having a satisfactory high molecular weight within acceptable reaction times, the apparatus required to carry out the three steps (A), (B) and (C), namely a forced motion heat transfer unit, a fixed bed type crystalliser and an agitated reactor, was regarded as too cumbersome.

In the light of this prior art disclosure, the problem underlying the application in suit may thus be seen in providing a simplified process without impairing the high molecular weight properties of the polyester or the duration of the preparation of the polymer.

This problem is solved by combining some of the above operative features with a certain moisture content of the prepolymer and specific temperature requirements within the polycondensation reactor. More specifically, the solution claimed by the Appellant involves feeding a polyester with a moisture content of between 0.005 and 0.01% into the moving bed reaction zone wherein there is a temperature gradient which increases the temperature of the polymer from an initial temperature between 170°C and 180°C to a final temperature between 200 and 225°C.

In view of the examples in the application in suit, especially of the comparison of Example 2 thereof with Table III of document (1), the Board is satisfied that the above technical problem is plausibly solved.

4. After examination of the cited documents the Board has reached the conclusion that this technical teaching is not disclosed in any of them and that the subject-matter of the application in suit is, therefore, novel. Since the issue of novelty has not been raised by the Examining Division, it is not necessary to consider this matter in detail.

5. It still remains to be examined whether the subject-matter of the application in suit involves an inventive step with regard to the teaching of the cited documents.

5.1 The differences between the process claimed in the application in suit and the process according to document (1) are such that the specific features of the prior art

process cannot be transposed to the context of the application in suit.

5.1.1 First, the technical meaning of the preheating step (A) in the prior art process should be considered. According to this teaching (column 4, lines 1 to 7), the preheater is any suitable agitated heat transfer unit which can raise the temperature of the amorphous polymer to the specified level without substantial polycondensation and with only a limited amount of crystallisation. It is further specified that the high agitation, high heat transfer apparatus disclosed in US-A-3 425 135 is quite effective for this purpose.

In this reference document, this apparatus is described as a rotary solids processing apparatus for processing solids, ideally suited for drying a solid particulate, especially granular, material by removing a liquid by evaporation on a continuous basis (column 1, lines 13 to 19). According to Examples 1 to 4 and 6, this granular material to be dried is a polymer.

It follows that step (A) in document (A) is basically a drying step; this means that the crystallisation step (B) is carried out on a granulated polyester already dried, unlike in the application in suit.

5.1.2 The critical feature in the prior art process, which was the basis of the whole argumentation of lack of inventive step in the impugned decision, is the existence of a temperature gradient in the crystallisation step (B). It was regarded as obvious to transfer this feature to the final polycondensation stage.

In the fixed bed crystalliser a non-reactive gas is passed in counter-current direction through the bed of gradually descending polymer. The rate of flow and temperature of the

gas is correlated with the polymer temperature, the polymer rate of descent in the fixed bed and the evolved heat of crystallisation to maintain the temperature throughout most of the fixed bed crystalliser in the temperature range of 180 to 215°C. Once crystallisation equilibrium is established, the considerable heat of crystallisation is carried by the non-reactive gas upward through the crystalliser to heat incoming and descending polymer, thereby substantially increasing the energy efficiency of the operation through utilisation of this evolved heat. This results in a temperature gradient of about 15°C from one end of the fixed bed to the other with the temperature increasing as the product descends through the crystalliser (column 3, lines 4 to 16 and column 4, lines 45 to 50).

In the prior art, thus, the temperature gradient occurs once the heat exchange equilibrium within the fixed bed crystalliser has been reached.

5.1.3 The differences between these steps of the process disclosed in document (1) and the corresponding features of the process according to the patent in suit are such that the prior art teaching cannot lead to the solution claimed by the Appellant.

First of all, nothing in document (1) would suggest to reverse the order of steps (A) and (B), i.e. to carry out the crystallisation step on a non-dried polymer like in the application in suit, in order to reduce the apparatus equipment. Further, the mere existence of a temperature gradient within the fixed bed crystalliser in the prior art cannot be regarded as an incentive to adopt the same feature inside the highly agitated polycondensation reactor as required in the application in suit, wherein it measures the difference between the temperature at which the crystalline prepolymer is introduced into the reactor

and the temperature at which it is polycondensed. Moreover, from a purely quantitative point of view, nothing would suggest to increase the known temperature gradient of 15°C to a range wherein the difference between the final temperature and the initial temperature is at least 20°C and extends up to 55°C.

5.2 The combination of operative features according to the application in suit cannot be derived from document (2) either.

The process described therein comprises first crystallising the polyester prepolymer at a temperature of 180 to 220°C under forced motion to a density of at least 1.385 g/cm^3 and a moisture content of less than 0.02 percent by weight, then continuously introducing the crystallised granulate to a continuous fixed bed reactor wherein polycondensation occurs at 210 to 235°C (column 2, lines 5 to 21). It follows, as conceded in the Statement of Grounds of Appeal, that a prepolymer having a moisture content of less than 0.02 percent by weight, which encompasses the range envisaged by the Appellant for this parameter, would not cause hydrolytic degradation and/or depolymerisation during the final stage. However, in this case, the skilled man would carry out the final stage in a fixed bed reactor instead of the moving bed reaction zone required in the application in suit. Moreover, the crystallised polymer would be discharged to the reactor according to a typical embodiment at a temperature of at least 210°C (column 4, lines 8 to 12) which cannot lead to the temperature features of the temperature gradient required in Claim 1.

5.3 Nor could a combination of documents (1) and (2) suggest the subject-matter of the application in suit.

Whereas the process according to document (1) can be schematically represented as a crystallisation step in a fixed bed crystalliser followed by a polycondensation step in an agitated reactor, the process according to document (2) comprises first crystallising under forced motion, then polycondensing in a fixed bed reactor. The two basic steps are thus carried out under different conditions.

In fact, as specified in both document (1) (column 1, lines 25 to 45 and column 2, lines 3 to 17) and document (2) (column 1, lines 16 to 68), these two processes should be regarded as different alternatives to overcome the problem of stickiness and agglomeration of the granulate polymer during solid phase polycondensation by carrying out, unlike previous attempts, the latter reaction at a higher temperature than crystallisation. For this reason, individual features of one process cannot be transposed as such to the other.

5.4 The comparison of intrinsic viscosity and residence time in document (1) and in the application in suit shows that the process presently claimed enables to prepare crystalline polyesters without detrimental effect on the molecular weight and the reaction time.

Table III of document (1) shows that polymers having an intrinsic viscosity between 0.7 and 0.85 can be obtained when the reactor temperature is in the range of 225 to 230°C with a residence time of four hours (Examples 1 to 4) and that viscosities as high as 1.4 can be obtained when the reactor temperature is between 220 and 245°C and the polymer residence time up to ten hours (Operative Ranges). These results are very similar to those indicated in Example 2 of the application in suit which was run to demonstrate the effect of prepolymer moisture content on solid state polymerisation; according to Figure 5, which

represents the intrinsic viscosity build-up curves for prepolymers with various moisture contents, viscosities of 0.7 can be obtained after four hours at 220°C; likewise, it appears from Figure 6 that, depending on the prepolymer moisture content, the solid state polymerisation time requirement varies broadly from five to nearly nine hours.

That such results, which are identical to a large extent, can be obtained by simplified operative means speaks in favour of an inventive step.

5.5 In conclusion, for the various reasons mentioned above, the subject-matter of the application in suit has to be regarded as involving an inventive step.

6. Claim 1 being allowable, the same applies to Claims 2 and 3 which merely represent preferred embodiments of Claim 1 and whose patentability is supported by that of the main claim.

Order

For these reasons, it is decided that:

1. The impugned decision is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 3 filed on 27 June 1985.

The Registrar: The Chairman:

F. Klein K. Jahn

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