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D E C I S I O N
of 5 September 1994

Case Number: T 0457/91 - 3.3.3

Application Number: 84112103.1

Publication Number: 0138187

IPC: C08F 2/50

Language of the proceedings: EN

Title of invention:
Photopolymerizable composition

Patentee:
MITSUBISHI KASEI CORPORATION

Opponent:
Hoechst Aktiengesellschaft

Headword:
-

Relevant legal provisions:
EPC Art. 56

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

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0457/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 5 September 1994

Appellant: (Opponent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office dated 14 March 1991, issued in writing on 18 April 1991 rejecting the opposition filed against European patent No. 0 138 187 pursuant to Article 102(2) EPC.**

Composition of the Board:

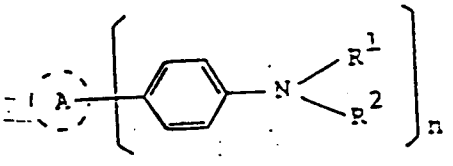
Chairman: C. Gérardin
Members: R. Young
W. M. Schar

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 138 187, in respect of European patent application 84 112 103.1, filed on 9 October 1984 and claiming a priority of 14 October 1983 (JP 192212/83), was announced on 11 January 1989 (cf. Bulletin 89/02). Claim 1 reads as follows:

"A photopolymerizable composition comprising an addition-polymerizable compound having at least one ethylenically unsaturated double bond and a photopolymerization initiator, characterized in that said initiator comprises:

(a) a compound represented by the general formula:



(I)

wherein R^1 and R^2 are an alkyl group, respectively, n is an integer of 1, 2 or 3 and ring A is an aromatic ring containing a nitrogen atom, in an amount of 0.1 to 15%, and

(b) hexaarylbiimidazole in an amount of 0.5 to 30%, the amounts of (a) and (b) being based on the weight of the compound with ethylenically unsaturated double bond."

Claims 2 to 5 are directed to further elaborations of the compositions of Claim 1.

II. Notice of Opposition was filed on 10 October 1989 on the ground of lack of inventive step. The opposition was supported by the following documents:

D1: US-A-3 479 185;
D2: US-A-3 888 672;
D3: US-A-3 647 467;
D4: US-A-4 282 309; and the later filed, but admitted
D5: Chemische Berichte, 93rd Year, No. 11 (1960), p. 2106.

III. By a decision which was given at the end of oral proceedings held on 14 March 1991 and issued in writing on 18 April 1991 the Opposition Division rejected the opposition.

According to the decision, the composition claimed was distinguished over the closest prior art document D3 by the heterocyclic sensitizer compound bearing 1 to 3 dialkylamino-phenylene groups. It was furthermore clear from experimental results provided by the proprietor that the claimed compositions showed higher sensitivity over a wide range of wavelengths than those of the prior art. Although the sensitizers used in the patent in suit were disclosed in D4, these belonged to another initiator system, and there was no suggestion that the dialkylamino substitution conferred improved sensitivity.

IV. On 25 June 1991 a Notice of Appeal against the above decision was filed, together with payment of the prescribed fee.

In the Statement of Grounds of Appeal filed on 27 August 1991 and a further submission dated 10 August 1992, the Appellant (Opponent) argued essentially as follows:

(i) According to the document D3 the heterocyclic sensitizer compounds extended the spectral sensitivity of the hexaarylbiimidazole to higher wavelengths, the substituents being so chosen that the absorption was in the spectral region corresponding to the light source used. One way of extending the absorption maxima of the heterocyclic sensitizers of D3 to still higher wavelengths, corresponding to the light sources disclosed in the patent in suit, would have been by the introduction of auxochromic substituents which did not interfere with energy transfer (Grounds of Appeal, page 2).

According to D4, compounds of similar structure to those known from D3 but having auxochromic substituents, including aromatic tertiary amino groups, were described as sensitizers for other photoinitiators (oxime esters).

It would have been *prima facie* obvious to try such compounds, instead of the unsubstituted compounds of D3, in combination with hexaarylbiimidazole (Grounds of Appeal, pages 2, 3).

(ii) Although the Respondent had shown by comparative experiments that the compounds with tertiary amino groups gave a mixture with substantially higher sensitivity than the unsubstituted ones of the prior art, this result could not be regarded as surprising for the skilled person.

In particular, there was a reference in D3 to a link between spectral sensitivity and light absorption. A sensitizer could advantageously be provided with appropriate substituents to shift

its absorption maximum to a desired spectral region. This was demonstrated in D5, in which absorption spectra of an unsubstituted and a dialkylamino-substituted sensitizer having a heterocyclic nucleus as disclosed in D3 were shown, the absorption maximum being shifted to a higher wavelength in the substituted variant. Furthermore, of three relevant heterocyclic sensitizer compounds described in D4, one was unsubstituted while the other two carried dialkylamino groups. The photosensitivity for the latter two was higher than for the former (Grounds of Appeal, pages 3, 4).

On the other hand, if the sensitizer and initiator both had their absorption maxima in the same spectral region they would compete for radiation, and a lower degree of sensitivity would result (cf. submission dated 10 August 1992, page 2).

In any case, the sensitizing effect was predictable.

(iii) The choice of substituent was also indicated in D3 itself, which taught that the compositions could additionally contain, as a coinitiator, a tertiary amine such as a leuco dyestuff, or, as a polymerization aid, an electron-donating free radical generator which, according to a reference, could also be an aromatic tertiary amine. Similar remarks could be found in D2, also relating to photopolymerizable mixtures with hexaarylbiimidazole, in which a rhodanine derivative very similar to the heterocyclic

group A in formula (I) of the patent in suit was taught as an additive (cf. Grounds of Appeal, pages 4, 5).

(iv) Although the Respondent had alleged a better storage stability of the claimed compositions as a further surprising effect, this had only been demonstrated relative to an initiator which had no relationship to the closest state of the art. Consequently, this effect should be left out of consideration (cf. Grounds of Appeal, page 5).

V. The Respondent (Patentee), on the other hand, argued substantially as follows:

(i) There was no disclosure in D3 that the heterocyclic sensitizer compounds were or could be substituted with dialkylamino-phenylene groups, or that the presence of such groups would improve photosensitivity of the photopolymerizable composition. Since, furthermore, the heterocyclic compounds in the examples were not substituted on the aromatic ring, there was no hint to direct the skilled person to the claimed sensitizers (cf. submission dated 5 March 1992, paragraph 4.1).

(ii) The comparative data already filed had shown that the specific combination of a hexaarylbiimidazole with the sensitizer compound of formula (I) yielded a photopolymerizable composition having a sensitivity improved over that obtained according to D3 by more than 100% in the worst case (cf. submission dated 5 March 1992, paragraph 4.2, and Table of results on page 5). The unsubstituted and substituted sensitizers mentioned in D4, on the other hand,

were not mutually comparable. Furthermore, D4 disclosed a completely different photoinitiator from the hexaarylbiimidazole used in the claimed composition. Thus any conclusions drawn from D4 could not be applied in relation to the photoinitiators of D3 (cf. submission dated 5 March 1992, paragraph 4.3).

(iii) Merely shifting the absorption maximum into a given range by adding a dialkylamino group did not mean that the result would necessarily be an increased photosensitivity of the composition. On the contrary, the comparative data already filed showed, for one of the prior art sensitizers which had its absorption maximum precisely coinciding with the wavelength of irradiation, that the degree of sensitization was actually reduced. The energy at the longer wavelength would in any case be less and its transfer to the photoinitiator could thereby be vitiated. (cf. submission dated 5 March 1992, paragraph 4.4, and submission dated 10 August 1993, paras. 2,2, 2,3).

Thus the sensitising effect could not have been expected.

(iv) The storage stability effect had been demonstrated in comparison to a photoinitiator composition which was broadly used in practice (cf. submission dated 5 March 1992, page 6, first and second paragraphs).

VI. The Appellant requests that the decision under appeal be set aside, and the patent revoked in its entirety for all Contracting states.

The Respondent requests:

(1) that the appeal be dismissed;

(2) that the patent be maintained as granted, with the provision that in line 2 of the English language Claim 1 the obvious error introduced when printing the patent is amended by substituting the word "inhibitor" by the word "initiator";

(3) as an auxiliary measure, oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. As regards the Respondent's request for the term "inhibitor" to be amended to read "initiator" in Claim 1, this was merely a printing error, the text forming the basis for consideration and decision by the Board in the sense of Article 113(2) EPC already being in the correct form. There is thus no requirement for such an amendment or correction in the official text of the patent. All that is necessary is the issue of an appropriate corrigendum in the printed version.

3. The patent in suit is concerned with a photopolymerizable composition comprising an addition-polymerizable compound having at least one ethylenically unsaturated double bond and a polymerization initiator, the initiator comprising:

(a) a heterocyclic compound comprising an aromatic ring containing nitrogen, and

(b) hexaarylbiimidazole, in stated quantities, based on the weight of the compound having the unsaturated double bond.

Such a composition was, however, known from D3, which according to the parties' and the Board's judgment is to be considered the closest state of the art.

3.1 According to D3, a photoactivatible composition comprised an admixture of:

A. a hexaarylbiimidazole that had its principal light absorption bands in the ultraviolet region of the electromagnetic-radiation spectrum, and was dissociable to triarylimidazolyl radicals on irradiation with such absorbable utraviolet light,

B. a heterocyclic compound of the formula Ar^1-G-Ar^2 wherein Ar^1 was an aryl group of 6 to 12 nuclear carbon atoms, Ar^2 was an aryl group of 6 to 12 nuclear carbon atoms or an arylene-G-Ar^1 group wherein arylene was of 6 to 10 carbon atoms, and G was a divalent furan, oxazole or oxadiazole ring, and which had its principal light absorption in the near ultraviolet or visible regions of the electromagnetic spectrum (preferably 290 to 420 mμ), and optionally

C. a leuco dye that was oxidizable to dye by triarylimidazolyl radicals and/or an addition-polymerizable, ethylenically unsaturated monomer (cf. Claim 1, and column 1, line 60 to column 2, line 19).

3.2 The substituents in the heterocyclic compounds were not critical provided the compound had the desired spectral properties. In general, the substituents were chosen

such that the sensitizer absorbed preferably above 300 mμ as measured in an inert solvent (column 2, lines 61 to 69). Representative sensitizer compounds included 2,5-diphenyloxazole, 2-phenyl-5-(alpha-naphthyl)oxazole and 1,4-bis[2-(5-phenyloxazolyl)] benzene (column 3, lines 38 to 42).

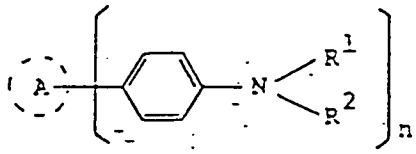
3.3 The sensitizer could be present in amounts of 0.01 to 0.5 mole per mole of hexaarylbiimidazole, which itself was present in amounts of at least 0.5 % of the composition (column 3, line 65 and column 10, lines 10 to 13).

3.4 While the sensitization mechanism was not known with certainty, it was considered that the heterocyclic compounds were activated to at least one excited energy transfer state, in which they transferred absorbed energy to the hexaarylbiimidazole, for example through collision or resonance interaction. The thus indirectly-activated hexaarylbiimidazole dissociated into triarylimidazole radicals (column 2, lines 21 to 30).

3.5 According to an embodiment relating to photopolymerization, the composition could comprise an addition-polymerizable ethylenically unsaturated compound, such as trimethylolpropane triacrylate, a solvent and a light-transparent binder, preferably together with a polymerization aid or coinitiator. The polymerization aid or coinitiator could be an electron-donating free radical generator as disclosed in Belgian patent No. 681 944 or a photooxidizable amine, such as an aminotriarylmethane leuco dye containing dialkylamino groups (column 12, lines 29 to 75; column 10, lines 52 to 62).

4. Compared with this state of the art, the technical problem could be seen in the search for such a photopolymerizable composition of improved photosensitivity without appreciable loss of storage stability.

The solution proposed according to Claim 1 of the patent in suit was to replace the heterocyclic compound B of D3 by a compound (a) represented by the formula (I):



(I)

wherein R^1 and R^2 were an alkyl group, respectively, n was an integer of 1, 2 or 3 and ring A was an aromatic ring containing a nitrogen atom.

4.1 It was admitted, and indeed was a major plank of the Appellant's argument, that the distinguishing feature of the sensitizers according to the patent in suit, namely the presence of a dialkylamino substituent in the benzene nucleus, was responsible for an increase of the sensitivity in the near ultraviolet region (cf. Grounds of Appeal, page 3, second paragraph, and page 5, last paragraph).

4.2 Such an effect was also supported by the experimental data filed by the Respondent with the submissions dated 30 March 1990 and 9 January 1991, the most relevant parts of which were reproduced in the submission dated 5 March 1992 (page 5, Table). According to the latter, the degree of sensitivity in the near ultraviolet (366 nm) using a 2,5-bis(dimethylaminophenyl)-1,3,4-oxadiazole sensitizer according to the patent in suit

(compound a-5) was more than six times greater than that obtained with a sensitizer differing from the compound a-5 only by the absence of the dimethylamino groups.

Although the latter species did not correspond exactly to a compound specifically disclosed in D3, the nearest such being the corresponding oxazole (cf. column 3, line 37), it was within the teaching of D3 and was considered by the Board to be sufficiently representative of the latter to constitute a fair comparison. The sensitizing effect itself was in any case not contested.

4.3 As regards the storage stability, it can furthermore be seen from the experimental data in the patent in suit, that the sensitivity of the claimed compositions after being heated in an oven at 55 °C for 150 hours was reduced by at most a third compared with the loss of sensitivity of a comparative composition based on Michler's ketone and benzophenone (cf. patent in suit, page 7, Table 2).

4.4 It is true that Michler's ketone did not correspond to the closest state of the art D3. It did, however, according to the uncontested submission of the Respondent, represent a widely used example of a high sensitivity initiator and therefore illustrated an acceptable level of storage stability for practical purposes.

4.5 There was, on the other hand, no mention whatever of storage stability in D3. Nor was any evidence offered on the matter by the Appellant, who at this stage had the onus of proof.

Consequently, there was no ground for assuming that a higher level of storage stability was obtained with the compositions according to D3 than was commonly accepted in practice, e.g. with those of the Michler's ketone based initiators. On the contrary, if anything, improved sensitivity had been found to be usually accompanied by poor stability in storage (cf. patent in suit, page 2, lines 29 to 32).

Thus, there was in the Board's view no reason for concluding that the storage stability of the compositions claimed, which was in any case significantly better than that of the commercially available compositions, was appreciably, if at all, poorer than those of D3.

In summary, it is credible to the Board that the claimed measures are effective to solve the stated problem.

5. No lack of novelty was alleged either in the opposition proceedings or in the appeal. The decision under appeal found that the subject-matter of the patent in suit was novel since none of the cited documents mentioned the composition of Claim 1. The Board sees no reason to diverge from this viewpoint.

Thus the claimed subject-matter is held to be novel.

6. As regards the issue of inventive step, the question arises as to whether the skilled person would have had reason to expect that the photosensitivity of a polymerizable composition according to D3 would be improved by providing, in the heterocyclic sensitizer, one to three aromatic dialkylamino substituents.

6.1 Although the disclosure of D3 opened the possibility that substituents might be present in the sensitizers, which were of the furan, oxazole or oxadiazole types, the nature of these substituents was stated to be "not critical", the only requirement being that the compound had "the desired spectral properties" (cf. column 2, lines 61 to 64). Furthermore, although certain of the specified heterocyclic compounds had a phenylene substituent, such as 2,5-diphenyloxazole, 2-phenyl-5-(alphanaphthyl)oxazole and 1,4-bis[2-(5-phenyl-oxazolyl)] benzene (cf. column 3, lines 38 to 42), there was no mention of a further substituent on the phenyl ring, let alone a dialkylamino such substituent.

6.2 As to the hexaarylbiimidazole component of the initiator, furthermore, the possibility of an alkylamino or dialkylamino group substituent was specifically excluded (cf. column 4, lines 23 to 28).

6.3 It is, however, true that the compositions of D3 could contain a further, optional ingredient having amino groups (component C.), which could be a polymerization aid such as an electron donating free radical generator (cf. section 3.5, above).

6.3.1 The latter species, according to the disclosure of D3 (in which the Belgian patent referred to had the same priority as D1), was in particular an aminotriarylmethane leuco dye containing dialkylamino groups, and was also termed a coinitiator (cf. column 12, lines 29 to 38 and 65 to 75).

6.3.2 It was, however, clear from the definition of component C. in D3 that the leuco dye species was oxidizable by **already formed** triazolylimidazole radicals (themselves arising from hexaarylbiimidazole) and not vice versa. This was furthermore confirmed by the reaction scheme

shown in D3 for the activation of hexaarylbiimidazole, in which the leuco dye reacted with the hexaarylbiimidazole **after** the latter had dissociated into radicals (cf. D3; column 2, lines 29 to 30 and 41 to 59; reactions (1) and (2)).

Thus, these amino group-containing species, although termed "polymerisation aids" or "coinitiators", did not have any sensitizing function in relation to the hexaarylbiimidazole, but on the contrary were evidently dependent on the previous dissociation of the latter for their own activity. Their structure could not therefore have been taken as a guide for designing improved sensitizers for promoting the dissociation of hexaarylbiimidazole.

6.3.3 The remaining additives were more remote in their structure and function from the sensitizers of the patent in suit.

Hence, there was no hint in D3 itself to make the modification to the sensitizers which formed the solution to the stated problem.

6.4 According to D1, there was disclosed a photopolymerizable composition comprising (1) an addition-polymerizable compound, (2) a free radical producing electron donor agent and (3) a 2,4,5-triphenylimidazolyl dimer (i.e. hexaarylbiimidazole). The agent, which had a reactive atom and was in particular a leuco dye, especially one having at least one dialkylamino group (cf. section 6.3.1, above), yielded, in the presence of a radical of the dimer, a further radical which reacted with the monomer to initiate the growth of polymer chains (cf. D1, Claims 1 and 2; column 2, lines 35 to 49; column 4, lines 51 to 55).

Thus the disclosure of D1 was similar to that of D3 in respect of the function of the leuco dye and consequently came no closer to a solution of the stated problem.

6.5 According to D2, which also related to photopolymerizable mixtures including an addition-polymerizable compound, a hexaarylbiimidazole and a free radical producing hydrogen- or electron-donor agent (cf. D2, Claim 1), but in quantities to provide reverse image working, it was stated that the donor agent could be, for instance, 5-(p-dimethylaminobenzylidine)-rhodanine (column 4, lines 46 to 62, especially lines 59, 60).

6.5.1 This rhodanine derivative, however, differed from the sensitizers of the patent in suit in that the dimethylamino substituent was linked to the heterocyclic nucleus through a benzylidene group. Thus the necessary direct phenylene linkage was absent (cf. patent in suit, page 3, lines 40, 41).

6.5.2 Furthermore, it was stated in D2 to be important that the donor compound (i.e. the rhodanine derivative in this case) should **not** itself be activatable by actinic radiation to produce free-radicals that would initiate polymerization, but that it should be reactive with the free-radical-producing agent (hexaarylbiimidazole) that was responsive to actinic radiation (D2; column 4, lines 30 to 45).

Thus, regardless of any incidental degree of similarity between the specific rhodanine derivative and the compounds of the patent in suit, it was evident from the teachings of D2 that it did not function to sensitize the hexaarylbiimidazole for dissociation into radicals.

6.5.3 Hence, the skilled person seeking compositions of improved sensitivity would have had no incentive to consider the rhodanine derivative of D2 as offering a solution to the stated problem.

6.6 According to D4 there were disclosed photosensitive compositions containing a photopolymerizable, e.g. photocrosslinkable ethylenically unsaturated compound and an oxime ester photopolymerization initiator in admixture with a sensitizer (column 1, lines 7 to 11). Two classes of sensitizers were disclosed, one of which included examples which fell within the terms of the solution of the stated problem (cf. D4, column 5, compounds 12 and 13, corresponding to compounds a-1 and a-5 respectively of the patent in suit) as well as an example according to the disclosure of D3 (cf. D4, column 5, compound 11). According to the results given in Tables 1 and 2 of D4, the effectiveness of the dialkylamino substituted compounds 12 and 13 in sensitizing an oxime initiator was higher than that of the unsubstituted compound 11 (cf. Examples 1 and 2, columns 13, 14).

6.6.1 The higher photosensitivity of the dialkylamino substituted compounds 12 and 13 compared with the unsubstituted compound 11 would in this connection not have had the alleged significance for the skilled person, however, because the compounds concerned differed in their structure by more than just the relevant dialkylamino substituent. Their sensitivity values were in any case not higher than those of other sensitizers disclosed in D4, in which neither the relevant dialkylamino substituents, nor the necessary aromatic heterocyclic nucleus were present (cf. compounds 9 and 10; column 5, lines 1 to 23; columns 13, 14, Tables 1 and 2).

Consequently, there was no teaching in D4 that the degree of sensitization was in any way associated with the presence or absence of an aromatic dialkylamino group on a heterocyclic nucleus of the kind forming the solution to the stated problem.

6.6.2 Furthermore, although the mechanism of sensitization of the heterocyclic compounds disclosed in D3 was not known with certainty (cf. column 2, lines 20 to 21), the energy transfer mechanism suggested would, in the light of the uncontested explanation provided by the Respondent (cf. submission dated 10 August 1993, paragraph 2.2), have required a particular energetic and therefore structural relationship between the initiator and the sensitizer. The oxime esters of D4 were, however, structurally quite different from the hexaarylbiimidazole initiators of D3.

Consequently, there would have been no reason for the skilled person to suppose that any of the sensitizers according to D4 would be suitable for use with the quite different type initiator of D3, regardless of their relative performance in the context of D4.

6.7 The argument of the Appellant based on the wavelength of the light source used (cf. section IV (i), above) was not convincing, because shifting absorption maxima of the sensitizers into a higher wavelength range was not itself a sufficient condition for achieving higher photosensitivity. In particular, the higher wavelength radiation would have had a lower energy which would not necessarily be transferable to the hexaarylbiimidazole, as shown in the uncontested explanation of the Respondent (cf. submission dated 10 August 1993, paragraph 2.2).

6.8 The supplementary argument, that the lower sensitivity of 2,5-diphenyl-1,3,4-oxadiazole, having an absorption maximum at 280 mμ, when irradiated at 290 mμ, was predictable in terms of competition with the initiator, which itself had an absorption maximum between about 235 and 285 mμ, was not convincing in the light of the more specific disclosure of D3. According to the latter, hexaarylbiimidazole absorbed maximally in the 255 to 275 mμ region (cf. column 3, lines 69 to 72), i.e. lower than the absorption maximum of the sensitizer, which in turn was lower than the wavelength of irradiation.

Consequently, the relationship of absorption maxima was broadly in conformity with the general concept, taught in D3, of extending the absorption maxima of the initiator to higher wavelengths (column 2, lines 8 to 19), and the poor performance recorded was therefore not predictable.

6.9 The argument was in any case irrelevant, however, because the spectral absorption characteristics alone were not an index to photosensitizing capability (cf. section 6.7, above), and consequently could not be used to predict the latter.

6.10 The argument that it would have been "obvious to try" the dialkylamino-substituted sensitizers of D4 with the hexaarylbiimidazole initiator of D3 was not convincing, because, for the reasons given above,

(a) no advantage was discernible for the relevant dialkylamino substituents among the many sensitizers of D4, which were in any case taught for use with a completely different initiator,

(b) the species taught as sensitizers in D3 did not have the relevant dialkylamino substituents, and

(c) those species in D3 which did have dialkylamino substituents were not taught as sensitizers.

Consequently, far from having a reasonable expectation of success with the proposed substitution, there was no teaching in any of the documents that would have established the slightest connection between the presence of aromatic dialkylamino substituents on the one hand and the degree of spectral sensitivity of a sensitizer for hexaarylbiimidazole on the other.

6.11 In summary, the solution of the stated problem did not arise in an obvious manner from the state of the art. On the contrary, the improved sensitivity performance with simultaneously good storage stability obtained using the dialkylamino-substituted sensitizers must be regarded as surprising.

7. Hence, the subject-matter of Claim 1 involves an inventive step. By the same token, Claims 2 to 5, which are dependent on Claim 1, are also directed to inventive subject-matter.

8. Since the Respondent's main requests are to be granted, there is no basis to grant its auxiliary request for oral proceedings and the final decision can therefore be directly made out in writing.

In view of the outstanding request of the Respondent concerning the correction of a printing error, in this connection, the Board has decided to make use of its power under Article 111(1) EPC to remit the case to the first instance for the issue of a corrigendum.

Order

For these reasons it is decided that:

1. The appeal is dismissed.

2. The case is remitted to the first instance with the order to issue a corrigendum in the sense of the request set out in section VI.(2), above.

The Registrar: The Chairman:

P. Martorana C. Gérardin