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Bezeichnung der Erfindung: Polyol(allyl carbonate) compositions and polymerizates prepared therefrom
Title of invention:
Titre de l'invention :

Klassifikation / Classification / Classement : C08F 18/24

ENTSCHEIDUNG / DECISION

vom / of / du 7 August 1990

Anmelder / Applicant / Demandeur : PPG Industries, Inc.

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (no) - analogy process - no inventive selection"

Leitsatz / Headnote / Sommaire

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Case Number : T 32/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 7 August 1990

Appellant : PPG Industries, Inc.
One PPG Place
Pittsburgh, Pennsylvania 15272
(US)

Representative : Hann, Michael, Dr.
Patentanwälte Dr. Michael Hann
Dr. H.-G. Sternagel
Sander Aue 30
D-5060 Bergisch Gladbach 2

Decision under appeal : Decision of Examining Division 011 of the European Patent Office dated 20 July 1988 refusing European patent application No. 84 113 443.0 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
J. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 84 113 443.0, filed on 7 November 1984, claiming priority of 9 November 1983 from an earlier application in the United States of America and published on 19 June 1985 under the publication No. 144 782, was rejected by a decision of the Examining Division dated 20 July 1988.

The rejection was based on a set of five claims filed on 7 July 1987, of which Claim 1 read as follows:

"Polymerizable, liquid, substantially gel-free, partially polymerized polyol(allyl carbonate) monomer, characterized by an allyl unsaturation of from 20 to 50 percent."

II. The ground for that decision was that the subject-matter of the application in suit as defined in Claim 1 did not involve an inventive step with regard to the teaching of mainly following documents:

(2) JP-A-57-133 106 (English translation)
(4) FR-A-1 122 427
(5) JP-A-65-9969 as Chemical Abstracts, 16494, Volume 63, 1965, Prepolymers from allyl polycarboxylates by M. Koshimura.

The decision stated that document (2) taught that up to 17% prepolymerisation of diethylene glycol bis(allyl carbonate) could be achieved without gelation. Since the beneficial effect of a solvent to reduce gelation was known from documents (4) and (5) in the case of diallyl phthalate, it was regarded as obvious to use such a solvent in the case of allyl carbonate for the same purpose, all the more so since both prepolymers had similar fields of applications, as a result of their roughly comparable properties.

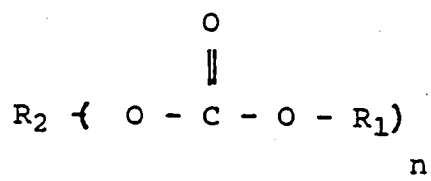
III. A Notice of Appeal was lodged against this decision on 6 September 1988 with payment of the prescribed fee. The arguments presented by the Appellant in the Statement of Grounds of Appeal filed on 21 November 1988 as well as during oral proceedings held on 7 August 1990 can be summarised as follows:

The essence of the teaching of document (2) and, similarly of a further document not mentioned in the decision under appeal, JP-A-51-9188 (document (3)) was that a high polymerisaton index was desired to prevent shrinkage, but gelation of the prepolymer, which occurred at a polymerisation degree of 17%, limited the increase of allylic utilisation. There was thus a prejudice against considering polymerisation degrees higher than 17%. Although document (4) suggested that the limit of gelation could be increased by the use of volatile solvents in the prepolymerisation step, this method required in practice either large amounts of solvent or specific polymerisation conditions, without being very effective. As to document (5), the mere fact that the polymerisation of diallyl phthalate could be controlled by addition of dodecylbenzene could not be an incentive to use specific halogenated solvents for the same purpose in the case of another monomer.

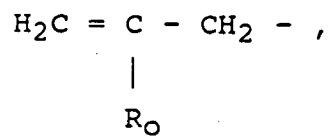
IV. At the beginning of oral proceedings on 7 August 1990, the Appellant informed the Board that the set of 10 claims filed on 21 November 1988 as main request as well as the set of 8 claims filed on 20 July 1990 as first auxiliary request were withdrawn, and that the appeal procedure should be prosecuted on the sole basis of the set of 7 claims filed on 20 July 1990 as second auxiliary request.

Claim 1 thereof reads as follows:

"Polymerizable, liquid, substantially gel-free partially polymerized aliphatic glycol bis(allyl carbonate) monomer composition,
characterized in
that it is obtainable by solution polymerization of said aliphatic glycol bis(allyl carbonate) monomer represented by the formula:



wherein R_1 is an allyl radical represented by the formula



wherein R_o is hydrogen, chlorine, bromine, or a C_1-C_4 alkyl, R_2 is a polyvalent radical derived from an aliphatic or aromatic polyol containing from 2 to 5 hydroxy groups, and n is a whole integer from 2 to 5, in the presence of from 0.1 to 1.5 weight percent based on said monomer of a polymerization initiator and an organic solvent selected from the group consisting of methyl chloride, methylene chloride, ethyl chloride, ethylene dichloride, 1,1,2-trichloro-1,2,2,-trifluoroethane and mixtures of such solvents until from 20 to 50 percent of the unsaturated allylic carbon-carbon linkages are consumed and separation from the solvent resulting in a kinematic viscosity at 25°C from 1000 to 40000 mm^2/s (1000 to 40000 centistokes)."

Whereas Claims 2 and 3 concerned preferred compositions according to Claim 1, Claim 4 was drafted as an independent process claim directed to the preparation of a monomer composition defined as in Claim 1 and Claims 5 to 7 were directed to specific embodiments of the process according to Claim 4.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the second auxiliary request filed on 20 July 1990.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. The wording of the claims does not give rise to objections under Article 123(2) EPC.

Claim 1 differs from original product Claim 1 in that it is drafted as a product-by-process claim incorporating various product and process features. The structural definition of the allyl carbonate monomer is specified in original Claim 2. The use of an organic solvent to carry out the polymerisation reaction, the list of solvents suitable for that purpose and, further, the separation therefrom, are mentioned respectively in original Claims 12, 18 and 15. The presence of a polymerisation initiator and the amount thereof are disclosed in original Claim 16. As to the amount of 20 to 50 percent of unsaturated allylic carbon-carbon linkages consumed and the resulting kinematic viscosity of from 1000 to 40000 centistokes, these features are supported by original Claim 6, the temperature of measurement of the latter parameter being indicated on page 6, line 8 of the application as originally filed.

Further, Claims 2, 3, 5 and 7 correspond respectively to Claims 5, 7, 21 and 20 as originally filed with their numbers and appendancies adjusted. In addition to the definition of the monomer, the list of specific solvents, the amount of unsaturated allylic carbon-carbon linkages consumed, the use of a polymerisation initiator and the amount thereof, already referred to in connection with Claim 1, Claim 4 incorporates the amount of solvent per gram of monomer, the general definition of the organic solvent and the temperature of the polymerisation reaction; these three features are mentioned respectively in original Claims 17, 14 and 16. As to the three categories of free radical initiators according to Claim 6, they correspond to the preferred compounds mentioned on page 13, lines 16 to 18 of the application as originally filed.

3. The application in suit concerns a polymerisable, liquid, substantially gel-free partially polymerised aliphatic glycol bis(allyl carbonate) monomer composition and a process for preparing such composition. A similar subject-matter is disclosed in document (2) which the Board, like the Examining Division, regards as the closest state of the art. According to the teaching thereof, diethylene glycol bis(allyl carbonate) or unsaturated monomers containing the latter as their major component are prepolymerised in the presence of a radical polymerisation initiator (page 4, paragraph 3 to page 5, paragraph 2; Application Examples 1 to 3). Although the use of organic peroxides for this purpose makes it possible to carry out this reaction at relatively low temperatures and thereby produce liquid prepolymers having linear bonds, gelation occurs as soon as the polymerisation degree reaches 17%, i.e. as soon as 17% of the unsaturated allylic carbon-carbon linkages have been consumed. Such low values result in high shrinkage and heat evolution during the course of the polymerisation of the prepolymer as well as in rather slow curing; the

combination of these detrimental effects makes these products not entirely satisfactory for the fabrication of shaped optical elements, especially lens blanks or ophthalmic lenses.

In the light of this prior art, the technical problem underlying the application in suit can be regarded as providing a prepolymer, for which the above effects would be reduced to a level compatible with the specific applications envisaged, as well as determining the operative conditions leading to such a prepolymer.

This problem is solved according to Claim 1 of the application in suit by schematically carrying out the prepolymerisation reaction in an organic solvent selected among specific halogenated compounds until 20 to 50 percent of the unsaturated allylic carbon-carbon linkages are consumed.

In view of the examples in the application in suit, especially Example 6 wherein good light transmission, low haze and yellowing index are achieved additionally to low shrinkage properties, the Board is satisfied that the above-defined technical problem has been effectively solved.

4. After examination of the cited documents the Board has reached the conclusion that the claimed solution is not disclosed in any of them and that the subject-matter of the application in suit is, therefore, novel. Since novelty has not been contested in the decision under appeal, it is not necessary to consider this matter in detail.

5. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the cited documents.

5.1 Documents (5) and (4), which teach that the tendency to gelation can be decreased by the presence of solvents in the polymerisation medium, provide a general solution to the above defined technical problem.

5.1.1 Document (5) is an experimental report concerning the radical polymerisation of allyl polycarboxylates in the presence of alkylbenzene solvents to control the polymerisation reaction. According to a first experiment, a mixture of 1 kg diallyl phthalate, 300 g dodecylbenzene and 2.6 g t-BuOOH is heated at 155°C for two hours to give 390 g prepolymer. By contrast, when the same reaction is carried out without dodecylbenzene, gelation occurs as the result of cross-linking.

From a quantitative point of view, the use of such solvent and monomer in the weight ratio of 0.3 to 1, which is partly below the range of 0.5 to 5 millilitres of solvent per gram of monomer as required in Claim 4 of the application in suit, makes it possible to obtain a rate of conversion of 39%, which represents an improvement of 56% with regard to the polymerisation index of 25% obtained in absence of solvent, as will appear hereinafter.

5.1.2 The introductory section of document (4) first recalls that total conversion of diallyl phthalate monomer to polymer cannot be achieved in one single operation, since gelation occurs as soon as 25% of the monomer has been polymerised (page 1, column 1, paragraph 2). Although quantitative conclusions cannot be drawn from the survey of various attempts made in the prior art to overcome this problem (page 1, column 1, paragraph 3 to column 2, paragraph 1), the beneficial influence of solvents on the polymerisation of diallyl phthalate cannot be denied.

It is first reported that easily removed volatile solvents, even used in large amounts, are not very effective in increasing the proportions of low molecular weight polymer prepared before gelation sets in. By contrast, the use of comonomers which do not homopolymerise easily and act thus like a so-called reactive diluent, gives a relatively high rate of conversion, the final product not being substantially different from diallyl phthalate homopolymer. Further, according to the general teaching of this document (page 1, column 2, paragraph 4 to page 2, column 1, paragraph 2), 70 to 90% of diallyl phthalate can be converted into a low molecular weight copolymer before gelation occurs by effecting a copolymerisation thereof with vinylidene chloride in an inert solvent and by adding part of vinylidene chloride at the beginning of the reaction and the remainder by stages as the polymerisation progresses. Although, admittedly, vinylidene chloride contributes to a certain extent to reduce the tendency to gelation (page 2, column 1, paragraph 7), the high polymerisation index mentioned above has to be regarded in the light of the general statement concerning copolymers, according to which, if diallyl phthalate is copolymerised with a rapidly homopolymerisable monomer, cross-linked polymers are formed before much of the diallyl phthalate has reacted (page 1, column 2, paragraph 2).

In the Board's view, these various teachings clearly show that the polymerisation index may be positively influenced by the use of non-volatile solvents.

5.1.3 In the Board's view, the prior art teaching, in particular document (5), would be a clear incentive for the skilled person to increase the prepolymerisation rate of diethylene glycol bis(allyl carbonate) in a similar manner. The fact that diallyl phthalate is less susceptible to gelation than diethylene glycol bis(allyl carbonate) does not alter this

conclusion, since the effect to be achieved in both cases is a relative improvement over a known figure. For this reason, the solution claimed by the Appellant is regarded as a mere analogy process based on the same operative feature to achieve a similar technical effect.

5.2 The argument that the claimed subject-matter in fact involves the selection of specific halogenated solvents cannot be accepted by the Board. Firstly, the use of inert solvents such as carbon tetrachloride, 1,2-dichlorethane and other common halogenated solvents is explicitly recommended in document (4) (page 2, column 1, paragraph 2); in the Board's view, this clearly points to the type of compounds enlisted in Claim 1 of the application in suit. Secondly, no comparative data has been provided by the Appellant showing unexpected results in terms of polymerisation index as the result of the use of the halogenated solvents specified in Claim 1.

5.3 The argument put forward by the Appellant (Statement of Grounds of Appeal, page 4, paragraph 1), according to which there was a prejudice against considering higher polymerisation degrees for the reason that gelling of the prepolymer occurs above 17% allylic utilisation, cannot be accepted either.

As emphasised by the Appellant, the teaching of document (3) is very similar to that of document (2) in that it confirms that the polymerisation index must be set at less than 17% in order to obtain a fluid prepolymer. The process disclosed in document (3) aims at the preparation of liquid prepolymers by heat polymerising diethylene glycol bis(allyl carbonate) at a temperature within the range of 65 to 100°C and in the presence of 0.05 to 0.17 mol% of a peroxide as a radical initiator (page 3, paragraphs 2 and 3; page 4, paragraphs 2 and 3). The amount of radical

initiator is critical in order to obtain a liquid prepolymer of uniform quality and polymerisation index lower than 17%, allowing long-term storage for even six months at room temperature (page 5, paragraph 1 and 2; page 7, paragraph 2).

The processes described in documents (2) and (3) differ, thus, by the amount of radical initiator - 0.3 to 2.3 weight percent respectively 0.05 to 0.17 mol percent - and by the temperature range - 5 to 35 respectively 65 to 100°C - at which the polymerisation should be conducted. In reality, both processes deal with a polymerisation reaction carried out in the liquid monomer as polymerisation medium, wherein the amount of radical initiator and the temperature are chosen and adjusted for kinetic purposes only, not for modification of the polymerisation index which is obviously unaffected by these parameters. These prior art processes can thus be regarded as particular embodiments of a single technical teaching, from which no prejudice can be inferred.

5.4 In conclusion, for the reasons given above, the subject-matter of Claim 1 does not involve an inventive step.

6. Claim 1 not being allowable, the same applies to Claims 2 and 3 which are directed to preferred polymerisable composition according to the main claim and thus fall with it, as well as to Claims 4 to 7 which concern combinations of process features which are not patentable for the same reasons as in Claim 1.

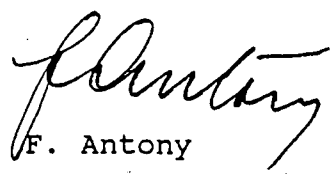
Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

M. Beer



F. Antony