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DECISION of 9 March 2005

Case Number:	T 0667/02 - 3.3.3
Application Number:	94925745.5
Publication Number:	0716664
IPC:	C08F 287/00

Language of the proceedings: EN

Title of invention: Improved rubber modified polystyrene

Patentee: THE DOW CHEMICAL COMPANY

Opponent: Enichem S.p.A.

Headword:

-

Relevant legal provisions: EPC Art. 54, 56, 100(b), 123(2)

Keyword:
"Amendments - added subject-matter (no)"
"Opposition grounds - insufficiency of disclosure (no)"
"Novelty (yes)"
"Inventive step - ex post facto analysis"

Decisions cited: T 0002/81, T 0969/91, T 0925/98

Catchword:

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Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number: T 0667/02 - 3.3.3

D E C I S I O N of the Technical Board of Appeal 3.3.3 of 9 March 2005

Appellant: (Opponent)	Enichem S.p.A. Piazza Boldrini, 1 I-20097 San Donato Milanese (Mi) (IT)	
Representative:	De Gregori, Antonella Ing. Barzano & Zanardo Milano S.p.A. Via Borgonuovo 10 I-20120 Milano (IT)	
Respondent: (Proprietor of the patent)	THE DOW CHEMICAL COMPANY 2030 Dow Center Midland Michigan 48674 (US)	
Representative:	Raynor, John Beck Greener Fulwood House 12 Fulwood Place London WC1V 6HR (GB)	
Decision under appeal:	Interlocutory decision of the Opposition Division of the European Patent Office dated 21 March 2002 and issued in writing on 24 April 2002 concerning maintenance of European patent No. 0716664 in amended form.	

Composition of the Board:

Chairman:	R.	Young
Members:	Α.	Däweritz
	н.	Preglau

Summary of Facts and Submissions

- I. The grant of European patent No. 0 716 664 in respect of European patent application No. 94 925 745.5, based on International patent application No. PCT/US94/08854 (which had been published as WO-A-95/06686), filed on 4 August 1994 and claiming priority of 2 September 1993 of an earlier application in the USA (116163), was announced on 15 October 1997 (Bulletin 1997/42). The patent contained 17 claims, including independent Claims 1 and 8 reading as follows:
 - "1. A method for the preparation of a rubber-modified monovinylidene aromatic polymer composition said composition comprising, on a total composition weight basis, from 90 to 55 weight percent of a rigid monovinylidene aromatic polymer matrix and from 10 to 45 weight percent of grafted and occluded 1,3-alkadiene-based rubber particles dispersed within said matrix and wherein from 25 to 80 weight percent of said rubber particles have a capsule particle morphology and a volume average particle size of from 0.1 to 0.4 μm and from 75 to 20 weight percent of said particles have an entanglement particle morphology and a volume average particle size of from 0.25 to 1 μ m; said method comprising:

a. dissolving from 5 to 30 parts by weight of a 1,3-alkadiene/monovinylidene aromatic block copolymer rubber in from 95 to 70 parts by weight of a monovinylidene aromatic monomer, said block copolymer rubber being one which has from 15 to 40 weight percent of monovinylidene aromatic monomer copolymerized therein;

b. polymerizing the resulting rubber/monomer solution at a temperature of from 50 to 210°C, in the presence of from 0 to 1000 parts per million by weight of a polymerization initiator and from 0 to 2000 parts per million by weight of a chain transfer agent and under agitation conditions sufficient to provide capsule morphology and entanglement morphology rubber particles in the above-specified size ranges, with a proviso that the amounts of said initiator and chain transfer agent are not both zero; and c. thereafter recovering the resulting rubbermodified monovinylidene aromatic polymer product.

8. A rubber-modified monovinylidene aromatic polymer composition which comprises, on a total composition weight basis, from 90 to 55 weight percent of a rigid monovinylidene aromatic polymer matrix and, dispersed within said matrix, from 10 to 45 weight percent of grafted and occluded 1,3alkadiene-based rubber particles, said particles being composed, on a rubber particle weight basis of:

a. from 25 to 80 weight percent of rubber particles having a capsule morphology and a volume average particle size of from 0.1 to 0.4 μ m; and b. from 75 to 20 weight percent of rubber particles having an entanglement morphology and having a volume average particle size of from 0.25 to 1 μ m.". The remaining dependent Claims 2 to 7 and 9 to 17 concerned particular elaborations of the subject-matter of Claims 1 and 8, respectively.

Products of this type are often referred to in the art as HIPS. This term will also be used herein below.

II. On 9 July 1998, a Notice of Opposition was filed in which revocation of the patent in its entirety was requested on the basis of the grounds according to Article 100(a) EPC, for non-compliance with the provisions of Articles 52(1), 54 and 56 EPC, and/or on the basis of the ground according to Article 100(b) EPC, for not complying with the provision of Article 83 EPC. The Opposition relied on

D1: EP-A-0 429 986.

In the course of the opposition proceedings, a third party filed observations under Article 115(1) EPC and submitted copies of further documents including

D2: EP-A-0 143 500,

D3: Adolf Echte, "Rubber-Toughened Styrene Polymers", American Chemical Society, 1989, and D4: EP-A-0 048 390.

The submitted copies had been numbered by the third party "D1", "D2" and "D7", respectively.

According to the minutes of oral proceedings held on 21 March 2002 (item 5), these further documents, with the exception of D2, were not, however, introduced by the Opposition Division in the proceedings (Article 114(2) EPC). Nevertheless, D3 was referred to also by the Patent Proprietor (minutes, item 7).

A new Main request (which had been filed by the Patent Proprietor by fax dated 18 March 2002 together with a new Auxiliary Request) was further amended in the oral proceedings to contain independent Claims 1 and 6 having the following wording:

A method for the preparation of a rubber-modified "1. monovinylidene aromatic polymer composition said composition comprising, on a total composition weight basis, from 90 to 55 weight percent of a rigid monovinylidene aromatic polymer matrix and from 10 to 45 weight percent of grafted and occluded 1,3-alkadiene-based rubber particles dispersed within said matrix and wherein from 25 to 80 weight percent of said rubber particles have a capsule particle morphology and a volume average particle size of from 0.1 to 0.4 μm and from 75 to 20 weight percent of said particles have an entanglement particle morphology and a volume average particle size of from 0.25 to 1 µm, wherein the total rubber content of the composition is from 10 to 30 weight percent, stated on a 1,3,-butadiene/total composition weight basis and wherein the overall volume averaged particle size of the said capsule morphology particles and entanglement morphology particles is less than 0.5 µm; said method comprising:

a. dissolving from 5 to 30 parts by weight of a 1,3-alkadiene/monovinylidene aromatic block copolymer rubber being a 1,3-butadiene/styrene block copolymer having from 20 to 30 weight percent of styrene copolymerized therein in from 95 to 70 parts by weight of a monovinylidene aromatic monomer;

b. polymerizing the resulting rubber/monomer solution at a temperature of from 50 to 210°C, in the presence of from 0 to 1000 parts per million by weight of a polymerization initiator and from 0 to 2000 parts per million by weight of a chain transfer agent and under agitation conditions sufficient to provide capsule morphology and entanglement morphology rubber particles in the above-specified size ranges, with a proviso that the amounts of said initiator and chain transfer agent are not both zero; and

c. thereafter recovering the resulting rubbermodified monovinylidene aromatic polymer product.

6. A rubber-modified monovinylidene aromatic polymer composition which comprises, on a total composition weight basis, from 90 to 55 weight percent of a rigid monovinylidene aromatic polymer matrix and, dispersed within said matrix, from 10 to 45 weight percent of grafted and occluded 1,3alkadiene-based rubber particles, said particles being derived from a 1,3-butadiene/styrene block copolymer having from 20 to 30 weight percent of styrene copolymerized therein, and the said

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particles being composed, on a rubber particle weight basis of:

a. from 25 to 80 weight percent of rubber particles having a capsule morphology and a volume average particle size of from 0.1 to 0.4 μ m; and

b. from 75 to 20 weight percent of rubber particles having an entanglement morphology and having a volume average particle size of from 0.25 to 1 $\mu\text{m};$

wherein the total rubber content of the composition is from 10 to 25 weight percent, stated on a 1,3,-alkadiene/total composition weight basis, and

wherein the overall volume averaged particle size of the said capsule morphology particles and entanglement morphology particles is less than 0.5 μ m.".

Dependent claims 2 to 5 of this further amended set of claims correspond to the granted version of Claims 2 and 5 to 7, and dependent Claims 7 to 14 of this set correspond to Claims 9, 10 and 12 to 17 as granted.

III. In an interlocutory decision orally announced at the end of the above oral proceedings and issued in writing on 24 April 2002, the patent in suit as amended according to the Main Request (section II, above) was held to meet the requirements of the EPC. More particularly, the ground for opposition pursuant to Article 100(b) EPC was rejected, because the Opponent had not convincingly demonstrated that the patent in suit could not be carried out over the whole range of the claims according to the above Main Request. Nor were the arguments found convincing, according to which (i) the processes known from D1 and D2 and the claimed process were the same and (ii) any alleged difference in the morphology of the respective products could only be explained by an insufficiency of disclosure.

Consequently, besides the rejection of the ground for opposition pursuant to Article 100(b) EPC, novelty was also acknowledged for the above reasons.

The technical problem underlying the claimed subjectmatter was seen in the provision of an alternative process which allowed the preparation of compositions having good impact strength and good gloss.

On the basis of the findings (a) that neither D1 nor D2 suggested a process as claimed in the patent in suit, (b) that neither of them explicitly disclosed products having the desired morphology, (c) that it was not sufficient to demonstrate that a skilled man <u>could</u> have taken certain measures, but that it was of decisive importance whether he <u>would</u> have done so in the expectation of some improvement or advantage, and (d) that the Opponent had not discharged the burden of proof to demonstrate lack of inventive step, the Opposition Division concluded that the claimed subjectmatter was also based on an inventive step. IV. Against this interlocutory decision, a Notice of Appeal was filed by the Appellant/Opponent on 21 June 2002, and the prescribed fee was paid on the same date.

> In the Statement of Grounds of Appeal, received on 26 August 2002, the Appellant further pursued the requests for revocation of the patent in suit in its entirety for all of the grounds previously raised, ie (a) lack of novelty over either D1 or D2, (b) lack of inventive step in view of either D1 or D2, each in combination with general knowledge, and (c) insufficient disclosure. Additionally, (d) an objection under Article 123(2) EPC was raised against Claim 6.

In addition to documents D1 and D2, the Appellant referred to documents "D2'" and "D7", equal to D3 and D4, respectively, as mentioned in section II, above.

With a letter dated 18 September 2002, the Appellant submitted experimental reports and photomicrographs (referred to by the parties as "microphotographs"), referred to as repetitions of Examples 12 and 13 of D1 and as four repetitions of Example C of D2. Some additional data concerning the products of the latter experiments were provided by the Appellant in a further letter dated 29 October 2002. Moreover, in this letter, two further documents were cited:

- D5: A partial translation of JP-A-3-162 407 into English ("Exhibit A") and
- D6: Adolf Echte, "Teilchenbildung bei der Herstellung von kautschukmodifiziertem Polystyol", Angewandte

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Makromolekulare Chemie, 58/59 (1977), pages 175 to 198 ("Exhibit B").

In order to overcome the objection under Article 123(2) EPC, the Respondent suggested, in a letter dated 26 February 2003, to amend Claim 6 (section II, above), by replacing the upper limit of "25" of the total rubber content by "30". However, contrary to a statement to this end in the letter, an amended version of the request was not enclosed. Beyond that, the Respondent disputed all the arguments and the experimental results of the Appellant.

- V. Oral proceedings were held on 9 March 2005 in the presence of both parties.
 - (a) At the beginning of these proceedings, the Respondent stated, when asked for its requests, that its Main Request was directed to the dismissal of the appeal. The amendment of Claim 6 mentioned in the last paragraph of section IV, above, should form part of an Auxiliary Request, not yet submitted in writing. This further request was, however, withdrawn by the Respondent when the debate, having dealt with all the objections raised by the Appellant against the above Main Request, had been closed with regard to the Main Request.
 - (b) The parties agreed to the following definitions of the particle morphologies as used in the patent in suit (page 3, lines 40 to 42 and 43 to 45), namely the capsule and entanglement particle morphologies: Thus, on the one hand, *labyrinth*, *coil*, *onion skin*

and concentric circle morphologies have, in general, the same meaning as entanglement, but are different from cellular morphology. On the other hand, the terms single occlusion and core/shell refer to the same morphology as the term capsule. Moreover, as set out in the patent in suit (page 2, line 29), cellular particles have been addressed in literature as salami particles. This latter term had, according to the submission of the Appellant dated 29 October 2002 (with reference to D5) and as accepted by the Respondent, been mistranslated in D1 as racemi (D1: page 6, line 5).

- (c) The Appellant maintained its objection to Claim 6 under Article 123(2) EPC with regard to the total rubber content of from 10 to 25 % by weight. In addition, it raised a similar objection with respect the corresponding range of from 10 to 30 % by weight of the corresponding feature in Claim 1. Although it was conceded that these numeric values per se, as used in Claims 1 and 6, had been disclosed in the application as filed, the new range limits were seen by the Appellant as new combinations of individual values selected from two lists. These selections should not be allowed.
- (d) According to the Appellant, the same starting compounds were used and treated under the same conditions in the patent in suit and in each of D1 or D2 so that the results should also be identical. Any alleged difference in the morphology of the respective products could, therefore, only be explained by an insufficiency of disclosure (Article 100(b) EPC), because the particulars

necessary to achieve this difference were missing from the disclosure of the patent in suit.

In contrast thereto, the Respondent argued, that the patent in suit (page 4, line 51 to page 5, line 7) disclosed the various influences which the initiator and the chain transfer agent imparted to the polymerisation reaction and the morphology of its product, as eg shown in the examples of the patent in suit. This information was consistent, according to the Respondent, with the statements in D1 (page 6, lines 3 to 10), on which the Appellant had relied for presenting its case with respect to the morphology of the rubber particles in the final product. Moreover, the Appellant's arguments in its letter dated 29 October 2002 (starting in the last paragraph of page 2 of the letter) showed that the skilled person knew how to vary the reaction conditions in order to obtain a product having a particular morphology. In view of the influence of the geometry of the production plant on the reaction and its product, a demand for a numeric definition of all the process features, eg the stirring conditions, in order to obtain the required product under all conceivable marginal conditions would, however, result in an undue restriction of the claimed subject-matter. The Respondent concluded that the patent in suit provided ample information about how to obtain the required microstructure of the product.

(e) With regard to novelty vis-à-vis D1 or D2, the Appellant again argued that the same starting compounds had been treated in the same way. This should, in consequence, have resulted in identical products. Moreover, specific reference was made by the Appellant to various particulars given in D1, concerning eg percentages, reaction temperatures, initiators, chain transfer agents and stirring (D1: page 2, lines 49 to 51; page 4, lines 14/15 and 17/18; page 6, lines 3 to 10) to show overlap with the patent in suit.

The Appellant saw this view also confirmed by the repetitions of Examples 12 and 13 of D1 (section IV, above), in particular by that of Example 13, because their products had been shown, in its view, to fall within the scope of the present claims. Thus, the HIPS product according to the latter repetition was said to contain capsule and entanglement rubber particles in a ratio of about 50:50 and to have an overall average size of below 0.5 µm as shown in the submitted photomicrograph (section IV, above). In order to identify the "entanglement particles" on the photomicrograph, the Appellant marked particles on a copy thereof, which was submitted in the oral proceedings.

Moreover, it had been known according to the Appellant that a 60/40 mixture of styrene/ butadiene block copolymer rubber ("SBR"), which contained 40 % by weight of styrene, and of polybutadiene homopolymer as used in the Example 13 of D1 would give the same results as SBR having a styrene content of 30 % by weight. However, this statement was disputed by the Respondent. Likewise, the products of the four repetitions of comparative Example C of D2 (section IV, above; with two chain transfer agents and in two polymerisation temperature profiles, respectively) also complied, according to the Appellant, with the definitions of the claimed product as shown in the table and on the photomicrographs submitted.

Moreover, the Appellant argued that it had generally been known how to control the morphology of the rubber particles. As indicated in Fig. 16 of D3 and on page 194 of D6, such a control would include not only the preparation of one occlusion (capsule) particles and of cellular ("racemi") particles (D1: page 6, lines 3 to 10) but also that of intermediate particle forms, *viz.* of coil (entanglement) particles (section V(b), above).

By contrast, the Respondent argued that the specific morphology of the rubber particles had been disclosed in neither D1 nor D2. In fact, neither of the two documents even mentioned "entanglement particles" in the sense of the patent in suit. Rather, D1 referred only to particles having "one occlusion structure" or "racemi structure", and D2 aimed only at "core/ shell" rubber particles.

Consequently, a valid novelty objection could have been based only on the argument that the products as claimed had been the inevitable result of true repetitions of the examples of D1 or D2. However, whilst accepting that the experimental report and D3 might be considered by the Board, the Respondent disputed that those additional experiments of the Appellant had been true ("photographic") repetitions of the prior art, and it objected to an introduction of the late-filed German language document D6 in the proceedings.

In both alleged repetitions of Examples 12 and 13, the styrene content in the SBR used was 40 % by weight, ie outside the range of 20 to 30 % by weight as defined in Claims 1 and 6. Moreover, the SBR was used in admixture with polybutadiene rubber. Both rubbers dissolved differently and separately in the styrene monomer solution and initiated the formation of separate forms of particles. Furthermore, the Respondent disputed that the photomicrograph of Example 13 showed that about half of the rubber particles had an entanglement morphology.

As regards the alleged repetition of Example C of D2, the Respondent argued that this product did not meet the overall particle size requirement according to the claimed subject-matter.

However, neither D1 nor D2, in general, nor their examples disclosed the particular combination of features as defined in the claims, let alone the combination of (i) the specific SBR, (ii) the total rubber content, (iii) the two rubber particle populations of (iv) particular morphology, (v) each in specific amounts and (vi) having specific volume average particle sizes, and (vii) the overall average particle size of the rubber.

(f) The technical problem underlying the patent in suit was seen by the Appellant in providing products showing a good balance of gloss and Izod impact strength. However, D1, assumed to represent the closest state of the art, provided a clear teaching in this direction. In particular, reference was made to page 6, lines 3 to 10.

Moreover, Examples 1, 5 and 8 of the patent in suit related no longer to subject-matter within the scope of the claims as amended, and amongst the remaining examples only Examples 2, 6, 9 and 10 provided experimental data concerning both the gloss and the Izod impact strength. However, only poor values for the latter property had been achieved in two of these remaining examples, ie in Examples 6 and 9. Only Examples 2 and 10 showed, according to the Appellant, a combination of the two properties in question similar to those achieved in Example 13 of D1. Consequently, the claims did not provide any surprising results, nor was the technical problem solved by all products within the scope of Claim 1.

The Respondent did not see any suggestion in either D1 or D2 to follow the road chosen in the patent in suit.

(g) Neither party wished to additionally comment on the product claims.

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

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

- 1. The appeal is admissible.
- 2. Since D3 had already been referred to by both parties (see sections II and V(e), paragraph 8, above), whilst D6 was only cited in the appeal proceedings and was indicated to present the same teaching as D3 (Appellant's letter dated 29 October 2002: page 2, paragraph 2), the Board came to the conclusion not to take D6 into account (Article 114(2) EPC).
- 3. Wording of the claims
- 3.1 The subject-matter of Claim 1 of the sole request under consideration differs from that of Claim 1 as granted by the limitations of
 - (i) the total rubber content of the composition to from 10 to 30 weight percent, stated on a 1,3butadiene/total composition weight basis, and
 - (ii) the overall volume averaged particle size of both rubber particle populations to less than 0.5 $\mu m.$ Moreover,

(iii)the 1,3-alkadiene-based rubber has been identified as being a 1,3-butadiene/styrene block copolymer having from 20 to 30 weight percent of styrene copolymerised therein.

Independent Claim 6 has been amended in the same way, except for the range of the total rubber content being 10 to 25 weight percent (feature (i)).

- 3.1.1 Whilst the amendments of the above features (ii) and (iii) were not disputed between the parties, the Appellant asserted that the definitions of the ranges of feature (i) contravened Article 123(2) EPC (section V(c), above).
- 3.1.2 With regard to features (ii) and (iii), reference can be made to page 8, lines 17 to 20, and to Claims 3 and 4, respectively, of the application as filed. The passage concerning feature (i) on page 6, lines 35 to 38, of the original application text read as follows:

"The total rubber content of the rubber modified monovinylidene aromatic polymer compositions hereof, stated on a 1,3-alkadiene weight basis only, is typically from 5 to 30 (preferably from 8 or 10 to 20 or 25) weight percent on a total rubber modified polymer composition weight basis.".

3.1.3 Besides a general range of from 5 to 30 % by weight, this passage, thus, discloses *expressis verbis* two alternatives for each of the lower and the upper limits of the preferred range. This fact was not disputed by the Appellant, who, however, contended that the ranges of "10 to 25" and "10 to 30" in Claims 1 and 6, respectively, were the result of selections from two lists and, therefore, not allowable.

- 3.1.4 According to established jurisprudence (cf. Case Law of the Boards of Appeal of the EPO, 4th Edition, 2001, III.A.3.3; in particular T 2/81: No. 3 of the reasons, OJ EPO 1982, 394; and T 925/98 of 13 March 2001: No. 2 of the reasons, not published in OJ EPO), the disclosure of a quantitative range of values together with an included preferred narrower range also directly discloses the possible part-ranges lying within the overall range (here the "typical" range) on either side of the narrow (preferred) range, and that, hence, a simple combination of the preferred narrower range and one of these part-ranges is also unequivocally derivable and is supported by the disclosure (cf. Headnote 2 of T 2/81, above).
- 3.1.5 This Board concurs with the above finding also with regard to the present situation. Thus, the range of 10 (preferred limit) to 30 (overall range limit) weight percent in Claim 1 is based on the original disclosure. Moreover, this conclusion is also valid for the range of 10 to 25 weight percent in Claim 6, since it is formed from one of only two preferred lower limits (close to each other) and one of only two preferred upper limits (close to each other), both disclosed *expressis verbis* in the passage quoted above.
- 3.2 Furthermore, the additional features (i), (ii) and (iii) further limit the scope of both independent claims.
- 3.3 Therefore, both the requirements of Article 123(2) and(3) EPC are met.

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4. Closest state of the art

- 4.1 The patent in suit relates to HIPS compositions showing a good balance of gloss and Izod impact strength and their preparation (page 3, lines 1 and 13 to 15).
- 4.2 The products obtained by the process of D1 are described to be compositions of this kind being "excellent in a physical balance". They have eg high impact strength, and further excellent gloss and stiffness (D1: page 2, lines 3 to 6). Therefore, the document, according to the Appellant, represents the closest state of the art.

The Board has no reason to take a different view.

4.2.1 The rubber used in the known process comprises a styrene-butadiene-based block copolymer rubber (SBR) having a styrene content of 20 to 50 % by weight (page 3, lines 14/15). According to D1, a styrene-based monomer solution containing 5 to 20 % by weight of the rubber component is supplied to a first polymerisation zone, in which the initial polymerisation is conducted to a conversion of 10 to 30 %. Thereafter, polymerisation is continued in a second polymerisation zone to a conversion of 30 to 70 %, wherein, in order to achieve the above required properties, it is necessary, at the same time, (i) to cause phase inversion to occur, (ii) the rubber particle diameter to be controlled to a optimum range, ie to from 0.2 to 1.5 μ m, and (iii) the particle size distribution, ie the ratio of the area average rubber particle diameter/number average rubber particle diameter Ds/Dn,

to be narrowed to not more than 2.5 (D1: Claim 1; page 2, lines 11 to 13). For controlling the rubber particle size and size distribution, it is suitable to apply a specific average shear speed in the second polymerisation zone (D1: page 4, lines 53 to 57).

According to Claim 2 (and page 3, lines 41/42) of D1, the rubber component preferably comprises, in addition to the above SBR, a polybutadiene (homopolymer) rubber in an amount of not more than 50 % by weight.

- 4.2.2 If necessary, chain transfer agents, antioxidants, solvents, mineral oils, silicone oils and organic peroxides may be added to the polymerisation mixture. The rubber particle diameter may be narrowed by using an organic peroxide (D1: page 4, lines 17 to 21). The presence of a chain transfer agent is, however, only referred to in Examples 10, 11 and 12, that of an initiator is only mentioned in Example 13.
- 4.2.3 The shape of rubber particles formed can be controlled appropriately from the one occlusion structure (ie capsule morphology) to the "racemi" structure (ie cellular morphology, cf. section V(b), above, and D5) by the following process features: rubber mixing ratio, the method of applying the shear force, the chain transfer agent and the amount of the catalyst (initiator) added (D1: page 6, lines 3 to 6).

Thus, an excellent gloss can be achieved by controlling the rubber particle diameter to 0.2 to 0.8 μ m and the particle size distribution to not more than 1.8 to give a product wherein at least 80 % of the rubber particles have a capsule morphology (D1: page 6, lines 7 to 10). 4.2.4 Four different SBR polymers and one butadiene homopolymer rubber were disclosed for use as rubbers in the examples and comparative examples of D1. The SBR polymers had styrene contents of 40 ("A-1"), 22 ("A-2"), 10 ("A-3") and 55 ("A-4") % by weight, respectively.

> Whilst, in Examples 5 and 8 and in Comparative Examples 5, 8 and 9, the SBR types ("A-1" and "A-4" having styrene-contents of 40 and 55 % by weight, respectively), were used as the sole rubber, in all the other examples and comparative examples of D1, the respective SBR types were used in admixture with the polybutadiene homopolymer rubber.

> Only the styrene content of SBR "A-2" as used in Examples 1, 2 and 14 and in Comparative Examples 1 and 2 lay in the range of from 20 to 30 % by weight. As indicated in the previous paragraph, this rubber was, however, only used in combination with additional polybutadiene homopolymer rubber. Moreover, the average diameters of the rubber particles in these latter examples and comparative examples exceeded, however, the limit of less than 0.5 μ m as required in Claim 1.

4.2.5 None of the examples and comparative examples in D1 contains any information concerning the morphology of the rubber particles in the final product. Only in Examples 5 (0.32 µm) and 13 (0.48 µm), an average particle diameter of less than 0.5 µm was disclosed. In Example 5, the SBR "A-1" (containing 40 % of styrene) was used alone, in Example 13, a 60:40 mixture of this SBR and polybutadiene was used; and Comparative

Example 8 (0.15 $\mu\text{m})$ was based on the use of SBR "A-4" (containing 55 % of styrene).

- 4.2.6 Further particulars of the polymer products given in the examples and comparative examples were measurements of the gloss and the Izod impact strength (determined according to JIS K-7105 and JIS K-7110, respectively), particle diameter distribution Ds/Dn, the drop weight strength, the flexural modulus and the weight average molecular weight (Table 3 of the document).
- 5. Novelty
- 5.1 In order to demonstrate the alleged identity of the subject-matter claimed in the patent in suit and of the subject-matter of D1, the Appellant had initially submitted a repetition of Example 5 of D1 (Notice of Opposition dated 7 July 1997) but did not provide the photomicrograph, which would have enabled the Patent Proprietor and the Opposition Division to verify the data presented in the Notice of Opposition for the volume average particle diameter Dv of the rubber particles and for their morphology. However, in reply to the above experimental data, the Patent Proprietor submitted on the basis of that Example 5 of D1 five new examples (in the absence of both initiator and chain transfer agent as in Example 5 of D1, or in the presence of one or both of these components) in order to investigate whether it was possible to prepare a composition having the morphology specified in the patent in suit. According to photomicrographs enclosed, the products of these additional examples showed rubber particles having core/shell, dot or combinations of core/shell and rod morphologies, but never a

combination of capsule and entanglement particles as required in the patent in suit (letter dated 8 March 2000). None of these experimental reports was further taken into consideration for the decision under appeal.

Nor does the Board see, under these conditions, any reason to decide otherwise or, therefore, to accept the repetition of Example 5 of D1, as filed by the Opponent, as a convincing proof for the alleged anticipation of the claimed subject-matter by D1. On the contrary, firstly, Example 5 of D1 did not comply with the requirements of the method of Claim 1, because of the absence of both initiator and chain transfer agent in that example, secondly, no proof had been provided by the Opponent for the asserted morphology of the product obtained in its experimental report, and, thirdly, the photomicrographs of the experiments of the Patent Proprietor showed that the required morphology as required in Claim 6 of the patent in suit was never achieved with or without initiator and/or chain transfer agent.

5.2 In letters dated 18 September 2002 and 29 October 2002, the Appellant specifically referred to Example 12 and more particularly (because of the rubber particle size of 0.48 µm disclosed in Table 3 of D1) to Example 13 of D1. Since in neither of these examples, however, mention was made of the morphology of the rubber particles, nor of a presence of different rubber particle populations in the final composition, an experimental report containing repetitions of these examples of D1 was filed with the first of the two above letters. In the further discussion, the Appellant focused its arguments, in particular, on Example 13 and its repetition as filed.

5.2.1 The photomicrograph provided with the report showed, according to the Appellant, the presence of both core/shell and entanglement rubber particles in amounts of "about 50/50". This was, however, disputed by the Respondent with regard to both the morphology and the quantitative distribution of those particles in the photomicrograph. Therefore, particles, which it considered to be entanglement particles were identified by the Appellant, in reply to an invitation of the Board, on a copy of the photomicrograph. The Respondent, however, maintained its above objections in view of this copy (cf. also section V(e), above).

> Nor can the Board derive from the above supplemented photomicrograph that a "visual analysis confirms the presence of both coreshell [*sic*] and entanglement rubber particles (about 50/50) and an average particle size diameter Dv of about 0.5 micron." (experimental report filed with the letter of 18 September 2002; page 9, last paragraph of "Example 13 of D1"). Consequently, it cannot concur with this argument of the Appellant.

5.2.2 Moreover, as already pointed out (section 4.2.4, above), in Examples 12 and 13 of D1 and in their above repetitions, mixtures of SBR having a total styrene content of 40 % by weight and polybutadiene were used.

> Both independent Claims 1 and 6 of the patent in suit require, however, the rubber used to be SBR having a styrene content of from 20 to 30 weight percent.

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When this difference was addressed, the Appellant asserted that it had been common general knowledge that a 60:40 mixture of SBR (containing 40 % by weight of styrene) and polybutadiene (as used in Example 13 of D1) would give the same results as SBR itself, which met the above relevant requirement in the patent in suit. This contention was, however, disputed by the Respondent, according to whom, each of these rubbers initiated the formation of separate forms of particles (section V(e), paragraph 9, above).

- 5.2.3 Consequently, this argument of the Appellant is not convincing for the Board, and neither of these experiments, therefore, convincingly demonstrates that the claimed subject-matter of Claims 1 and 6 lacks novelty over D1.
- 5.3 Another novelty objection was raised, by the Appellant on the basis of Comparative Example C of D2. As this comparative example does not disclose all features to prove lack of novelty either, the above experimental report filed by the Appellant additionally contained a "Table 1 : E.P. 143500: repetition of EXAMPLE C" listing the properties of the products of four experiments and four photomicrographs relating to the "T.E.M. Analysis" of the products of the four experiments of Table 1. The table was further supplemented by letter dated 29 October 2002. (section IV, paragraph 4, above).
- 5.3.1 Unlike the above repetition of Example 13 of D1, the latter photomicrographs of the products of this Table 1 were accepted by the Respondent to show the presence of

both capsule and entanglement particles. However, the data in the above Table 1, as argued by the Respondent, show that none of the experiments fulfilled *all* the requirements of the present claims (section V(e), last two paragraphs, above). In particular, the "overall morphology" values in all the experiments exceeded the limit of "less than 0.5 μ m" as required in the claims of the patent in suit.

- 5.3.2 Furthermore, the table in D2 (page 15) describing the properties of the products obtained in the examples and comparative examples of the document clearly defines the morphology of the dispersed rubber phase in the product of comparative Example C as "shell/core". This is clearly contrary to the photomicrographs of the experiments in Table 1 submitted by the Appellant.
- 5.3.3 Apart from the above findings, the general description of the experiments, the results of which are shown in Table 1, mentioned above, deviates from the text describing Example C in D2 in that reference is additionally made to the presence of an antioxidant and of zinc stearate.

Moreover, according to Example 1 of D2, "The polymerization temperatures are controlled from 115 to 175 °C and the conditions controlled such that at phase inversion, rubber particles of a shell/core morphology having a desirably small particle size are formed.". In Comparative Example C, it is said that "A rubberreinforced polystyrene is prepared using the techniques described in Example No. 1 except that the initial monomer/rubber solution contained 0.1 percent of a chain transfer agent and only 0.012 percent of the tert-butylperoxybenzoate initiator which, at the other polymerization conditions employed, did not give the desired rubber particle size."

As admitted by the Appellant, the description of the examples in D2 does not provide all details which have or may have an influence on the properties of the final product. Therefore, four different variants of Example C were carried out in the experimental report filed by the Appellant, using two different mercaptan chain transfer agents (TDM and NDM) at different temperatures in the two polymerisation stages. Thus, "The polymerization temperatures were controlled from 125°C to 170°C.", and in Table 1 the polymer temperatures obtained in the two stages were given as 160 and 170°C, and 150 and 160°C, respectively.

- 5.3.4 Therefore, the above repetitions of (comparative) Example C suffer from the deficiencies that (i), their results, as shown in the photomicrograph, are not comparable with those described in D2, whether as to size or morphology of the products, (ii) the procedure used deviated from the text of Example C in D2, and (iii) details of the procedure in D2 had to be supplemented by Appellant, thus demonstrating that Example C could not be repeated as disclosed in D2.
- 5.3.5 The conditions to be met by repetition of examples in order to be valid duplications of prior art are referred to in case law. Thus, deficiencies of the kind as mentioned in the previous paragraph led to the conclusion by the Board, in the case of decision T 969/91 of 8 February 1995 (not published in OJ EPO; Nos. 3.2.1.2 and 3.2.1.3), that experiments allegedly

repeating a Run 9 disclosed in a document (8), referred to in that case, had not been "true duplications, partly because it did not exactly follow the procedure according to document (8) and partly because (8) does not disclose all information necessary for an exact reworking." and "In order to be valid as novelty anticipation, there must not be any doubt that the product obtained and tested is identical to the one duplicated.".

- 5.3.6 Hence, it follows that the experiment submitted by the Appellant as a "Repetition of Comparative Example C" is not a true duplication of Comparative Example C of D2. Nor is it, consequently, prior art which could serve to prove lack of novelty vis-à-vis this document.
- 5.3.7 Furthermore, D2, itself, does not contain any hint to HIPS type compositions containing two populations of capsule and entanglement rubber particles, let alone two rubber populations which would comply with the limitations as defined therefor in the independent claims of the patent in suit.
- 5.4 In view of the above facts and findings, the Board takes the view that lack of novelty has not been convincingly demonstrated. Therefore, the subjectmatter complies with the requirements of Article 54 EPC.

6. Insufficiency of disclosure

Having assessed novelty on the basis of the arguments and evidence provided by the Appellant as set out above, it is now possible to decide on a further ground for opposition, maintained by the Appellant in the appeal, ie an alleged insufficiency of disclosure.

- 6.1 The key argument of the Appellant in this respect had been that, since the same starting materials had been treated in D1 and D2 in the same way as in the patent in suit, any asserted differences between the final product of the patent in suit and those of those two documents could only be based on some feature not disclosed in the patent in suit. This would however, amount to an insufficiency of disclosure in the sense of Article 100(b) EPC (section V(d), above, paragraph 1).
- 6.2 The Respondent, by contrast, referred to the examples in the patent in suit and to page 4, line 43 to page 5, line 7 of the patent in suit (page 6, lines 10 to 28 of the application text), which provided the teaching necessary for the skilled person to adjust the polymerisation parameters for a particular design of the production plant used to produce the desired product. This teaching was even consistent with the information in D1, which had been found sufficient by the Appellant (cf. section V(d), above, paragraph 2).
- 6.3 Having regard to the above diverging arguments of the parties, document D1 (page 6, lines 3 to 10), as well as the arguments of the Appellant based thereon (letter dated 29 October 2002: page 2, last paragraph) show, in the Board's view, that the person skilled in this art has been familiar with functional limitations of the kind to be found in Claim 1 under consideration, ie the definitions in step b referring to the amounts of initiator and/or chain transfer agent and to the

polymerisation conditions (temperature and stirring). This view is also supported by D2 (Example 1: page 12, lines 30 to 33).

- 6.4 Furthermore, as shown in the above passages relating to the question of novelty (sections 5 to 5.4, above), it was not convincingly demonstrated by the Appellant that, in fact, the same starting materials had been reacted in the above prior art in exactly the same way as in the patent in suit (section 6.1, above). Rather, it was established that neither the rubber component used in the repetitions of D1 as starting material was identical to that required in the patent in suit, nor that the procedure followed in D2 was the same as the method according to the patent in suit.
- 6.5 Consequently, the Board has come to the conclusion that the Appellant has not convincingly demonstrated that the decision under appeal was wrong with respect to the objection under Article 100(b) EPC.

The ground for opposition under Article 100(b) EPC is, therefore, rejected.

- 7. Problem and solution
- 7.1 In the decision under appeal, the technical problem was formulated as being directed to the provision of an alternative process which allowed the preparation of compositions having good impact strength and good gloss (item II.6.2 of the decision under appeal).

7.2 Since it considered the results according to the patent in suit to be better than those of D1, the technical problem, according to the Respondent, related to the provision of HIPS composition having very high gloss and improved impact strength.

- 7.3 However, in the context of the questions of which technical problem was underlying the patent in suit and of whether this problem had been solved by the subjectmatter as claimed, the Appellant argued that former Examples 1, 5 and 8 no longer fell within the scope of the claims and had been indicated to be comparative, and that only Examples 2, 6, 9 and 10 gave measurements for both gloss and impact strength.
- 7.3.1 Therefore, Examples 3, 4 and 7 could not be taken into consideration. Only Examples 2 and 10 included properties of the respective products which were similar to those of Example 13 of D1, whereas the impact strength results in Examples 6 and 9 were poor. Therefore, the Appellant concluded that the claims encompassed embodiments which did not solve the above problem. Nor did the claims provide any surprising results as shown by Examples 1, 5 and 8 (section V(f), above).
- 7.3.2 These arguments were disputed by the Respondent, who stated that the patent in suit, in general, was directed to high gloss HIPS products. Furthermore, the measurements of the Izod impact strength reported in D1 (JIS K-7110 standard) were carried out on injection moulded specimen whereas the "rest of the world" used compression moulded specimen, and this was true, in particular, for the patent in suit (page 7, line 20).

The Izod measurements on injection moulded specimen would, however, give higher Izod impact strength values than the compression moulded specimen. The Appellant, who stated that it did not know the particulars of the Japanese standard, did not dispute that different standards used in measurements of a parameter might give different results not directly comparable with each other, namely when specimen of different sizes were used.

7.3.3 It has not been shown by the Appellant, on whom, as the Opponent, the onus of proof lay, that Examples 3, 4 and 7 of the patent in suit, in fact, failed with respect to gloss.

> A comparison of the particulars of the Examples 2 and 3 in Table I of the patent in suit shows that the initial monomer/rubber solutions were nearly the same, with only a slight difference in the content of mineral oil. This resulted in only small variations in morphology of the respective dispersed rubber phases of the final compositions and their particle sizes, ie the average sizes of the two particle populations as well as the respective overall volume average particle size. Since the rubber particle sizes are generally known to have a decisive influence on gloss (cf. D1: page 2, lines 16 and 17; page 6, lines 7 to 10), the Board has no reason to assume that Example 3 failed in this respect.

> As regards the differences in the Izod impact strength between these examples, it must be noted that the rubber phase in Example 6 has a distinctly smaller overall volume average particle size as well as smaller

average particle sizes of both rubber particle populations.

Although not directly comparable with the above examples, due to a different quantitative composition, the above considerations are also corroborated by the results of Example 9 in Table II of the patent in suit.

The above considerations concerning the interference between particle size parameters and impact strength find further support in Example 7 of the patent in suit, which contained rubber particles having distinctly smaller particle sizes, as compared to the rubber particles in Examples 1 and 4 of the patent in suit. The comparison of the particle sizes reported in these three examples cannot, however, convincingly lead, in view of the above considerations concerning the gloss, to the assumption that Examples 4 and 7 in comparison to (comparative) Example 1 (reporting a gloss value of 84) had failed with regard to this property.

Nor can (comparative) Example 8 showing rubber populations, each having larger particle sizes in comparison to those previously mentioned (showing a minority having a size average of 0.36 and a majority having an average of 1.02 μ m, respectively), invalidate the above considerations, since it corresponds to those compositions mentioned on page 2, lines 49 to 52 of the patent in suit, according to which it had been generally accepted before, that at least some minimum portion of the dispersed rubber particle populations was to have a size of from 1 to 2 μ m in order to obtain a good toughness (eq Izod impact strength). 7.4 Although, for the reasons given above, the Board does not accept that only Example 2 can be considered as having dealt with the relevant technical problem (see sections 7.1 or 7.2, above), it is evident that the experimental data in the patent in suit (namely, the impact strength values ranging from 25.8 to 149.9 J/m; eg Example 6: gloss 97, Izod 37.3 J/m) extend over a large range of values and that there is some overlap in this respect with the closest prior art (see Table 3 of D1, eg Example 5: gloss 97, Izod 9.0 kg-cm/cm, equal to approximately 90 J/m).

> In view of these data it is somewhat doubtful for the Board whether *improvements* of the combination of gloss and impact strength are achieved in the whole range covered by the claims (cf. section 7.2). Therefore, the above formulation of the technical problem as adopted by the Opposition Division is also used by the Board for the assessment of inventive step (section 7.1, above).

In this context, it must be borne in mind that the achievement of a technical advantage or improvement is not, according to the EPC, a prerequisite for grant of a patent.

7.5 In view of the results of the examples in the patent in suit, considered above, the Board is satisfied that the above technical problem (section 7.1, above) has been solved by the subject-matter of the two independent Claims 1 and 6.

8. Inventive step

It remains to be decided whether the claimed solution of this problem derives in an obvious way from the cited documents.

- 8.1 In the Appellant's opinion, D1 gave a clear teaching in the direction of controlling the shape of the rubber particles from capsule (one occlusion) to "racemi" (salami = cellular) structures, and intermediate structures at phase inversion, in order to achieve a HIPS having a good balance of gloss and impact strength. With respect to the different rubber particle structures, the Appellant referred to Fig. 16 of D3, against the consideration of which the Respondent raised no objection, in order to demonstrate general common knowledge.
- 8.1.1 In D1 (cf. sections 4.2.1 and 4.2.4, above), emphasis is repeatedly put on the importance of a rubber particle diameter in the range of from 0.2 to 1.5 µm and a narrow particle size distribution Ds/Dn, as well as on their control during the polymerisation (cf. Claim 1; page 2, line 55 to page 3, line 2; page 3, lines 18 to 20, 45 to 47; page 4, lines 20/21 and 53; page 6, lines 7 to 10). This can also be seen in the results of the examples in comparison to the broadened particle size distributions in those comparative examples, in which the polymerisation had to stopped because of lack of control.
- 8.1.2 Although on page 6, lines 3 to 6, mention is made of the possibility to control these features and, in addition, also to control the "shape of the particle

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formed", the document never mentions, nor suggests to control the particle size in such a way so as to obtain a product containing two distinct rubber particle populations each fulfilling the requirement of having a specific volume average size and, at the same time, showing an overall volume average particle size of all the dispersed rubber particles of less than 0.5 μ m. Reference can be made in this respect to the results in Table 3 only referring to "Shape of Rubber" expressed in terms of one particle diameter and the particle diameter distribution Ds/Dn.

8.1.3 Moreover, in view of the photomicrograph marked by the Appellant in the oral proceedings (cf. section 5.2.1, above), the Board takes the view that, in the repetition of Example 13, the shape of the rubber particles apparently complies with the statement on page 6 of D1. This latter passage describes the possibility of achieving high gloss when controlling the particle diameter to 0.2 to 0.8 µm and the particle size distribution Ds/Dn to not more than 1.8, so that a HIPS results in which at least 80 % of the rubber particles have a one occlusion structure (ie core/ shell or capsule morphology; D1: page 6, lines 7 to 10).

> With regard to the shape of the remainder of rubber particles, the document refers only to the "racemi" structure. Other morphologies, let alone the simultaneous presence of particles having capsule and entanglement structures, respectively, have never and nowhere been considered or suggested in the document.

- 8.1.4 Therefore, this document itself does not give the slightest hint to modify its teaching in such a way so as to arrive at something within the scope of the claims under consideration, for any reason, let alone in order to solve the relevant technical problem.
- 8.2 Therefore, it remains to be investigated whether any one of the further documents relied upon by the Appellant provides such an incentive.
- 8.2.1 Having regard to the argument of the Appellant on the basis of D3, Figure 16, it has to be examined whether this disclosure or the common general knowledge would have suggested to the skilled person to prepare a HIPS composition containing rubber particles of a shape different from that of D1, containing in particular an "intermediate" particle form, viz. entanglement (coil) particles (section V(e), paragraph 5, above).

Figure 16 of D3 describes graphically and in very general terms that it had been found that polybutadiene and styrene/butadiene block copolymers of different styrene contents used in HIPS preparation have an influence on the morphology of the resulting dispersed rubber particles. According to the photographs displayed in Figure 16 and the 1 µm indicator at the lower right corner of the right-most picture, SBR initially having a cylindrical morphology can be converted into HIPS having a coil structure with particles sizes of distinctly more than 1 µm in the absence of grafting, or into HIPS having a capsule structure with grafting. No reference is made in this context to any influence those particles may have on the properties of the respective compositions. Furthermore, no basis is found therein for the argument of the Appellant that the coil structure as disclosed in Fig. 16 of D3 should be understood as being an intermediate morphology or structure to be achieved when controlling the polymerisation before and at phase inversion from "one occlusion" to "racemi" structure as indicated in D1.

Therefore, on the basis of D3, any conclusion reaching beyond the acknowledgement of the prior art on page 2, lines 49 to 52 of the patent in suit ("In connection with the foregoing, it has heretofore been generally accepted, conventional wisdom that it was essential in order to obtain good toughness (e.g., Izod impact strength) properties in mass, solution or mass/ suspension polymerized high impact polystyrene (HIPS) resin compositions that there be present therein at least some minimum portion of a dispersed rubber particle population in the size range from 1 to 2 μ m or greater.") can only be made on the basis of an *ex post facto* analysis, ie in the knowledge of the solution presented by the patent in suit.

In the Board's view, the general information of Figure 16 in D3 does not provide any hint to modify the clear teaching of D1 so as to arrive at something within the scope of the claims in order to solve the relevant technical problem.

8.2.2 When considering document D2 in respect of the desired gloss of the HIPS composition, it is noteworthy to remember (section 8.1.3, above) that, according to the recommendation in D1, at least 80 % of the rubber particles should have a capsule morphology. This is consistent with the teaching of D2, which requires that the discrete rubber particles of its HIPS compositions have a core/shell morphology and a volume average particle size of from 0.1 to 0.25 µm. Furthermore, D2 clearly discourages to consider any morphology other than core/shell (one occlusion, capsule) as demonstrated by the less good results of the comparative examples, the compositions of which included cellular or rod/sphere rubber particle morphologies. Like D1, D2 does not even mention entanglement morphology.

Consequently, this document does not, in the Board's view, provide any incentive either to modify the teaching of D1 in such a way so as to solve the relevant technical problem and thereby to arrive at something within the scope of the claim.

Moreover, as regards the argument of the Appellant concerning D2 in conjunction with the disclosure of D3, the same findings as described in section 8.2.1, above, are valid. D3 does not provide any incentive to deviate from the clear teaching of D2 in respect of the particle morphology. Moreover, in respect of the rubber particle size, Fig. 16 of D3 is even inconsistent with D2 (requiring a particle size of from 0.1 to 0.25 µm).

8.2.3 Any argument that the skilled person would, on the basis of D1 alone or in combination with D2, with or without consideration of D3, be led to arrive at something within the scope of the present claim would, in the Board's view, lack support by these documents.

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9. It follows that, besides the requirements of novelty and sufficiency of disclosure, the subject-matter of present Claims 1 and 6 according to the request of the Respondent also fulfils the requirement of Article 56 EPC, it involves an inventive step.

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10. By the same token this finding is also valid for the remaining dependent claims which include the same features as discussed above with respect to either Claim 1 or Claim 6.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

R. Young