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D E C I S I O N
of 27 May 1999

Case Number: T 0822/96 - 3.3.3

Application Number: 87115279.9

Publication Number: 0264885

IPC: C08G 64/02

Language of the proceedings: EN

Title of invention:
Washing method for a solution dissolving a polycarbonate resin in an organic solvent

Patentee:
Idemitus Petrochemical Co. Ltd.

Opponent:
Bayer AG, Leverkusen Konzernverwaltung RP Patente Konzern
General Electric Company

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - ex post facto analysis"

Decisions cited:
T 0002/83

Catchword:
-

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Case Number: T 0822/96 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 27 May 1999

Other party: (Opponent)
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 26 August 1996 concerning maintenance of European patent No. 0 264 885 in amended form.

Composition of the Board:

Chairman: C. Gérardin
Members: A. Däweritz
A. Lindqvist

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 264 885 in respect of European patent application No. 87 115 279.9 filed on 19 October 1987 and claiming priority of 22 October 1986 of an earlier application in Japan (251277/86), was announced on 3 March 1993 (Bulletin 93/09) on the basis of 9 claims.

Claim 1 as granted read as follows:

"A method for washing an organic solution of a crude polycarbonate resin in an organic solvent, which comprises preparing a mixture of said organic solution with an aqueous washing liquid, so that the aqueous phase is in an amount of 5 to 30 vol.% based on the total volume of said solution and said liquid, stirring said mixture at a stirring power per unit flow rate of 0.1 kw/m^3/hr or more, to form a water-in-oil type emulsion, and subjecting said dispersion to centrifugal separation to separate the purified organic solution of the polycarbonate resin from the aqueous washing liquid, thereby removing impurities from said crude polycarbonate resin, wherein the method is conducted without causing a phase inversion of the water-in-oil-type emulsion."

Claims 2 to 9 concerned preferred embodiments of the method according to Claim 1.

II. On 19 November 1993 and 25 November 1993, Notices of Opposition were filed by two Opponents in which revocation of the patent in its entirety was requested on the grounds of lack of novelty within the meaning of

Article 54(1) and (2) EPC, lack of inventive step within the meaning of Article 56 EPC and extension of the subject-matter beyond the content of the application as originally filed within the meaning of Article 123(2) EPC.

The objection under Article 100(c) EPC was withdrawn during the opposition proceedings.
The objections were supported essentially by the following documents:

E1: CA-A-0 747 994,

E2: English translation of JP-A-51-112897 (Application Sho 50-38955), and

E3: US-A-4 323 519.

III. By interlocutory decision announced orally on 9 July 1996 and issued in writing on 26 August 1996, the Opposition Division held that the grounds for opposition did not prejudice the maintenance of the patent in amended form, based on Auxiliary request I submitted during oral proceedings on 9 July 1996. The amendments in the claims as maintained consisted in a more specific definition of the claimed method according to Claim 1 and in the deletion of dependent Claims 3 and 9 as granted. The other claims were maintained unamended and renumbered when appropriate. Claim 1 reads as follows:

"A method for washing an organic solution of a crude polycarbonate resin in an organic solvent obtained by the phosgene process, optionally including a

centrifugal separation step, and containing alkali halides, caustic alkali, alkali carbonates, unreacted dioxy compounds and tertiary amines as impurities, which consists of preparing a mixture of said organic solution, which solution is produced in the polycarbonate resin-producing process, with an aqueous washing liquid, so that the aqueous phase is in an amount of 5 to 30 vol.% based on the total volume of said solution and said liquid, stirring said mixture by means of a line stirring mixer at a stirring power per unit flow rate of 0.1 kw/m^3/hr or more, to form a water-in-oil type emulsion, and subjecting said dispersion to centrifugal separation to separate the purified organic solution of the polycarbonate resin from the aqueous washing liquid, thereby removing impurities from said crude polycarbonate resin, wherein the method is conducted without causing a phase inversion of the water-in-oil type emulsion."

(i) In its decision, the Opposition Division first acknowledged that the claimed subject-matter was novel over E3, because the document did not disclose a washing method of crude polycarbonate resin consisting of the process steps as claimed.

(ii) Starting from E2, which - according to the minutes of the oral proceedings (page 5, paragraph 1 of point 5.2) - was unanimously regarded as representing the closest prior art, it was not considered obvious to modify the method claimed in E2 in accordance with the requirements of the patent in suit in order to improve extraction efficiency compared to the known purification method. Consequently, an

inventive step was also acknowledged.

IV. On 31 August 1996, a Notice of Appeal was lodged by Opponent II (Appellant) against this decision with simultaneous payment of the prescribed fee.

In the Statement of Grounds of Appeal filed on 19 December 1996, the Appellant again denied that the subject-matter claimed involved an inventive step. To that end it relied on E1 and E3. In substance, it argued essentially as follows:

(i) Example IX of E1 described a process for the preparation of polycarbonate, wherein the polymer solution was separated by settling, washed with water and then stabilised with hydrochloric acid. After removal of the aqueous layer, the polymer solution was mixed with water in a vibrating blade ultrasonic mixer instead of using a stirred mixer. The so obtained water-in-oil emulsion was broken by coalescence filtration instead of centrifugation. These two distinguishing features did not justify the grant of a patent. The use of in line stirring mixers was suggested on page 6, line 19 of E1, and to replace coalescent filtration by centrifugation was obvious to a person in the art.

(ii) It was well known from E3 to improve the quality of polycarbonate by removing impurities. Reference Example 1 essentially differed only in two features: the use of an in-line stirrer at a power per unit flow rate of 0.1 kw/m^3/h or more and centrifugal separation.

Both of these features did not impart inventive character to the claimed method because there were no data supporting the inventive character of the minimum power and centrifugation could not be the basis for inventive step either.

(iii) It belonged to the normal routine of a person skilled in the art to seek optimal conditions for mixing the polycarbonate solution and water for purification purposes. The use of a compact stirring device with a short residence time which could be operated at high speed was obvious in view of E1.

V. In its Counterstatement of Appeal, the Respondent (Proprietor) supported the findings of the decision under appeal substantially as follows:

(i) Example IX of E1 differed from the claimed method not only by the two features mentioned above, but also by the fact that it did not define a minimum stirring power and additionally required the use of an aqueous stabiliser. Moreover, the known process also required a separation step by gravitation, because coalescing filtration was not a separation step.

Due to the additional mandatory steps of coalescing filtration and stabilisation in E1 this document represented a more remote prior art. Example IX was merely comparative, because it did not illustrate the general method according to E1, as it did not include a washing step under high shear. The poor results obtained

would support this assumption.

(ii) A person skilled in the art would not try to improve the results obtained on the basis of a comparative example, but rather follow the basic teaching of the document.

(iii) This would also be true for Reference 1 in E3 which was also comparative. Moreover, the method in E3 differed completely from the claimed one in that the water-in-oil emulsion was subjected to a phase inversion.

(iv) The Appellant's conclusions were based on a mere hindsight analysis of the prior art.

(v) Figure 4 annexed to the Counterstatement demonstrated the criticality of the minimum stirring power used in the claimed method irrespective of the further conditions required.

(vi) The method as claimed was not a liquid-liquid extraction because the impurities from the phosgene process were essentially dissolved in water droplets which were dispersed in the polycarbonate. Therefore the claimed method comprised dilution and separation steps rather than extraction or rinsing.

VI. Oral proceedings were held on 27 May 1999.

(i) The Appellant emphasised its previous submissions essentially as follows:

1. The comparative examples in a prior art document had to be regarded as a positive disclosure. Therefore, a comparative example as disclosed e.g. in E3 was also relevant, and it was an adequate starting point for the consideration of obviousness and inventive step.

2. E3 was a highly relevant document which summarised the prior art. Thus it described the purification of polycarbonate resin. The invention aimed at an optimisation of this process by selecting a specific mixing device including a specific power input and a specific separation technique.

3. E1 clearly invited the skilled person to use the same mixing device as required in the contested Claim 1.

4. Any improvement in purity could only be regarded as a so-called bonus effect.

(ii) The Respondent contested the arguments presented by the Appellant. In particular, it expressed the opinion that the Appellant had argued on a hindsight basis by selecting specific features from a cited document out of their context and combining them with other features from another known process. Such patchwork was clearly based on the knowledge of the teaching of the patent in suit. But even such an argumentation did not lead to the improvement of purification efficiency, i.e. improved purity in a shorter time.

VII. Opponent I, which had not lodged an appeal nor submitted any arguments in writing, was duly summoned as a party as of right to the oral proceedings. By letter of 19 May 1999 it informed the EPO that it would attend the hearing.

Although expressly invited by the Board during the hearing to present arguments to the various aspects of the issue of inventive step, it declared it would refrain from doing so and expect the decision.

VIII. The Appellant requested that the interlocutory decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. In the Statement of Grounds of Appeal, inventive step has been the only issue raised by the Appellant. The Board concurs with the findings of the Opposition Division in the decision under appeal that novelty is given vis-à-vis the cited prior art and that the requirements of Article 123(2) and (3) EPC are met. There is thus no need to consider these matters in further detail.

3. The patent in suit concerns a washing method for a solution dissolving a polycarbonate resin in an organic solvent.

3.1 Such a method is known from E2 which the Board, like the parties and the Opposition Division, regards as representing the closest state of the art.

3.1.1 This citation describes "a method of purification for a polycarbonate solution using an orifice-mixer characterized by comprising steps of contacting a solution of crude polycarbonate containing an impurity matter in an organic solvent with a washing liquid using an orifice-mixer while adjusting the pressure drop in said orifice-mixer to a value within the range of 2 to 12 kg/cm^2 and keeping the proportion of said washing liquid to the whole fluid at 5 to 45% by volume." (Claim 1). In its example, E2 additionally describes the separation of the aqueous phase from the polymerisation reaction solution by means of a centrifuge after dilution of the solution with further organic solvent (page 4, last line to page 5, line 2).

3.1.2 This known method aims at an improved, easy and effective method for the purification of polycarbonate resin obtained in a phosgene process, thereby avoiding, first, a complicated sequence of operations, and, secondly, the problem of antinomy brought about by the difficulty of perfect separation of the resin solution and the washing liquid at the end of the purification caused by the vigorous high speed mixing in order to achieve efficient washing (introduction of the description on page 1 and page 2, paragraph 1; Reference Example 2).

3.1.3 The examples and comparative (reference) examples (pages 5 and 6) based on repeated washing steps including (a) mixing by means of an orifice-mixer under

controlled pressure conditions and controlled ratios of washing liquid and resin solution and (b) separation by still standing, show that, on the one hand, the purity of polycarbonate resin expressed in terms of light transmittance greatly depends on the mixing conditions, and, on the other hand, mixing by intensive mechanical stirring causes difficulties as regards the separation of the resin solution and the washing liquid.

3.2 In the light of these shortcomings and in line with the introductory statements in the patent specification (page 2, lines 5 to 9, 15 to 23 and 35 to 37), the technical problem underlying the patent in suit may thus be seen as the definition of a process not only more efficient in removing impurities from polycarbonate resin solutions as directly obtained in the phosgene process, which optionally includes a centrifugal separation step prior to the purification, but also more simple so as to rapidly purify solutions with even high concentrations of polymer causing high viscosities.

3.3 According to the patent in suit, this problem is solved by (i) using a line stirring mixer at or above a specific minimum stirring power to prepare a water-in-oil type dispersion containing a limited amount of an aqueous washing liquid dispersed in an organic solution of polycarbonate resin, and (ii) subjecting that water-in-oil dispersion to centrifugal separation, as specified in Claim 1.

3.4 In both tables on pages 5 and 7 of the patent in suit, as well as in the further experiments submitted during examination (received on 29 October 1991), it has been

demonstrated that a significantly improved reduction of the contents of inorganic and organic impurities, *viz.* of sodium and chloride ions and bisphenol A, is achieved by operating in accordance with Claim 1 in comparison to prior art methods or modified prior art methods (see in particular Comparative Examples 5 and 6 which meet the requirements as defined in the claim of E2).

In accordance with the method as claimed, the flow rate of the 12.1 wt.% resin solution in the examples is kept at 38 l/h, compared to 35 and 27 l/h, respectively, in the experiments of E2. The assessment of the polymer concentration for the experiments of E2 results in a concentration in the resin solution used in the purification which is not higher than that in the examples in the patent in suit.

Consequently, the two aspects of the above defined technical problem are effectively solved by the method as defined in Claim 1 of the patent in suit.

4. It remains to be decided whether this solution was obvious to a person skilled in the art having regard to the state of the art relied upon by the Appellant.

4.1 It is evident from the above considerations that document E2 by itself cannot render the claimed invention obvious. Not only it does not provide any incentive to replace the orifice mixer by a line stirred mixer, but it even teaches away from using vigorous stirring said to be generally unsatisfactory (see the top of page 2 and Reference Example 2). Despite the fact that a centrifuge is used to remove

the aqueous phase of the reaction mixture (page 5, line 2), the document does not suggest at all to use such a device in the actual separation step of the purification.

4.2 Document E1 relates to a process for the removal of impurities from highly viscous polymer masses, in particular, of organic and inorganic residues from highly viscous polymeric solutions and melts (page 2, paragraph 1). According to page 3, paragraph 1, these impurities are present directly in the polymer mass or dissolved in droplets of an aqueous phase which are in turn suspended or emulsified in the viscous polymer mass.

4.2.1 In a list of polymers that can be treated accordingly (see the paragraph bridging pages 4 and 5), polycarbonate resin is also mentioned. The polymer solutions or melts preferably have viscosities of about 200 to 7000 cP (mPas).

4.2.2 According to page 4, lines 4 to 18 and Claim 1 of the citation, a polymer mass containing impurities dissolved in droplets of an aqueous phase, which are in turn dispersed in the viscous polymer mass, is at first passed through a coalescing filter bed, then the coalesced aqueous phase is removed therefrom. Thereafter the polymer mass is admixed with an aqueous stabiliser (page 8, line 3 *et seq.*) and the stabilised polymer mass is washed with water under high shear while maintaining the polymer mass as the continuous phase. Finally, the purified polymer mass is continuously removed. According to page 5, lines 14 to 17, the coalesced aqueous phase can be rapidly

separated from the polymer mass by settling under gravitation.

It has not been a matter of dispute between the parties that the additional stabilisation step was a difference between the methods of E1 and the patent in suit.

4.2.3 If the impurities are dissolved directly in the polymer or the organic solvent phase, a washing step has to be carried out prior to coalescing the dispersion, wherein high shear is applied by a suitable device such as a line-type-impeller agitated mixer or an ultrasonic vibrating blade mixer. In this washing step, the amount of aqueous washing liquid has to be limited so as to maintain the organic polymer solution as the continuous phase. For any given mixer design, at a fixed shear rate, the energy input is determined by the effective viscosity (page 6, lines 12 to 25).

4.2.4 For further purification and removal of residual impurities, the polymer is thereafter subjected to high shear washing whereby the polymer is again maintained as the continuous phase. For this step a rotating disc contactor, Rushton or Scheibel columns are recommended, while most (other) countercurrent washing units have been found to be unsatisfactory (page 9, paragraph 2). When using the rotating disc contactor in this step, the polymer mass can be maintained as the continuous phase at a wash water/polymer mass ratio of 2 or 3 or more (page 10, lines 8 to 11); when using a countercurrent column, the said ratio in the feed streams can be less or more than about 1:1, the higher ratio being preferred (page 10, lines 21 to 23). The two phases are separated in settling zones (page 10,

line 27).

4.2.5 Thereafter, one or more high shear washing steps in autoclaves or using a high shear line mixer in combination with a coalescing device can additionally be carried out (page 11, paragraph 2).

4.2.6 Only Examples VIII and IX deal specifically with the purification of polycarbonate resin. In Example VIII the aqueous phase originating from the polymerisation is first decanted after settling, the polymer solution is then pumped through a coalescing bed and the effluent is allowed to settle by gravity, and finally the aqueous layer is removed ("essentially 10% by weight of the total effluent"). In Example IX, after the water present in the polymerisation mixture has been decanted and the polymer has been washed with water and thereafter stabilised, the polymer solution is additionally treated with water in a vibrating blade ultrasonic mixer. The resulting emulsion is then broken by coalescing filtration as in Example VIII. 87.7% of the impurities are removed in this way.

4.2.7 This degree of removal of the impurities is clearly below the figures (at least 96%) obtained in the examples of the patent in suit; it rather corresponds to those in the comparative examples reported in Tables 1 and 2 of the patent in suit.

4.2.8 The document by itself thus does not provide any incentive to dispense with the pretreatment, which includes a coalescing filtration and separation, and with the stabilisation step, to select a line stirring mixer and to use a centrifugal separation as the final

purification step in order to improve the purification efficiency, including an improved removal of impurities.

4.3 E3 relates to a further method to remove impurities from a polycarbonate solution in methylene chloride as obtained from the phosgene process, the impurities being incorporated with a small amount of water in the form of an emulsion (column 1, lines 15 to 22). According to Claim 1, the known method comprises the steps of (1) mixing the crude polycarbonate solution with a certain amount of aqueous washing liquid to form a water-in-oil dispersed phase, (2) adding thereto a further amount of aqueous washing liquid to cause a phase inversion of the dispersion obtained in step (1), (3) then separating the dispersed organic phase, i.e. the purified polycarbonate solution, from the continuous aqueous phase, i.e. the diluted aqueous washing liquid containing the impurities.

4.3.1 In column 1, lines 41 to 68, reference is made to problems which occur when either aqueous washing liquid is added in small amounts to form a water-in-oil emulsion, in particular under alkaline conditions and at high solids contents and viscosities of the organic solution, or when it is added in large amounts to form a oil-in-water dispersion. In the first case, the time for separating the continuous organic phase from the dispersed aqueous phase is said to be too long to be

suitable for an industrial operation, in the second case, the elution of impurities from the continuous organic phase is said to be so slow that it is difficult to attain a satisfactory washing effect (see also column 3, lines 41 to 45).

4.3.2 According to the first two paragraphs in column 2, the method of E3 does not only allow to effectively remove impurities from polycarbonate solutions of high concentrations and viscosities which may even be alkaline "by the combination of certain steps", but "a satisfactory result can be attained even through the use of a simple apparatus such as a combination of stirring vessel and a settler separator".

The known method requires a thorough mixing of the two organic and aqueous solutions in both steps (1) and (2) by using e.g. a stirring vessel, a multi-plated tower or other vessels or towers having a stirrer (column 3, lines 46 to 50 and 64 to 68).

Due to the phase inversion in step (2), the phase separability of the mixture is improved, whereby the phases can be easily separated by a conventional method using a settler separator, tower type separator or a centrifugal separator (column 4, lines 1 to 7).

In the drawings as well as in the examples and references, stirring vessels (equipped e.g. with a turbine blade mixer) and settlers are shown and used, respectively.

4.3.3 In the comparative examples ("references") of the document, the unsatisfactory results of embodiments

without phase inversion are demonstrated (see e.g. Table 1, References 1 and 2). These poor results in the prior art have been confirmed by Examples 7 and 8 as submitted on 20 October 1991 during the examination proceedings (Document (1) referred to in these examples corresponds to E3). Moreover, there is no hint that by modifying the method, let alone by departing from the explicit teaching of E3, even further improved results would be obtained, as shown by the above additional examples.

4.3.4 It is evident that the gist of the method of E3 is the combination of a washing step in water-in-oil dispersion and its phase inversion before the separation of the organic and aqueous phases rather than the selection of particular mixing and separation devices. It is clear from the teaching in E3 that it had to be expected that dispensing with the phase inversion would result in a very poor removal of impurities. In any case, E3 on its own does not provide any incentive to dispense with the phase inversion and to select a line stirring mixer at a specific power input to improve the purification efficiency.

4.4 Even a combination of the documents relied upon by the Appellant would not render obvious the claimed subject-matter.

4.4.1 From the above discussion it is evident that the known processes are each characterised by a combination of specific features and that in each case the desired level of purification can only be achieved when all the required conditions are fulfilled. A given feature,

whether it is a device used in a particular step of the purification method or the physical state of the system, is thus only important within the framework of a specific technical context. In the absence of any indication, a skilled person would thus have no reason to depart from the corresponding teaching and, consequently, would not consider an isolated feature from one process to combine it with another method.

4.4.2 It is not disputed that the solution proposed in the patent in suit is based on well-known technical features and that a combination of features appropriately selected from the various disclosures could result in a process within the terms of the method as claimed. As pointed out by the Respondent, however, the question is not whether a skilled person could have considered such a combination at the priority date of the patent in suit, but whether this skilled person faced with the above defined technical problem would have done so with a reasonable expectation of success (cf. T 2/83, OJ EPO 1984, 265).

4.4.3 In the Board's view, the Appellant clearly failed to demonstrate that there was an incentive to modify the process known from E2 in accordance with the requirements of Claim 1 of the patent in suit in order to simultaneously improve the efficiency of purification and simplify the method. In the absence of such a link between the technical problem and the features of its solution, the Appellant's arguments can only be made with the benefit of the contribution of the patent specification and amount thus to hindsight analysis.

4.5 It follows that the method as defined in Claim 1 would not be obvious to a person skilled in the art having regard to the state of the art relied upon by the Appellant, whether considered in isolation or in combination and, therefore, involves an inventive step.

5. Claims 2 to 7, which relate to preferred embodiments of the method according to Claim 1, are supported by the patentability of the main claim and thus also allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin