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DECISION of 10 December 1998

Case Number:

т 0275/96 - 3.3.3

Application Number:

84201705.5

Publication Number:

0143500

IPC:

C08F 279/02

Language of the proceedings: EN

Title of invention:

Rubber-reinforced polymers of monovinylidene aromatic compounds having a unique balance of gloss and physical strength properties and a method for their preparation

Patentee:

The Dow Chemical Company

Opponent:

BASF Aktiengesellschaft, Ludwigshafen

Headword:

Relevant legal provisions:

EPC Art. 54, 56, 100(b)

Keyword:

"New ground of opposition - no consent of patentee"

"Novelty-(yes) purposive selection from broader range"

"Inventive step-(yes) unobvious selection"

Decisions cited:

G 0010/91, G 0009/92, T 0198/84, T 0279/89

Catchword:

Europäisches Patentamt

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Beschwerdekammem

Boards of Appeal

Chambres de recours

Case Number: T 0275/96 - 3.3.3

DECISION of the Technical Board of Appeal 3.3.3 of 10 January 1998 December

Appellant:

(Proprietor of the patent)

The Dow Chemical Company

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Representative:

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Respondent: (Opponent)

BASF Aktiengesellschaft, Ludwigshafen

-Patentabteilung - C6-Carl-Bosch-Strasse 38 67056 Ludwigshafen (DE)

Decision under appeal:

Interlocutory decision of the Opposition Division of the European Patent Office posted 23 January 1996 concerning maintenance of European patent

No. 0 143 500 in amended form.

Composition of the Board:

Chairman:

Members:

C. Gérardin
P. Kitzmantel

J. A. Stephens-Ofner

Summary of Facts and Submissions

- I. European patent application No. 84 201 705.5 in the name of THE DOW CHEMICAL COMPANY, which had been filed on 22 November 1984, claiming priority from a NL application filed on 23 November 1983, resulted in the grant of European patent No. 0 143 500 on 22 January 1992 on the basis of 4 claims, independent Claims 1 and 4 reading as follows:
 - "1. A rubber-reinforced polymer comprising a continuous phase of a polymer consisting essentially of units derived from one or more polymerizable monovinylidene aromatic compounds having discrete particles dispersed throughout the polymer matrix, wherein the discrete rubber particles are formed in a mass-polymerization process, having a shell/core morphology and have a volume average particle size from 0.1 to 0.19 µm."
 - A method for preparing the rubber-reinforced polymer product of claim 1, the method comprising the steps of mass polymerizing a solution of one or more polymerizable monovinylidene aromatic compounds and a rubber at conditions such that upon the phase inversion of the mass polymerization mixture and the subsequent sizing of the rubber the rubber is formed as particles of a shell/core morphology having a volume average particle size of from 0.1 to 0.19 µm, wherein these conditions are achieved by bonding sufficient blocks of monovinylidene aromatic polymer to the alkadiene rubber either prior to or simultaneously with the mass polymerization step and by the appropriate modification of the polymerization mixture matching the viscosity of the rubber phase and the viscosity of the continuous phase at phase inversion, subsequently completing the polymerization using either mass or suspension

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polymerization techniques and thereafter subjecting the polymerization mixture to conditions sufficient to remove the unreacted monomers and to cross-link the polymerized alkadiene of the shell/core rubber particles."

Claims 2 and 3 related to preferred embodiments of the polymer according to Claim 1.

II. Notice of Opposition was filed by BASF
Aktiengesellschaft on 20 October 1992 requesting
revocation of the patent in its entirety, on the ground
that the claimed subject-matter was not novel over the
disclosure, respectively, of documents

D1: GB-A-1 422 208 and

D2: GB-A-1 180 085.

Further documents that have been considered in the opposition proceedings were

- D3: G.E. Molau, W.M. Wittbrodt, "Colloidal Properties of Styrene-Butadiene Block Copolymers",

 Macromolecules, vol. 1, No. 3, May June 1968,
 pages 260 to 264, and
- D4: A. Echte, "Teilchenbildung bei der Herstellung von kautschukmodifiziertem Polystyrol", Die Angewandte Makromolekulare Chemie 58/59 (1977) 175 to 198.
- III. By its interlocutory decision announced orally on 13 December 1995 and issued in writing on 23 January 1996 the Opposition Division decided that the grounds of opposition did not prejudice the maintenance of the patent in amended form, the amendments consisting (i) in the substitution in the independent Claims 1 and 4 as lower limit of the volume average particle size

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range of the figure "0.15 μ m" for the granted lower limit of "0.1 μ m" and (ii) in the consequential deletion of the particle size feature in dependent Claim 3.

It was held in that decision that the subject-matter of granted Claim 1 (then Main Request), comprising the presence of rubber particles having a volume average size of 0.1 µm, was not novel over D1 because this document disclosed rubber-reinforced polystyrene polymers containing shell/core particles having the same average size of 0.1 µm. As opposed thereto, the subject-matter of amended Claim 1 (Auxiliary Request) was considered to be novel, because, with respect to the particle size range of 0.1 to 0.7 µm disclosed in D1, the range of 0.15 to 0.19 µm according to said Claim 1 was narrow and was also the result of a purposive selection.

The subject-matter of Claim 1 of the Auxiliary Request was also held to be inventive, because the internal evidence contained in the patent in suit showed that the restricted particle size range provided a superior balance of gloss and mechanical properties.

Furthermore, the Opposition Division, in this decision, refused to admit into the opposition proceedings any arguments concerning the ground of Article 100(b) EPC, which had not been invoked during the opposition period.

IV. Notice of Appeal against the above decision, with simultaneous paying of the appeal fee, was filed by the Proprietor of the patent (Appellant) on 14 March 1996.

The Statement of Grounds of Appeal was submitted on 24 May 1996.

V. With the Statement of Grounds of Appeal the Appellant submitted, as Main Request, a set of four claims, which are identical to the claims as granted but for the lower limit of the volume average particle size range in Claims 1 and 4 which has been amended to "more than $0.1~\mu m$ ".

As an Auxiliary Request the Appellant maintained the version of the claims which was accepted in the decision under appeal.

In reply to a specific request by the Board, the Appellant in his submission dated 19 October 1998 declared that he did not agree to the introduction into the appeal proceedings of the ground of opposition under Article 100(b) EPC raised by the Respondent/Opponent (cf. last paragraph of point VI below).

In his written submissions and during the oral proceedings held on 10 December 1998 the Appellant argued that the subject-matter of Claim 1 of the Main Request was novel, since the rubber particle size range defined therein met the criteria for a selection invention. In particular he argued that this range, with regard to the range of 0.1 to 0.7 µm disclosed in D1, was narrow, was sufficiently far removed from the values exemplified in D1 and was also the result of a purposive selection, as evidenced by the comparison of the properties (gloss, Izod impact strength, elongation at break and tensile yield) according to Samples No. 1 and No. E (comparative) in the Table on page 8 of the patent in suit.

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In the Appellant's view the subject-matter of Claim 1 was also inventive, because the prior art contained no pointer towards the claimed solution of the existing problem, i.e. the provision of high impact strength polystyrene-type resins (hereinafter "HIPS") having an improved balance of gloss and physical properties.

VI. In his written submissions of 11 October 1996 and during the oral proceedings the Respondent argued that the claimed subject-matter was not novel over D1 because it did not meet the indispensable criterion for a selection invention, namely that of an abrupt technical progress existing only in the selected area. In particular, he argued, the lower limit of "more than 0.1 µm" of the particle size range now claimed was not sufficiently far removed from the value of 0.1 µm disclosed in D1 as lower limit of the particle size range.

In the Respondent's opinion, the claimed subject-matter was also not inventive, because, being aware from D1 and D4 of the importance of the rubber particle size for the properties of HIPS, the skilled person would not be surprised to find a particularly advantageous sub-range. This was even more obvious in view of the disclosure in document D3, Table 2 which demonstrated the availability of styrene-butadiene block copolymers having rubber particles in the size range "selected" according to present Claim 1.

Having been made aware by the Board that in the absence of the Proprietor's consent the ground of Article 100(b) EPC could not be admitted into the appeal proceedings, the Respondent refrained from further commenting at the oral proceedings upon these previously raised objections (cf. G 10/91 OJ EPO 1993, 420).

VII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims submitted with the Statement of Grounds of Appeal by way of Main Request.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

- 1. The appeal is admissible.
- 2. Prodecural matters
- 2.1 Since the Proprietor of the patent in suit is the sole Appellant against the interlocutory decision under appeal, according to G 9/92 (OJ EPO 1994, 875), neither the Board nor the non-appealing Opponent may challenge the maintenance of the patent as amended in accordance with this interlocutory decision.
- 2.2 The ground of opposition of Article 100(b) EPC has not been invoked during the opposition period and the Opposition Division refused to admit this ground into the first instance opposition proceedings (cf. point 2.1 of the Reasons of the decision under appeal). Therefore, at the appeal stage, this ground could only be introduced with the Proprietor's consent (G 10/91 cf. supra). Such consent was, however, withheld and the ground of Article 100(b) EPC is therefore not to be considered in this appeal.
- 3. Article 123(2) and (3) EPC

The rubber particle size range of "from more than 0.1 to 0.19 μm " according to present Claim 1 is different from the wording of the granted Claim 1 by the words

"more than". On the one hand this definition of the particle size range combines the lower limit of 0.1 μm of the range according to Claim 1 as filed with the upper limit of 0.19 μm of the range according to Claim 4 as filed, and on the other hand it introduces the new information "more than".

Since the combination of the limits of the ranges according to Claims 1 and 4 as filed does not add any new matter going beyond the original disclosure and since the words "more than" are used to distinguish the claimed subject-matter from that of document D1 by disclaiming the previously overlapping value of "0.1 μ m", Claim 1 does not contravene Article 123(2) EPC.

By virtue of this disclaimer, the scope of the new Claim 1 is narrower than that of granted Claim 1; thus, the provision of Article 123(2) EPC is also met.

The same conclusions apply to the subject-matter of Claim 4, which differs from that of Claim 4 as granted by the same disclaimer.

4. Novelty

4.1 Document D1

This document relates to a thermoplastic composition which comprises (a) a polyphenylene ether resin and (b) a rubber modified polystyrene resin, comprising a polystyrene matrix in which there is uniformly dispersed a discontinuous phase comprising particles of a styrene homopolymer, alkylstyrene or halostyrene polymers, or a mixture thereof englobulated in a diene rubber membrane, said particles having an

average size ranging from 0.1 to 0.7 μ m, the membrane thickness being not in excess of 1/4 the average particle diameter, and the diene rubber content being from 1 to 10% by weight of the rubber modified polystyrene (cf. Claim 1).

There is no disclosure in D1 of a concrete value of the average rubber particle size of any specific HIPS, save the information that can be deduced from Figure 2 (cf. page 2, lines 84 to 93). According to the uncontested calculations of the Proprietor in his letter to the Opposition Division dated 12 April 1994 the volume average particle size of the material of Figure 2 of D1 was $0.48~\mu m$.

There was agreement between the parties, and the Board concurs with this conclusion, that the only difference between the polymer compositions of the patent in suit and those according to D1 resides in the rubber particle size.

4.2 Selection criteria

It was not contested by the Respondent that the particle size range of "more than 0.1 to 0.19 μm " according to Claim 1 was not as such disclosed in D1. He argued, however, that this range was not novel over the broader range of "from 0.1 to 0.7 μm " disclosed in D1.

4.2.1 It is well established in law that nothing should be patented that was within the realm of the state of the art, i.e. that has been made available to the public before the filing of the European patent application (Article 54(2) EPC).

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In the case of an overlap or partial anticipation of ranges of parameters it has thus to be established whether the overlapping or partially anticipated area has been made available in an enabling way.

- 4.2.2 In decisions T 198/84 (OJ EPO 1985, 209) and T 279/89 of 3 July 1991 (not published in the OJ EPO) the Boards of Appeal developed the following three criteria, which have to be met together, in order to establish the novelty of a selected sub-range:
 - (i) the selected sub-range should be narrow,
 - (ii) the selected sub-range should be sufficiently far removed from the known range illustrated by means of examples, and
 - (iii) the selected area should not provide an arbitrary specimen from the prior art, but something equivalent to another invention (purposive selection).
- 4.2.3 In following this analysis, this issue of novelty, in the present case, turns on the question whether the sub-range "from more than 0.1 to 0.19 μm" was made available in D1, or more particularly, was made available by the range of 0.1 to 0.7 μm disclosed therein.
- 4.2.3.1 Since the said sub-range amounts to only about 15% (= 100 x [0.19 0.1]/[0.7 0.1]) of the range according to D1, it can be considered as "narrow" (afore-mentioned requirement (i)).
- 4.2.3.2 In addition, the afore-mentioned requirement (ii) is met, because the only concretely disclosed particle size is the one to be calculated on the basis of Figure 2 and which is 0.48 µm; the lower limit of

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"0.1 μ m" of the range "0.1 to 0.7 μ m" cannot be considered as an **exemplified** value and does not qualify, therefore, as a value critical to the appreciation of a "true selection".

4.2.3.3 As to the purposiveness of the selected sub-range, (afore-mentioned requirement (iii)), it can be concluded from the data in the Table on page 8 of the patent in suit that the pattern of the properties: Izod impact strength, gloss, elongation at break and tensile yield deviates in a significant way from the pattern of the same properties of similar HIPS compositions comprising the same amount (8.5%) of rubber particles of the same shell/core morphology, but having particle sizes within the range of 0.1 to 0.7 µm as disclosed in D1:

Sample No.	particle size [µm]	Izod Impact Strength [J/m]	Gloss [%]	Elonga= tion At Break [%]	Tensile Yield [N/mm²]
1	0.17	29	89	13	26.9
E	0.25	24	79	10	24.5
С	0.6	66	66	15	23.4

This table shows that the inventive Sample No. 1 does not follow the trend to be expected from a reduction of the rubber particle size, as evidenced by samples No. C and E, namely that of the achievement of better gloss at the expense of deteriorated physical properties, but - quite surprisingly - exhibits a completely different quality, i.e, a property pattern comprising not only enhanced gloss but also, with respect to sample E, consistingly enhanced physical properties. The purposiveness of the selection of the composition according to Sample No. 1 can thus be recognized.

The Board is not aware of any circumstances, which could cast doubt on the Appellant's contention that the purposiveness of the selection experimentally demonstrated for (only) one composition (Sample No. 1) within the defined range, is to be judged differently for the whole sub-range specified in present Claim 1. The Respondent, on whom the burden of proof rests, although asking in his counterstatement of appeal of 11 October 1996 for a time limit to submit comparative examples, with his letter of 28 October 1996 abandoned this request and has not in fact filed any evidence able to refute the Appellant's case.

It follows that the sub-range "from more than 0.1 to 0.19 μm " in Claim 1 can be considered to also meet the afore-mentioned criterion (iii), i.e. the purposiveness of the selection.

4.2.3.4 In consequence, the rubber particle size range defined in Claim 1 fulfils all the criteria (i), (ii) and (iii) set out in the decisions T 198/84 and T 279/89, both mentioned supra.

4.3 Conclusion

From this it follows that the compositions according to Claim 1 are clearly distinguishable from the compositions according to D1 comprising rubber particles of a size of from 0.1 to 0.7 μm .

The subject-matter of Claim 1 is therefore novel over the disclosure of document D1.

The same conclusion applies to the subject-matter of Claim 4 which comprises the same sub-range feature and to the subject-matter of Claims 2 and 3, which are dependent on Claim 1.

5. Inventive step

5.1 State of the art

The present invention is concerned with the provision of rubber-reinforced polystyrene (including other monovinylidene aromatic compounds) materials having a combination of good gloss and good physical properties (cf. original application: page 2, lines 21 to 25 and page 4, lines 7 to 20; patent in suit: page 2, lines 37 to 40 and page 3, lines 24 to 33).

None of the documents D1 to D4 cited in the opposition proceedings relate to this concrete technical field:

5.1.1 Document D1

D1 (cf. point 4.1 supra) relates to polyphenylene ether compositions, which contain HIPS, but does not expand on the intrinsic properties of the HIPS component.

5.1.2 Document D2

D2 relates to transparent impact-resistant thermoplastic compositions comprising a matrix of a copolymer of a mono-alkenyl aromatic compound and an alkyl methacrylate wherein particles are dispersed having a butadiene rubber shell englobulating a copolymer of the composition of the matrix (Claim 1). This document is only concerned with compositions comprising alkyl methacrylate units in the matrix and in the rubber phase core. It is not, therefore, an appropriate starting point for the assessment of the obviousness of HIPS compositions not comprising alkyl

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methacrylate units; it is furthermore an important condition of D2 that the difference in refractive index between the copolymer and the butadiene rubber does not exceed 0.005 (cf. Claim 1, feature (ii)), a condition which cannot be fulfilled in the absence of alkyl methacrylate units: cf. page 10, Table 2, "Polymerizable solutions" F and G exhibiting a refractive index difference of 0.079, in conjunction with page 15, Table 5, showing that "Solution" F exhibits indeed inferior Izod impact and Haze qualities.

5.1.3 Document D3

This document relates to the colloidal properties of styrene-butadiene block copolymers, including styrene polymer compositions comprising small micellar rubber particles (cf. page 262, Figure 2 and right-hand column penultimate paragraph; page 363, left-hand column, second paragraph and right-hand column, Table I and Figure 3). This document is mainly concerned with problems of compatibility (cf. page 260, Abstract) and is silent on the optical and physical properties of the compositions.

5.1.4 Document D4

This document discusses the morphology of rubber particles formed during the polymerization of solutions of polybutadiene in styrene (cf. "Summary" on pages 175 to 176). On page 180, last paragraph it is stated that the "use" properties ("Gebrauchseigenschaften") of a rubber-modified material depend not only on the volume proportion of the disperse phase but, to a large extent, on the particle spectrum ("Teilchenspektrum"), i.e. particle size distribution. However, D4 does not refer to particular properties and is not, therefore an

appropriate starting point for the assessment of the obviousness of a technical solution concerned with the improvement of gloss and physical properties.

5.1.5 Closest state of the art

During the oral proceedings the Appellant argued that the starting point of the present invention was the state of the art acknowledged in the patent in suit itself, thus including prior art as represented by DE-A-2 613 352 (= D5). With respect to this document page 2, lines 17 to 25 of the patent in suit (page 1, line 22 to page 2, line 2 of the application) refers to HIPS compositions having improved transparency and mechanical properties, which comprise rubber particles of less than one μm , preferably from 0.2 to 0.6 μm , and reports that these compositions, while having relatively good physical strength properties, do not always exhibit the desired gloss.

It can, thus, be concluded that this document represents a state of the art, which qualifies as an appropriate starting point in the concrete technical field the invention is concerned with (cf. point 5.1 supra) and is thus considered as the closest prior art.

5.2 Problem and solution

With respect to the afore-mentioned closest state of the art the problem underlying the present invention is the provision of rubber-reinforced polymers consisting essentially of units derived from one or more polymerizable monovinylidene aromatic compounds, which polymers exhibit an improved balance of good gloss and good physical properties. According to Claim 1 of the patent in suit this problem is solved by the provision of HIPS materials comprising discrete rubber particles, which have a shell/core morphology and a volume average particle size from more than 0.1 to 0.19 μm .

The evidence reported in the Table on page 8 of the patent in suit demonstrates that this problem has effectively been solved (cf. point 5.3.1 below).

5.3 Obviousness

The material according to inventive Sample No. 1 of the Table on page 8 of the patent in suit provides a balance of the properties gloss, Izod impact strength, elongation at break and tensile yield, which is surprisingly superior to the balance of the same properties of (i) Samples E and C, whose materials comprise rubber particles having sizes above the upper limit of 0.19 µm of the particle size according to present Claim 1, as well as (ii) Sample D, which comprises rubber particles of a size below the lower limit of "more than 0.1 µm" of present Claim 1 and which rubber particles have a morphology of the type "rod/sphere" different from the type "shell/core" used according to present Claim 1:

Sample No.	particle size [µm]	Izod Impact Strength [J/m]	Gloss [%]	Elonga= tion At Break [%]	Tensile Yield [N/mm²]
1	0.17	29	89	13	26.9
E	0.25	24	79	10	24.5
C	0.6	66	66	15	23.4
D	0.06	15	95	3	35

As set out in point 4.2.3.3 supra, the superiority of "inventive" Sample No. 1 over the "comparative" Samples E and C is surprising, because according to the latter samples, and in line with his general common knowledge, the skilled person would have expected that a further reduction of the rubber particle size would not only cause an improvement of the gloss, but also a deterioration of the physical properties Izod impact strength, elongation at break and tensile yield. However, the data in the table prove that not only the gloss but also the physical properties are enhanced over the values according to Sample E which uses particles of a size of 0.25 µm. Sample D shows that this suprisingly improved balance of the mentioned properties is not just the result of a smaller particle size of the rubber particles, but also requires a specific morphology, namely a shell/core morphology, of these particles.

None of the documents which have been cited in these 5.3.2 proceedings comprise any hint that the solution of the existing technical problem can be achieved by the combination of features specified in present Claim 1 (cf. point 5.2 supra): D1 and D2 do not relate to HIPS, but D1 to a combination of HIPS with polyphenylene oxide and D2 to alkyl methacrylate modified HIPS-like materials; D3 and D4 are concerned with certain aspects of HIPS compositions, but are silent on their gloss, Izod impact strength, elongation at break and tensile yield; as to the closest state of the art discussed in the patent in suit itself (D5), it is clearly shown by the data contained in the above table that the particle size and the morphology of the rubber particles contribute in an unexpected fashion to the improved balance of the properties gloss, Izod impact strength, elongation at break and tensile yield. The Respondent

has not brought forward any argument in favour of the obviousness of the subject-matter of present Claim 1 over this state of the art and the Board is also unaware of any relevant information on the basis of which the surprising character of the balance of properties evidenced by the data in the Table on page 8 of the patent in suit could be questioned.

- 5.3.3 The arguments brought forward by the Respondent in favour of the obviousness of the subject-matter of present Claim 1 cannot be accepted by the Board:
- 5.3.3.1 The fact that D1 discloses that the HIPS component contributes to the transparency of its blend with polyphenylene ether (cf. page 2, lines 42 to 49) cannot suggest the surprisingly improved balance of gloss and some physical properties in the selected sub-range of the rubber particle size.
- 5.3.3.2 The same conclusion is valid with respect to the disclosure in Table I, page 263 of D3 of rubber particle sizes within the selected sub-range. Even if the disclosure of the "Micellar Core Diameters" of from 750 to 4000 Å (= 0.075 to 0.4 μ m) could be equated with the volume average particle sizes required by the patent in suit (which is not the case, because there is no information concerning the corresponding shell thickness), the absence in D3 of a correlation of the particle size with the specifically required properties obviates any speculation of the possible obviousness of a selected sub-range of the rubber particle size. The Respondent's argument that it was only necessary to determine the properties of the particles is therefore beside the point; there was no incentive in D3 for the skilled person to measure the gloss, the Izod impact strength, the elongation at break and the tensile yield properties of these particles.

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- 5.3.3.3 The information which is missing in D3 cannot be found in D4 either. The reference on page 180 of this document to the dependence of the material properties on the rubber particle size only confirms what is self-evident. This statement cannot, however, serve as a guidance to the skilled person wishing to solve the existing technical problem, because nothing is said about the interdependence of the rubber particle size and certain specific properties. The only measure the person skilled in the art could envisage on the basis of this information, would be to engage in a random screening program involving a huge number of experiments. On the basis of said statement in D4 the skilled person could not, therefore, by a few orienting experiments and without inventive skill arrive at the claimed solution of the existing technical problem.
- 5.3.4 It follows that the subject-matter of present Claim 1 involves an inventive step.
- 5.3.5 The same conclusion applies a fortiori to the subject-matter of Claim 4 which relates to a method for preparing the rubber-reinforced product of Claim 1 and comprises the same features of rubber particle size and particle morphology as Claim 1.

The unobviousness of the subject-matters of Claims 2 and 3 results from their dependency on Claim 1.

Order

For these reasons it is decided that:

- 1. The Opposition Division's decision is set aside.
- 2. The case is remitted to the Opposition Division with the order to maintain the patent with the claims filed as the Main Request and after any necessary amendments to the description.

The Registrar:

The Chairman:

C. Gérardin

