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**Datasheet for the decision  
of 18 June 2013**

**Case Number:** T 0404/10 - 3.3.10

**Application Number:** 00906676.2

**Publication Number:** 1174406

**IPC:** C07C27/00, C07C27/28

**Language of the proceedings:** EN

**Title of invention:**

METHOD FOR CONTINUOUSLY PRODUCING A DIALKYL CARBONATE AND A  
DIOL

**Patent Proprietor:**

Asahi Kasei Chemicals Corporation

**Opponent:**

Bayer MaterialScience AG

**Headword:**

**Relevant legal provisions:**

EPC Art. 100(a), 54(2), 56

**Keyword:**

Novelty (yes); inventive step (yes)

**Decisions cited:**

**Catchword:**



**Beschwerdekammern  
Boards of Appeal  
Chambres de recours**

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Case Number: T 0404/10 - 3.3.10

**D E C I S I O N  
of Technical Board of Appeal 3.3.10  
of 18 June 2013**

**Appellant:** Bayer MaterialScience AG  
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Patents and Licensing  
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**Representative:** BIP Patents  
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**Respondent:** Asahi Kasei Chemicals Corporation  
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**Representative:** Strehl Schübel-Hopf & Partner  
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**Decision under appeal:** **Decision of the Opposition Division of the  
European Patent Office posted on 11 December  
2009 rejecting the opposition filed against  
European patent No. 1174406 pursuant to Article  
101(2) EPC.**

**Composition of the Board:**

**Chairman:** P. Gryczka  
**Members:** R. Pérez Carlón  
C. Schmidt

## Summary of Facts and Submissions

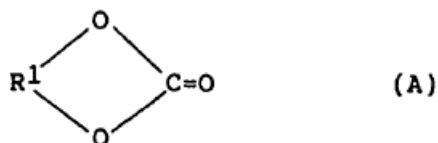
- I. The appeal lies from the decision of the opposition division to reject the opposition against European patent EP 1 174 406.
- II. An opposition had been filed requesting revocation of the patent on the grounds of lack of novelty and inventive step (Article 100(a) EPC) and that the invention was not disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Article 100(b) EPC).
- III. *Inter alia*, the following documents were submitted during the opposition proceedings.
- D1: EP 0 569 812 A1  
D2: EP 0 530 615 A1  
D3: DE 27 40 243 A1
- IV. The opposition division decided that the contested patent contained sufficient information about the suitable reaction temperatures so that the skilled person could find suitable conditions without undue burden, and the amount of cyclic carbonate content defined in claim 1 could be determined by known methods, routinely used in the art. Document D1 failed to explicitly disclose whether the high boiling stream (9) of D1 contained unreacted ethyl carbonate, and the opponent had not shown beyond reasonable doubt that it was the case, so the subject-matter of the patent as granted was novel. Document D1 was the closest prior art, and the problem underlying the claimed invention was the provision of an alternative method for continuously producing a dialkyl carbonate. The solution was to allow unreacted cyclic carbonate to be

present in the high boiling point mixture withdrawn from a lower portion of said multi-stage distillation column and to etherify said cyclic carbonate and a diol, and this solution was not obvious neither in the light of D1 alone nor in combination with document D2, which mentioned such an etherification as a reaction to be avoided. The subject-matter claimed was, therefore, inventive.

V. Claim 1 of the patent as granted reads as follows:

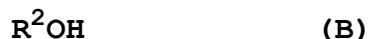
*"A method for continuously producing a dialkyl carbonate and a diol from a cyclic carbonate and an aliphatic monohydric alcohol, comprising:*

*(1) continuously feeding a cyclic carbonate represented by the following formula (A):*



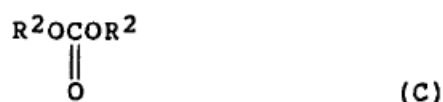
*wherein  $R^1$  is a divalent group which is represented by the formula  $-(CH_2)_m-$ , wherein  $m$  is an integer of from 2 to 6, and which is unsubstituted or substituted with at least one substituent selected from the group consisting of a  $C_1$ - $C_{10}$  alkyl group and a  $C_6$ - $C_{10}$  aryl group,*

*and an aliphatic monohydric alcohol represented by the following formula (B):*



*wherein  $R^2$  is a monovalent aliphatic  $C_1$ - $C_{12}$  hydrocarbon group which is unsubstituted or substituted with at least one substituent selected from the group consisting of a  $C_1$ - $C_{10}$  alkyl group and a  $C_6$ - $C_{10}$  aryl group, to a continuous multi-stage distillation column,*

and continuously effecting a transesterification between said cyclic carbonate and said aliphatic monohydric alcohol in the presence of a transesterification catalyst in said multi-stage distillation column, thereby continuously producing a dialkyl carbonate represented by the following formula (C):

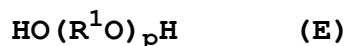


wherein  $\text{R}^2$  is as defined for formula (B) above, and a diol represented by the following formula (D):



wherein  $\text{R}^1$  is as defined for formula (A) above, while continuously withdrawing a low boiling point mixture in a gaseous form containing the produced dialkyl carbonate (C) and the unreacted aliphatic monohydric alcohol (B) from an upper portion of said multi-stage distillation column and continuously withdrawing a high boiling point mixture in a liquid form containing the produced diol (D) and the unreacted cyclic carbonate (A) from a lower portion of said multi-stage distillation column, and

(2) continuously feeding said high boiling point mixture withdrawn from the lower portion of said multi-stage distillation column in step (1) to a continuous etherification reactor, to thereby effect a continuous etherification reaction between said unreacted cyclic carbonate (A) and a part of said produced diol (D) and produce a chain ether represented by the following formula (E):



*wherein  $R^1$  is as defined for formula (A) above and  $p$  is an integer of from 2 to 4, and carbon dioxide, while continuously withdrawing the resultant etherification reaction mixture containing the remainder of the diol (D) produced in step (1) and the produced chain ether (E) from said continuous etherification reactor, said etherification reaction mixture having a cyclic carbonate content of from 0 to  $10^{-2}$  in terms of the weight ratio of the cyclic carbonate (A) to the diol (D)."*

- VI. The appellant (opponent) argued that the subject-matter of claim 1 as granted was not novel over D1. Although D1 did not explicitly disclose alkylene carbonate as a component of bottoms feed (9) of figure 5, this compound must have been part of this feed in the light of the experimental evidence submitted and taking into account that the reactions involved were equilibria. D1 did not explicitly mention an etherification reaction of bottoms (9) as required by step (2) of claim 1, but said reaction took place unavoidably in apparatuses (III), (IV) and (VII) and in the lines of figure 5. D1 disclosed, therefore, all the features of claim 1.

With respect to inventive step, document D1 was the closest prior art. If an additional etherification step of the bottoms of the transesterification step was the distinguishing feature, the problem underlying the claimed invention was providing an alternative process for obtaining a dialkyl carbonate and a diol. The etherification of diols and cyclic carbonates was well known in the art as shown for example in document D2, and including such a step over the unreacted cyclic carbonate of a transesterification procedure was obvious for the skilled person in order to achieve an

alternative.

- VII. The respondent (proprietor) argued that D1 failed to disclose the presence of alkylene carbonate in the bottoms (9) of reactor (I) and an additional etherification step of the bottoms of the transesterification reactor, with the consequence that the subject-matter of the patent as granted was novel.

Document D1 was the closest prior art. Considering the etherification step (2) of claim 1 as the sole distinguishing feature, the problem underlying the claimed invention was providing a method for continuously producing a dialkyl carbonate and a diol which enabled a high purity diol to be obtained without a need for a complicated distillation-separation step, as defined in paragraph [13] of the patent in suit. Said problem was credibly solved in the light of the results of example 1 and comparative example 1 by introducing an additional etherification step of the bottoms of the transesterification reactor, and this solution was not obvious in the light of the prior art.

- VIII. Oral proceedings before the board took place on 18 June 2013.

- IX. The final requests of the parties were as follows:

- The appellant requested that the decision under appeal be set aside and that the patent be revoked.
- The respondent requested that the appeal is dismissed and to maintain the patent as granted or, subsidiarily, within the scope of the auxiliary requests 1, 2 or 3 filed with a letter

dated 8 November 2010.

- X. At the end of the oral proceedings, the decision was announced.

### **Reasons for the Decision**

1. The appeal is admissible.

Sufficiency of disclosure, Article 100(b) EPC:

2. The opposition division decided that the patent in suit contained sufficient information so that the invention could be carried out by the person skilled in the art. Sufficiency of disclosure has not been contested in appeal and the board sees no reason to take a different view than the opposition division.

Novelty, Article 100(a) and 54(2) EPC:

3. Document D1 discloses (example and figure 5) a process for producing dimethyl carbonate (C) and ethylene glycol (D) by reacting ethylene carbonate (A) with methanol (B) in a multi-stage distillation column (I), from which a mixture of methanol and dimethyl carbonate is removed as a top feed (7), and a bottoms (9) containing ethylene glycol is obtained. It has not been disputed that the relative amount of ethylene carbonate with respect to the ethylene glycol in line (9) is already within the limits defined by claim 1 (0 to  $10^{-2}$  weight ratio) and hence will also remain within said limits after an eventual etherification step.

Document D1 fails to explicitly disclose the following features:



- whether feed (9), withdrawn from the lower portion of the continuous multi-stage distillation column (I) of D1, contains unreacted cyclic carbonate (A), and
- the etherification of bottoms feed (9), as required by step (2) of claim 1.

In order to decide whether the subject-matter of claim 1 of the patent in suit is novel, it needs to be examined whether these features are the unavoidable result of the process disclosed in D1.

*Presence of unreacted cyclic carbonate (A) in the bottoms (9) of the multi-stage distillation column (I):*

4. Line (9) exiting reactor (I) in figure 5 of D1 corresponds to the bottoms of the multi-stage distillation column defined in claim 1.

Document D1 discloses (page 8, lines 1 and 2) that feed (9) contains ethyleneglycol, small amounts of lights (methanol and dimethylcarbonate), heavies such as diethyleneglycol, and the catalyst. Ethylene carbonate is, however, not mentioned.

5. The respondent relied on replications of the example of D1 provided during the appeal proceedings for proving that by using a sufficient number of stages, "the amount of ethylene carbonate (if any) was lower than the detection limit". The respondent considered that the detection limit of 1 ppm, disclosed on page 20, line 25 of the description, should be read within claim 1. As the skilled person could choose conditions in which no ethylene carbonate was present in the bottoms of column (I), and document D1 only disclosed the top and bottom temperatures of the column but not the

pressure or the number of stages, D1 failed to disclose the presence of cyclic carbonate in the bottoms of the transesterification reactor.

The description of the patent in suit acknowledges that "even when it is possible to achieve a substantially 100% conversion of a cyclic carbonate, it is impossible to prevent a trace amount of the cyclic carbonate from remaining unreacted in a produced diol" (see paragraph [21]). This sentence is in line with the general technical knowledge that a total conversion cannot be achieved in an equilibrium reaction. The detection limit relied upon by the respondent is mentioned in the examples of the patent in suit, but claim 1 is not limited by any minimum amount of cyclic carbonate. Since the patent in suit acknowledged that "unavoidable trace amounts" will always be present in the bottoms of a transesterification reactor and said trace amounts fulfill the conditions set in claim 1, the process of D1 discloses, as its unavoidable result, the presence of cyclic carbonate in line 9 of figure 5.

6. The respondent had further argued that in reactor (I) of D1, in addition to a transesterification, an etherification had also taken place, which was proven by the presence of diethylene glycol in bottoms (9) (page 8, line 2 of D1). This etherification reaction must have completely consumed any remaining unreacted ethylene carbonate, so that no ethylene carbonate could be present in (9).

However, said etherification is, as agreed by the parties, also an equilibrium reaction and as such cannot lead to a complete conversion, either. For this reason cyclic carbonate must be present in the bottoms

(9), as required by claim 1.

7. As the presence of unreacted cyclic carbonate in the bottoms (9) of reactor (I) in figure 5 of D1 is the unavoidable result of the process therein disclosed, it is concluded that this feature is not a distinguishing feature vis-à-vis D1.

*Etherification of the bottoms (9) of the transesterification reactor (I).*

8. The appellant argued that an etherification as required by claim 1 would necessarily take place in apparatuses (III), (IV) and (VII) of D1, and in any of the lines after reactor (I). The mixture in line (9) of D1 contained transesterification catalyst (KOH), which was also an etherification catalyst according to paragraph [74] of the patent in suit, diethyleneglycol was formed already in reactor (I) of D1 (page 8, line 2), and the etherification reaction must have been simply completed in the subsequent apparatuses of the system, in the light of the pressure and temperature conditions in said apparatuses, and of the presence of catalytic KOH.
9. Document D1 neither discloses the operating conditions of the apparatuses (III), (IV) and (VII), nor the temperature or pressure of any of the feeds. In the absence of said information, it needs to be examined whether at least one of these apparatuses or feeds acts as an etherification reactor under every operating conditions compatible with a transesterification process.
10. Apparatuses (III) and (IV) are falling film evaporators. In comparative example 1 of the patent in suit, which contained a falling film evaporator (28)

(page 20, line 45 in combination with page 20, line 55), no etherification took place. Therefore, it cannot be concluded that an etherification necessarily takes place in falling film evaporators (III) and (IV) of D1 since, as shown by comparative example 1, there are operating conditions at which no reaction occurs in such a falling film evaporator.

11. In the examples of the patent in suit, it was measured whether any etherification reaction had taken place by detecting the presence of CO<sub>2</sub> in line (24), since said CO<sub>2</sub> is a product of said etherification. The appellant has challenged this method by arguing that this CO<sub>2</sub> could also remain, dissolved, in the bottoms of column (17), and concluded that comparative example 1 failed to prove that no etherification took place in the tested conditions.

However, the appellant has not provided any evidence in this respect. Moreover, line (24) of the working examples of the patent in suit does contain CO<sub>2</sub> originating from the etherification step. This argument of the appellant is, hence, dismissed.

12. The appellant argued that an etherification as required by claim 1 necessarily took place in D1 in the hot feeds containing catalyst linking the different apparatuses after reactor (I).

However, in comparative example 1 of the patent in suit no etherification has been detected, and therefore such an etherification reaction did not take place in the lines of said example. As document D1 is silent about the pressure and temperature in the feeds disclosed, and the patent in suit proves that at least under some conditions no reaction takes place, an etherification

reaction in the feeds after reactor (I) is not the unavoidable result of the process of D1.

13. With respect to distillation column (VII) of document D1, comparative example 4 of the patent in suit provides evidence that such a distillation column, (column (17) in figure 3), at the operating conditions chosen, did not act as an etherification reactor, as no carbon dioxide had been detected in (24). Since D1 is silent about the operating conditions of (VII) and at least under some conditions compatible with a transesterification system it does not act as an etherification reactor, such an etherification is not the unavoidable result of the distillation column (VII) in the process of D1.

14. For these reasons, it is concluded that an etherification step of the bottoms (9) of column (I) is not the unavoidable result of the process of D1, with the consequence that the subject-matter of claim 1 is novel.

Inventive step, Article 100(a) and 56 EPC:

15. Closest prior art:

It is agreed with the parties and the opposition division that document D1 is the closest prior art.

D1 differs from the subject-matter of claim 1 in that it fails to disclose an etherification step of the bottoms of the transesterification reactor (see points 8. to 14.).

16. Technical problem underlying the invention:

The problem underlying the invention is providing a method which enables a high purity diol to be easily obtained without a need for a complicated distillation-separation step (see paragraph [13] of the patent in suit).

17. Solution:

The solution proposed by the subject-matter of claim 1 is an additional etherification of the bottoms of the transesterification reaction.

18. Success:

18.1 Example 1 of the patent in suit discloses a process comprising a transesterification in reactor (1) followed by an etherification in reactor (15). After a distillation in column (17), 99.8% ethyleneglycol was obtained in line (33) and no other product was detected.

In comparative example 1, the bottoms of the transesterification reaction (1) are directly fed into distillation column (17) without the additional etherification step. Although the yield of ethyleneglycol obtained in (33) was the same as in example 1, it contained 0.27% of diethyleneglycol.

Therefore, an additional etherification step, which is the distinguishing feature of the invention, allows obtaining very pure alkylene glycol only after one distillation step. The problem underlying the claimed invention, namely providing a method which enables a high purity diol to be easily obtained without a need for a complicated distillation-separation step, is thus

considered to be solved by the method of claim 1.

- 18.2 The appellant argued that the process of D1 required less separation steps than some embodiments of the patent in suit, such as example 6, and for this reason, the problem underlying the claimed invention could not be considered to be solved over the whole scope of the subject-matter of claim 1.

Example 1 and comparative example 1 show the effect of the feature distinguishing the claimed process from D1. The patent in suit does not provide any poly(diols) detection limit, but amounts in the order of the tens of parts per million have been disclosed, so that the level of diethylene glycol in the ethylene glycol obtained in line (33) of example 1 must be well below 0.27%. Although additional separation steps could still be carried out, by using the claimed method one distillation is sufficient to obtain diol in which no poly(diols) is detected and, therefore, the distinguishing feature of the invention vis-à-vis D1, namely including an etherification step of the unreacted cyclic carbonate, enables a high purity diol to be easily obtained without a need for a complicated distillation-separation step, and solves therefore the problem underlying the invention defined in point 16.

This argument of the appellant is therefore dismissed.

- 18.3 The appellant also argued that the number of purification steps was not a feature of claim 1, so that the alleged effect of simplifying the separation of the diol obtained with respect to the process of document D1 was not achieved over the whole scope of the subject-matter claimed.

The invention seeks to provide a method for obtaining a high purity diol easily and without complicated distillation-separation steps. Avoiding complicated distillation-separation steps is thus the result achieved by the claimed invention. The feature which needs to be specified in the claim for achieving this effect, namely an etherification step of the bottoms of the transesterification reaction, is defined in said claim.

This argument of the appellant is thus rejected.

- 18.4 The appellant has further argued that the quality of the product obtained in D1 was already good according to the experiments provided during the appeal proceedings, so that no improvement with respect to D1 had been achieved.

However, the problem underlying the claimed invention is providing a method in which high purity diol can be easily obtained without a need for a complicated distillation-separation step. As such, it is not necessary to prove an improvement with respect to D1 in terms of the purity of the final diol in order to consider said problem solved.

This argument of the appellant is, therefore, also dismissed.

19. Finally, it remains to be examined whether the claimed solution was obvious for the person skilled in the art:

The etherification of cyclic carbonate with the diol produced in the reaction is a known, undesired side reaction in a transesterification process, see for example document D2, page 2, lines 15-17 or document



D3, page 3, lines 1-5 and 18-19. Such a reaction necessarily leads to a lower diol yield, and the poly(diols) formed is an undesired impurity.

Although in reactor (I) of document D1 some etherification is already obtained, no indication has been found that an additional etherification step on the bottoms from reactor (I) could lead to an easy separation of the diol.

This is even more surprising taking into account that the product of such an etherification is the main impurity in the final diol according to the data in the patent in suit. When trying to obtain a method which allowed a simple purification of glycol obtained by a transesterification process, the skilled person would not attempt to increase the amount of a compound which is a known impurity of the final product.

No indications towards this solution had been thus found in the art, with the consequence that the subject-matter of claim 1 is inventive in the sense of Article 56 EPC.

20. The subject-matter of claim 1 is thus novel and inventive. Since claims 2-12 are all dependent from claim 1, the subject-matter of these claims is, hence, novel and inventive for the same reasons.

## **Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:



C. Rodríguez Rodríguez

P. Gryczka

Decision electronically authenticated