Case Number: T 0713/01 - 3.3.3
Application Number: 96112946.7
Publication Number: 0742232
IPC: C08F 36/04

Language of the proceedings: EN

Title of invention:
Solubilized anionic polymerization initiators and products therefrom

Applicant:
Bridgestone Corporation

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54

Keyword:
"Novelty (no) - "product-by-process" feature not distinguishing"

Decisions cited:
T 0205/83, T 0206/83, T 0279/84, T 0124/87, T 0464/94, T 0727/95

Catchword:
-
Case Number: T 0713/01 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 7 October 2003

Appellant: Bridgestone Corporation
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Tokyo 104-8340 (JP)

Representative: Albrecht, Thomas, Dr
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Decision under appeal: Decision of the Examining Division of the European Patent Office posted 22 January 2001 refusing European application No. 96112946.7 pursuant to Article 97(1) EPC.

Composition of the Board:
Chairman: R. Young
Members: P. Kitzmantel
E. Dufrasne
Summary of Facts and Submissions

I. This appeal, which was filed on 23 March 2001, lies against the decision of the Examining Division dated 12 January 2001 and issued in writing on 22 January 2001, refusing European patent application No. 96 112 946.7, filed on 22 September 1993 in the name of Bridgestone Corporation, and published under No. 0 742 232. The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 25 May 2001.

II. The decision under appeal was based on Claims 1 to 12 filed with the Applicant's submission dated 4 January 2001, independent Claims 1, 5, 8 and 10 reading as follows:

"1. A vulcanizable rubber composition comprising:
a polymer carrying at least one amine functional group A, wherein A is derived from the reaction product of an organolithium compound and an amine;
and a tin-carbon bond:
said amine functional group A being a cyclic amine having the general formula

\[ \begin{array}{c}
\text{R}_2
\end{array} \text{N} - \]

where \( R_2 \) is selected from the group consisting of an alkylene, oxy- or amino-alkylene group having from 3 to 7 methylene groups,
and being derived from a polymerization initiator having the general formula

\[ (A)\text{Li(SOL)}_y \]
where \( y \) is [of] from 1 to 3; SOL is a solubilizing component selected from the group consisting of hydrocarbons, ethers, amines or mixtures thereof; and A is as defined above."

"5. A tire having at least one vulcanizable elastomeric component comprising:
   a multifunctional polymer as defined in one of Claims 1 to 4; and from 5 to 80 parts by weight of carbon black, per 100 parts of said polymer."

"8. A rubber composition comprising:
   a polymer carrying at least one amine functional group A, wherein A is derived from the reaction product of an organolithium compound and an amine;
   said amine functional group A being a cyclic amine having the general formula

\[
\text{R}_2 \text{N}^-
\]

where \( \text{R}_2 \) is selected from the group consisting of an alkylene, oxy- or amino-alkylene group having from 3 to 7 methylene groups,
and being derived from a polymerization initiator having the general formula

\[
(\text{A})\text{Li(SOL)}_y
\]

where \( y \) is [of] from 1 to 3; SOL is a solubilizing component selected from the group consisting of hydrocarbons, ethers, amines or mixtures thereof; and A is as defined above,
and a second functional group selected and derived from the group consisting of terminating agents, coupling agents and linking agents."

"10. A tire having at least one vulcanizable elastomeric component comprising:
a multifunctional polymer as defined in one of Claims 8 and 9; and from 5 to 80 parts by weight of carbon black, per 100 parts of said polymer."

The further claims were dependent: Claims 2 to 4 on Claim 1; Claims 6 and 7 on Claim 5; Claim 9 on Claim 8, and Claims 11 and 12 on Claim 10.

III. The decision under appeal refused the application for the following reasons:

(a) The subject-matter of Claim 8 was anticipated by the disclosure of document D2 (EP-A-0 316 255) because the use of a polymerisation initiator having the general formula (A)Li(SOL)\_y in lieu of one of the formula (A)Li, as according to D2, did not contribute a distinguishing feature to the claimed compositions whose method of preparation was otherwise identical to that disclosed in D2. The Applicant's counter-argument that this difference led to a narrower molecular weight distribution of the "inventive" polymers was not accepted by the Examining Division because Example 5 of the application showed that similarly narrow molecular weight distributions could also be achieved with an unsolubilised initiator (A)Li. Accordingly, this characteristic was unable always
and unequivocally to distinguish the claimed polymers from those obtained according to D2.

(b) Nor was the Applicant's argument considered convincing that D2 did not anticipate the claimed subject-matter because it lacked an enabling disclosure as to how reliably to achieve a narrow molecular weight distribution. In the Examining Division's opinion, for a disclosure to be novelty destroying, it was sufficient that the result of its reworking could, by chance, meet the requirements of the claimed invention (here: a narrow molecular weight distribution). This conclusion was arrived at in spite of the Applicant's argument that D2 failed to disclose the preparation method of the initiator and that this deficiency led, in an unpredictable way, to the different molecular weight distributions of the polymers according to "comparative" Examples 3, 5 and 7 of the application in suit notwithstanding that all these examples used identical reaction conditions and the "same" non-solubilized initiator (i.e. an initiator according to D2).

(c) The subject-matter of Claims 10 to 12 was considered obvious by the Examining Division because the skilled person being aware of the high affinity for carbon black of the polydienes according to D2 required no inventive effort to select amounts of carbon black which were appropriate for the manufacture of tires.
(d) The same conclusion applied to the subject-matter of Claims 1 and 5 to 7 because nothing inventive could be seen in substituting the terminating agents used according to D2 by others known to be common in the tire industry from D1 (EP-A-0 451 603) including those establishing a tin-carbon bond used according to the claimed invention.

IV. The Appellant's arguments submitted with the Statement of Grounds of Appeal, the letter dated 30 September 2003 (filed in response to the Rapporteur's communication of 21 July 2003) and at the oral proceedings held on 7 October 2003 may be summarised as follows:

(a) The conclusions of the Examining Division were not in agreement with the findings of T 124/87 (OJ EPO 1989, 491) and T 206/83 (OJ EPO 1987, 5) according to which the criterion for a prior art disclosure to be novelty destroying was that the claimed subject-matter could be directly and unequivocally inferred from an enabling disclosure therein.

(b) Document D2, however, did not disclose polymers which had been prepared in the presence of solubilised initiators, nor did it comprise an enabling disclosure for the preparation of polymers whose molecular weight and molecular weight distribution was predictable as demonstrated by the divergent results of Examples 3, 5 and 7 of the application in suit which were representative of this prior art.
(c) In this context the Appellant also referred to T 464/94 of 21 May 1997 (not published in the OJ EPO) and T 727/95 (OJ EPO 2001, 1) according to which the mere probability ("by chance") for the achievement of an embodiment in a prior art document was not prejudicial to novelty.

(d) In the Appellant's view it was clear from the evidence in the application that the use of a solubilised initiator \((A)Li(SOL)\) instead of a non-solubilised initiator \((A)Li\) led not only to polymers having narrower molecular weight distributions but also to lower vinyl contents of these polymers and to lower tan \(\alpha\) values of rubber compositions prepared therefrom. These effects were derivable by comparison of the data for ("comparative") Examples 3 and 5 and those for ("inventive") Examples 9, 16 and 18.

(e) Concerning the issue of inventive step the Appellant inter alia argued that D2 was not a proper springboard for obviousness considerations because it was not concerned with the problem underlying the present invention.

V. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the Claims 1 to 12 submitted with the letter dated 4 January 2001.
Reasons for the Decision

1. The appeal is admissible.

2. Novelty of the subject-matter of Claim 8

2.1 Claim 1 of document D2 relates to a process for preparing a polydiene having a high level of affinity for carbon black which is characterised by reacting a metal terminated polydiene with a capping agent selected from the group consisting of (a) halogenated nitriles having the structural formula $X\cdot A\cdot C\equiv N$ wherein $X$ represents a halogen atom and wherein $A$ represents an alkylene group containing from 1 to 20 carbon atoms, (b) heterocyclic aromatic nitrogen containing compounds, and (c) alkyl benzoates.

Organolithium compounds are the preferred polymerisation initiators for the preparation of the "metal terminated polydiene" (page 2, line 61).

On page 3, lines 7 to 10 it is stated: "Lithium amides are also highly preferred initiators because they can be used to prepare polydienes which are terminated with polar groups at both ends of their polymer chains. The extra polar functionality provided by lithium amides results in increased interaction with carbon black resulting in better polymer-carbon black dispersion. The lithium amides that can be used include lithium pyrrolidide."

Examples 5 and 6 exemplify the use of lithium pyrrolidide as polymerisation initiator.
2.2 D2 thus discloses a method for preparing polydienes in the presence of an initiator whose amine portion A is within the definition of present Claim 8 and whose further polymerisation conditions, the introduction of a "second functional group" inclusive, meet the respective requirements of Claim 8 of the application-in-suit. D2 does not disclose a contact/reaction of the lithium pyrrolidide initiator, prior to its combination with the polymerisation mixture, with any "solubilising" agent (SOL). This difference, however, is not a characteristic of the claimed product per se but of the manner of its preparation contained in Claim 8 in the form of a "product-by-process" feature.

2.3 Since it is established practice of the Boards of Appeal that the subject-matter of "product-by-process" claims must be novel per se, i.e. independently of the process feature(s) (cf. Case Law of the Boards of Appeal, 4th edition 2001, page 173, section 6.2), the issue of novelty of the subject-matter of present Claim 8 turns on whether the use of a solubilised lithium pyrrolidide initiator contributes one or more features to the claimed polymerisation product which distinguish(es) it from analogous polymerisation products prepared in the presence of non-solubilised lithium pyrrolidide initiators as used according to D2.

2.4 The "solubilising" agent (SOL) is not comprised by the polymer resulting from the polymerisation reaction and its use cannot therefore be discovered by analysis of the polymer. This was not contested by the Appellant.
2.5 In the Board's judgment, the use of a solubilised initiator can also not retrospectively be found out by resorting to the properties of the resulting polymerisation product. The Appellant's contention that the evidence in the application-in-suit would demonstrate that, as compared to the use of a non-solubilised lithium pyrrolidide initiator, the polymerisation in the presence of an analogous solubilised initiator would lead to polymers having narrower molecular weight distribution (M<sub>w</sub>/M<sub>n</sub>), lower vinyl content and, ultimately, to compositions having lower tan δ is not supported by the facts.

This results from the following considerations:

2.5.1 Concerning the distinguishing contribution of the "inventive" initiator it has to be borne in mind that the relevant question is whether the claimed products themselves comprise any characteristics derived from the solubilised lithium pyrrolidide initiator and not, as argued by the Appellant, whether, by comparing the properties of polymers prepared, respectively, with a solubilised initiator and with a non-solubilised initiator, indirectly some differences can be construed whose significance as characteristics of the use of the solubilised initiator could not be discovered by a skilled person being confronted with this polymer alone.

2.5.2 The experimental evidence in the application-in-suit shows that the "inventive" polymers which have been prepared in the presence of solubilised lithium pyrrolidide initiators may exhibit values of molecular weight distribution, vinyl content and tan δ which may
also be achieved by polymers prepared in the presence of non-solubilised lithium pyrrolidide initiators.

2.5.3 It is noted in this context that present Claim 8 does not distinguish between polymers having "a second functional group" derived from a "terminating" agent like isopropanol (covered by Examples 1, 3, 5, 7, 9, 11, 14, 16 and 18) and those derived by "end-capping" with a "coupling" or "linking" agent (covered by Examples 2, 4, 5, 8, 10, 12, 13, 15, 17 and 19) and that also the application-in-suit does not contain any information on the basis of which different values of molecular weight distribution, vinyl content and tan δ should be attributed to these two types of polymers having different "second functional groups". For the purpose of establishing whether the use of a solubilised initiator leaves a characteristic "mark" on the resulting polymer, both groups of examples are therefore equally relevant.

2.5.4 The conclusion drawn in paragraph 2.5.2 above is inter alia based on the following data taken from Tables V, VII and VIII of the application-in-suit. From this summary (examples marked with an asterisk are "comparative", i.e. relate to polymers prepared in the presence of a non-solubilised initiator) it can be inferred that the molecular weight distribution of the "comparative" polymer according to Example 5 is in the same range as the lower values of the "inventive" polymers,
- that the vinyl content of the "inventive" polymer according to Example 11 is in the same range as that of the "comparative" polymers according to Examples 5 and 6, and
- that the tan ä value of a rubber composition comprising the "comparative" polymer according to Example 6 is within the range of the "inventive" polymers:

<table>
<thead>
<tr>
<th>Example</th>
<th>5*)</th>
<th>6*)</th>
<th>11</th>
<th>12</th>
<th>13</th>
<th>14</th>
<th>16</th>
</tr>
</thead>
<tbody>
<tr>
<td>M_w/M_n</td>
<td>1.1</td>
<td>2.2</td>
<td>1.14</td>
<td>1.79</td>
<td>2.09</td>
<td>1.15</td>
<td>1.10</td>
</tr>
<tr>
<td>vinyl</td>
<td>52.9</td>
<td>52.9</td>
<td>53.3</td>
<td>53.3</td>
<td>53.3</td>
<td>50.1</td>
<td>42.7</td>
</tr>
<tr>
<td>tan ä</td>
<td>.144</td>
<td>.078</td>
<td>.080</td>
<td>.076</td>
<td>.078</td>
<td>.129</td>
<td>.100</td>
</tr>
</tbody>
</table>

2.5.5 It is thus established that none of the polymer properties (M_w/M_n, vinyl content, tan ä) alleged by the Appellant to be characteristic for polymers prepared in the presence of a solubilised lithium pyrrolidide initiator are appropriate for distinguishing these polymers from others prepared in the presence of a non-solubilised lithium pyrrolidide initiator because in both cases the same properties may be obtained.

2.5.6 Consequently, the only difference between the content of the disclosure of D2 and the wording of present Claim 8, i.e. the presence or not of a solubilising agent as part of the lithium pyrrolidide initiator, does not qualify as a distinguishing feature of the claimed product because this "product-by-process" feature is not detectable on the product by a reliably distinguishing property.
2.5.7 No other conclusion can be arrived at on the basis of
the available evidence. As emphasised in paragraph
2.5.1 above, the relative improvement of some
properties which can be inferred by comparing
"comparative" and "inventive" polymers on which the
Appellant wants to rely (cf. section IV (d) above) is
to no avail with respect to the aim of establishing a
characteristic distinguishing feature on the
"inventive" polymer itself.

2.5.8 It is noted in this context that in proceedings before
the Examining Division the burden of proof for an
allegedly distinguishing "product-by-process" feature
lies with the Applicant (cf. T 205/83, OJ EPO 1985, 363,
Reasons 3.2.1; T 279/84 of 29 June 1987 (not published
in the OJ EPO), Reasons 4.1 and 4.2).

2.5.9 In view of the afore-mentioned considerations, the
arguments of the Appellant denying the existence of an
"enabling" disclosure in D2 of polymers prepared in the
presence of a solubilised lithium pyrrolidide initiator
by reference to T 124/87 and T 206/83 are irrelevant;
and the same applies to the question whether lack of
novelty can be decided on the basis of the mere
probability of an anticipatory disclosure addressed in
T 464/94 and T 727/95 (cf. paragraphs IV (a) and (c)
above).

2.6 In the circumstances, the disclosure of D2 is novelty
destroying for the subject-matter of present Claim 8
because it directly and unequivocally comprises all
features of the claimed rubber composition which are
apt for its characterisation.
3. The non-compliance of a single claim with one of the requirements of the EPC entails the non-allowability of the application as a whole.

4. In this situation there is no need to deal with any further arguments of the Appellant or to address further deficiencies of the application-in-suit.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:  The Chairman:

E. Görgmaier  R. Young