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D E C I S I O N
of 6 April 2004

Case Number: T 1167/00 - 3.3.7

Application Number: 92302781.7

Publication Number: 0569624

IPC: B01J 23/52

Language of the proceedings: EN

Title of invention:

Catalysts and processes for the manufacture of vinyl acetate

Patentee:

BP Chemicals Limited

Opponent:

CELANESE LIMITED

Headword:

-

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step (no) - problem and solution - obvious combination"

Decisions cited:

T 0219/83

Catchword:

-



Case Number: T 1167/00 - 3.3.7

D E C I S I O N
of the Technical Board of Appeal 3.3.7
of 6 April 2004

Appellant: BP Chemicals Limited
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 9 November 2000
revoking European patent No. 0569624 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: R. E. Teschemacher
Members: B. J. M. Struif
P. A. Gryzcka

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 569 624 with respect to European patent application No. 92 302 781.7 filed on 30 March 1992 was published on 12 August 1998. The granted patent was based on sixteen claims. Independent claims 1, 6, 9 and 14 read as follows:

"1. A shell impregnated catalyst for use in the production of vinyl acetate from ethylene, acetic acid and an oxygen containing gas, said catalyst having a productivity of greater than 661 grams of vinyl acetate per hour per litre of catalyst at 150 °C and consisting of:

(1) a catalyst support having a particle diameter from about 3 to about 7 mm and a pore volume of 0.2 to 1.5 ml per gram

(2) palladium and gold distributed in the outermost 1.0 mm thick layer of the catalyst support particles, and

(3) from 3.5 to 9.5% by weight of potassium acetate wherein the gold to palladium weight ratio in said catalyst is in the range 0.60 to 1.25 and, the palladium content is greater than 3.9 grams per litre of catalyst."

"6. A shell impregnated catalyst for use in the production of vinyl acetate from ethylene, acetic acid and an oxygen containing gas consisting of:

(1) a catalyst support having a particle diameter from about 3 to about 7 mm and a pore volume of 0.2 to 1.5 ml per gram,

(2) palladium and gold distributed in the outermost 1.0 mm thick layer of the catalyst support particles, and

(3) from about 3.5 to about 9.5% by weight of potassium acetate wherein the gold to palladium weight ratio in said catalyst is greater than or equal to about 0.9 and the palladium content is greater than 3.9 grams per litre of catalyst."

"9. A process for preparing vinyl acetate which comprises reacting ethylene with acetic acid in the presence of an oxygen containing gas at a temperature in the range 100 to 200 °C in the presence of a shell impregnated catalyst having a productivity at greater than 661 grams of vinyl acetate per hour per litre of catalyst at 150 °C and consisting of:

(1) a catalyst support having a particle diameter from about 3 to about 7 mm and a pore volume of 0.2 to 1.5 ml per gram,

(2) palladium and gold distributed in the outermost 1.0 mm thick layer of the catalyst support particles, and

(3) from about 3.5 to about 9.5% by weight of potassium acetate wherein the gold to palladium weight ratio in said catalyst is in the range 0.60 to 1.25 and, the palladium content is greater than 3.9 grams per litre of catalyst."

"14. A process for preparing vinyl acetate which comprises reacting ethylene with acetic acid in the presence of an oxygen containing gas at a temperature in the range 100 to 200 °C in the presence of a shell impregnated catalyst consisting of:

(1) a catalyst support having a particle diameter from about 3 to about 7 mm and a pore volume of 0.2 to 1.5 ml per gram

(2) palladium and gold distributed in the outermost 1.0 mm thick layer of the catalyst support particles, and

(3) from about 3.5 to about 9.5% by weight of potassium acetate wherein the gold to palladium weight ratio is greater than or equal to about 0.9 and the palladium content is greater than 3.9 grams per litre of catalyst."

II. A notice of opposition was filed against the granted patent, in which the revocation of the patent in its entirety was requested on the grounds of Article 100(a), (b) and (c) EPC with respect to lack of novelty, lack of an inventive step, insufficient disclosure, and extension beyond the content of the application as originally filed, respectively. The opposition was supported *inter alia* by the following documents:

D1: US-A-4 048 096

D2: US-A-4 087 622

III. In a decision posted on 9 November 2000, the opposition division revoked the patent. That decision was based on the patent as granted as the sole request.

The opposition division held that:

(a) Having regard to sufficiency of disclosure, the feature "said catalyst having a productivity of greater than 661 grams of vinyl acetate per hour

per litre of the catalyst at 150 °C" was not a product feature itself but merely the result obtained when using the defined catalyst. The description provided sufficient technical information, such as pressure and the composition of the feed, to enable the skilled person to reproduce the claimed subject-matter.

- (b) The patent in suit did not violate Article 123(2) EPC, because the term objected to "the palladium content is greater than 3.9 g/l of the catalyst" could be derived from the originally filed documents.
- (c) The claimed subject-matter was novel over surface impregnated catalysts of D1 and D2.
- (d) Having regard to inventive step, D1 and D2 were considered to represent the closest state of the art. Since those catalysts were used for the same purpose and had a composition very close to that of the claimed subject-matter, an inventive step could only be recognized, if the claimed composition ranges, which were different from those of the prior art, provided an improvement.

The problem to be solved over that prior art was to find catalysts having a productivity in excess of 661 grams of vinyl acetate per hour per litre of catalyst at 150°C. However, such a high productivity was also obtained when using a gold/palladium ratio outside the claimed range as shown by the examples of the patent in suit. Thus, the results given in the examples of the contested

patent showed discrepancies. Figures 1 and 4 were not in line with the technical data shown in Tables 1 and 2. Consequently, the opposition division came to the conclusion that a surprising technical effect had not been shown and that an inventive step could not be acknowledged.

- IV. On 7 December 2000 the proprietor (appellant) filed a notice of appeal against the above decision, the prescribed fee being paid on the same day. In the statement setting out the grounds of appeal filed on 9 March 2001, the appellant submitted a new main request, two auxiliary requests and an experimental report.

These requests contained the following amendments to the granted version:

In the main request the term "about" was deleted. In claims 1 and 9 of the first auxiliary request additionally the phrase "at an oxygen conversion of 30 %" was inserted after the phrase "... vinyl acetate per hour per litre of catalyst at 150 °C ...". In claim 13 of that request the word "catalyst" was amended to read "process". The amendments of the second auxiliary request corresponded to those of the first auxiliary request, with the exception that granted claim 13 remained unamended.

- V. In a communication dated 26 January 2004, the board addressed the points to be discussed during the oral proceedings, in particular novelty and inventive step.
- VI. By letter of 5 March 2004 in reply to the above communication, the respondent (opponent) submitted an

evaluation of the experimental data reported in the patent in suit, D1, D2 and in the further prior art document US-A-5 179 056, based on mathematical models in form of tables.

- VII. In a reply dated 11 March 2004, the appellant requested that the oral proceedings be postponed or that the material submitted by the respondent on 5 March 2004 be considered as inadmissible.
- VIII. In a further communication dated 15 March 2004, the board announced that the date of the oral proceedings was maintained and that the relevance of the respondent's submissions should be discussed in the oral proceedings, if that became necessary.
- IX. By letter of 18 March 2004, the appellant maintained his position and argued that in view of the volume of material filed by the respondent, a fair and proper defence as required by Article 113(1) EPC was not possible. He maintained his requests on the basis of the written records and announced that he would not attend the oral proceedings scheduled for 6 April 2004.
- X. Oral proceedings were held on 6 April 2004 in the absence of the appellant in accordance with Rule 71(2) EPC.
- XI. The appellant argued in writing in substance as follows:
- (a) As regards novelty, D1 did not explicitly disclose the claimed weight ratio of gold to palladium in the catalysts. Furthermore, the concentration of gold in the claimed catalyst, which should be

implicitly greater than 2.3 g/l was not disclosed in D1. Although in D2 the alkali metal acetate concentration covered a range of 1 to 30 weight percent, the potassium acetate concentration in all examples was 3%, and thus outside the claimed range of 3.5 to 9.5%.

- (b) As regards inventive step, D1 could be used as a suitable starting point since it disclosed the highest catalyst activity of 661 g of vinyl acetate per hour per litre of catalyst at 150°C or space time yield (abbreviated as STY). Figure 1 of the patent in suit showed that at a particular palladium content of the catalyst, the activity passed a maximum within the claimed Au/Pd range and surpassed that having a Au/Pd ratio outside the claimed range. Furthermore, if the palladium content was greater than 3.9 g/l the activity at 30% oxygen conversion was greater than 661 g of vinyl acetate per hour per litre of catalyst at 150°C. In addition, the data in Table 1 of the opposed patent were measured at different oxygen conversions and must be compared in a consistent manner by using the procedure set out on page 5, lines 29 to 56 of the patent in suit. When comparisons were made, the same palladium and potassium acetate content must be observed by using equations (1) to (3) at a consistent oxygen conversion as set out in the patent in suit. The improvements were shown in Figure 1. At an Au/Pd ratio higher than 0.9, a superior selectivity was observed as shown in Figure 3 of the opposed patent.

The improved effect of the potassium acetate concentration on the catalyst activity had been shown by the experimental report filed with the statement setting out the grounds of appeal.

- (c) The appellant agreed to the decision under appeal as far as it had rejected the objections under Article 100(b) and (c) EPC.

XII. The arguments of the respondent can be summarized as follows:

- (a) The palladium content explicitly claimed was only disclosed with respect to typical catalysts having a sodium content of about 0.5% by weight. The appellant had detached this teaching from its proper context which amounted to a violation under Article 100(c) EPC.
- (b) As regards insufficiency, the productivity was governed by process conditions not mentioned in the claims, for example the pressure and the composition of the feed.
- (c) As to novelty, D1 disclosed shell impregnated catalysts used in the production of vinyl acetate from ethylene, acetic acid and oxygen. The catalyst of D1 contained gold in an amount of 2.25 g/l which did not provide a distinction over the calculated gold amount according to claim 1, when taking into account normal measurement errors. Furthermore, palladium and gold were distributed in a weight ratio of 0.91:1 as calculated from the explicitly disclosed amounts of both metals.

Finally, the exemplified potassium acetate content was within the range of from 4.7 to 7% by weight. Furthermore, the productivity was an intended result of the claimed catalyst which could be ignored when evaluating novelty and inventive step.

D2 disclosed shell impregnated catalysts, which, according to the decision under appeal, differed from the claimed subject matter only with respect to the amount of potassium acetate. Since in D2 the exemplified potassium acetate content was 3% by weight, the claimed potassium acetate content of 3.5 to 9% by weight provided no novel selection over a content of 1 to 30% by weight disclosed in D2.

Thus, the subject-matter of claims 1, 6, 9 and 14 lacked novelty.

- (d) As to inventive step, D1 and D2 were both suitable starting points. Since several samples of Table 1 of the patent in suit showed that also outside the claimed range a productivity above 661 STY could be achieved, the claimed subject-matter did not show any advantage. Thus, the problem was to provide an alternative catalyst composition over that of the cited prior art. Since the claimed subject-matter differed from D2 only by the potassium acetate content and, since it was known that this compound enhanced the catalytic activity, it was obvious to use higher amounts than those suggested by D1 and other prior art documents on file.

When starting from D1, the examples thereof showed that increasing the amounts of palladium and gold in the finished catalyst led to an enhanced productivity. Since D2 suggested that the use of higher ratios of gold to palladium had an impact on the productivity, it was obvious to apply that teaching to D1.

- (e) In the auxiliary requests, the additional feature "at an oxygen conversion of 30%" represented added subject-matter under Article 123(2) EPC or lacked clarity under Article 84 EPC. Furthermore, the auxiliary requests did not provide any further distinction over the cited prior art which contributed to an inventive step. Thus, the claimed subject-matter of all requests lacked an inventive step.

XIII. The appellant requested in writing that the decision under appeal be set aside and that the patent be maintained on the basis of the main request, or, alternatively, on the basis of the first or second auxiliary request, all filed with the statement setting out the grounds of appeal.

XIV. The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

Main request

Amendments

Objection under Article 100(c) EPC

2. According to the application as filed, claims 17 and 18 are directly referred back to claim 13 defining the same shell impregnated catalyst as in original claim 1. Consequently, the palladium content greater than 3.9 grams per litre of catalyst is originally disclosed in a general way without any reference to the sodium content. In view of the above, the objection under Article 100(c) EPC is not justified. Other formal objections have not been raised.

Objection under Article 100(b) EPC

3. The question, whether the patent in suit disclosed the invention sufficiently clearly and completely for it to be carried out by a person skilled in the art, can be left open, since irrespective of how this question is answered, the claimed subject-matter lacks an inventive step.

Objections under Article 100(a) EPC

Novelty

4. According to the decision under appeal, the claimed subject-matter differed from D2 only in that the catalyst composition comprises from 3.5 to 9.5% by weight of potassium acetate. The appellant has neither pointed out any further difference, nor argued that the decision under appeal was not correct in respect of novelty. The board has no reason to take a different view from that of the Opposition Division.

Since in D2, the exemplified amount of palladium of 3.0% by weight appears to be close to the lower claimed limit of 3.5% by weight, and since furthermore the selected sub-range of 3.5 to 9.5% by weight does not appear to be narrow in relation to the known range of 1 to 30% by weight disclosed in D2, it is highly questionable whether the claimed potassium acetate amount meets the requirements established by the boards of appeal for selection inventions (Case Law of the Boards of Appeal of the European Patent Office, 4th edition, 2001, I.C.4.2.1). However, as the claimed subject-matter lacks inventive step, the board did not need to take a decision on this question.

Inventive step

5. The patent in suit concerns a shell impregnated catalyst for use in the production of vinyl acetate having a productivity greater than 661 g of vinyl acetate per hour per litre of catalyst at 150°C (page 2, lines 41 to 43 and 54 to 56). Furthermore, a high

selectivity towards the production of vinyl acetate at the expense of by products such as carbon dioxide is envisaged (page 3, lines 7 and 8). Both parties and the opposition division regarded D1 and D2 as appropriate starting points for assessing inventive step.

- 5.1 D1 discloses a catalyst having a specific activity of at least about 83 grams of vinyl acetate per gram of precious metal per hour measured at 150°C, consisting essentially of (1), a catalyst support having a particle diameter of from about 3 to about 7 mm and a pore volume of from about 0.2 to about 1.5 ml/g, a 10% by weight water suspension of the catalyst support having a pH of from about 3.0 to about 9.0; (2) a palladium-gold alloy distributed in a surface layer of the catalyst support, the surface layer extending less than about 0.5 mm from the surface of the support, the palladium in the alloy being present in an amount of from about 1.5 to about 5.0 grams per litre of catalyst, and the gold being present in an amount of from about 0.5 to about 2.25 grams per liter of catalyst, and (3) from about 5 to about 60 grams per liter of catalyst of alkali metal acetate (column 3, lines 52 to 68). The quantity of the different catalyst components is such, that the amount of gold present in the catalyst is from 9 to 60% of the combined amount of palladium and gold (column 5, lines 26 to 29). From the highest percentage it can be derived that the maximum ratio of gold to palladium is $0.60 : 0.40 = 1.5$.

- 5.1.1 According to Example III, a surface impregnated catalyst is prepared by using a silica support in the form of 5 mm diameter spheres having a density of 540 g/l and a pore volume of 0.68 ml per gram

(Example III, in connection with Example I, column 6, lines 59 to 62). The support is impregnated with HAuCl_4 , Na_2PdCl_4 and an aqueous solution of potassium acetate having a potassium acetate content of 30 gram per litre catalyst. The obtained catalyst contains 5.06 g/l of palladium, 2.10 g/l of gold, 5.2% by weight of potassium acetate and produces 610 g (corrected by the appellant to read 661 g, which corrected value is indicated in the following; see patent in suit, page 2, lines 38 to 40) vinyl acetate per litre catalyst at 150°C . The weight ratio of gold to palladium in that catalyst is consequently about 0.42 (2.10 : 5.06).

5.1.2 According to the prior art cited in D1, which also makes reference to D2 in form of its Japanese priority document No. 73713/74 (see column 2, lines 67 to column 3, lines 33), the space time yield of known surface impregnated catalysts is low. Thus, D1 aims at an increase in activity to approximately 500 g of vinyl acetate per litre per hour at 150°C (column 4, lines 53 to 56).

5.2 D2 discloses a method of producing vinyl acetate, which comprises reacting ethylene, oxygen and acetic acid, in vapor phase, in the presence of a catalyst comprising a particulate, porous carrier, at least predominantly comprised of a member selected from the group consisting of silica, alumina, and mixtures thereof supporting 0.3 to 3.0 percent by weight of palladium metal, 0.0157 to 1.8 percent by weight of gold metal, and 1 to 30 percent by weight of an alkali metal acetate, all based on the weight of said carrier, and wherein and on said carrier at least 90 percent by weight of each of the supported palladium and gold is

distributed proximate to the external surfaces of the carrier particles, extending to depths no greater than about 30% of particles radius as measured from the exterior surface to the center of such particles, said porous carrier has a particle radius 1.0 to 5.0 millimeters, a pore volume of 0.1 to 2.0 cubic centimeters per gram and an internal surface area of 10 to 350 square meters per gram (claim 1).

5.2.1 In the catalysts of Example 2, the proportions of palladium and gold are varied so as to provide *inter alia* a weight ratio of gold to palladium of 0.67:1 and 1:1 (Table 1, samples 4-4 and 4-5). Since the catalysts according to Example 2 have a specific weight of 572 g/l, the catalyst samples 4-4 and 4-5 have a palladium content of 5 to 6 g/l and a gold content of 3.4 to 9.0 g/l. Thus, samples 4-4 and 4-5 also provide a palladium and gold content being within the range specified in claim 1.

5.2.2 According to D2, when use is made of the known catalyst systems comprising a porous carrier impregnated with palladium and/or gold, the metal components deposited at or about the support interiors or central regions do not contribute significantly to the reaction mechanism, since the reactants are scarcely able to diffuse into the central or inner regions of the porous network of the catalyst and hence, the reaction occurs substantially only at the outermost or surface regions of the catalyst. Therefore, the catalyst components in large part do not contribute to the reaction scheme, resulting in a reduction in catalytic efficiency per unit weight of the catalyst components. Furthermore, the use of a highly active catalyst at times gives rise

to side reactions and, therefore, leads to a reduced selectivity for the contemplated reaction product. On the other hand, a catalyst wherein the palladium metal and/or gold metal has not substantially penetrated into the carrier, but rather bears most of the metallic component deposited on the carrier surface only, displays a limited catalyst life, and does not permit the production of vinyl acetate in high yield (column 1, lines 25 to 50).

5.2.3 Thus, D2 aims to produce a catalyst in the production of vinyl acetate, wherein most of the catalyst components are deposited at the exterior surface of the carrier, or wherein the same have penetrated far into the central regions of the carrier (column 1, lines 53 to 60).

5.3 Both prior art documents relate to shell impregnated catalyst, comprising gold, palladium and potassium acetate for use in the production of vinyl acetate. Whilst D1 aims at improving the catalyst activity to achieve a high productivity, D2 relates more to the reduction of side reactions and the improvement of selectivity and catalyst life (see D2, column 1, lines 38 to 50).

5.4 The appellant has not presented any arguments that the catalyst support (feature(1)) or the distribution of the palladium and gold in the outermost layer (feature (2)) provides any further distinction of the claimed subject-matter over D1 or D2, and contributes to an inventive step. Thus, the claimed catalysts differ from those of D1 only by the amount of gold, since the highest amount of gold in D1 is 2.25 g/l and the lowest

gold amount claimed is 2.3 g/l (calculated on the basis of the palladium content of 3.9 g/l and the ratio of gold to palladium of 0.60). The claimed catalysts differ from those of D2 only by the specified weight percentage of the potassium acetate. Thus, both documents disclose catalysts which are structurally close to those claimed in the patent in suit. Since D1, in comparison with D2 relates more closely to the improvement of the productivity also aimed at in the patent in suit and furthermore has been used to formulate the problem in the patent in suit, it is appropriate to use D1 as starting point for assessing inventive step (Case Law of the Boards of Appeal of the European Patent Office, 4th Edition 2001, I.D.3.1 and I.D.4.3).

Problem and solution

5.5 In the examples of the patent in suit, the productivity and selectivity of the claimed catalysts were measured after 20 hrs for different amounts and weight ratios of gold and palladium and at different oxygen conversions. Table 1 comprises 23 tests, in which catalyst compositions within and outside the composition of claim 1 were used. If the catalyst compositions in Table 1 are numbered from the top to the bottom of the list, among the 23 samples only the samples 9, 12, 13, 17 and 22 fulfil the required catalyst composition of claim 1, while the remaining catalysts compositions do not. The measured productivity for the catalysts, which fall under the present claim 1, is from 693 to 993 STY when using an oxygen conversion between 16 and 35.7%. Since in Table 2 of the patent in suit the ratio of gold to palladium is outside the claimed range, none of

these samples falls under the claimed subject-matter. However, catalysts C, D, and E shown in the appellant's test report filed with letter of 3 April 2001 meet the required composition and provide a productivity of 970 to 1038 STY when using an oxygen conversion of 23 to 40.1%.

5.5.1 The respondent argued that productivity values above 661 STY did not provide any distinction over the cited prior art and thus could be ignored when assessing inventive step. Furthermore, such a high productivity could also be achieved by using catalysts having a weight ratio of gold and palladium outside the claimed range so that no improvement over the prior art had been shown.

5.5.2 There are several results in Table 1 which indeed show that outside the claimed composition range a higher activity than 661 STY can also be achieved (see samples No. 1, 6 to 8, 10, 11, 14 to 16, 19, 20 and 23). However, these samples have not been marked as comparative and the respondent has not shown that these samples represent a reproduction of prior art catalysts.

Moreover, the respondent has not filed any experimental evidence of its own, to show that the catalyst composition according to claim 1 does not provide a productivity above 661 STY or that any of the prior art catalysts do that. The onus of proof in this respect lies, however, with the opponent (respondent) (T 219/83, OJ EPO 1986, 211).

5.5.3 From the above it follows, that all catalyst compositions which fulfil the requirements of claim 1

also show a productivity above 661 STY under the specified process conditions, when using usual oxygen conversions. Hence, the problem solved over D1 may be seen in providing shell impregnated catalyst for use in the production of vinyl acetate having a productivity greater than 661 g of vinyl acetate per hour per litre of catalyst at 150°C by maintaining a high selectivity (page 2, lines 41 to 43 and 53 to 56).

- 5.5.4 That problem is solved by a catalyst according to present claim 1 comprising from 3.5 to 9.5% by weight of potassium acetate and having a gold to palladium weight ratio within the range of 0.60 to 1.25 and, wherein the palladium content is greater than 3.9 grams per litre of catalyst.

Obviousness

- 5.6 It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.
- 5.6.1 According to Examples I to III of D1, catalysts have been prepared by using the same catalyst support, in the surface layer of which different amounts of palladium and gold are precipitated. In these examples, the gold to palladium weight ratio is held at about 0.42 to 0.45 (patent in suit, page 2, line 36). The reported productivity is enhanced from 275 to 610 (corrected 661) g/l.hr with increasing palladium content, since palladium is the active catalyst metal and gold is the catalyst promoter, so that the specific activity can also be based on the amount of palladium present (column 4, lines 64 to 67). Thus, D1 already

provides an indication that a higher productivity can be achieved by simply increasing the amount of palladium and that the best productivity can be achieved at the highest possible amount of palladium used in D1.

- 5.6.2 Furthermore, D1 refers to D2 as a relevant prior art document, according to which "by depositing the necessary amount of palladium and gold to be employed in the catalyst which will then adhere to the surface layer of the catalyst support" ... "catalytic efficiency as compared to conventional catalysts" is improved (D1, column 3, lines 20 to 25). In Example 2 of D2, catalysts are prepared in which only the amount of gold is changed (0.03 to 2.3% by weight), whilst the amount of palladium is constant (1 percent by weight). The weight ratio varied from 0.25:1 to 1.5:1 by using 0.03:1 and 2.3:1 as comparison.

In Table II the productivity measured at 110°C and the percent drop in activity after 90 days of reaction are indicated. From these results it can be seen that the highest production rate (158 and 150 g/l/hr) and the lowest percent drop in activity (0%) is achieved with samples 4-4 and 4-5 having a weight ratio of gold to palladium of 0.67:1 and 1.0:1, respectively, whilst weight ratios higher and lower than in samples 4-4 and 4-5 provide a lower production rate and a higher drop in activity.

Since the production rate in those samples is measured at a low reaction temperature of 110°C and since according to general technical knowledge a higher temperature of 150°C as used in D1 will enhance the

reaction rate, the increase in the productivity of the catalyst at higher reaction temperatures would be considerably pronounced.

5.6.3 From the above it follows that the weight ratio of gold to palladium is critical for catalyst activity and that weight ratios being within the claimed range have an enhanced effect on productivity. Thus, the skilled person would have an incentive to increase in Example III of D1 (already providing a productivity of 661 STY) the weight ratio of gold to palladium according to D2, by holding the content of palladium at a high level, in order to further improve the productivity above 661 STY.

5.6.4 Thus, the subject-matter of claim 1 is rendered obvious when following the teaching of D1 and D2 in combination and does not involve an inventive step.

5.6.5 The appellant argued that there is nothing in D2 that suggests the criticality of the gold to palladium ratio over a wide range of palladium loadings (see also patent in suit page 2, lines 52 and 53).

However, in D2 the exemplified ratios of gold to palladium providing the best results fall within the claimed range of 0.6 to 1.25. Those ratios are not only illustrated by samples 4.4 and 4.5 but also by Examples 1 and 3 of D2, in which catalysts containing 1.5% by weight of gold and 2.2% by weight of palladium corresponding to a ratio of gold to palladium of 0.68 are used. Since 2.2 % by weight of palladium metal used in Example 1 corresponds to about 10.4 to 13.2 g/l, a preferred ratio of gold to palladium (0.67 and 0.68) is

exemplified for a wide range of palladium amounts (about 5 to 13.2 g/l). Thus, the appellant's argument is not convincing.

- 5.6.6 The appellant furthermore argued that Figure 1 of the patent in suit showed an maximum effect in activity, when using a gold/palladium ratio as claimed.

Figure 1 is however based on data calculated from mathematical models which best fit with the experimental results of Table 1 and the variation in catalyst composition and oxygen conversion (page 5, lines 29 to 56). Thus, Figure 1 only provides a mathematical illustration showing that within the claimed ratio of gold to palladium a maximum productivity may be achieved. The relation between productivity dependent on the gold to palladium weight ratio similar to Figure 1 can however already be derived from experimental data shown in Table II of D2 so that Figure 1 of the patent in suit cannot contribute to an inventive step.

- 5.6.7 From the above it follows that claim 1 of the main request does not involve an inventive step. Since D1 and D2 both relate to processes, which comprise reacting ethylene with acetic acid in the presence of an oxygen containing gas at a temperature of 100 to 200°C (see D1, examples; D2 examples), the considerations as outlined under point 6.6.3 above apply *mutatis mutandis* to process claim 9. Thus, also process claim 9 is not inventive either.

- 5.6.8 Since the main request as a whole is not allowable, it is not necessary to decide, whether or not the independent claims 6 and 14 involve an inventive step.
- 5.6.9 In view of the above, the respondent's objections to the test results of the patent in suit (Figures 1 to 4) are not relevant to this decision and it is not necessary to decide on the question, whether or not the late filed submissions of the respondent dated 5 March 2004 should be admitted. Hence, the appellant's objection under Article 113(2) EPC is without effect.

Auxiliary requests

6. Claims 1 and 9 of both auxiliary requests have been amended by the feature "at an oxygen conversion of 30 %". The amendment only provides a clarification under which oxygen conversion the test results best fit with the mathematical equation 3) specified in the patent in suit (page 5, lines 52 to 56). Thus, that amendment does not change the composition of the catalyst. Since the appellant has not provided any new argument, which may support an inventive step based on that amendment and since the board sees no additional argument either, it concludes that the same considerations as outlined with respect to claims 1 and 9 of the main request must apply *mutatis mutandis* for the claims 1 and 9 of the auxiliary requests (points 6.6.3 and 6.6.7 above).
- 6.1 Consequently, the claimed subject-matter of auxiliary requests 1 and 2 are not inventive either.

6.2 From, the above it follows that none of the requests meets the requirements of Article 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

C. Eickhoff

R. Teschemacher