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Datasheet for the decision of 17 July 2008

Case Number:	T 1469/05 - 3.3.03
Application Number:	97305727.6
Publication Number:	0822222
IPC:	C08K 5/05
Language of the proceedings:	EN

Title of invention:
Method for decolorizing polyphenylene ether resins

Applicant:
GENERAL ELECTRIC COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Relevant legal provisions (EPC 1973):
-

Keyword:
-

Decisions cited:
T 0013/84

Catchword:
-

Case Number: T 1469/05 - 3.3.03

DECISION
of the Technical Board of Appeal 3.3.03
of 17 July 2008

Appellant:	GENERAL ELECTRIC COMPANY 1 River Road Schenectady NY 12345 (US)
Representative:	Bublak, Wolfgang Bardehle, Pagenberg, Dost, Altenburg, Geissler Galileiplatz 1 D-81679 München (DE)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted 31 May 2005 refusing European application No. 97305727.6 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman:	R. Young
Members:	C. Idez
	E. Dufrasne

Summary of Facts and Submissions

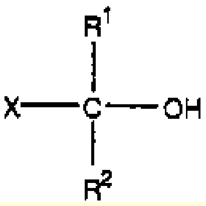
I. European patent application No. 97 305 727.6, filed on 30 July 1997 in the name of General Electric Company and claiming the priority of two earlier applications in the United States of America (US 689122 of 30 July 1996 and US 788666 of 24 January 1997) was refused by a decision of the Examining Division issued in writing on 31 May 2005.

II. The decision of the Examining Division was based on a main request consisting of Claim 1 as filed with letter dated 25 April 2003, and of Claims 2 to 7 as filed with letter dated 22 October 2002, and on an auxiliary request consisting of Claim 1 as filed with letter dated 30 July 2004 and of Claims 2 to 7 as filed with letter dated 22 October 2002.

Claim 1 of the main request read as follows:

"A method for decolorizing a polyphenylene ether which comprises mixing it in solution in an organic solvent at a temperature in the range of about 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound having the formula

(I)



$$X-\overset{\displaystyle R^1}{\underset{\displaystyle R^2}{C}}-OH$$

,

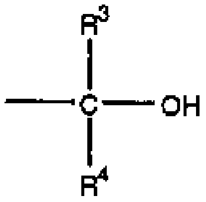
wherein:

each of R^1 and R^2 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,

X is

(II) -CZ or

(III)



$$-\overset{R^3}{\underset{R^4}{\overset{|}{\underset{|}{C}}}}-OH$$

,

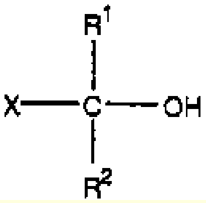
Z is at least one moiety linked to C by an aliphatic double bond or an aromatic bond, and
each of R^3 and R^4 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical.

Claims 2 to 7 were dependent claims.

Claim 1 of the auxiliary request read as follows:

"A method for decolorizing or suppressing color formation a polyphenylene ether during melt processing which comprises mixing the polyphenylene ether in solution in an organic solvent at a temperature in the range of about 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound having the formula

(I)



$$X-\overset{\overset{R^1}{|}}{\underset{\underset{R^2}{|}}{C}}-OH$$,

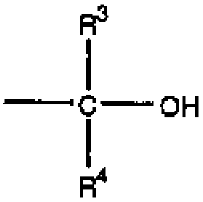
wherein:

each of R^1 and R^2 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,

X is

(II) -CZ or

(III)



$$-\overset{\overset{R^3}{|}}{\underset{\underset{R^4}{|}}{C}}-OH$$,

Z is at least one moiety linked to C by an aliphatic double bond or an aromatic bond, and
each of R^3 and R^4 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical, prior to melt processing."

Claims 2 to 7 corresponded to Claims 2 to 7 of the main request.

III. According to the decision of the Examining Division, the subject-matter of Claims 1 to 7 of the main request was obvious starting from either document D5 (US-A-4 060 514), D7 (EP-A-0 148 800), or D13 (US-A-3 681 285) which all disclosed a method for decolorizing polyphenylene ethers (PPE) by mixing them with a thio compound in an organic solvent at a temperature of 20-

50°C. The subject-matter of Claims 1 to 7 of the auxiliary request was considered as obvious starting from document D1 (US-A-4 695 601) which disclosed a method for decolorizing PPE during melt processing by mixing PPE with an alphahydroxyketone such as benzoin.

IV. Notice of Appeal was filed on 1 August 2005 by the Appellant (Applicant) with simultaneous payment of the prescribed fee. In the Statement of Grounds of Appeal filed on 5 October 2005, the Appellant contested the findings of the Examining Division concerning inventive step of the subject-matter of the main request and of the auxiliary request.

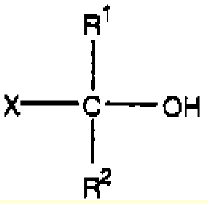
V. A communication was issued on 22 April 2008 by the Board, in which the Board gave its preliminary view concerning issues under Articles 84, 54 and 56 EPC concerning the main request and the auxiliary request.

VI. With its letter dated 17 June 2008, the Appellant submitted a new main request and two new auxiliary requests.

Claim 1 of the main request read a follows:

"A method for decolorizing a polyphenylene ether which comprises mixing it in solution in an organic solvent at a temperature in the range of 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound having the formula

(I)



$$X-\overset{\displaystyle R^1}{\underset{\displaystyle R^2}{\overset{|}{\underset{|}{C}}}}-OH$$

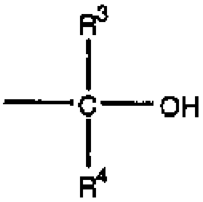
,

wherein:

each of R^1 and R^2 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,

X is
(II) -CZ or

(III)



$$-\overset{\displaystyle R^3}{\underset{\displaystyle R^4}{\overset{|}{\underset{|}{C}}}}-OH$$

,

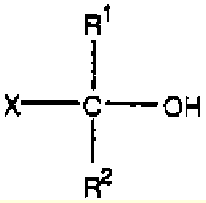
wherein -CZ may be any acyclic or cyclic radical in which at least one moiety is linked to the depicted carbon atom by an aliphatic double bond or an aromatic bond, and
each of R^3 and R^4 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical.

Claims 2 to 7 were dependent claims.

Claim 1 of the first auxiliary request read as follows:

"A method for decolorizing a polyphenylene ether which comprises mixing it in solution in an organic solvent at a temperature in the range of 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound having the formula

(I)



$$X-\overset{\overset{R^1}{|}}{\underset{\underset{R^2}{|}}{C}}-OH$$

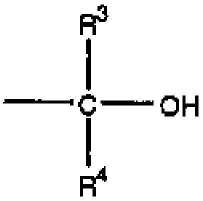
,

wherein:

each of R^1 and R^2 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,

X is

(III)



$$-\overset{\overset{R^3}{|}}{\underset{\underset{R^4}{|}}{C}}-OH$$

,

wherein each of R^3 and R^4 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,
or of at least one of the hydroxy compounds of the following group:
benzoin, acetoin, allyl alcohol, acetol, 2-butene-1,4-diol.
Claims 2 to 7 of the first auxiliary request corresponded to Claims 2 to 7 of the main request

Claim 1 of the second auxiliary request read as follows:

"A method for decolorizing a polyphenylene ether which comprises mixing it in solution in an organic solvent at a temperature in the range of 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound of the group consisting of benzopinacol, benzoin, acetoin, allyl alcohol, acetol, 2-butene-1,4-diol, benzyl alcohol or hydrobenzoin, benzhydrol, methylphenylcarbinol, pinacol or 2,3-diphenylbutane-2,3,diol."

Claims 2 to 6 of the second auxiliary request corresponded to Claims 2 to 6 of the main request.

The Appellant also argued essentially as follows:

(i) Concerning Article 84 EPC:

(i.1) Claim 1 of the main request stated that "the radical —CZ may be any acyclic or cyclic radical in which at least one moiety is linked to the depicted carbon atom by an aliphatic double bond or an aromatic bond."

(i.2) This made clear that the group —CZ was in its entirety a cyclic or acyclic radical which was substituted at least once with Z. The remaining substituent(s) could be arbitrarily selected.

(i.3) The term "aromatic bond" in that context meant that CZ was part of an aromatic system wherein the bond between C and Z was aromatic, i.e. both C and Z were sp^2-hybridized.

(ii) Concerning inventive step:

(ii.1) The term "decolorization", as stated in the specification on page 2, lines 45-47, had to be interpreted as substantial reduction of the color index of the freshly polymerized PPE resin.

(ii.2) The Board had considered in its Preliminary Opinion that none of the examples reflected the claimed invention and that there was no comparison of color before and after treatment.

(ii.3) Example 4 however reflected the claimed invention. The omission of the temperature at which the treatment was carried out implied that the treatment was carried out at ambient temperature, i.e. within the range 20 to 50°C.

(ii.4) Although a direct comparison of the coloring before and after treatment in the solvent was not given in Example 4, a reduction of the initial coloring of the freshly polymerized PPE in said Example 4 was demonstrated if one would take the entire disclosure of the application in suit into consideration.

(ii.5) Reference was made to Example 1 of the application in suit in that respect. It showed that a slight coloration occurred when PPE containing benzoin was heat processed. It could thus be deduced that the reduction in coloring observed in Example 4 from 4.32

to 2.90 had to be attributed to reduction of coloration during solution treatment.

(ii.5) Furthermore, it was known that rapid decolorization of freshly polymerized PPE occurred in solution at ambient temperatures with dithionate (cf. D5) or with thiols (cf. D7). This supported that the alcohols according to the application in suit were capable of reducing color in solution at ambient temperature.

(ii.6) Thus, Example 4 demonstrated the claimed technical effect of decolorization upon treatment of PPE in solution with the hydroxyl compounds according to theapplication in suit.

(ii.7) The closest prior art was represented by teachings related to solution treatment of PPE, i.e. D5 and D7.

(ii.8) In these documents a different decolorizing agent was employed.

(ii.9) D1 related to the prevention of color formation during heat treatment.

(ii.10) Dl was not at all concerned with the removal of coloration in freshly polymerized PPE.

(ii.11) Thus, the objective technical problem solved by the application in suit invention was the provision of an improved solution-based method for the removal of colored PPE polymerization by-products at low temperatures.

(ii.12) There was no indication in the prior art that the claimed alcohols could serve such a purpose.

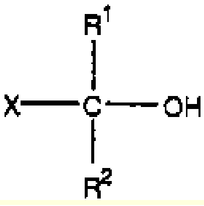
VII. Oral proceedings were held before the Board on 17 July 2008.

(i) At the oral proceedings the discussion firstly focussed on the question of clarity of Claim 1 of the main request submitted with letter dated 17 June 2008, in particular in respect of the presence of the group CZ in the definition of the hydroxy compound to be used as decolorizing agent in the claimed method. Following that discussion, the Appellant withdrew the main request and the first auxiliary request both filed with letter dated 17 June 2008, and it submitted a new main request consisting of 7 claims.

Claim 1 of the main request read as follows:

"A method for decolorizing a polyphenylene ether which comprises mixing it in solution in an organic solvent at a temperature in the range of 20-50°C with 0.2 to 50 parts by weight per 100 parts of polyphenylene ether of at least one hydroxy compound having the formula

(I)



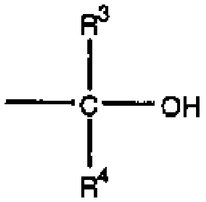
$$X-\overset{R^1}{\underset{R^2}{C}}-OH$$

,

wherein:

each of R^1 and R^2 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical, X is

(III)



$$-\overset{R^3}{\underset{R^4}{\overset{|}{\underset{|}{C}}}}-OH$$,

wherein each of R^3 and R^4 is hydrogen or a C_{1-4} alkyl or C_{6-10} aryl radical,
or of at least one of the hydroxy compounds of the following group:
benzoin, acetoin, allyl alcohol, acetol, 2-butene-1,4-diol, benzyl alcohol, benzhydrol, methylphenylcarbinol.

Claims 2 to 7 corresponded to Claims 2 to 7 of the previous main request.

(ii) The discussion then moved to the question of inventive step of the subject-matter of Claim 1 of the main request. The arguments presented in that respect by the Appellant may be summarized as follows:

(ii.1) The aim of the claimed method was to reduce the coloration of freshly polymerized PPE.

(ii.2) Example 4 of the application in suit demonstrated that this problem was effectively solved by the claimed method. While admitting that there was no comparison between the color of the starting PPE ether resin and its color after treatment with benzoin in toluene, it was evident from the comparison between the yellowness index of the PPE resins after extrusion that the treatment in the toluene solvent had been effective for reducing the color of the fresh polymerized PPE resins.

(ii.3) This was further confirmed by the statement at the bottom of Table IV according to which "it is apparent from the yellowness index that color is effectively decreased".

(ii.4) It was not obvious to replace the thiol compounds used in D7 by the present alcohol compounds, since alcohols and thiols were totally different chemical entities. It could not have been expected that alcohols would behave as thiols.

(iii) The Board, after deliberation, informed the Appellant of its view that Example 4 of the application in suit did not appear to show that the color of the freshly polymerized PPE had been reduced by the treatment with benzoin in toluene, and that Example 4 only showed that the increase of coloration after melt processing was reduced when the PPE resin had been mixed with the hydroxy compound (i.e. benzoin). In this connection, the Appellant was invited to present its comments on document D1.
The Appellant essentially submitted that D1 clearly taught to mix the benzoin at high temperature and that decomposition of the benzoin would take place. It did not hence suggest treating the PPE with benzoin in solution. The same line of argument would apply in its view to the subject-matter of the second auxiliary request as submitted with letter dated 17 June 2008.

VIII. The Appellant requested that the decision under appeal be set aside, and a patent be granted on the basis of the main request (claims 1 to 7) filed during the oral proceedings, or, in the alternative, on the basis of the 2nd auxiliary request (claims 1 to 6) filed with letter dated 17 June 2008.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. *Wording of the claims*

The Board is satisfied that Claims 1 to 7 of the main request meet the requirements of Article 123(2) and 84 EPC.

3. *Novelty*

3.1 In its decision the Examining Division has considered that the subject-matter of the main request then on file was novel over the cited prior art. In its communication dated 22 April 2008, the Board indicated that it saw no reason to depart from that view.

3.2 Since the subject-matter of Claim 1 of the present main request has been further restricted, the Board is satisfied that the requirements of Article 54 EPC are met by Claims 1 to 7 of the main request.

4. *Problem and solution, closest state of the art*

4.1 According to the Appellant the aim of the claimed method according to Claim 1 is to reduce the color of freshly polymerized PPE resins.

4.2 In that context, document D7 which is concerned with the problem of reduction of coloration in freshly polymerized PPE resins would, in the Board's view, represent the closest state of the art.

4.3 According to D7, a substantial decolorization of PPE resins obtained by oxidative polycondensation of alkylphenols in the presence of copper-amine catalysts can be achieved by treating a solution of the polymer, that may be the same mixture coming directly from the polymerization of the alkylphenols, with one or more compounds containing at least a sulphydrylic SH function (thiols) in an acid medium.

4.4 As disclosed in Examples 1 to 6, 11, 12, 14, 24, 29, 30 and 31 of D7 the treatment can be carried out in toluene at a temperature of 35°C.

4.5 According to these examples the color number of the treated PPE resins is between 2.2 (Example 29) and 4.6 (Example 5) while the untreated PPE (cf. control example, page 10, lines 25 to 26) exhibits a color number of 10, i.e. the treatment according to D7 effectively reduces the color of the fresh polymerized PPE resin.

4.6 In this connection the Board firstly notes that there is no comparison between the method disclosed in D7 and the method according to Claim 1 in the application in suit. Furthermore, no such comparison has been submitted by the Appellant in the course of the examination and appeal proceedings.

4.7 Thus, starting from D7, the technical problem would hence be seen in the provision of an alternative process for reducing the color of fresh polymerized PPE resins.

4.8 According to the Appellant, this problem is solved by using the specific alcohol components as defined in Claim 1 instead of thiols as taught in D7.

4.9 In that context, the Board, however, firstly notes that none of the Examples 1, 2, and 3 of the application in suit fall under the scope of Claim 1, because in these examples the PPE resin is only submitted to heat treatment in the melt (i.e. not in an organic solvent) with a specific hydroxy compound as defined in Claim 1.

4.10 While in Example 4 of the application in suit, the freshly polymerized PPE resins are treated with benzoin in toluene before extruding at 280°C the thus obtained mixtures of PPE resin and benzoin, the Board observes that the temperature at which the treatment in toluene has been carried out has not been indicated in that example.

4.11 Nevertheless, even if one would consider that, in Example 4, the treatment in toluene has been indeed carried out at a temperature between 20 and 50°C, the Board further observes that absolutely no indication is given in Example 4 concerning the color number either of the fresh polymerized PPE resin or of the PPE resin after treatment in toluene and before extrusion.

4.12 While it is true that in Table IV the yellowness index values of the PPE resins after extrusion are indicated, the Board can only state that there is no indication either of the yellowness index value of the fresh polymerized PPE resin or of the PPE resin after treatment with benzoin in the toluene solution and before extrusion.

4.13 Under these circumstances, the only evidence provided by Example 4 is that extruded PPE resins containing benzoin exhibit better yellowness index values than the same extruded PPE resins not containing benzoin (cf. comparison between Runs 1 to 2 (no benzoin) and Runs 3 to 5, and comparison between Run 7 (no benzoin) and Runs 8 to 10).

4.14 In other words, in view of the known problem of color formation in PPE resins during melt processing (cf. page 2, lines 26 to 27 of the application in suit), Example 4 shows only that color formation is less during melt processing of PPE resins containing benzoin than during melt processing of PPE resins not containing benzoin, but in no way it provides evidence that the color of the fresh polymerized PPE resins has been effectively decreased by contacting them with benzoin in toluene.

4.15 This conclusion cannot be altered by the statement at line 56 on page 6 concerning Example 4 that "it is apparent from yellowness index values that color is effectively decreased" for the following reasons:

4.15.1 As submitted by the Appellant the color number of fresh polymerized PPE resins obtained using copper/DBEDA catalyst system is generally of the order of 10 but their color number might increase to about 30 after melt processing (cf. application in suit page 2, lines 26 to 27).

4.15.2 On that basis, it can be considered that Table I shows that the color number of a PPE resin melt processed in the presence of a decolorizing agent increases from 10 to 11.6 in the case of benzoin to up to 23,5 in the case of 2,3 diphenylbutane-2,3-diol while the color number of a PPE resin melt processed without decolorizing agent increases to 25.

4.15.3 Nevertheless, despite the increase in color number of PPE resin containing the decolorizing agent, it is concluded at the bottom of Table I that "it is apparent from the results in Table I that the method of this invention is effective to reduce the color of polyphenylene ether resins".

4.15.4 Thus, under these circumstances, this statement can only make sense when interpreted as expressing that the color of PPE resins melt processed in the presence of the decolorizing agent is lower than the color of the polyphenylene resin melt processed in the absence of that decolorizing agent, i.e. that the color formation during melt processing is less in the presence of the specific hydroxy compounds (e.g. benzoin).

4.15.5 As indicated above in paragraphs 4.13 and 4.14, the results in Table IV only show that the yellowness index and hence the color of PPE resins extruded in the presence of a decolorizing agent (benzoin) are lower than the yellowness index and hence the color of the polyphenylene ether extruded in the absence of such decolorizing agent.

4.16 Consequently, there is no reason for the Board not to interpret the reduction of color referred to at the bottom of Table IV in the same manner as the reference at the bottom of Table I, i.e. as expressing only the fact that the color of PPE resins extruded in the presence of benzoin is lower than the color of the PPE resins extruded in the absence of such decolorizing agent, or, in other words, that the color formation during melt processing of PPE resins is less in the presence of the specific hydroxy compounds (e.g. benzoin).

4.17 The further argument of the Appellant in view of the low increase in color of PPE resins containing benzoin in Example 1 (cf. Section VI above, points (ii.4) to (ii.5)) that the reduction of yellowness index from 4.32 (Run 2 in Example 4) to 2.90 (Run 5 in Example 4), had to be attributed to the solvent treatment also cannot be accepted for the following reasons:

4.17.1 Independently of the fact that the heat treatment in Example 1 (compression molding at 270°C) is not the same as in Example 4 (extrusion molding at 280°C), and that the color changes are expressed in totally different manners in these examples (color number in Example 1 versus yellowness index in Example 4), so that a valid comparison between color changes in Example 1 and in Example 4 is more than questionable, this is because the respective contributions of the solvent treatment step and of the melt processing step in the color change of the PPE resin cannot be estimated in the absence of indication of the yellowness index of the fresh polymerized PPE, i.e. of any evidence that this index was very close to or greater than 2.90.

4.18 Nor could the further argument of the Appellant be accepted that the rapid decolorization of freshly polymerized PPE resins occurring in solution at ambient temperatures with thiols as shown in D7 would support that alcohols according to the application in suit were capable of reducing color in solution at ambient temperature (cf. Section VI above, point (ii.5)), because this argument is contradicted by the Appellant's own submissions that, alcohols and thiols being totally different chemical entities, it could not be expected that alcohols would behave as thiols (cf. Section VII above, point (ii.4)).

4.19 Consequently, there is no convincing evidence that the claimed measures provide an effective solution of the technical problem defined in paragraph 4.7 above. It thus follows that this technical problem cannot be used for the assessment of inventive step, and that the technical problem must be hence reformulated.

4.20 Nevertheless, it still remains that Example 4 shows that the claimed method leads to a reduced color formation during melt processing of PPE resins.

4.21 In that connection, the Board observes that document D1 is the only document which deals with the reduction of color formation in PPE resins during melt processing.

4.22 In Examples 1 and 3 of D1, a discoloration preventing additive (benzoin) is melt compounded with a PPE resin and the obtained mixture is injection molded. The obtained injection molded articles show a reduced color (higher lightness) than injection molded articles of PPE resin without benzoin (57 versus 41 in Example 1, 73 versus 59 in Example 3).

4.23 D1 teaches that the PPE and the discoloration additive could be mixed by any of a variety of techniques (cf. D1, column 6, lines 5 to 9) but it does not disclose the mode of incorporation of the discoloration preventing additive by mixing it with a solution of PPE in an organic solvent.

4.24 Thus, taking document D1 as the closest state of the art, the technical problem underlying the application in suit can be formulated as the provision of a further method of reducing color formation in PPE resins during melt processing, since this problem can be clearly inferred by the person skilled in the art from the experimental evidence in the application as originally filed (cf. also T 13/84, OJ EPO 1986, 253).

4.25 The solution proposed according to Claim 1 of the main request is to mix an hydroxy compound as defined in Claim 1 (e.g. benzoin) with the PPE resin in an organic solvent before melt processing.

5. *Inventive step*

5.1 It remains to be decided whether the proposed solution was obvious in view of the cited prior art.

5.2 As indicated above D1 clearly discloses that the discoloration preventing agent (e.g. benzoin) can be incorporated into the PPE resin by any methods.

5.3 In this connection, the Board however notes that there is no evidence on file of any specific effect of the method of incorporation according to Claim 1 of an hydroxy compound such as benzoin in the PPE resin.

5.4 Consequently, the subject-matter of that claim can only be considered as an obvious alternative to the method disclosed in D1.

5.5 It thus follows that the subject-matter of Claim 1 of the main request does not meet the requirements of Article 56 EPC, and hence, that the main request must be refused.

Second auxiliary request

6. *Wording of the claims*

6.1 Claim 1 of the second auxiliary request differs from Claim 1 of the main request only in that the hydroxy compounds have been restricted to benzopinacol, benzoin, acetoin, allyl alcohol, acetol, 2-butene-1,4-diol, benzyl alcohol or hydrobenzoin, benzhydrol, methylphenylcarbinol, pinacol or 2,3-diphenylbutane-2,3-diol.

6.2 It is thus evident that Claim 1 of the second auxiliary request still encompasses the use of benzoin as decolorizing agent.

7. *Inventive step*

7.1 Since Claim 1 of that request still encompasses the use of benzoin, the same considerations as for the main request inevitably apply.

7.2 Consequently, the subject-matter of Claim 1 of the second auxiliary request must be considered as not meeting the requirements of Article 56 EPC.

7.3 It thus follows that the second auxiliary request must be refused.

8. Since none of the requests of the Appellant can be granted, the appeal must be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:	The Chairman:
M. Kiehl	R. Young