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DECISION of 25 October 1995

Case Number:	T 0913/92 - 3.3.3
Application Number:	85102065.1
Publication Number:	0156170
IPC:	C08F 2/02

Language of the proceedings: EN

Title of invention:

Bulk polymerization process for preparing high solids and uniform copolymers

Patentee:

S. C. JOHNSON & SON, INC.

Opponent:

BASF Aktiengesellschaft, Ludwigshafen

Headword:

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Relevant legal provisions: EPC Art. 56

Keyword:

"Inventive step - affirmed; omission of feature taught as essential in prior art"

Decisions cited: T 0339/91; T 0237/93; T 0229/85

Catchword:

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Case Number: T 0913/92 - 3.3.3

D E C I S I O N of the Technical Board of Appeal 3.3.3 of 25 October 1995

Appellant:BASF Aktiengesellschaft, Ludwigshafen(Opponent)-Patentabteilung- C6 -
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Representative:

Respondent: S. C. JOHNSON & SON, INC (Proprietor of the patent)1525 Howe Street Racine Wisconsin 53404-5011 (US)

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Representative:	Baillie, Iain Cameron
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Decision under appeal: Decision of the Opposition Division of the European Patent Office dated 8 July 1992, issued in writing on 20 August 1992 rejecting the opposition filed against European patent No. 0 156 170 pursuant to Article 102(2) EPC.

Composition of the Board:

Chairman:	С.	Gé	rardin
Members:	R.	Young	
	w.	М.	Schar

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 156 170, relating to "Bulk polymerization process for preparing high solids and uniform copolymers", in respect of European patent application No. 85 102 065.1, filed on 25 February 1985 and claiming a US priority of 29 February 1984 (US 584661), was announced on 11 January 1989 (cf. Bulletin 89/02). Claim 1 read as follows:

> "A continuous bulk polymerization process for preparing enhanced yields of high solids, non-styrenic acrylic polymer product having a number average molecular weight of about 1000 to 2500, a polydispersity ratio of less than about 3; a dispersion index of up to about 5 and a low chromophore content characterized by the steps of continuously:

(a) charging into a continuous mixed reactor zone containing a molten resin mixture consisting essentially of:

(i) at least one acrylic monomer;

(ii) a polymerization initiator in amounts to provide a molar ratio of said initiator to said acrylic monomer from about 0.0005 to 0.06:1, (iii) from about 0 to 25 percent based on the weight of acrylic momomer of a reaction solvent, wherein said molten resin mixture comprises unreacted acrylic monomers and the acrylic polymer product; (b) maintaining a flow rate through said reaction zone sufficient to:

(i) provide a residence time of said charged acrylic monomer in said reaction zone of from about 1-30 minutes; and
(ii) maintain a predetermined level of reaction mixture in said reaction zone, and;

(c) maintaining the molten resin mixture at an elevated temperature with the range of from about 180°C. to 270°C. sufficient to provide accelerated conversion to a readily processable, uniform, concentrated polymer product."

Claims 2 to 8 were dependent claims directed to elaborations of the process of Claim 1.

- II. Notice of Opposition was received on 11 October 1989 on the grounds of Article 100(a) and 100(b) EPC. The opposition was supported inter alia by the documents:
 - D1: DE-A-2 728 459;
 - D2: DE-B-2 502 172;
 - D3: DE-A-2 534 603;
 - D4: DE-A-3 140 383;
 - D5: WO-A-82 02387; and the later filed, but admitted D7: US-A-4 117 235.
- III. By a decision which was given at the end of oral proceedings held on 8 July 1992 and issued in writing on 20 August 1992, the Opposition Division rejected the opposition.

According to the decision, the description, which included worked examples satisfying the requirements of Claim 1, was sufficient to comply with Article 100(b) EPC. As to novelty, none of the cited documents disclosed both the monomers and the process steps of Claim 1. With regard to inventive step, there was no reason to modify the only one of these documents, D5, relating to a similar problem in respect of the three admittedly distinguishing features, since this would constitute an ex post facto analysis.

IV. On 30 September 1992 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 20 November 1992 as well as in a further written submission filed on 7 December 1993, the Appellant (Opponent) argued essentially as follows:

- (a) D5 was concerned with a continuous bulk polymerisation process as claimed in the patent in suit, except that styrene was essential and no initiator was used, the reaction temperature range overlapping that claimed.
- (b) The skilled person was aware from the teachings of D1 to D4, which disclosed corresponding bulk polymerisation processes carried out with or without initiator and with or without styrene monomer, that styrene was not essential. Consequently, there could be no prejudice against its omission.

- (d) Hence, it would be obvious for the skilled person, confronted with the problem of carrying out the process of D5 in the absence of styrene, to utilise a certain amount of initiator instead.
- V. The Respondent (Patentee) argued in essence as follows:
 - (a) Only D1 and D5 were relevant to the problem of the patent in suit because none of the other documents mentioned molecular weight distribution in terms of polydispersity and distribution index. It was clear, however, that both D1 and D5 were essentially concerned with styrenic polymers, whereas the patent in suit was concerned with nonstyrenic polymers.
 - (b) On the question of prejudice, it was believed, at the priority date of the patent in suit, that styrene-type monomers were indeed essential if the required properties were to be obtained, as had been stated in the patent in suit itself.
 - (c) Accordingly, it could not be a mere optional step to remove the styrene. No evidence had been brought to show that the skilled person would have expected the desired narrow molecular weight distribution to be obtained if styrene were omitted, let alone at the high yields disclosed.
- VI. In a communication issued on 25 August 1995, the Board informed the parties that the document D8: EP-A-96 901 was a more relevant state of the art than D5, and would be considered during the oral proceedings to be held on 25 October 1995.

- VII. With a letter dated 25 September 1995, the Respondent submitted an amended set of Claims 1 to 8 corresponding to Claims 1 to 8 as granted, except that in Claim 1, step (a) the expression "continuous mixed reactor zone" had been replaced by "continuous stirred reactor zone", as well as amended pages 4 and 9 of the description of the patent in suit.
- VIII. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the patent be maintained with the amendments filed on 25 September 1995.

Reasons for the Decision

- 1. The appeal is admissible.
- 2. Allowability of amendments
- 2.1 The amendment in Claim 1 (see section VII, above) is supported by the description on page 8 at lines 64 and 65 of the granted patent (application as filed, page 22, final paragraph) and does not involve any broadening in scope.
- 2.2 The amendment to the description on page 4 corresponds to that made in Claim 1, and on page 9 the sentence at lines 12 and 13 has been deleted. The latter deletion merely concerns an alternative means of providing a suitable apparatus for carrying out the claimed process.

No objection to the amendments was raised under Article 123 EPC by the Appellant. Nor does the Board see any such objection.

Consequently, the amendments are allowable under Article 123 EPC.

- 3. Interpretation of Claim 1
- 3.1 It was questioned by the Appellant during the oral proceedings whether the term "non-styrenic" acrylic polymer product in Claim 1 was intended to exclude polymers containing monoalkenyl aromatic monomer units in general or only those containing styrene itself.

The Respondent declared during the oral proceedings, however, that the term "non-styrenic" in Claim 1 was intended to be interpreted adjectivally, i.e. as meaning "non-styrene-like" and therefore to exclude all monoalkenyl aromatic monomers and not just styrene itself.

This interpretation is in the Board's view in any case consistent with the phrase "consisting essentially of... (i) at least one acrylic monomer" defining the mixture charged into the continuous stirred reactor zone. The phrase "consisting essentially of" could not, in the Board's view, be construed as excluding, say, only styrene itself and not other styrene-like (monoalkenyl aromatic) monomers.

Consequently, the Board takes the view that Claim 1 can only be interpreted in the sense of the declaration by the Respondent. 3.2 As to the term "continuous stirred reaction zone" (CSTR) in Claim 1 and elsewhere, the definition of such a zone was the subject of deliberation by the Board in earlier, related proceedings involving the same parties (T 0339/91 and T 0237/93, both of 12 July 1995) and was held, in relation to the subject-matter of the patents then in suit (corresponding to D5 and D8, respectively), to be a zone having a mixing profile with no concentration or temperature gradients, and in which back mixing was essential (cf. T 0339/91, Reasons for the Decision, paragraph 4.4).

The term does not have a different meaning in the present case, and hence the same interpretation is applicable.

4. The closest state of the art

The patent in suit is concerned with a bulk polymerisation process capable of selectively providing high yields of high purity, low molecular weight acrylic polymers suitable for high solids applications, the term "acrylic polymers" referring to the addition polymer formed by polymerising acrylic monomers (page 3, line 64 to page 4, line 2).

4.1 Such a process is known from D7, which discloses the polymerisation of acrylic monomers to give a nonstyrenic acrylic polymer product of low molecular weight and low viscosity and thus relates to the same problem as the patent in suit. A further line of argument starting from D8 as closest state of the art was presented at the oral proceedings by the Appellant and will be considered in turn. From a systematic point of view, however, D7 is considered to form the closest state of the art.

4.2 According to D7, a low molecular weight liquid, polymeric, thermally removable coating vehicle is prepared by polymerising at least one acrylic monomer, e.g. an alkyl methacrylate, at elevated pressure (2 to 10 atmospheres) and at a temperature of about 200°C to 300°C., and close to but not exceeding the "ceiling temperature", i.e. the temperature at which the rate of polymerisation becomes equal to the rate of depolymerisation, the resulting polymeric material having a number average molecular weight (Mn) less than about 5 000 and a viscosity at 25°C of less than about 200 000 centipoise (Claim 1; column 1, lines 40 to 45).

> The process may be carried out in the presence or absence of a polymerisation catalyst (column 2, lines 1 to 5).

> According to the examples, 10 ml samples of n-butyl methacrylate (Examples 1, 3) or methyl methacrylate (Example 2) were heated under nitrogen in a sealed tube for 16 h at 262°C (Example 1), at 230°, 240°, 248° and 280°C (Example 3) or for 18 h at 230°C (Example 2). The resulting polymers were liquid, with polymerisations at higher temperatures yielding products of lower viscosity (column 5, lines 29 to 63).

This process is thus a batch process carried out on a laboratory scale and has the disadvantage that excessively long reaction times of 16 to 18 h are necessary (cf. Examples). No information is given about

- 8 -

molecular weight distribution in terms of a polydispersity ratio or a distribution index.

- 4.3 Compared with this state of the art, the technical problem is to be seen as providing an improved process of preparing such wholly acrylic polymers more rapidly and on a larger scale, in high (commercial) yield, in particular to provide a non-styrenic acrylic polymer product having a number average molecular weight of about 1000 to 2500, a polydispersity ratio of less than about 3, a dispersion index of up to about 5 and a low chromophore content (low colour).
- 4.4 The solution proposed according to Claim 1 of the patent in suit is to replace the pressurised discontinuous (batch) process of D7 by a continuous bulk polymerisation process which may be carried out at atmospheric pressure and involves the use of a continuous stirred reactor zone (CSTR), into which acrylic monomer mixture containing a small amount of initiator (molar ratio of initiator to acrylic monomer mixture 0.0005:1 to 0.06:1) and optionally some solvent (0 to 25 wt% of the acrylic monomers) is continuously charged, and through which the flow rate is maintained to provide a residence time of the monomer mixture of about 1 to 30 minutes and a predetermined level of the reaction mixture in the reaction zone.
- 4.5 That this process results in enhanced yields (well over 90% of theoretical) of high solids, non-styrenic acrylic polymer product having the required molecular weight and molecular weight distribution in terms of polydispersity ratio and distribution index as well as low colour is evidenced by the large number of examples

- 9 -

in the patent in suit, especially, for instance, Examples 1 to 3.

Consequently, the Board finds it credible that the claimed measures provide an effective solution of the technical problem.

5. Novelty

- 5.1 The Board concurs with the finding in the decision under appeal, according to which none of the documents D1 to D7 discloses a process where both the monomers used and the process steps taken are the same as in Claim 1.
- 5.2 Whilst D8 admittedly relates to a process identical in almost every essential respect with that of the process of the patent in suit, nevertheless, in D8, both Claim 1 and the statement of invention on page 7 require the presence of "at least one monoalkenyl aromatic monomer", as do all the worked examples.

Although there is an isolated statement on page 9 of D8, that "The mixture of vinyl monomers employed to form the vinylic polymer product preferably includes at least one monoalkenyl aromatic monomer and at least one acrylic monomer", it is clear from the commentary on D8 in the description of the patent in suit, that it had previously been thought that the presence of styrenetype monomers was necessary to the overall kinetics of bulk polymerisation to produce a polymer having a tight molecular weight distribution, low viscosity and low colour (page 6, lines 25 to 28). This is corroborated by the description in D8 itself, according to which "The reaction is postulated to be primarily thermally initiated with the polymerization initiator (catalyst) acting to help define the purity, weight distribution, molecular weight and yield of the resulting polymer." (page 8, lines 8 to 12).

In this connection, it was accepted by both parties that styrene was generally known to be typically thermally polymerised (Statement of Grounds of Appeal, page 2, pre-penultimate paragraph).

Thus, the argument of the Respondent, at the oral proceedings, that the word "preferably" should be construed in the light of the claims and statement of invention, was not contested by the Appellant. On the contrary, the Appellant was also of the opinion that D8 required, as an essential feature, the presence of a monoalkenyl monomer (submission filed on 24 August 1995, page 2).

Consequently, it is accepted by the Board that D8, properly construed, must be regarded as requiring the presence of a monoalkenyl aromatic monomer.

Even if independent account were taken of the isolated statement on page 9, it in any case links the acrylic monomers unambiguously with monoalkenyl aromatic monomers.

Consequently, D8 fails to disclose a process where both the monomers used and the process steps taken are the same as in Claim 1 of the patent in suit. Thus the subject-matter of Claim 1 is novel.

6. Inventive Step

- 6.1 To assess the question of inventive step, it is necessary to consider whether the skilled person, starting from D7 would have considered making the combined modifications of the solution specified in section 4.4 above, in the expectation of achieving an improved, commercial yield of acrylic polymer product of the required low molecular weight, narrow molecular weight distribution and low colour.
- 6.2 There is no suggestion to make these modifications in D7 itself, because this document discloses only a discontinuous batch process which is exemplified as taking many (16 to 18) hours reaction time to produce a yield inferior to that of the patent in suit. In particular, there is no disclosure of a CSTR.
- 6.3 As to the process of D8, this requires the presence of a monoalkenyl aromatic (styrenic) monomer (section 5.2, above).

The argument of the Appellant, that the skilled person would consider replacing the styrene and would find it obvious, in the absence of styrene, to add an initiator is unconvincing, for the following reasons.

6.3.1 On the one hand, it is not at first sight obvious to omit something which is taught as essential in a document. Consequently, for the skilled person considering the disclosure of D8 in relation to the - 13 -

stated problem, the question of omitting styrene would not arise.

In any case, whilst D8 describes a copolymer of two types of monomers, these two categories are in practice not equivalent, since styrene has a dual function in that it also acts as a thermal initiator (see section 5.2, above). Thus, even a broad interpretation of D8, according to which the two types of monomers would not be compulsory, would still not lead the skilled person to dispense with styrene, but if anything rather with the acrylic monomer.

6.3.2 On the other hand, according to the uncontested submission of the Respondent at the oral proceedings, the half-life of a typical peroxide initiator at the temperatures utilised in the claimed process (180° to 270°C) was short compared with the necessary residence time in the reaction zone, and further depletion of the initiator compared with the input concentration in any case occurred during the continuous flow through the reaction zone.

> Thus, to replace styrene would have required excessively large quantities of initiator (according to the Appellant at the oral proceedings at least 10 to 15 wt%).

In the Board's view, the skilled person, faced with this situation, in which styrene was considered essential, and the peroxide-type initiator was not considered capable surviving in sufficient quantities to sustain the reaction, let alone to assure adequate control over the molecular weight and molecular weight distribution of the product, would have regarded such replacement either as not feasible at all, or else as requiring addition of the initiator in such large quantities as would have risked prejudicing the quality (colour) of the product. Such quantities would in any case have exceeded the limits permitted by the solution of the technical problem (section 4.4, above).

- 6.4 The further argument of the Appellant, that such polymerisation reactions were nevertheless known from D1 to D4 to have been carried out in the absence of styrene and still using only small quantities of initiator (0.1 to 3 wt%) is not supported by the relevance or the disclosures of the documents themselves.
- 6.4.1 Of the documents D1 to D4, which relate to the bulk preparation of copolymers from mixtures including inter alia acrylic monomers, D1 to D3 have been the subject of scrutiny by the Board in the earlier, related proceedings already referred to (section 3.2, above). Indeed, according to these earlier decisions of the Board, the documents here numbered D1, D2 and D3 were found not to disclose the use of a CSTR in the sense of the term used in D5 or D8. Furthermore, according to the first-mentioned of these decisions, these documents were found, except in the case of D1, also to fail to disclose a polymer product having a narrow molecular weight distribution, expressed in numerical terms corresponding to polydispersity ratio (see also T 0339/91, Reasons for the decision, paragraph 6.1.6.3, 6.2 and 6.3.6 in relation to the documents there numbered D1, D2 and D3 respectively).

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- 14 -

Since no further submissions in relation to these documents are on file which would throw new light on the matter in the present case, the Board sees no reason to alter its previous findings in this connection.

Hence D1, D2 and D3 are held not to disclose the use of a continuous stirred reactor zone, nor, in the cases of D2 and D3, polymer products of the low polydispersity ratio etc., which, according to D5 and D8 are achievable with such a zone.

6.4.2 Although D4 discloses a bulk polymerisation process which is continuous in the sense that a molten copolymer mixture is removed from a pressure vessel at the same rate that fresh monomer mixture of monomer is charged (page 10, Examples 1 to 6), there is no mention of a stirrer of any kind. Even if the presence of a stirrer were inferred from the safety point of view, however, there is no indication that such stirring would be of a kind to establish temperature and gradient free conditions corresponding to a CSTR.

> This is borne out (i) by the nature of the products contemplated by D4 (organic acid builders for washing powders) in which the viscosity properties attainable with narrow molecular weight distributions are of no interest, and (ii) the molecular weight of such products, as shown by the results in Table 1, in which the K-values are indicative, according to the uncontested submission of the Respondent at the oral proceedings, of a molecular weight in excess of about 15 000, i.e. far above the maximum allowed by the solution of the technical problem. Hence, there is no

evidence that the conditions for a low molecular weight and narrow molecular weight distribution corresponding to a CSTR zone have implicitly been fulfilled.

Consequently, D4 is also held not to disclose, explicitly or implicitly, the use of a CSTR or the preparation of polymer products of low molecular weight and narrow molecular weight distribution.

In summary, whatever the recommendations of D1 to D4, they are evidently not made in the context of a CSTR process for obtaining a polymer product of the kind having a low molecular weight and a narrow molecular weight distribution with which the solution of the technical problem is concerned.

6.4.3 In any case, D1 and D2 contain no specific teaching to omit styrene whilst maintaining a low level of initiator and indeed all the examples of preparation of acrylic monomer-containing polymers in both these documents include the presence of styrene.

> Consequently, the skilled person would not have derived from the teaching of either D1 or D2 that styrene could be dispensed with where only a small amount of initiator is present.

6.4.4 Although all the examples of D4 and certain of the examples of D3 disclose acrylic monomer-containing polymers without styrene, and furthermore contain general references to a small amount of initiator being present, closer examination of these examples reveals that maleic acid anhydride is invariably present as a monomer. According to an uncontested submission of the Respondent at the oral proceedings, maleic acid anhydride was known to have a propensity to dimerize and then polymerise quickly by an ionic mechanism.

Consequently, the skilled person would have understood from such examples that they were not examples of conventional radical polymerisation in the sense of D8, but rather of some other mechanism of polymerisation. Hence the examples referred to have no relevance for the question of replaceability of styrene in D8.

Hence, there is nothing in the disclosures of D1 to D4, whether considered from the point of view of the general character of the processes involved, or the combinations of ingredients specified, which would lead the skilled person to suppose that the styrene component could be omitted from such polymers produced by radical bulk polymerisation using a CSTR type process as disclosed in D8.

Consequently, a combination of the disclosure of D7 with that of D8 would not, even in the light of the disclosures of D1 to D4 lead, to the solution of the technical problem.

6.5 Conclusions similar to those arrived at in relation to D8 also apply in relation to D5, the disclosure of which is similar to that of D8, except that no initiator is disclosed and the range of temperature of the polymerisation reaction is somewhat higher, since such a reaction in the absence of initiator must also be thermally initiated and the same essential requirement for the presence of a styrene-type monomer also applies.

Consequently, a combination of the disclosure of D7 with that of D5 would also not, even in the light of the disclosures of D1 to D4, lead to the solution of the technical problem.

- 6.6 As to the argument, put forward by the Appellant at the oral proceedings, concerning a combination of the disclosures of D1 to D4 directly with that of D7, the result would not be something corresponding to the solution of the technical problem, because, for the reasons given above, none of the documents in question discloses the use of a CSTR.
- 6.7 In summary, the subject-matter of Claim 1 does not arise in an obvious way starting from D7 as closest state of the art.
- 7. In view of the line of argument followed by the Appellant at the beginning of the oral proceedings, the Board regards it appropriate to investigate whether a different result would have been obtained starting from D8 as closest state of the art (cf. section 4.1, above).
- 7.1 As indicated in section 5.2 above, the disclosure of D8 differs from what is claimed in the patent in suit essentially only in its requirement for the presence of a styrene-type monomer.
- 7.2 Although the statement of problem arising from this state of the art, as presented by the Appellant at the

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- 18 -

oral proceedings, was "to produce further acrylic polymers in the absence of styrene", this statement of problem is not appropriate. According to the jurisprudence of the Boards of Appeal, it is not permissible to formulate the technical problem in such a way as to contain pointers to the solution (T 0229/85, OJ EPO 87, 237). In this case, the statement of problem not only contains pointers to its solution, but to all intents and purposes is the solution.

- 7.3 A more neutral statement of problem starting from D8 is in the Board's view "to provide further applications of the process of D8 for preparing acrylic polymers of low molecular weight (in particular, a number average molecular weight of about 1000 to 2500), narrow molecular weight distribution (in particular a polydispersity ratio of less than about 3 and a dispersion index of up to about 5), and having a low chromophore content (low colour)".
- 7.4 The solution proposed according to Claim 1 is to replace the styrene-type monomer with a monomer mixture consisting essentially of at least one acrylic monomer, i.e. to omit the styrenic monomer, and thus arrive at a non-styrenic polymer product of the same molecular weight and molecular weight distribution characteristics as in D8.
- 7.5 It is credible that the proposed measure provides an effective solution of the technical problem, for the reasons given under section 4.5, above.

- 20 -

- 7.6 The solution of the technical problem is novel, for the reasons given under section 5.1, above.
- 7.7 To determine the issue of inventive step, it is necessary to consider whether the skilled person, starting from the disclosure of D8, would expect to get wholly acrylic, non-styrenic polymer products of the same quality and molecular weight characteristics if the styrene-type monomer were omitted from the monomer charge to the CSTR reaction zone.

This question has to be answered in the negative, for the same reasons as given under section 6.3 etc., above, in relation to D8 itself and under section 6.4 etc, above, in relation to a combination of its disclosure with that of D7, with or without any of D1 to D4.

Hence the solution to the technical problem does not arise in an obvious way starting from D8 as closest state of the art, either.

- 7.8 On the contrary, as the prime source of initiation in D8 was believed to be the styrene-type monomer, it appears in the Board's view surprising that such a relatively small amount of initiator in the absence of styrene could produce products having a narrow molecular weight range and a low molecular weight as is found according to the patent in suit, let alone in the high yields disclosed.
- 8. In view of the above, the subject-matter of Claim 1 is not only novel, but involves an inventive step. The subject-matter of Claims 2 to 8, which are directly or

indirectly dependent on Claim 1 is by the same token also novel and involves an inventive step.

Order

For these reasons it is decided that:

- 1. The decision under appeal is set aside.
- 2. The case is remitted to the first instance with the order to maintain the patent on the following basis:

Claims:	1 to 8 as filed on 25 September 1995;
description:	pages 1 to 3, 5 to 8 and 10 to 14 of the
	patent as granted and pages 4 and 9 as
	filed on 25 September 1995.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin