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D E C I S I O N
of 18 May 2000

Case Number: T 0925/97 - 3.3.3

Application Number: 92305885.3

Publication Number: 0524731

IPC: C08L 69/00

Language of the proceedings: EN

Title of invention:
Polymer blends of polycarbonate-polysiloxane block copolymers with polycarbonates and polyestercarbonate copolymers

Applicant:
GENERAL ELECTRIC COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - problem and solution approach - ex post facto analysis"

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0925/97 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 18 May 2000

Appellant: GENERAL ELECTRIC COMPANY
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 25 February 1997 refusing European patent application No. 92 305 885.3 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
J. De Preter

Summary of Facts and Submissions

I. The European patent application No. 92 305 885.3 filed on 25 June 1992, claiming the priority of 1 July 1991 of an earlier application in the United States of America (724023) and published under No. 0 524 731 on 27 January 1993 (Bulletin 93/04), was refused by a decision issued in writing on 25 February 1997.

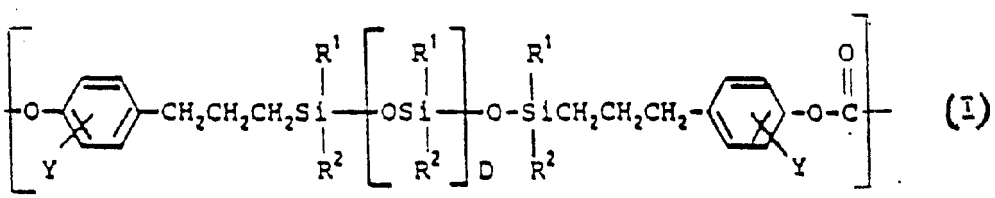
The decision was based on a set of 11 claims forming the main request and on an alternative version of Claim 1 forming the basis of an auxiliary request.

The claims according to the main request, i.e. Claims 1 to 3 as filed on 10 October 1995 and Claims 4 to 11 corresponding to amended page 24 as filed on 6 August 1996, read as follows:

"1. A thermoplastic composition which comprises a blend of:

(A) 5 to 50 weight percent of a thermoplastic block copolymer comprising:

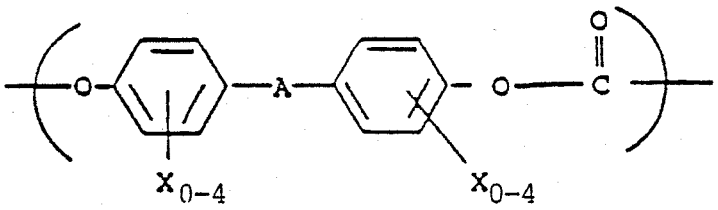
(1.) 1 to 50% by weight of siloxane comprising polysiloxane blocks of the formula:



(I)

where R^1 and R^2 are each independently selected from hydrogen, hydrocarbyl and halogen-substituted hydrocarbyl, D is an integer of from 10 to 120; Y is hydrogen or alkoxy; and

(2.) 50 to 99% by weight of the block copolymer of a polycarbonate-block comprising recurring units of the formula:

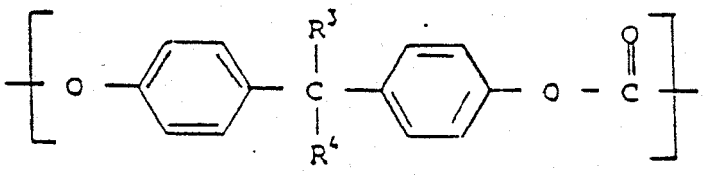


(II)

wherein A is a divalent hydrocarbon radical containing from 1 to 15 carbon atoms; a halogen substituted divalent hydrocarbon radical containing from 1 to 15 carbon atoms or a divalent moiety selected from; -S-; -SS-; -S(O)-; $-S(O)_2-$; -O-; or C-; each X is independently selected from the group consisting of halogen and a monovalent hydrocarbon radical, and

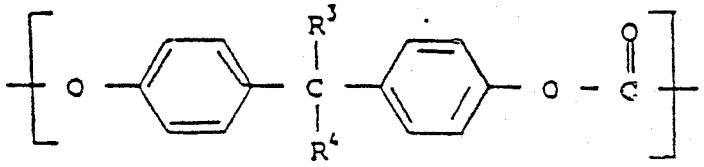
(B) from 50 to 95 weight percent of an aromatic carbonate polymer, wherein the proportions of thermoplastic copolymer (A) and aromatic carbonate polymer (B) are such that the siloxane moieties comprise 4 to 8 weight percent of the total blended composition.

2. The blend of Claim 1 wherein said aromatic carbonate polymer (B) is a polycarbonate comprising repeating units of the formula:

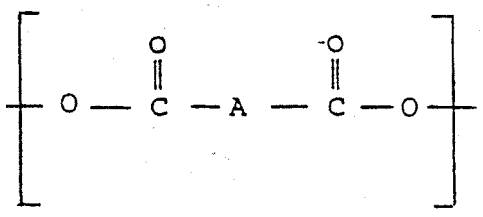


where R^3 and R^4 are independently selected from hydrocarbyl and halogen substituted hydrocarbyl.

3. The blend of Claim 1 wherein said aromatic carbonate polymer is a polyester-carbonate comprising recurring carbonate units of the formula:



where R^3 and R^4 are selected from the group consisting of hydrocarbyl and halogen-substituted hydrocarbyl, copolycondensed with 1 to 20 mole %, relative to total carbonate and ester units, of recurring ester units of the formula:



where A is alkylene of 6 to 18 carbon atoms or phenylene.

4. The blend of Claim 3 wherein said alkylene is straight chain alpha-omega alkylene.

5. The blend of Claim 4 wherein said alkylene is hexamethylene.

6. The blend of any preceding claim wherein R^1, R^2, R^3 and R^4 are methyl and Y is methoxy.

7. The blends of any preceding claim wherein D is 40-60.

8. The composition of Claim 1 which further comprises an effective flame retarding amount of a flame retardant.

9. The composition of Claim 8 wherein there is present from 0.3 to 0.6 percent by weight of an alkali metal or alkaline earth metal salt of an organic sulfonate fire retardant.

10. The composition of Claim 9 wherein the fire retardant is potassium diphenylsulfone-3-sulfonate.

11. The composition of Claim 8 wherein there is present from 0.5 to 2.0 percent by weight of a halogenated fire retardant compound."

Claim 1 according to the auxiliary request is directed to a thermoplastic composition which consists of a blend of (A) ...(as defined above), (B) ...(as defined above), and optionally (C) a flame retardant.

II. The reason for the decision was lack of inventive step with regard to the teaching of EP-A-376 052

(document (1)).

(i) It was first stated that the wording of Claim 1 complied with the requirements of Articles 123(2) and 84 EPC as well as with the provisions for correction of errors pursuant to Rule 88 EPC. This applied to Claim 1 according to both requests on file.

(ii) Document (1) described blends comprising (a) 6 to 90% by weight of a polycarbonate/polyorganosiloxane copolymer, (b) 10 to 60% by weight of glass fibers, and (c) 0 to 84% by weight of a polycarbonate resin, the amount of polyorganosiloxane accounting for 0.5 to 40% by weight of the resin components. Novelty could be acknowledged on a general basis, since the amount of 4 to 8% by weight of polysiloxane in the now claimed composition represented a selection within the broader range in document (1); novelty over the specific copolymer F was also given, since the latter derived from a 2-allylphenol-capped polysiloxane.

(iii) Although claimed as ternary compositions the blends according to document (1) were in fact mere binary resin compositions, since the polycarbonate was mixed with the block copolymer before the glass fibers were added to the mixture. In the absence of any evidence of a technical effect, neither the selection of a narrower range for the amount of polysiloxane, nor the choice of 4-allylphenol-capped siloxanes could be regarded as inventive features. These

considerations applied equally to the compositions according to both requests.

III. On 21 April 1997 a Notice of Appeal against that decision was lodged by the Appellant (Applicant) together with payment of the prescribed fee. The arguments submitted in the Statement of Grounds of Appeal filed on 1 July 1997 can be summarised as follows:

(i) Glass fibers were an essential ingredient of the compositions described in document (1), which had good impact resistance, rigidity and dimensional stability. By contrast, the polymer compositions according to the application in suit displayed good flame ignition resistance and flame retardancy, while at the same time having good impact strength over a wide temperature range. These differences in both the compositions and the object of the respective teachings cast some doubt on the relevance of this citation for the assessment of inventive step.

(ii) Examples 2 to 4 of the application in suit provided evidence of the improved ignition resistance and impact performance achieved by the polymer compositions as defined in Claim 1.

(iii) The replacement of 2-allylphenol used in document (1) as the end capper in the preparation of polydimethylsiloxane by eugenol (2-methoxy-4-allylphenol) in the examples of the application in suit was not arbitrary, since a para-substituted phenol was less sterically

hindered, which made the hydrosilation step easier.

(iv) The fact that document (1) stated that in the examples glass fibers were added downstream of the hopper of the extruder through which the resin stock was fed could not be equated with a teaching regarding compositions of block copolymer and polycarbonate which would not contain any glass fibers.

IV. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of Claims 1 to 11 according to the main request or of amended Claim 1 forming the basis of the auxiliary request, alternatively that oral proceedings be arranged.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. *Wording of the claims*

The Board concurs with the Examining Division that the wording of the claims as amended satisfies the requirements of Articles 123(2) and 84 EPC as well as the provisions for correction of errors pursuant to Rule 88 EPC.

3. *Document (1)*

This document, which was the only citation considered in the decision to refuse the application, will be discussed from the viewpoint of successively (i) its general teaching, (ii) the specific embodiments, and (iii) the properties and applications of the resin compositions.

3.1 The resin compositions described in document (1) are defined as polycarbonate-based resin compositions comprising (a) 6 to 90% by weight of a polycarbonate/polyorganosiloxane copolymer, (b) 10 to 60% by weight of glass fibers, and (c) 0 to 84% by weight of a polycarbonate resin, the amount of the polyorganosiloxane accounting for 0.5 to 40% by weight of the resin components (Claim 1). In view of this definition the glass fibers (b) are a compulsory ingredient of the known resin compositions and the polycarbonate resin (c) is only an optional component thereof.

As to copolymer (a) it is defined in Claim 2 as a block copolymer comprising a polycarbonate segment having repeating units which correspond to a large extent to formula (II) in the application in suit and a polysiloxane segment having repeating units corresponding to a large extent to the recurrent units D in formula (I) in the application in suit. According to the description of document (1) (page 3, lines 45 to 50) the block copolymer may be prepared by an interface reaction of a polycarbonate oligomer with a polyorganopolysiloxane having terminal reactive groups in the presence of an alkaline solution of bisphenol and a catalyst.

3.2 According to a typical embodiment the

polyorganosiloxane segment derives from a reactive polymer, e.g. a phenol end group containing polydimethylsiloxane obtained by reacting 2-allylphenol with a mixture of octamethylcyclotetrasiloxane and 1,1,3,3-tetramethyldisiloxane (Preparation Example 1-4). The resulting polydimethylsiloxane is then adducted with a polycarbonate oligomer (Preparation Example 2) to yield a block copolymer, e.g. copolymer F containing 3.5% by weight of dimethylsiloxane moieties (Preparation Example 3-6 in conjunction with Preparation Example 3-1). Copolymer F is subsequently mixed with glass fibers in the weight ratio of 70:30 (Table 1, Example 16).

Ternary compositions, i.e. compositions containing a block copolymer (a), glass fibers (b) and additionally a polycarbonate resin (c) are also reported (Table 1, Examples 9 to 14). In these compositions the block copolymer is identified as being a copolymer A, B or C, which are all obtained from a different reactive polyorganosiloxane.

As to the blending process it is specified (i) that the polymer component(s) and the glass fibers are blended together at the given proportions, (ii) that the blends are formed through a 30 mm vented extruder into pellets, which are in turn injection-moulded, and (iii) that the glass fibers are supplied downstream of the hopper of the extruder through which the resin stock is fed in (page 7, lines 45 to 52; Examples 1 to 18).

3.3 Regarding the properties the prior art review in the introductory section of document (1) (page 2, lines 12 to 29) first reports in general terms that polycarbonate resins excel in mechanical strength,

electrical characteristics and transparency, which explains their various applications in the form of engineering plastics, then indicates that the need to improve both rigidity and dimensional stability for specific uses has led to reinforce these resins with glass fibers, which in turn tends to give rise to a drop of Izod impact strength due to brittle fracture.

The attempts to overcome that shortcoming, which consisted in the further addition of an organopolysiloxane or an organopolysiloxane/polycarbonate copolymer, were not entirely satisfactory in that either the electrical characteristics were adversely affected or the improvement in impact resistance was still not compatible with the needs for specific applications, such as chassis and electrically powered tools (page 2, lines 19 to 29).

The compositions disclosed in D1 must thus be regarded as a further attempt to optimize the balance between impact resistance, rigidity and dimensional stability of glass reinforced polycarbonate polymer compositions (page 2, lines 34 to 37; page 4, lines 55 to 57), as apparent from the properties reported in Table 1: Izod impact strength, tensile modulus and bending strength.

4. *Novelty*

Although no objection was raised having regard the novelty of the compositions as claimed, the Board deems it appropriate to deal with this issue in detail.

4.1 The first question to examine is whether the mention in D1 (page 7, lines 45 to 52) that the glass fibers are

supplied downstream of the hopper of the extruder through which the resin stock is fed in, can be interpreted as a disclosure of binary mixtures of a polycarbonate/polyorganosiloxane block copolymer and a polycarbonate resin within the terms of the application in suit.

Although this passage suggests that the polycarbonate resin is mixed with the block copolymer before addition of the glass fibers, this must in fact be regarded as the first step of a continuous process, the subsequent steps being (i) the addition of the glass fibers, (ii) the blending of the various ingredients, (iii) the forming of pellets, and (iv) the injection moulding of samples for the determination of their physical properties. There is no isolation of blends of the two polymer components, let alone any experimental data reported in Table 1 concerning such blends.

In the Board's view, thus, the processing feature relied upon in the decision under appeal cannot be equated with the availability of a blend of the sole polymer components. It follows that the compulsory presence of 10 to 60% by weight of glass fibers represents a distinguishing feature over the claimed compositions.

4.2 The second feature to consider is the polysiloxane segment of the thermoplastic block copolymer.

In document (1) the organopolysiloxane is characterised (i) by repeating units of formula (I) (cf. Claim 2), which correspond to a large extent to the recurrent units D in formula (II) in the application in suit, and (ii) by reactive end groups, e.g. terminal phenolic

hydroxyl groups derived in particular from 2-allylphenol (cf. Preparation Example 1-4).

This should be compared with formula (III) in the application in suit, which represents the reactive polyorganosiloxane, i.e. bisphenolsiloxane, from which the polysiloxane segment of formula (I) is derived. According to the description as originally filed (page 4, line 22 to page 5, line 15) such bisphenolsiloxanes are prepared by the addition of a polydiorganosiloxane to an optionally substituted para-alkenylphenol of formula (VI), e.g. eugenol.

Thus, even if one considers the most favourable combination of compositional and structural features in document (1), the bisphenolsiloxane giving rise to the polysiloxane segment of formula (I) in the application in suit differs from its counterpart in the citation as being an isomeric derivative thereof.

4.3 A further feature to deal with is the amount of siloxane moieties in the total blended compositions, which is required to be between 4 and 8% by weight in the application in suit.

By contrast, in the composition according to document (1) the polyorganosiloxane accounts for 0.5 to 40% by weight of the resin components, which is a much broader range.

Regarding the specific compositions reported in Table 1, only those according to Examples 12, 14 and 15 have amounts of poyorganosiloxane within the required range. However, even if one leaves out of account the presence of 30% by weight of glass fibers in the three

compositions and the absence of polycarbonate resin in the composition according to Example 15, the figures indicated for the amount of siloxane moieties, e.g. 5.7, 4.1 and 4.8 respectively, are not directly comparable with the amount required in the application in suit because of the differences in the composition of the respective block copolymers B, C and E.

It follows that, whether one considers the general teaching or the specific embodiments of document (1), the amount of siloxane moieties in the application in suit represents a distinguishing feature over the prior art.

4.4 For these reasons novelty can be acknowledged on the basis of the three features discussed above.

5. *Problem and solution*

The application in suit concerns thermoplastic blends of polysiloxane-polycarbonate block copolymers with polycarbonate resins.

5.1 From the discussion of the background art in the application in suit (page 1, line 14 to page 2, line 5) it appears that polysiloxane-polycarbonate block copolymers are well known, but that these copolymers require that a number of grades which differ in siloxane level, must be manufactured to meet the various market requirements for various balances of low and high temperature properties. A first aspect of the invention may thus be seen in the need to have means for meeting these diverse requirements by manufacturing only a single grade of siloxane-modified polycarbonate copolymer.

A second aspect of the invention resides in the fact that good impact strength is difficult to combine over a wide temperature range with good flame ignition resistance and flame retardancy without a degree of compromise (page 8, lines 17 to 26).

Accordingly, the experimental data in the application in suit concern not only a number of mechanical properties measuring the impact performance, but also the results of the ignition resistance performance as measured by the test UL94.

5.2 By contrast, as stated in point 3.3 above, the compositions disclosed in document (1) are concerned with an optimized balance between impact resistance, rigidity and dimensional stability of glass fiber reinforced compositions, which corresponds to a different pattern of properties.

5.3 This difference between the object of the application in suit and the object of document (1) casts serious doubts on the suitability of this citation for assessing inventive step on the basis of the problem-solution approach.

According to the established case law of the boards of appeal a document serving as the starting point for evaluating the inventive merits of an invention should relate to the same or a similar technical problem or, at least, to the same or a closely related technical field as the application or the patent in suit. A document not mentioning a technical problem which is at least derivable from the original application or patent specification does not normally qualify as the closest prior art for inventive step purposes, however many

technical features it may have in common with the subject-matter of the application or patent concerned (cf. Case Law of the Boards of Appeal of the European Patent Office, 3rd edition 1998, English edition, pages 110 to 113).

5.4 In the present case, thus, the mere fact that document (1) discloses compositions which are based on polysiloxane-polycarbonate block copolymers, the latter being compositionally and structurally very similar to component (A) in the application in suit, but which aim at achieving a totally different pattern of properties, is not sufficient to qualify this citation as the closest state of art. It follows that the technical problem underlying the application in suit cannot be derived objectively from that prior art and that, consequently, the missing link between the known compositions and the claimed subject-matter can only be established by means of an artificial chain of assumptions and reasoning involving *ex post facto* analysis.

For the sake of the present decision, however, the Board will follow the approach of the Examining Division and regard the teaching of document (1) as a suitable starting point.

5.5 On that basis the technical problem may be seen as the provision of compositions based on polysiloxane-polycarbonate block copolymers having an improved balance of impact strength and flame ignition resistance over a wide range of temperature; moreover that improvement should be achieved with a single grade of siloxane in the copolymer.

5.6 According to the application in suit this problem is solved by blends of specific polysiloxane-polycarbonate block copolymers and aromatic polycarbonates, as defined in Claim 1.

5.7 The experimental data in the application provide evidence that this problem is effectively solved by the required combination of features. In particular, (i) Example 2 shows that at comparable impact performance the compositions as claimed have a superior ignition resistance performance, (ii) Example 3 shows that at comparable ignition resistance performance the compositions as claimed have a superior impact performance over a wide range of siloxane degree of polymerisation, and (iii) Example 4 shows that at equal glass fiber loading the compositions as claimed have both superior ignition resistance performance and impact performance.

6. *Obviousness*

It remains to be decided whether the novelty-conferring features, i.e. (1) the absence of glass fibers, (2) the bisphenolsiloxane giving rise to the polysiloxane, and (3) the selection of a narrow siloxane range, can be regarded as obvious to a person skilled in the art having regard to the disclosure of document (1).

6.1 The prior art review in the introductory section of document (1) (cf. point 3.3 above) clearly shows that this citation is concerned with glass fiber-reinforced polycarbonate-based resin compositions aiming at a certain pattern of mechanical properties, including high impact resistance. The solution taught in document (1) resides in the use of a specific

polycarbonate-polysiloxane block copolymer reinforced with glass fibers. It follows that the glass fibers must be regarded as having a major contribution to the known pattern of properties and that a skilled person aiming at compositions based on such block copolymers with a different pattern of properties, but also requiring high impact strength, would have no reason to depart from that teaching and use instead an aromatic polycarbonate resin.

As noted above (cf. point 4.1), the passage in document (1) (page 7, lines 51/52) relied upon by the Examining Division, according to which the glass fibers are supplied downstream of the hopper of the extruder through which the resin stock is fed in, does not suggest thermoplastic compositions containing only polymer ingredients which would have by themselves the desirable pattern of mechanical properties in accordance with the general teaching of the document. This passage only shows that glass fibers are supplied separately in order to prepare and process compositions by a continuous multi-step process. Thus the possible existence during the preliminary mixing step withing the extruder of a polymer blend comprising a polyorganosiloxane-polycarbonate block copolymer and a polycarbonate resin cannot be equated with the disclosure of an unfilled composition liable to be isolated and tested.

This also appears from Table 1 in document (1), where only the properties of filled compositions without polycarbonate (Examples 1 to 8 and 15 to 18) and filled compositions without block copolymer (Comparative Examples 1, 2, 4 and 5) are reported, but not of unfilled compositions.

6.2 Regarding the end groups of the organopolysiloxane used for the preparation of the block copolymer, the argument in the decision under appeal (cf. Reasons for the decision, point 2.4.4), that a skilled person aware of the commercial availability of eugenol, i.e. 2-methoxy-4-allylphenol, would consider capping the polysiloxane with this compound instead of 2-allylphenol used in Preparation Example 1-4 in document (1) (cf. point 3.2 above) "without the need for inventive skills", cannot be accepted for the following reasons.

In contrast to the repeating units of the polyorganosiloxane segment of the block copolymer, which are represented by a specific formula, the end groups are defined as terminal phenolic hydroxyl groups (cf. point 4.2 above), examples thereof being mentioned in Preparation Examples 1-1 to 1-4. Whilst the repeating units must thus be regarded as essential features, the actual structure of the end group is only of minor importance, provided it contains a terminal phenolic group. Assuming that a skilled person would nevertheless recognize the necessity to specify the structure of the end group, any "obvious" modification should occur on the basis of the information available from document (1), e.g. from the Preparation Examples 1-1 to 1-4, not on the basis of the commercial availability of a compound not even envisaged in this citation. Even if there were theoretical reasons justifying the choice of a 4-allylphenol derivative for the preparation of the polyorganosiloxane, in particular a lower steric hindrance leading to better reactivity and higher yields, the experimental results in the only example in which a block copolymer derived from a polyorganosiloxane and 2-allylphenol is used

(Example 16), do not speak particularly in favour of such a structure, so that in practice this skilled person had no reason at all to consider a solution along that line.

6.3 Similar considerations apply to the amount of polyorganosiloxane defined in the form of a selection of a narrow weight range.

According to document (1) (page 4, lines 18 to 22) the quantity of polyorganosiloxane contained in the resin composition should be 0.5 to 40% by weight. If the quantity of polyorganosiloxane is below 0.5% by weight, there is no improvement in impact resistance; if the quantity exceeds 40% by weight, any copolymer having sufficient molecular weight cannot be obtained. The experimental data in Table 1 confirm that good mechanical properties, particularly in terms of Izod impact strength, can indeed by obtained over the whole range. It was thus not to be expected that within a narrow weight range the amount of polyorganosiloxane could also serve to improve the ignition resistance performance of the composition without impairing the impact performance.

6.4 It follows from these considerations that the above distinguishing features and, consequently, their combinations do not arise in an obvious manner from document (1), so that the subject-matter of Claim 1 involves an inventive step.

6.5 The same conclusion applies to the subject-matter of dependent Claims 2 to 11, which are directed to preferred compositions and blends and equally involve an inventive step.

7. *Auxiliary request*

Since the main request is allowed, it is not necessary to consider the auxiliary request.

Order

For these reasons it is decided that:

1. The decision under appeal be set aside.

2. The case is remitted to Examining Division with the order to grant a patent on the basis of Claims 1 to 11 according to the main request after any consequential amendment of the description.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin