A		B		C	X

File Number: T 341/90 - 3.3.3

Application No.: 83 304 492.8

Publication No.: 0 104 731

Title of invention: 3-Hydroxybutyrate polymers

Classification: D01F 6/62

DECISION
of 22 January 1993

Applicant: IMPERIAL CHEMICAL INDUSTRIES PLC

Opponent: Petrochemie Danubia Gesellschaft m.b.H.

Headword:

EPC Article 56

Keyword: "Inventive step (affirmed) - application of conventional processing technique neither obvious in fact nor feasible in practice"

Case Number : T 341/90 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 22 January 1993

Appellant : (Opponent)	Petrochemie Danubia Gesellschaft m.b.H. Danubiastr. 21-25 A - 2323 Schwechat-Mannswörth (AT)
Representative :	Kunz, Ekkerhard, Dr. c/o Patentwesen der Chemie Linz GmbH St. Peter-Strasse 25 A - 4021 Linz (AT)
Respondent : (Proprietor of the patent)	IMPERIAL CHEMICAL INDUSTRIES PLC Imperial Chemical House Millbank London SW1P 3JF (GB)
Representative :	Locke, Timothy John ICI Group Patents Services Dept. PO BOX 6 Shire Park Bessemer Road Welwyn Garden City Herts AL7 1HD (GB)
Decision under appeal :	Decision of the Opposition Division of the European Patent Office of 6 February 1990, issued on 27 February 1990 rejecting the opposition filed against European patent No. 0 104 731 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : F. Antony
Members : R. Young
F. Benussi

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 104 731 in respect of European patent application No. 83 304 492.8 filed on 3 August 1983 and claiming a GB priority of 27 August 1982 (GB-8 224 683) was published on 25 November 1987 (cf. Bulletin 87/48).

Claim 1 as granted reads as follows:

"A process for the production of an oriented shaped article from a polymer containing at least 40 mol% of 3-hydroxybutyrate residues in the polymer chain comprising uni- or bi-axially drawing a preform of said hydroxybutyrate polymer in a partially crystalline state at a temperature between the glass transition temperature Tg, and the melting point of the hydroxybutyrate polymer into said shaped article, and then increasing the degree of crystallinity of the hydroxybutyrate polymer by subjecting the drawn preform, while in the drawn condition, to a temperature between the glass transition temperature and the drawing temperature until the drawn article is non-rubbery, the amount of said uni- or bi-axial drawing being such that, after said step of increasing the degree of crystallinity, at least one dimension of the preform has increased by at least 200%, said hydroxybutyrate polymer having a degree of crystallinity, at the time of drawing, such that the preform is not so brittle that it breaks before achieving said increase in said at least one dimension, and such that the drawn article does not break during the step of increasing the degree of crystallinity.".

The remaining Claims 2 to 8 are directed to further elaborations of the above process.

II. Notice of Opposition was filed on 25 August 1988 on the grounds of lack of novelty and lack of inventive step. The Opposition was supported by the documents:

D1: DE-A-2 519 075;
D2: DE-A-2 328 639;
D3: DE-A-2 722 087;
D4: DE-A-2 061 418;
D5: DE-A-1 504 251;
D6: US-A-2 586 820;
D7: "Modern Plastics International", June 1981, page 6, and the later filed, but admitted
D8: Chemistry and Industry, 7 August 1982, pages 508 to 511.

III. By a decision which was given at the end of oral proceedings held on 6 February 1990 and issued in writing on 27 February 1990 the Opposition Division rejected the opposition.

According to the decision, it was held that the claimed subject-matter was novel and inventive. D1 to D6 related to processes for producing oriented shaped articles from thermoplastic polymers such as polypropylene (PP), but none of them mentioned poly-3-hydroxybutyrate (PHB) as a possible working material; there were a number of differences between PHB and PP in intrinsic properties such as the flexural modulus and extension to break percentage as represented by the data in D7 and in D8 as well as in the behaviour under stress and the rate of crystallisation; and none of the available prior art documents suggested the claimed combination of steps by which the resulting problem of finding process conditions which would allow the preparation of uni- or bi-axially drawn PHB articles was solved.

IV. On 26 April 1990 a Notice of Appeal against the above decision was filed, together with payment of the prescribed fee.

In the Grounds of Appeal filed on 5 July 1990, the Appellant (Opponent) argued that the assessment of the differences in properties on the basis of the data in D7 and D8 gave a false picture, and that drawing processes known to be suitable for PP could also be applied to the processing of PHB with the same success. In any case the acknowledged fact that the degree of crystallisation could not be determined during the claimed process meant that this parameter was superfluous and the skilled person, in experimenting with the parameters conventional in PP processing, would in practice only have required routine tests to arrive at the optimal conditions for processing PHB.

With the Grounds of Appeal, the Appellant filed further documents in support of his case, as follows:

D9: Decision of Opposition Division revoking patent No. 0 078 609 concerning producing a shaped article of PHB by subjecting a solvent-free preform of the polymer to compression;

D10: "Die Pharmazie", publication of Humboldt University, 44 (8) 545 (1989);

D11: "Kunststoffe", Vol. 51, 1961, page 183, Figure 2; a plot of fraction of crystallinity of polyethylene as a function of temperature, and

D12: "Journal of Polymer Science", Vol. 58, 1962, page 1098, Figure 4; a plot of fraction of crystallinity of isotactic polypropylene as a function of temperature;

and with a letter dated 22 December 1992 still further documents:

D13: "Propylene", H. P. Frank, published by Gordon & Breach Science Publishers, (New York), 1968, pages 110 to 111, and

D14: Test of "Daplen FM 553" polymer according to DIN 53455, dated 11 November 1991.

V. The Respondent disagreed with the above arguments regarding D7 and D8 and cited a further document (which will be referred to as the "King" article, and not by number). He pointed out furthermore that the rate of crystallisation, and the variation of that rate with temperature, was one property in which PP and PHB critically differed and which underlay the invention; none of the documents cited, however, taught the need to control the time of treatment to get the PHB-peculiar extent of pre-crystallisation (see letter dated 11 May 1992, pages 1, 2, paragraphs 4 to 7).

The Respondent was also opposed to the introduction of the newly filed documents D9 to D14 of the Appellant.

VI. The Appellant requests that the decision under appeal be set aside and the patent in suit be revoked in its entirety.

The Respondent requests that the appeal be dismissed and the patent be maintained as granted (main request) or on the basis of any of the three auxiliary requests submitted in writing on 20 January 1993.

VII. Oral proceedings were held before the Board on 22 January 1993. At the proceedings the Board informed the parties that the newly filed documents D9 and D13 would be admitted. The documents D10, D11, D12 and D14 as well as the King article were, however, excluded from the proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. There are no formal objections under Article 123(2) and (3) EPC to the claims of the main request since the latter correspond to the claims as filed.

3. <u>Closest state of the art: the technical problem and its solution</u>

3.1 The patent in suit is concerned with providing a degree of (molecular) orientation to shaped articles made of a particular class of polymers - i.e. those containing at least 40 mol%, preferably at least 80 mol% of 3-hydroxybutyrate residues ("PHB"). Molecular orientation is desirable in the production of some shaped articles, such as fibres, ribbons, films, and bottles.

Although conventional orientation techniques involved mono- or bi-axially drawing polymers either (a) just above the glass transition temperature (Tg), used for polyesters and polyamides, or (b) just below the crystalline melting point (Tm), used for olefin polymers, e.g. PP, problems of breakage and lack of reproducibility arose when these techniques were applied to PHB (patent in suit, page 2, lines 45 to 58).

3.2 Of the documents cited by the Appellant and forming part of the proceedings, D1 to D6 and D13 concern various techniques of drawing conventional olefin polymers, especially PP, but make no mention at all of PHB, whereas D7 and D8, whilst mentioning PHB, contain no reference to specific drawing techniques.

3.2.1 According to D13, which is regarded as more pertinent than any of D1 to D6, PP filaments are melt spun from a round die with 60 to 80 orifices, at a temperature of 260° to 290°C., fed downwards into a quench tank (water filled) and from there to a draw oven (125 to 160°C); to an annealing oven (95 to 120°C), and finally to a wind up station (see Figure 6.7). A draw ratio of 9/1 may be applied.

3.2.2 According to D8, which relates to ICI's work on PHB and is slightly more comprehensive than D7, the problem with PHB was that it had no particular mechanical property advantages to offer; it was broadly similar to PP and polyethyleneterephthalate (page 510, left column). Reference was made in this connection to a Table of comparisons between PP and PHB (page 511, left column). The Table is reproduced below.

Table 2 PHB compared with propylene homopolymer (PP)

	PHB	PP
Crystalline melting point (°C)	175	176
Crystallinity (per cent)	80	70
Molecular weight (Daltons)	$5x10^5$	$2x10^5$
Glass transition temperature (°C)	15	-10
Density (g/cm^3)	1.250	0.905
Flexural modulus (GPa)	4.0	1.7
Tensile strength (MPa)	40	38
Extension to break (per cent)	6	400
Ultraviolet resistance	good	poor
Solvent resistance	poor	good

3.2.3 D9 is a reference to an EPO decision which does not itself form prior art. The Board does not feel bound to follow this decision, which not only originated from a lower instance, but also relates to subject-matter which is different from that claimed in the patent in suit.

It thus appears to the Board that the closest prior art, in the sense of the most suitable starting point for the skilled person concerned with the orientation of PHB, is not any of the above citations, but that corresponding to technique (b) referred to in section 3.1 above.

The argument of the Appellant that the description of technique (b) in the patent in suit was inaccurate in that PP was conventionally drawn at a temperature clearly below, rather than below, but near to, the crystalline melting point Tm (see Grounds of Appeal, page 6, last paragraph) is literally correct, but does not make any practical difference in the opinion of the Board. On the basis of the figures given in the submission (melting point of PP = 176°C; draw temperature = 125° to 160°C.) the average draw temperature is about 80% of the melting temperature. There can thus be no confusion with technique (a) where drawing takes place close to the glass transition temperature Tg (see patent in suit, page 2, lines 48 to 50), in the case of PP corresponding to a temperature of -10°C. (see D8; Table 2).

3.3 The problem arising from the state of the art represented by technique (b) would then seem to be the provision of a practically feasible method for the orientation of PHB preforms.

3.4 The solution, according to Claim 1 of the patent in suit, involves modifying the technique (b) by controlling the degree of crystallinity of the PHB at the time of drawing, so that the PHB is in a partially crystalline state in which it is not so brittle as to break, and subsequently increasing the degree of crystallinity until the drawn article is non-rubbery and yet does not break during the latter step, the amount of drawing being such that, after the step of increasing the degree of crystallisation, at least one dimension of the preform has increased by at least 200%.

3.5 It is credible that the claimed process effectively solves the technical problem arising, as can be seen from the results, in terms of draw ratio, of melt extruding, quenching/crystallising, drawing and further crystallising PHB in the apparatus illustrated in the patent in suit, (cf. Table on page 4).

4. Novelty

The claimed solution is novel, for the reasons given under section 3.2 above. The novelty of the claimed subject-matter was in any case not in dispute between the parties at the appeal stage.

5. Inventive Step

5.1 In deciding whether the claimed solution involves an inventive step, it is relevant to consider, on the one hand, processes common for the orientation of PP as represented by D13, and, on the other hand, a comparison of the properties of PHB and PP such as is given in D8.

5.2 Would the notional skilled person, confronted with the technical problem arising, have turned to the claimed solution or not?

5.3 From a consideration of the properties of PHB and PP compared in the Table referred to in D8 (see section 3.2.2 above) it can be seen that, whereas some intrinsic properties, such as the melting point, the tensile strength and even the degree of crystallisation (at equilibrium), are quite similar, other properties, such as flexural modulus, molecular weight, and extension to break are quite different.

5.3.1 The Appellant's objection that the data in the Table related to already shaped articles and that parameters such as flexural modulus and extension to break thereof were themselves dependent on the way the polymer had been processed is unconvincing in the absence of more specific evidence (which it was the onus of the Appellant to provide in sufficient time) and would in any case apply equally to both types of polymer (see Grounds of Appeal, page 3, last paragraph). The broader implication behind such an argument, that no significance at all can be attached to such data, is unacceptable because it is at variance with the Appellant's own reliance, as expressed at the oral proceedings, on the "general structure" of the Table to support his assertion of a strong similarity in properties between PHB and PP in the first place.

On the contrary, it can be said that the skilled person would deduce from the Table, consistently with the commentary in D8, that PHB is "broadly similar" to PP, in that it has (as stated in D7), "several properties similar to" those of PP. The skilled person would not in this connection read more into this value judgment, regardless of its source, than what was objectively justified by the factual content of the Table itself. That PHB is not similar in all respects to PP is self evident to the skilled person reading the Table. It is furthermore clear that precisely those parameters in which PHB is least similar to PP happen to be those which are most important in assessing the capability of the polymer of being drawn. The most crucial of these is the extension to break percentage. This is given as 6% in the case of PHB and 400% in the case of PP.

5.4 It must be borne in mind in this connection that it is a requirement of the method claimed in the patent in suit that "the amount of drawing is such that ... at least one dimension of the preform has increased by at least 200%" (cf. Claim 1).

5.5 The skilled person would consequently deduce that, although one might expect to be able to draw PP, with an extension to break of 400%, to the extent required by Claim 1 of the patent in suit, there was no likelihood of being able apply the same treatment to PHB, with an extension to break of only 6%. The same conclusion would still apply, in the Board's view, even if the two values had differed by only one order of magnitude, instead of by almost two.

By reason of this knowledge alone, therefore, it would not have been obvious to the skilled person to apply a conventional drawing technique used for PP to solve the technical problem.

5.6 If, however, the skilled person had, notwithstanding the above, nevertheless chosen to apply a conventional PP processing technique to the orientation of PHB, the question arises as to what might in practice have been the result, and, in particular, whether the technical problem would thereby have been solved.

5.6.1 It is true that D13 discloses an apparatus for drawing PP analogous in many respects to that used for PHB in the preferred embodiment of the patent in suit. Certain of the process parameters used in drawing PHB in the apparatus of the patent in suit are also somewhat similar to those disclosed in D13 for PP, e.g. the draw temperature (120°C, compared with 125° to 160°C for PP) and the annealing temperature (60°C, compared with 95° to 120°C for PP), although the extrusion temperature is much lower (185°C compared with 260° to 290°C for PP) and the draw ratio falls short of that disclosed in D13 (maximum 8 compared with 9/1 commonly applied for PP). The document is, however, completely silent as to the temperature in the quench tank, and as to the processing time for the PP filaments.

Thus there is no information given in D13 which is relevant to the possibility of controlling the degree of crystallisation with time, let alone a pointer to the desirability of such control.

5.7 Such control is, however, necessary for the solution of the technical problem, because PHB in its fully developed (80%) crystalline state is a stiff, brittle material which can only be drawn to a very small extent before it breaks (cf. submission of Respondent, dated 6 February 1989, page 2). The brittle nature of PHB in its state of equilibrium crystallisation was incidentally confirmed by tactile examination of a sample of PHB supplied by the Respondent at the oral proceedings, and contrasted with the compliant nature of a sample of PP which was also supplied.

5.7.1 In particular:

- if, at the time of drawing, the PHB is too highly crystallised, then the preform can only be drawn to a very small extent before it breaks, and

- if, at the time of drawing, the PHB has a too low degree of crystallinity, then the preform is rubbery and, on release of the drawing force, the product relaxes on release of the drawing force, or even breaks if held under tension while further crystallisation occurs (cf. patent in suit, page 2, line 61 to page 3, line 4).

5.7.2 The argument of the Appellant that the degree of crystallisation was a parameter which was superfluous on account of the difficulty of measuring it during processing is illogical. The difficulty of continuously measuring a particular parameter in no way prevents that parameter from being a critical variable in the operation of a process. Conversely, it may well be possible effectively to control such a variable without the necessity of continuously obtaining measured values.

5.7.3 The assertion of the Appellant in this respect, however, that adjustment of the conventional processing parameters (e.g. the rotation speed of the draw rollers and the processing temperature), until the polymer was neither "rubbery" nor such that the drawn part would break, would result in a solution of the technical problem was not supported by any evidence (see submission of 22 December 1992, especially page 4, last paragraph to page 6). In particular was never alleged, let alone proved, that anyone had ever tried to put a sample of PHB on to a conventional PP processing line.

5.8 As to what would have happened in practice had the skilled person applied the PP processing technique of D13, for instance, to PHB, it is relevant to consider the information in items 3.2 and 3.3 of the minutes of the oral proceedings held before the Opposition Division.

These items made reference to certain slides and figures, of which the graph of Figure 7 (which was again referred to and discussed during the oral proceedings before the Board) in particular was of interest. Inspection of this graph showed, in a plot of time to reach 50% crystallisation ($T_{1/2}$, seconds) against temperature (°C.), that PP and PHB behave very differently from each other. Whereas with reducing temperature the crystallisation half time of PP declines rapidly and somewhat asymptotically to an extremely low value ($T_{1/2}$ is around 2 seconds for all temperatures below about 70°C), the corresponding values for PHB rapidly decrease to a minimum at around 50 to 70°C. and then equally rapidly increase as the temperature is lowered still further, giving a 'U' shaped curve in this region. The minimum value of the half crystallisation time on this curve is, however, about 72 seconds, i.e. at least 30 times greater than that of PP, in the same temperature region.

5.8.1 Without actually contesting the above figures, the objection of the Appellant that the values of $T_{1/2}$ varied with the quenching conditions chosen, so that it was doubtful what the skilled person would derive from such data, was not supported by any specific evidence and is thus unconvincing to the Board for reasons analogous to those set out under section 5.3.1 above, first sentence.

5.8.2 The further objection, that the time to full crystallisation was not given in Figure 7, but in the case of PP could be several months, so that the curves did not reflect speed of crystallisation at all, but rather a parameter referred to as "crystallisability" ("Kristallisierbarkeit"), does not in the Board's view detract from the qualitative message of Figure 7 concerning the difference in crystallisation behaviour with time between PP and PHB.

In this connection, and apart from the fact that the significance of the "crystallisability" parameter was not explained, there was no suggestion from the Appellant that thermoplastic polymers such as PP shared with PHB the important characteristic of a narrowly defined minimum in the crystallisation $T_{1/2}$/temperature curve.

It is therefore evident that there existed a "processing window", i.e. a narrowly defined set of processing conditions peculiar to PHB, under which its degree of crystallisation could be adjusted to a level which would enable the technical problem to be solved.

5.9 The Appellant admitted at the oral proceedings before the Board, that although it might be said in principle that there was a "processing window" for every polymer, nevertheless access to such a window was not in all cases in practice technically feasible, and furthermore that the "window" for the processing of PHB itself was substantially smaller than any "window" which might be represented by the conditions appropriate to processing PP, the latter being described by the Respondent without contradiction as "a very forgiving material".

This implies that there would be very little likelihood that trial and error experimentation with unrelated conventional parameters such as draw roller speed and drawing temperature - even with the latter in the claimed region between the glass transition temperature and the melting point - would have resulted in this window being found; the more so since the state of the art was completely silent on the necessity of controlling the relevant parameter (degree of crystallisation with time) and therefore on the existence of such a window.

5.10 On the contrary, according to the uncontested and convincing argument of the Respondent, the skilled polymer processing engineer will in general wish to minimise the number of variables which must be continuously controlled during the processing of a polymer. For this reason he will choose the process conditions so that as many of the polymer parameters as possible are at equilibrium. In particular he would choose, in the case of the conventional processing of PP, that the degree of crystallisation was at equilibrium, i.e. substantially complete. If he were to follow this principle in applying the PP processing technique to PHB, as canvassed by the Appellant, then it would be logical to ensure that the PHB was in its fully (equilibrium) crystallised state before starting. This would by the same token ensure that the processing window for PHB was never approached, let alone traversed.

In other words, the application of the conventional technique would, for the reasons given above, deprive the process of success.

5.11 The remainder of the prior art cited is more remote and in particular does not give any pointer to the solution of the technical problem.

6. In summary, therefore, the application of a conventional PP processing technique successfully to draw PHB was neither obvious in fact, nor feasible in practice. Rather, it was the initiative of the Respondent to have succeeded

in determining the conditions allowing the technical problem to be solved.

The subject-matter of Claim 1 therefore involves an inventive step.

The remaining Claims 2 to 8 being directly or indirectly dependent on Claim 1 by same token also relate to subject-matter which is both novel and inventive.

7. This being the case, it is not necessary to consider further the auxiliary requests of the Respondent.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier F. Antony