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# Datasheet for the decision
# of 15 May 2007

**Case Number:** T 0371/00 - 3.3.07

**Application Number:** 94105086.6

**Publication Number:** 0618009

**IPC:** B01J 31/40

**Language of the proceedings:** EN

**Title of invention:**
Method for stabilizing rhodium compound

**Patent Proprietor:**
DAICEL CHEMICAL INDUSTRIES, LTD.

**Opponents:**
BP Chemicals Ltd

**Headword:**
-

**Relevant legal provisions:**
EPC Art. 84, 123(2), 54, 56

**Keyword:**
"Amendments - added subject-matter: yes - first and second auxiliary requests"
"Novelty: no - main request; yes - third and fourth auxiliary requests"
"Inventive step: no - third auxiliary request; yes - fourth auxiliary request"

**Decisions cited:**
-

**Catchword:**
-

**Case Number:** T 0371/00 - 3.3.07

# DECISION
**of the Technical Board of Appeal 3.3.07**
**of 15 May 2007**

**Appellants:**
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(02) (Patent Proprietors) DAICEL CHEMICAL INDUSTRIES, LTD.
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**Representative:** Grünecker, Kinkeldey,
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**Decision under appeal:** **Interlocutory decision of the Opposition Division of the European Patent Office posted 1 March 2000 concerning maintenance of European patent No. 0618009 in amended form.**

**Composition of the Board:**

**Chairman:** S. Perryman
**Members:** B. ter Laan
G. Santavicca

## Summary of Facts and Submissions

I. The appeals by the proprietors and the opponents lie against the interlocutory decision of the opposition division posted on 1 March 2000 deciding that the patent in amended form met the requirements of the EPC, but the patent as granted did not.

II. Mention of the grant of European patent No. 0 618 009, based on application No. 94105086.6, filed on 30 March 1994 and published on 5 October 1994, was published on 30 July 1997. The patent was granted on the basis of eight claims with claims 1, 2 and 7 reading:

"1. A method for stabilizing a rhodium compound contained in a catalyst solution contaminated with tar formed as a by-product in the carbonylation reaction in which methyl acetate or dimethyl ether is chemically bonded with carbon monoxide in the presence of a catalyst system comprising a rhodium compound and an alkali metal iodide to produce acetic anhydride, characterized in that the catalyst solution as such or alternatively a catalyst solution mixture formed by adding a diluent comprising at least one of the compounds present in the carbonylation reaction system to the catalyst solution is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution.

2. The method for stabilizing a rhodium compound according to claim 1, wherein said diluent is a solvent containing methyl iodide.

7. A method for regenerating a catalyst solution contaminated with tar formed as a by-product in the carbonylation reaction in which methyl acetate or dimethyl ether is chemically bonded with carbon monoxide in the presence of a catalyst system comprising a rhodium compound and an alkali metal iodide to produce acetic anhydride, characterized by comprising the step that the catalyst solution as such or alternatively a catalyst solution mixture formed by adding a diluent comprising at least one of the compounds present in the carbonylation reaction system to the catalyst solution is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

III. A notice of opposition against the patent was filed on 30 April 1998, in which the revocation of the patent in its entirety was requested on the grounds of Article 100(a) EPC, specifically lack of novelty and of inventive step.

The opposition was, *inter alia*, supported by:

D1 US-A-4 476 238,
D2 Journal of Chemical Education 63(3), 1986, pages 206-209,
D3 EP-A-0 064 989,
D4 Journal of Molecular Chemistry 1987, 39, pages 115 to 136,
D6 US-A-4 735 749,
D8 US-A-4 976 947.

IV. At the oral proceedings on 15 February 2000, the opposition division decided that the patent in the amended form based on six claims of the fifth auxiliary request fulfilled the requirements of the EPC, but the main and the first to fourth auxiliary requests did not. The main request related to the claims as granted. The auxiliary requests were all filed during the oral proceedings. The reasoning of the decision can be summarized as follows:

(a) The main request was not allowable since the subject-matter of independent claims 1 and 7 as granted was not novel over D3, which disclosed a process for the recovery of rhodium and tar separation by a liquid-liquid extraction with aqueous hydrogen iodide solution and organic methyl iodide solution. The rhodium catalyst solution from which the tar was extracted came from the carbonylation of methyl acetate (D3, Claim 1). Hence, once the carbonylation reaction was started and tar began to form, the catalyst solution of the carbonylation reaction of D3 could not be distinguished from that claimed in the contested patent. It comprised a rhodium compound, tar, methyl iodide (18-20% by weight), methyl acetate, acetic acid, acetic anhydride and lithium (D3, page 4, lines 30-53). As the carbonylation reaction continued, a catalyst solution as defined in claims 1 and 7 of the contested patent was treated with carbon monoxide. The wording of claims 1 and 7 did not clearly define that the carbonylation of the methyl acetate had to be interrupted before the carbon monoxide or carbon monoxide and hydrogen treatment of the catalyst solution started.

(b) The additional feature added in claim 1 of the first auxiliary request "wherein said catalyst solution or said catalyst solution mixture has a methyl iodide concentration of 5 to 50 percent by weight" did not serve to establish novelty over D3 which also showed this feature.

(c) The amendment made in the second and third auxiliary requests, that now contained the feature "... a catalyst solution mixture formed by adding a solvent containing methyl iodide comprising..." was, in the absence of an indication when the actual addition thereof took place, not suited to make a distinction between the catalyst solution claims and what was disclosed in D3.

(d) The subject-matter of claim 1 of the fourth auxiliary request was limited by the feature that "... said catalyst solution comprises a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction." This made the claim novel as neither D1 nor D3 disclosed the treatment of a catalyst solution provided by conducting the flash vaporization after the carbonylation reaction, nor did they disclose the addition of a solvent containing methyl iodide to that catalyst solution.

As regards inventive step of the process of claim 1 of the fourth auxiliary request, the closest prior art document was D1, which disclosed a process for the carbonylation of methyl acetate to acetic anhydride in the presence of a rhodium catalyst, methyl iodide and lithium iodide. The tar formed in the reactor was removed by a liquid-liquid extraction with a solvent substantially immiscible with the carbonylation reaction mixture, and the extracted catalyst solution was then recycled to the carbonylation reaction.

The problem to be solved was to stabilize the rhodium in the catalyst solution in order to prevent it from settling, thus making it possible to return it to the carbonylation system without removal of the alkali metal and/or iodine therefrom.

That problem had become apparent when carrying out the process of D1. Once formulated, its solution was obvious from D4, which taught that either an increase of iodide or an increase in the carbon monoxide partial pressure could stabilize the rhodium complex in the methanol carbonylation system, which was similar to the carbonylation of methyl acetate.

Therefore, the process of the fourth auxiliary request was not inventive.

(e) In the fifth auxiliary request, the process was further distinguished from the prior art by the addition of methyl iodide after the carbonylation reaction. The examples showed that that prevented the sedimentation of the rhodium compound in a subsequent heat treatment. Although D3 and D4 taught that iodide could stabilize the rhodium compound, neither of them disclosed increasing the iodide concentration by the carbonylation of methyl iodide. Nor did any of the other documents suggest doing so with a view to stabilize the rhodium compound. Therefore, the process of the fifth auxiliary request was inventive.

V. On 10 April 2000, the opponent lodged an appeal against the above decision. The prescribed fee was paid on the same day. With the statement setting out the grounds of appeal filed on 30 June 2000, arguments were submitted and three further documents were cited. With a letter dated 1 November 2000, further arguments were filed. In response to a communication by the Board pointing out the issues to be discussed during oral proceedings, the opponent informed the Board of their intention not to attend the oral proceedings (letter of 19 March 2007) asking for a decision on the basis of the written record.

On 5 May 2000 the patent proprietor also filed an appeal against the above decision, the prescribed fee being paid on the same day. With the statement setting out the grounds of appeal filed on 7 July 2000, the proprietor filed four sets of six claims each as the first to fourth auxiliary requests, the main request being maintenance of the patent as granted. By letter of 19 January 2001, a set of five claims as the fifth auxiliary request, as well as further comments were submitted. By letter of 11 April 2007, a new main request of eight claims as well as four further sets of four claims each were filed, headed Fifth to Eighth

Auxiliary Request, but in fact being the sixth to ninth auxiliary requests.

VI. Oral proceedings before the Board were held on 15 May 2007 in the absence of the opponents, in accordance with Rule 71(2) EPC. During the oral proceedings the proprietor filed a new fourth auxiliary request to replace the fourth auxiliary request then on file.

The independent claims of the requests then on file included the following (amendments vis-à-vis the corresponding granted claim being indicated in bold by the Board, deletions by striking out and dots indicating unchanged passages):

*Main request:*

"1. A method for stabilizing a rhodium compound ... in the catalyst solution, **wherein the catalyst solution to be treated is provided by removing acetic anhydride from the reaction mixture.**"

*First auxiliary request:*

"1. A method for stabilizing a rhodium compound ... characterized in that the catalyst solution as such **containing methyl iodide** or alternatively a catalyst solution mixture formed by adding a ~~diluent comprising at least one of one of the compounds present in the carbonylation reaction system~~ **solvent containing methyl iodide** to the catalyst solution**, wherein said catalyst solution comprises a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction**, is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

*Second auxiliary request*

"1. A method for stabilizing a rhodium compound ... characterized in that the catalyst solution as such **containing 3.3 to 50% by weight methyl iodide** or alternatively a catalyst solution mixture **containing 5 to 50% by weight of methyl iodide** formed by adding a ~~diluent comprising at least one of one of the compounds present in the carbonylation reaction system~~ **solvent containing methyl iodide** to the catalyst solution, **wherein said catalyst solution comprises a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction,** is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

*Third auxiliary request:*

"1. A method for stabilizing a rhodium compound ... characterized in that the catalyst solution as such **containing 5 to 50% by weight methyl iodide** or alternatively a catalyst solution mixture **containing 5 to 50% by weight of methyl iodide** formed by adding a ~~diluent comprising at least one of one of the compounds present in the carbonylation reaction system~~ **solvent containing methyl iodide** to the catalyst solution,

**wherein said catalyst solution comprises a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction,** is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

*Fourth auxiliary request:*

"1. A method for stabilizing a rhodium compound contained in a catalyst solution contaminated with tar formed as a by-product in the carbonylation reaction in which methyl acetate or dimethyl ether is chemically bonded with carbon monoxide in the presence of a catalyst system comprising a rhodium compound and an alkali metal iodide to produce acetic anhydride, characterized in that ~~the catalyst solution as such or alternatively~~ a catalyst solution mixture formed by adding a ~~diluent comprising at least one of one of the compounds present in the carbonylation reaction system~~ **solvent containing methyl iodide** to the catalyst solution**, wherein said catalyst solution is a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction,** is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

"5. A method for regenerating a catalyst solution contaminated with tar formed as a by-product in the carbonylation reaction in which methyl acetate or dimethyl ether is chemically bonded with carbon monoxide in the presence of a catalyst system comprising a rhodium compound and an alkali metal iodide to produce acetic anhydride, characterized by comprising the step that ~~the catalyst solution as such or alternatively~~ a catalyst solution mixture formed by adding a ~~diluent comprising at least one of the compounds present in the carbonylation reaction system~~ **solvent containing methyl iodide** to the catalyst solution, **wherein said catalyst solution is concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction**, is treated with carbon monoxide or a mixture of carbon monoxide and hydrogen before the catalyst solution or the catalyst solution mixture is applied to a process for separating the tar contained in the catalyst solution."

VII. The opponent's arguments given in writing, in so far as they are relevant to the requests needing consideration, can be summarized as follows:

(a) Regarding the main request, if the catalyst solution to be treated was construed as including the tar-containing reaction composition, then D3 was novelty destroying for the reasons given by the opposition division in their decision.

If the catalyst solution however was not so construed but was seen as a catalyst composition treated outside the carbonylation reactor, as the proprietor maintained, then the claims lacked an inventive step in view of the prior art documents already on file as well as three additional documents, in various combinations.

Dl disclosed a process for the selective removal of tars from the reaction composition or a concentrate thereof, which tars were produced in rhodium-catalysed, iodide-promoted carbonylation of methyl acetate or dimethyl ether. The concentrate was obtained by flashing the reaction composition. Addition of an alkyl halide solvent to a rhodium- and tar-containing composition was also disclosed in Dl. D4 taught that in rhodium-catalysed carbonylation processes, an increase in iodide concentration stabilised the rhodium catalyst against precipitation, as did carbon monoxide. To increase the iodide concentration by the carbonylation of methyl iodide, for which there was in fact no special requirement in the claims, was general common knowledge, as shown by D2.

(b) The claimed subject-matter of the main request was also not inventive in view of D6, which disclosed a process for the stabilization of a rhodium carbonylation catalyst, involving the separation of a catalyst solution from a carbonylation reaction composition. As according to D6 a methyl iodide as well as an accelerator may be used, it would be obvious to use a methyl iodide/alkali iodide system. D6 also contemplated treatment of the catalyst solution with carbon monoxide and hydrogen, so that the claims of the main request were not inventive.

(c) For the same or similar reasons, the subject-matter claimed according to the first auxiliary request was also not inventive and the addition, in the process according to the second auxiliary request, of concentration ranges for methyl iodide did not provide any contribution to an inventive step, especially in view of the statement in the patent specification that there was no particular limitation on the composition. The claimed concentrations were known from D1 and carbonylation reactions with methyl iodide concentrations in that range were part of the common general knowledge. Similar reasons were valid regarding the process of the third auxiliary request.

(d) Regarding the fourth auxiliary request, claims 1 and 5 of which were the same as those of the fifth auxiliary request before the Opposition division, on the basis of which the latter had decided to maintain the patent, the opponent had argued in its grounds of appeal that D1 was the closest prior art and disclosed a process of tar removal from the reaction composition or a concentrate thereof in a rhodium-catalysed, iodide-promoted carbonylation of methyl acetate or dimethyl ether. If, as stated by the proprietor, D1 suffered a problem of rhodium instability, then the solution was obvious in view of D4, which taught that in rhodium–catalysed carbonylation processes, an increase in iodide concentration stabilised the rhodium catalyst against precipitation, and which also disclosed the addition of alkyl halide solvent to a rhodium- and tar-containing composition, including one concentrated by flashing. As it was common general knowledge of the skilled person, and also stated in document D11 (EP-A-0132391, page 11, lines 21 to 24) that hydrogen iodide is produced by the carbonylation of methyl iodide in the presence of a carbonylation catalyst, it would be obvious to increase the iodide ion concentration using a methyl iodide solvent and carbon dioxide. Common general knowledge supporting this argument was also found in documents D2, D8 and D12 (Eby & Singleton Applied Industrial Catalysis, 1983, Vol. 1 pages 275 to 296).

VIII. The proprietor's arguments can be summarized as follows:

(a) The invention concerned the stabilisation of a rhodium catalyst solution that had to undergo a heat treatment in order to remove tar. The catalyst solution to be treated resulted from the production of acetic anhydride followed by at least partial removal of the acetic anhydride. It was therefore not identical to the solution as present in the reactor. The stabilisation was achieved by treating the catalyst solution as such or after addition of one or more compounds as present in the reactor, with carbon monoxide or a carbon monoxide/hydrogen mixture before the tar was removed. That was reflected in the wording of the claims of the main request. The difference between the independent claims 1 and 7 was that one was directed to the stabilization of the catalyst, whereas the claim directed to the regeneration process concerned the prevention of precipitation of the catalyst during subsequent steps.

The wording of the present claims, support for which could be found in the original claims and the description, excluded the treatment of the reaction mixture as such with carbon monoxide or carbon monoxide/hydrogen. The requirements of Articles 84, 123(2) and 123(3) EPC were therefore complied with.

(b) As none of the documents on file disclosed the present combination of features, in particular the present mandatory removal of acetic anhydride from the catalyst solution before its treatment, the claimed subject-matter was novel.

(c) D1 was the closest prior art document. The problem to be solved was the stabilization of the rhodium compound in a process for tar removal and could be seen as an improvement over D1. The examples showed that that problem had been solved by the claimed process, involving the treatment of the rhodium containing solution with carbon monoxide before it was submitted to the tar removal heat treatment.

(d) D3, D4 and D6 addressed the stabilization of rhodium catalysts.

D3 however referred to the stabilization of rhodium catalysts in the water used for extracting it from the tar, hence not before the removal of the tar, but during extraction of the rhodium. D4 concerned a different aqueous reaction system - methanol carbonylation at low water concentrations, whereas the present system was free of water. Therefore, the combination of any of D3 or D4 with D1 did not render the claimed subject-matter obvious.

D6 did refer to the same reaction system as now claimed, but it did not mention stabilization of the rhodium catalyst before tar removal. It rather aimed at improving the selectivity and activity of the catalyst. Precipitation of the catalyst during tar removal was not mentioned and no teaching was present relating to the addition of carbon monoxide to the catalyst solution. For those reasons, a combination of D6 with D1 would also not render the claimed subject-matter obvious so that the presence of an inventive step should be acknowledged.

(e) As to the first to fourth auxiliary requests, the amendments limited the claimed subject-matter. They found their basis in the original application and were intended to overcome the objections that had been raised. In particular, the wording indicated that the treatment of the catalyst solution took place outside the reactor. Other limitations moved the subject-matter now being claimed further away from the prior art. In particular, no document disclosed the addition of methyl iodide to the flashed catalyst solution before the treatment with carbon monoxide/hydrogen.

IX. The appellants (opponents) requested that the decision under appeal be set aside and the patent be revoked.

The appellants (patent proprietors) requested that the decision under appeal be set aside and that a patent be maintained on the basis of the claims of the main request filed on 11 April 2007 or the claims of the first, second or third auxiliary requests filed on 7 July 2000, or on the basis of claims 1 to 6 of the fourth auxiliary request filed on 15 May 2007, page 2 of the description of the patent specification, pages 3 and 5 to 8 of the description filed during oral proceedings on 15 February 2000 before the opposition division, and page 4 of the description filed at oral proceedings on 15 May 2007, or on the basis of the claims of the fifth auxiliary request filed 19 January 2001, or on the basis of the claims of the sixth to ninth auxiliary requests filed headed Fifth to Eighth Auxiliary Requests on 11 April 2007.

## Reasons for the Decision

1. The appeals are admissible.

*Main request - novelty*

2. Claim 1 refers to the treatment with carbon monoxide or carbon monoxide/hydrogen of two alternatives, either "the catalyst solution as such" or "a catalyst solution mixture formed by adding a diluent comprising at least one of the compounds present in the carbonylation reaction system to the catalyst solution".

2.1 Although tar-separation is mentioned in claim 1, this is only by way of indicating the context of when the rhodium stabilizing process actually claimed should be applied. Tar separation itself is not a technical feature required by the claim.

2.2 D6 discloses a process for producing an O-acetyl compound by contacting compounds selected from the group of methyl carboxylates, dimethyl ether and mixtures thereof with methanol, with carbon monoxide gas in the presence of a rhodium carbonylation catalyst and an iodine material that provides methyl iodide in the reaction liquid, whereby a liquid reaction product is produced comprising volatile components and a non-volatile, rhodium catalyst-containing solution, after which the reaction mixture formed in the carbonylation reaction step is separated into the volatile component and the rhodium catalyst-containing solution which is then fed into a treatment zone and heated in the presence of a hydrogen-containing gas and then recirculating the hydrogen-treated catalyst solution by feeding it from said treatment zone into the carbonylation reactor (claim 1). The presence of carbon monoxide in the hydrogen gas is preferred (column 3, line 60 to column 4, line 4). In Example 1, an autoclave was charged with rhodium chloride trihydrate, aluminium metal powder, methyl iodide, acetic acid and methyl acetate, which mixture was reacted with carbon monoxide and then released from the reactor and flash-distilled until half the volume. The remaining solution contained 0.45 weight% methyl iodide, 0.5 weight% methyl acetate, 38.7 weight% acetic acid and 51.2 weight% acetic anhydride. In example 2, the catalyst solution obtained in example 1 was contacted with a gas mixture of 4 parts by volume of hydrogen and 1 part by volume of carbon monoxide and then heated to 140°C and heat-treated in the presence of hydrogen.

D6 therefore discloses all the features of the stabilization of the rhodium compounds according to present claim 1: the carbonylation reaction mixture is flashed and the remaining catalyst solution is contacted with a mixture of hydrogen/carbon monoxide. Hence, the method according to claim 1 is not novel over D6 (Article 54 EPC). Since for this reason alone the main request has to be refused it is not necessary to consider whether the main request should also be refused on the grounds of other objections, raised in the communication sent by way of preparation of the oral proceedings, regarding compliance with the requirements of Articles 84 and 123(2) EPC.

*First auxiliary request*

3. The first auxiliary request differs from the main request in the requirements that the catalyst solution as such should contain methyl iodide and the catalyst solution mixture should be formed by adding a solvent containing methyl iodide to the catalyst solution and that "said catalyst solution comprises a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction."

3.1 According to the appellant, the basis for the first amendment would be found on page 16, last paragraph of the original application. The paragraph bridging pages 16 to 17 in fact states "Although there is no particular limitation on the composition of the concentrated catalyst solution to be subjected to a carbonylation treatment, it is preferred that the methyl iodide concentration be 5 to 50% by weight, preferably 10 to 30% by weight, from the viewpoint of the reaction rate. In order to meet this requirement, the carbonylation treatment is preferably conducted after methyl iodide as such or a solution containing methyl iodide is added to the concentrated catalyst solution ..." . The passage discloses adding methyl iodide in order to achieve a specific methyl iodide concentration. The claim seeks to generalize this beyond what is actually disclosed, and so does not meet the requirements of Article 123(2) EPC.

3.2 The first auxiliary request must therefore be refused and the question of novelty over D6, according to which the catalyst solution as such contains methyl iodide, need not be answered.

*Second auxiliary request*

4. The second auxiliary request differs from the first auxiliary request in the additional requirement that the catalyst solution obtained as such should contain 3.3 to 50% by weight methyl iodide and the catalyst solution mixture should contain 5 to 50% by weight methyl iodide and should be formed by adding a solvent containing methyl iodide to the catalyst solution.

4.1 As pointed out above in point 3.1, the paragraph bridging pages 16 and 17 as filed refers to a preferred methyl iodide concentration of 5 to 50% by weight. There is however no disclosure for the lower limit of 3.3 weight% methyl iodide in the concentrated catalyst solution. The only instance where that amount can be found is in Example 1, which is limited to a specific situation resulting in a specific catalyst solution. There is no disclosure from which the conclusion could be drawn that that amount could be a lower limit having any technical significance. Therefore, claim 1 of the second auxiliary request does not comply with the requirements of Article 123(2) EPC, and this request must be refused.

### *Third auxiliary request*

5. The third auxiliary request is the same as the second auxiliary request apart from the lower limit of now 5% by weight methyl iodide in the catalyst solution as such. This does have basis in the above-cited paragraph (cf. point 3.1) bridging pages 16 and 17 of the application as filed, already cited in connection with the second auxiliary request.

### Novelty

6. The addition of a solvent containing methyl iodide to the catalyst solution prior to the treatment with carbon monoxide(/hydrogen) is not disclosed in any of the cited documents either for the catalyst solution as such or for the catalyst solution mixture.

6.1 In D6 the reaction mixture is flashed and the catalyst solution is then fed into a treatment zone and heated in the presence of a hydrogen-containing gas, preferably containing carbon monoxide, without any addition of methyl iodide, after which it is recirculated to the carbonylation reactor (column 3, lines 35 to 47).

6.2 In Example 1 of D1 the product, acetic anhydride is removed by flashing a withdrawn stream of the reaction mixture and the remaining liquid is recycled to the reactor without any methyl iodide addition. In Examples 3 and 4, the liquid remaining after flashing is subjected to a liquid-liquid tar removal extraction without any prior addition of methyl iodide.

6.3 In D3 methyl iodide is added to the catalyst solution as an extractant and as part of the tar removal system (examples). No contacting with methyl iodide prior to tar removal is described.

6.4 For the alternative in claim 1 of this request relating to the catalyst solution as such, novelty over D3 can be acknowledged as in D3 the methyl iodide is added as an extractant in the tar-removal system, not to the the catalyst solution after the carbonylation reaction. Novelty over D6 can be accepted in view of the feature relating to the methyl iodide concentration being between 5 to 50%, whereas D6 discloses a far lower value (see point 2.2 above). D1, too, does not disclose any value of 5% or higher for the methyl iodide concentration (see point 7. below).

Inventive step

7. For the purpose of assessing inventive step, D1 needs consideration as a potential starting point.

7.1 D1 discloses a process for the selective removal of high molecular weight rhodium-containing tars containing organic carbonyl and acetate function produced in rhodium-lithium catalyzed carbonylation reactions in which methyl acetate or dimethyl ether are combined with carbon monoxide in the presence of iodides to form acetic anhydride or ethylidene diacetate comprising extracting said tars from carbonylation reaction mixtures or concentrates thereof with a liquid solvent substantially immiscible with said mixtures and concentrates and capable of preferentially dissolving said tars relative to the contained rhodium while removing a minimum of carbonylation products and by-products and comprising at least one member of the group consisting of alkanes, cycloalkanes, and solvents belonging to Groups I, VIb, and VII as defined by Snyder, J. of Chromatography, 92, p. 223-230, 1974 and thereafter separating the tar-containing solvent from said mixtures and concentrates, recovering said solvent, discarding said tar, and recycling said separated mixtures or concentrates to said carbonylation reaction (claim 1). The solvent may be contacted with the tar-containing liquid concentrate resulting from the flashing of a carbonylation reaction mixture from reaction pressure to a lower pressure to remove a portion of low-boiling reactants and reaction products (claim 3).

In Example 1, methyl acetate, methyl iodide, carbon monoxide and hydrogen are present as reactants and the reaction is catalyzed by a mixture of rhodium trichloride trihydrate and lithium iodide. The product, acetic anhydride, is removed by flashing a withdrawn stream of the reaction mixture. The remaining liquid contains 4 weight% methyl iodide, 7 weight% methyl acetate, 32 weight% acetic anhydride, 24 weight% acetic acid and about 1 to 2 weight% heavy residues. A slipstream is withdrawn from the recycle stream at a rate sufficient to maintain the residues in the autoclave at an acceptable level. The tar-containing liquid which remains after the gases have been separated is treated with solvents to remove the tar content while excluding the rhodium.

In Examples 2 to 4 of D1 the heavy residue is extracted from the flashed liquid, leaving most of the rhodium for recycling to the carbonylation reaction.

7.2 According to the specification of the patent in suit, several methods existed for separating tar from rhodium contained in the solution resulting from the carbonylation reaction to produce acetic anhydride (page 2, lines 14 to 32), the process according to D1 not having the disadvantages of the other methods and enabling the tar/rhodium separation to be conducted in a very simple manner. However, the process of D1 necessitated the removal of components having a low-boiling point contained in the catalyst solution, such as methyl iodide and methyl acetate, because they could form azeotropes with the solvents used for tar removal. They should be separated, e.g. by distillation, before the tar and rhodium containing solution was contacted with the extractants (page 2, lines 14 to 43). It had been found that some rhodium complexes contained in the catalyst solution had a tendency of precipitating when heated during such a distillation step and it was the object of the invention to stabilize the rhodium compound (page 2, line 44 to page 3, line 3 and page 3, lines 43 to 49).

8. In view of that passage, there can be no doubt that D1 is a proper starting point for the assessment of inventive step and that, according to the patent in suit, the problem to be solved is to improve the process described in D1, i.e. to stabilize the rhodium compound present in the concentrated catalyst solution, in particular so that it will not precipitate when it is submitted to a heat-treatment step such as distillation in order to remove low-boiling point components before the actual tar-removal.

8.1 Hence, the problem described in the patent in suit wholly refers to a process including the removal of low-boiling components from the concentrated catalyst solution such as by a distillation step before tar-removal, as in the process of D1. However, present claim 1 is completely silent about any such removal, let alone by a distillation step. The process according to present claim 1 may or may not involve the removal of low-boiling components by distillation. If no distillation prior to tar-removal is carried out, the specific problem of precipitation of the rhodium compound will not occur, so that the prevention of such precipitation cannot be taken as the problem to be solved by the subject-matter of claim 1. The only remaining problem that can be stated is the stabilization of the rhodium compound in the catalyst solution.

8.2 Although the examples in the patent in suit only provide information regarding the precipitation of the rhodium compound in a heat-treatment, it has not been contested that a stabilizing effect occurred. Therefore, the Board can accept that the treatment with carbon monoxide/hydrogen will effectively result in the desired stabilization.

9. The problem of the stabilization of rhodium compounds is addressed in D6 (cf. point 2 above). According to D6, treating the concentrated catalyst solution with a mixture of hydrogen and carbon monoxide not only results in the formation of less methane, but also in a rapid recovery of the activity of the rhodium catalyst (column 3, line 60 to column 4, line 1) due to the formation of more active complexes. The formation of the active form of the rhodium/carbon monoxide-complex - indicated in the patent in suit as complex (a) (page 2 line 50)-, as a consequence of the treatment, is specifically described (column 4, lines 11 to 26).

Therefore, D6 steers the skilled person looking for stabilizing the rhodium compounds contained in the concentrated catalyst solution, toward treating the concentrated catalyst solution with a mixture of hydrogen/carbon monoxide. While the specific example of D1 has a methyl iodide concentration of 4% by weight, there is no evidence that treating a catalyst solution as such having a lower limit of 5 % by weight methyl iodide would lead to any special effect. The teaching of D6 must be thus taken as applicable to any concentrated catalyst solution having a methyl iodide concentration of a level found in known processes. It is therefore obvious following the teaching of D6 to treat concentrated catalyst solutions with a mixture of carbon monoxide/hydrogen, and thereby arrive at something falling under claim 1 of the third auxiliary request. Therefore, that claim does not meet the requirements of Article 56 EPC.

In view of the objections given above, the third auxiliary request must be refused.

*Fourth auxiliary request*

10. The fourth auxiliary request differs from the previous one in the deletion of the limits for the methyl iodide concentration and the treatment of the catalyst solution as such. According to claim 1 of the fourth auxiliary request, the catalyst solution to be treated is a mixture formed by adding a solvent containing methyl iodide to a solution defined as "a concentrated catalyst solution provided by conducting flash vaporization after the carbonylation reaction". From the present wording it is clear that the reactor contents are flashed, then a solvent containing methyl iodide is added, and that mixture is then treated with carbon monoxide(/hydrogen). The requirements of Article 84 EPC are fulfilled.

11. The above-mentioned passage bridging pages 16 and 17 of the application as filed provides an adequate basis for the present claims, so that the requirements of Article 123(2) EPC are met.

*Novelty*

12. As detailed in point 6 above regarding the third auxiliary request, the addition of a solvent containing methyl iodide to the catalyst solution prior to the

treatment with carbon monoxide(/hydrogen) is not disclosed in any of the cited documents. Claim 1 of the fourth auxiliary request is therefore novel (Article 54 EPC).

*Inventive step*

13. The patent in suit concerns a method for stabilizing a rhodium compound. D1 is, also for the fourth auxiliary request, a proper starting point for assessing inventive step for the same reasons as given for the third auxiliary request (see point 7 above), the problem being the same as well: to improve the process described in D1, i.e. to stabilize the rhodium compound (point 8 above). For the same reasons as for the third auxiliary request, the Board can accept that adding methyl iodide to the concentrated catalyst solution followed by a treatment with carbon monoxide/hydrogen will effectively result in the desired stabilization of the rhodium compound (point 8.2 above).

14. The question remains to be answered whether the claimed subject-matter is obvious in view of the prior art documents on file.

14.1 As none of the documents discloses to add a solvent containing methyl iodide to the catalyst solution prior to the treatment with carbon monoxide(/hydrogen), there is no teaching as to the effects of such addition.

14.2 D1 concerns only the extraction process itself and the properties of the extractants, without any indication of how to improve the stability of the rhodium compounds in the process.

14.3 D4, to which the opponent referred in combination with D1, is a scientific article describing reaction mechanisms for methanol carbonylation. According to D4, the addition of iodide salts to rhodium catalyst solutions promotes the rate of carbonylation and stabilizes the rhodium catalyst in an aqueous system (Page 115, Summary). Water also plays a role in stabilizing the rhodium catalyst (page 115, Introduction). D4 therefore concerns a chemical system that differs from that of the patent in suit which does not relate to an aqueous system. Also, there is no indication in D4 to add methyl iodide after flashing the reactor contents and before carbon monoxide/hydrogen treatment.

14.4 D6 aims at prevention of the loss of activity of the catalyst and the lowering of the selectivity of the reaction and repeated recycling of the catalyst solution (column 1, line 64 to column 2, line 3). To that end, it proposes to treat the flashed reactor contents with hydrogen or hydrogen/carbon monoxide (claim 1; column 4, lines 11 to 25; example 1). It does not however teach that the addition of methyl iodide before that treatment would lead to an enhanced stabilization of the rhodium catalyst.

14.5 D3 aims at the stabilization at elevated temperatures of a rhodium catalyst in the aqueous liquid used for separating the tar from the rhodium in an extraction procedure (page 2, lines 27 to 28). It proposes to solve that problem by adding hydrogen iodide to the aqueous extraction liquid separating the rhodium from the tar. Methyl iodide is used as the organic phase. D3 does not suggest adding methyl iodide at an earlier stage, followed first by treatment with carbon monoxide or carbon monoxide/hydrogen, and only then proceeding with tar removal.

14.6 It is true that if the skilled person had known of the influence of a high iodide ion concentration on the stability of rhodium complexes, as taught in the patent in suit, he might have realized that the known carbonylation reaction involving methyl iodide would be a way of achieving this. But the prior art did not point him in this direction and the Board is not satisfied that the subject matter of the claims of this request, i.e. adding methyl iodide to the concentrated catalyst solution followed by a treatment with carbon monoxide/hydrogen, could have been derived in an obvious manner from any of the documents cited or any combination of these documents which the problem of stabilizing rhodium would have caused the skilled person to consider.

15. The same considerations apply *mutatis mutandis* to independent claim 5 as well as to the dependent claims.

16. In view of the above, the Board comes to the conclusion that the subject-matter of the fourth auxiliary request meets the requirements of the EPC.

17. The opponent did not raise any objections against the amendments in the description and the Board sees no reason to take a different view.

## Order

### For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent on the basis of claims 1 to 6 of the fourth auxiliary request filed on 15 May 2007, page 2 of the patent specification, pages 3 and 5 to 8 of the description filed during oral proceedings on 15 February 2000 before the opposition division, and page 4 of the description filed at oral proceedings on 15 May 2007.

| Registrar | Chairman |
|---|---|
| C. Eickhoff | S. Perryman |