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D E C I S I O N
of 5 January 1998

Case Number: T 0178/93 - 3.3.3

Application Number: 86116712.0

Publication Number: 0226112

IPC: C08F 210/08

Language of the proceedings: EN

Title of invention:
Highly amorphous olefin terpolymer

Patentee:
EASTMAN CHEMICAL COMPANY

Opponent:
Hüls Aktiengesellschaft

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes) - non-obvious combination of known features"

Decisions cited:
-

Catchword:
-

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Case Number: T 0178/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 5 January 1998

Appellant: (Opponent)
Hüls Aktiengesellschaft
Patentabteilung/PB 15
D-45764 Marl (DE)

Representative:
-

Respondent: (Proprietor of the patent)
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 23 December 1992 concerning maintenance of European patent No. 0 226 112 in amended form.

Composition of the Board:

Chairman: C. R. J. Gérardin
Members: H. H. R. Fessel
A. C. G. Lindqvist

Summary of Facts and Submissions

I. European patent No. 0 226 112 in respect of European patent application 86 116 712.0 filed on 2 December 1986 in the name of Eastman Kodak Company was granted on 6 June 1990 (cf. Bulletin 90/23) on the basis of a set of 3 claims. Claim 1 was directed to an amorphous terpolymer comprising repeating units from about 0.1-10% b.w. ethylene, about 50-78% b.w. propylene and about 12-63% b.w. butene-1, said terpolymer showing a singlet at about 730 cm-', when examined in the solid state by IR spectrum analysis, being greater than 99% soluble in boiling hexane, having a melt viscosity when measured by the Termosel method of about 0.1 to about 100 Pa s (100 to about 100,000 cp) at 190°C, a ring and ball softening point (RBSP) of about 75°C to about 120°C and a Tg of about -20°C to about -35°C.

Claim 2 concerned a process and read as follows:

"A process for producing a highly amorphous olefin terpolymer according to Claim 1 comprising continuously polymerising a mixture of about 0.1-10% by weight ethylene, about 50-78% by weight propylene and about 12-63% by weight butene-1, in the presence of a catalyst comprising an anionic coordination catalyst and in a solvent medium, at a temperature of about 140°C-250°C and at a pressure of about 689-34,450 kPa (100-5,000 psig) (7-351.6 kg/cm^2)."

Dependent Claim 3 related to a preferred embodiment of the subject-matter of Claim 2.

II. A Notice of Opposition was filed by Hüls AG on 28 December 1990. In its Statement of Grounds of Opposition filed on 5 March 1991 the Opponent alleged lack of novelty of the subject-matter claimed in Claim 1 and considered the subject-matter of Claims 2 and 3 not to involve any inventive step.

These objections were based essentially on:

D1: EP-A-0 023 249 (equivalent to US-A-4 309 522, acknowledged on page 2, line 18 of the patent in suit) and

D2: US-A-3 923 758 (acknowledged on page 2, line 9 of the patent in suit).

III. By its interlocutory decision of 15 September 1992 issued in writing on 23 December 1992, the Opposition Division held that the grounds of opposition did not prejudice the maintenance of the patent in amended form on the basis of a set of 2 claims filed on 19 November 1992 of which the independent Claim 1 reads as follows:

"A process for producing a highly amorphous olefin terpolymer comprising repeating units from about 0.1-10% by weight ethylene, about 50-78% by weight propylene and about 12-63% by weight butene-1, said terpolymer showing a singlet at about 730 cm^{-1}, when examined in the solid state by infrared spectrum analysis, being greater than 99 percent soluble in boiling hexane, having a melt viscosity when measured by the Termosel method of about 0.1 to about 100 Pa.s (100 to about 100,000 cp) at 190°C, a ring and ball softening point of about 75°C to about 120°C and a glass transition temperature of about -20°C to about -35°C, comprising continuously polymerising a mixture of about 0.1-10% by weight ethylene, about 50-78% by weight propylene and about 12-63% by weight butene-1,

in the presence of a catalyst comprising an anionic coordination catalyst and in a solvent medium, at a temperature of about 140°C-250°C and at a pressure of about 689-34,450 kPa (100-5,000 psig) (7-351.6 kg/cm^2)."

Claim 2 relates to a preferred embodiment of the subject-matter of Claim 1.

In substance, the Opposition Division took the view that D2, which described a continuous polymerisation process of propylene and butene-1 involving the same process features as required in the patent in suit, was the closest state of the art. Although a terpolymer with additional ethylene units was already known from D1, a skilled person could not have expected an improvement of ring and ball softening point (RBSP) by preparing such terpolymer with the process disclosed in D2.

IV. On 16 February 1993 an appeal was lodged by the Appellant (Opponent) together with payment of the prescribed fee. The Statement of Grounds of Appeal was received on 17 April 1993. The arguments presented in that statement can be summarised as follows:

(i) The Opposition Division considered D2 to represent the most relevant prior art. That document taught the preparation of a propylene/butene-1 copolymer under the same conditions (temperature, catalyst, pressure and continuous polymerisation) as defined in Claim 1. The problem underlying the patent in suit was considered to be the definition of a process for preparing a substantially amorphous propylene/butene-1/ethylene terpolymer having a singlet at about 730 cm^{-1} instead of a doublet and an improved RBSP compared with the propylene/butene-1/ethylene terpolymer obtainable

with the continuous process of D1. Although the latter citation taught to use rather low reaction temperatures during the copolymerisation of ethylene, a skilled person would have also considered higher temperatures in view of their beneficial effect on stereospecifity.

(ii) If on the contrary one regarded D1 as the closest state of the art, one would reach the same conclusion, since nothing spoke against the use of the polymerisation conditions known from D2 for the polymerisation of the monomer combination known from D1.

V. The Respondent (Proprietor), now by transfer of rights Eastman Chemical Company (cf. letter of 23 August 1994), only acknowledged the EPO notification enclosing the Statement of Grounds of Appeal , but did not provide any arguments (cf. reply of 30 August 1994).

VI. The Appellant requested that the decision under appeal be set aside and the patent be revoked entirely.

The Respondent requested that the appeal be dismissed, alternatively oral proceedings be arranged.

Reasons for the Decision

1. The appeal is admissible.

2. The claimed subject-matter meets the requirement of Article 123(2) and (3) EPC since Claim 1 combines the product features of Claim 1 with the process features of Claim 2, both as granted. Claim 1 as granted resulted from the combination of Claims 1 and 2 as originally filed and the incorporation of the range of

glass transition temperatures mentioned on page 4, lines 28/29 of the original application; Claim 2 as granted corresponded to Claim 6 as originally filed. As to present Claim 2, it corresponds to Claim 3 as granted which is identical with Claim 7 as originally filed.

3. Novelty of the claimed subject-matter was not under dispute.

4. *Inventive step*

4.1 Since the Statement of Grounds of Appeal considers D1 and D2 as possibly representing the closest state of the art, the teaching of these citations must be discussed first.

4.1.1 D1 relates to a process for producing amorphous propylene/butene-1/ethylene terpolymers which are completely soluble in boiling heptane, which have a RBSP of 90 to 130°C and a melt viscosity of 1,000 to 100,000 mPa.s at 190°C (cf. page 4, lines 29 to 36). According to the most preferred embodiment a mixture of e.g. 0.5-10% b.w. ethylene, 40-60% b.w. propylene and 32-40% b.w. butene-1 is continuously or discontinuously polymerised at temperatures of 50 to 100°C and at pressures of 5 to 20 bars (cf. page 3, lines 1 to 8 and 18/19; page 4, lines 25 to 27). As correctly acknowledged on page 2, lines 18 to 28 of the patent specification this process is very similar to that of D2 with the exception of temperature and pressure.

4.1.2 D2 describes a process for preparing an amorphous propylene/butene-1 copolymer under the same process conditions (T = 140-250°C, p = atmospheric to about 2,000 psig, catalyst, cf. claim 1) as required in the patent in suit. The copolymers are produced in a continuous manner.

When a batch process is used the polymers are less random at a given butene-1 content as e.g. demonstrated by a RBSP of 113°C instead of 103°C (cf. column 4, line 60 - column 5, line 24). In general, the melt viscosity of the copolymers is 100 to about 100,000 cp at 190°C (column 5, lines 39 to 43), the solubility in boiling hexane greater than 99 weight % (column 2, lines 33 to 35) and the RBSP is 78-120°C (Table II). That document too is already acknowledged as prior art on page 2, lines 9 to 17 of the patent in suit.

4.1.3 Since the patent in suit is directed to a process for the preparation of a specific terpolymer, it is evident that the closest state of the art should be another process giving rise to that kind of product, e.g. D1.

4.2 The question of the definition of the technical problem underlying the patent in suit, e.g. either in positive or in alternative terms, arises if one considers the experimental evidence provided by the Patentee during opposition proceedings (reply of 1 October 1991).

4.2.1 In the absence of an exact reworking of Example 2 of D1 by the Opponent, involving exactly the same monomer composition and the same polymerisation conditions, a deficiency which the Opponent did not dispute (statement of 13 December 1991, paragraph bridging pages 2/3), it would be correct to rely on the results of the test report provided by the Patentee which represents the only truly comparative data in the file.

4.2.2 According to that submission (paragraph bridging pages 8 and 9), although it is customary to distinguish amorphous fractions from crystalline fractions on the basis of a solubility test in boiling hexane, the solubility test based on ether extraction is a more accurate method to differentiate between predominantly amorphous olefin polymers. In practice, thus, extraction with refluxing ether removes the truly amorphous fraction of the terpolymer, leaving as insoluble residue a partially crystalline material which would be normally soluble in refluxing hexane.

4.2.3 From the test report submitted by the Patentee it appears that a terpolymer obtained by the process as claimed is more amorphous than the corresponding terpolymer obtained by the process according to D1. Structurally this means that the bulk of ethylene is in the soluble fraction, where it has been effectively utilized to disrupt crystallinity, in other words that ethylene is more randomly distributed throughout the terpolymer chains (page 9, third full paragraph).

4.2.4 On that basis it would thus be legitimate to define the technical problem underlying the patent in suit in positive terms, namely the definition of a process giving rise to more amorphous terpolymers.

4.2.5 However, in view of the arguments in the Statement of Grounds of Appeal (in particular the passage bridging the pages 3/4), from which it would follow that the terpolymers according to the patent in suit and the terpolymers according to D1 could not be distinguished on the basis of their RBSP, the Board will also define the technical problem in alternative terms.

4.3 The problem to be solved may thus be seen in the definition of an alternative process to that of D1.

4.4 According to the patent in suit this problem is said to be solved by a continuous polymerisation process at a temperature of about 140°C - 250°C, as specified in Claim 1.

4.5 In view of the description, the examples and the evidence provided during Opposition Proceedings the Board is satisfied that the above problem is effectively solved with the given means.

5. It remains to be decided if this solution is obvious to a person skilled in the art having regard to the state of the art relied upon by the Appellant.

5.1 In the polymerisation process disclosed in D1 the most preferred monomer combination which comprises 0.5 -10% b.w. ethylene, 40 - 60% b.w. propylene and 32 - 40% b.w. butene-1 may be continuously or batchwise polymerised in a solvent medium in the presence of an anionic coordination catalyst (cf. whole page 3) at temperatures of 50 - 100°C and pressures of 5 - 20 bar (cf. page 4, lines 25 to 27). No hint is given therein to a polymerisation temperature of 140 - 250°C and a preference to a continuous process (all examples use a batchwise process).

From D1 alone it would thus not be obvious to use the higher process temperatures required in the patent in suit.

5.2 D2 relates to a process to produce amorphous propylene/butene-1 copolymers. The polymerisation process comprises contacting propylene and butene-1 with an anionic coordination catalyst at a temperature of 140-250°C and a pressure in the range of atmospheric to 2,000 psig (cf. Claim 1). It is taught that propylene/butene-1 copolymers prepared with the

specified aluminium triethyl/aluminium reduced titanium trichloride catalyst activated by trituration (AA-$TiCl_3$) at temperatures below 140°C have higher RBPS and higher hexane insoluble contents, i.e. fewer random propylene and butene-1 segments than copolymers made at high temperatures (cf. column 3, lines 13 to 21 in conjunction with column 4, lines 8 and 9). Moreover, although hexane soluble copolymers can be prepared in a batch solution process at high temperatures, a less random copolymer is obtained at a given butene-1 rate than in a continuous process (cf. column 4, lines 60 to 65).

The person skilled in the art would thus learn from that document that some process features such as catalyst, temperature and kind of process are important factors in the preparation of highly amorphous propylene/butene-1 copolymers, since the amount of random segments is influenced thereby. However, if a direct influence of temperature on the amorphous properties of the copolymer cannot be denied, the teaching of D2 is clearly limited to the situation where the two monomers, in that case propylene and butene-1, have about the same reactivity ratios at both the low and the high temperatures considered. At higher temperatures, the higher rate of chain propagation in the polymer, which affects the two monomers to the same extent, results in the incorporation of shorter and more random segments in the polymer chain. In the present case, by contrast, where propylene and butene-1 are copolymerised in the presence of ethylene, the latter having a much higher reactivity ratio, one would rather expect on the basis of that teaching the

formation of ethylene segments than a statistical distribution of these units. For these reasons, the skilled person would not have considered an increase in temperature for the solution of the above defined technical problem.

5.3 It follows that on the basis of D1 as the closest state of the art the claimed subject-matter cannot be regarded as obvious, which confirms the conclusion of the Opposition Division.

6. But even if starting with D2 as the most relevant prior art the result will be the same.

6.1 The problem may then be seen as the extension of the process known from D2 for the production of further amorphous copolymers.

6.2 The solution is to add ethylene as a third monomer to the mixture of propylene/butene-1 in the amounts specified in Claim 1 of the patent in suit.

6.3 In view of the patent specification and the evidence produced before the Opposition Division the Board considers that problem to be plausibly solved with the indicated means.

6.4 It has now to be considered whether it was obvious to add ethylene to that mixture in the expectation to get a highly amorphous terpolymer.

6.4.1 From the above considerations it is evident that the teaching of D2 is very specific in that all the process features are tailored for the copolymerisation of propylene and butene-1, e.g. compounds having equivalent monomer reactivity ratios; in particular,

the temperature of the polymerisation of at least 140°C, which is said to be critical (column 4, lines 19 to 23), is obviously chosen to ensure a random incorporation of short segments of propylene and butene-1 and, thereby, the formation of an amorphous copolymer having specific recurrent units. In view of that close relation between monomer combination and temperature of polymerisation the skilled person would have no information whatsoever from D2 about other monomers or monomer combinations compatible with such reaction conditions.

6.4.2 The next question is whether the skilled person would have considered that the monomer system disclosed in D1 could be suitably polymerised at the high temperatures used in D2. In the Board's view, the skilled person would have had no reason to contemplate such a modification which entails working conditions against the essence of the teaching of D1. As explained in the introduction of that citation (page 1, line 6 to page 2, line 18), one of the difficulties encountered in the preparation in solution of propylene polymers, including copolymers and terpolymers, is that the final product usually contains undesired amounts of crystalline fractions; this is overcome (page 3, line 7 to 17) by the process discussed above (point 4.1.1), which requires a temperature limited to the range of 50 to 100°C (page 4, lines 25 to 27). This teaching clearly speaks against higher temperatures for the copolymerisation of propylene and other monomers having different reactivity ratios, such as ethylene.

6.4.3 The argument that higher temperatures would nevertheless have been considered by a skilled person in view of their beneficial effect on stereospecifity cannot be accepted.

First, there is nothing in D1 which may suggest that the terpolymers described therein may be unsatisfactory in that respect; nor has the Appellant provided any evidence that this might be the case. Secondly, the presence of ethylene cannot be an incentive to use higher temperatures since, unlike propylene and butene-1, ethylene units are not concerned with stereospecifity.

6.4.4 It follows that also on the basis of D1 as the closest state of the art the claimed subject-matter cannot be regarded as obvious.

7. The process according to Claim 1 involving an inventive step the same applies to the subject-matter of Claim 2 which is directed to a preferred embodiment of the former and the patentability of which is supported by that of the main claim.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin