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D E C I S I O N
of 24 February 1994

Case Number: T 0658/90 - 3.3.2

Application Number: 85307380.7

Publication Number: 0218766

IPC: B01J 35/02

Language of the proceedings: EN

Title of invention:
Silver catalyst for production of ethylene oxide and method for manufacture thereof

Patentee:
Nippon Shokubai Kagaku Kogyo Co., Ltd.

Opponent:
Bayer AG, Leverkusen

Headword:
Saddles/NIPPON

Relevant legal norms:
EPC Art. 56

Keyword:
"Inventive step (yes) - Claimed features responsible for the improvement - Comparative examples- Contrary assertions of the parties - Benefit of doubt"

Decisions cited:
T 0219/83

Headnote/Catchword:
-

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Case Number: T 066[illegible] - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 24 February 1994

Appellant: (Opponent)
Bayer AG, Leverkusen
Konzernverwaltung RP
Patente Konzern
Bayerwerk
D-51368 Leverkusen (DE)

Representative:
-

Respondent: (Proprietor of the patent)
Nippon Shokubai Kagaku Kogyo Co., Ltd.
1, 5-chome, Koraibashi
Higashi-ku
Osaka-shi, Osaka-fu 541 (JP)

Representative:
Serjeants
25, The Crescent
King Street
Leicester, LE1 6RX (GB)

Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office dated 19 June 1990 concerning maintenance of European patent No. 0 218 766 in amended form.**

Composition of the Board:

Chairman: P.A.M. Lançon
Members: M.M. Eberhard
S.C. Perryman

Summary of Facts and Submissions

I. European patent No. 0 218 766 based on application No. 85 307 380.7 was granted on the basis of twelve claims.

II. The Appellant (Opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty and lack of inventive step. Of the documents cited during the opposition procedure only the following were relied upon at the appeal stage:

(2) DE-A-1 936 233
(3) Römpp, Chemie Lexikon, Vol. II (F-L), 1966, columns 2231/2232 and 2957
(4) Ullmanns Encyklopädie der technischen Chemie, 4th edition, Vol. 2, 1972, page 529
(5) Dissertation Böhring, Darmstadt, 1984
(6) Catal. Rev.-Sci. Eng. 23, 163-165, 1981, page 176
(7) Applied Industrial Catalysis, Vol. 1, 1983, pages 221-226.

III. In response to the notice of opposition, the Respondent (Patentee) filed an amended set of claims on 4 December 1989. Claim 1 reads as follows:

"A silver catalyst for the production of ethylene oxide by catalytic vapor phase oxidation of ethylene with molecular oxygen in the presence of a halogenated inhibitor, having silver and at least one accelerator selected from the group consisting of alkali metals and alkali metal compounds deposited on a porous inorganic refractory carrier, characterised in that the carrier is a shaped body of the porous inorganic refractory material in the shape of Intalox saddles or Berl saddles and has a specific pore volume in the range of 0.06 to

1.0 cc/g, an outer peripheral length (A) in the range of 3 to 70 mm, an inner peripheral length (C) in the range of 1.5 to 68 mm, a thickness (W) in the range of 0.1 to 4 mm, an outside diameter (D) in the range of 0.5 to 20 mm, and a length (E) in the range of 0.5 to 65 mm." (Obvious errors in this text have been corrected by the Board in accordance with the granted request for correction: see points VIII and 3 below.)

IV. By an interlocutory decision the Opposition Division decided to maintain the patent in the amended form as filed on 4 December 1989. In this decision the Opposition Division considered that document (2) did not suggest that the shape of the carrier might have any particular influence over the selectivity to ethylene oxide. The possible use of saddles and rings was merely mentioned in (2) to solve different problems, such as the pressure loss. In this respect, saddles and rings were regarded as equivalent. In the Opposition Division's view the teaching of document (2) could not make obvious the claimed catalyst since the Patentee had demonstrated in the comparative examples that saddles surprisingly afforded improved selectivity. The claimed catalyst was also not made obvious by a possible combination of (2) with the additional documents cited during the opposition procedure as none of them was concerned with the problems of the invention.

V. The Appellant lodged an appeal against this decision. In a communication pursuant to Article 110(2) EPC, the parties were informed that either US-A-4 248 740, document (8), or US-A-4 389 338, document (9), both cited in the patent in suit, seemed to represent the closest prior art and that it was questionable whether the slight improvement in selectivity shown in the examples was attributable to the particular shapes defined in Claim 1. Reference was also made to Römpps

Chemie Lexikon, 1973, pages 1198 to 1200, document (3a).
Oral proceedings were held on 24 February 1994.

VI. The Appellant's arguments submitted in writing and at the oral proceedings can be summarised as follows:

The comparison of Example 1 with control Example 2 and of Examples 4 and 5 with control Example 8 shows that the problem of minimising the pressure loss in the catalyst bed with respect to the known supports has not been solved. Furthermore, one cannot agree with the Respondent's argument that the higher selectivity in the examples 1, 4 and 5 over the control Examples 2 and 8 is attributable to the shape of the carrier. It is well known to the skilled person that the chemical composition of the carrier and of the impregnated active components as well as the conditions of preparation of the catalyst affect the selectivity to ethylene oxide much more than does the shape of the carrier. The carriers of example 1 and of control example 2 exhibit a different porosity and a different ratio of apparent surface/apparent volume and were prepared using different operating conditions, for example as regards the concentration of the impregnation solution. It is well known that the distribution of the silver and promoters on the support depends upon such conditions. In control Example 8 the activation temperature was not the same as in the examples 4 and 5. Furthermore, the catalysts were tested at different reaction temperatures. These differences explain the different selectivities obtained in the compared examples and do not make possible any comparison concerning the shape of the support. In the present case, the submission of comparative examples to support these arguments is not necessary since it is clearly a *prima facie* case.

According to document (2) not only pellets or rings can be used as catalyst support for the production of ethylene oxide but also saddles, and engineering factors such as the pressure drop may influence the choice of the geometrical configuration. Saddles are mentioned in this context. Furthermore, the properties of Intalox or Berl saddles are disclosed in the documents (3) or (3a) and (4) as well as their use as catalyst support. The skilled person was aware of the fact that these saddles have to be employed in order to minimise the pressure drop. As regards the selectivity it is noted that if the pressure drop decreases, then the contact time of the gases with the catalyst is shorter and, thus, the selectivity to ethylene oxide increases as an inherent effect of the lower pressure drop. Therefore, in view of the teaching of documents (2), (3) and (4) the positive effects of the saddles were known to the skilled person and it was obvious to replace the rings of the catalysts according to document (8) or (9) by Berl or Intalox saddles. Further, the skilled person could find the relationship between the size of carriers and its influence on the pressure drop in the documents (5), (6) and (7).

VII. The Respondent put forward i.a. the following arguments:

There is no evidence that the slight improvement in selectivity over the comparative catalysts is not due to the shapes and dimensions set out in Claim 1 and so it should be accepted without question that the given shapes and dimensions are responsible for the improvement.

Between Example 1 and control Example 2 not only the shape of the support had been changed but also the pore volume and the concentration of the impregnation solution. However, both concentrations of the final

catalyst are identical. Catalytic activities depend mainly on the concentration of the final catalyst rather than that of the solution to be impregnated. Regarding pore volumes, in this technical field, the activity of a catalyst is due to the surface area of the support rather than its pore volume. Thus, the inventors selected the supports in order mainly to harmonise surface area rather than pore volume. The inventors believe that the difference between their invention and the controls lies in the shape of the support.

The improvement in selectivity is admittedly small, however even a small improvement is worthwhile in this technical field. Document (2) does not suggest that the selectivity can be improved by using saddles instead of pellets or rings. The replacement of rings by saddles gave an unexpected effect which the Opposition Division rightly held to be inventive. In this respect reference is also made to document (5) which points out that the development of the catalyst for the production of ethylene oxide is so far advanced that the high conversions and selectivities can hardly be further increased.

VIII. The Appellant requested that the decision under appeal be set aside and the patent be revoked. The Respondent requested that Claim 1 submitted on 4 December 1989 be amended by correction of "ehtylene" in line 2 to "ethylene", of "siler" in line 3 to "silver" and of "cc" in line 9 to "cm^3", that the appeal be dismissed and that the decision under appeal be set aside and the patent be maintained on the basis of the claims submitted on 4 December 1989 with Claim 1 corrected as above, and a description to be adapted.

Reasons for the Decision

1. The appeal is admissible.

2. The amendments in the claims meet the requirements of Article 123(2) and (3).

3. The request for correction pursuant to Rule 88 in Claim 1 is allowable since it is immediately apparent that the terms "ehtylene", "siler" and "cc" are erroneous and that the only possible correction the skilled person would think about when reading the patent in suit is the following: "ethylene", "silver" and "cm^3" respectively.

4. None of the cited documents discloses a silver catalyst for the production of ethylene oxide containing an alkali metal promoter and having a carrier in the shape of Intalox saddles or Berl saddles with the dimensions defined in Claim 1. Therefore the catalyst according to Claim 1 is novel. This not being in dispute there is no need to give further details.

5. The problem underlying the invention was defined in the patent in suit taking the processes and catalysts of document (8) or document (9) as the starting point in the prior art. These documents disclose silver catalysts for the production of ethylene oxide by catalytic vapour phase oxidation of ethylene with molecular oxygen in the presence of a halogenated inhibitor, comprising silver and at least one accelerator selected from the group consisting of alkali metals and alkali metal compounds deposited on a porous inorganic refractory carrier, i.e. silver catalysts as defined in the preamble of Claim 1. The silver catalyst of document (8) is prepared by a method including the succession of steps as defined in

the amended Claim 7 of the patent in suit whereas the method of manufacture of the silver catalyst according to document (8) comprises the succession of steps defined in the amended Claim 8. The carrier used in the silver catalysts of document (9) is in the form of pellets or rings, the average equivalent diameter of which lies with the range of 3 to 20 mm, preferably 3 to 10 mm (cf. column 4, lines 46-50).

The catalyst of document (9), contrary to the catalyst of document (2) considered by the Opposition Division as closest prior art, contains the same promoter as the claimed catalyst. Furthermore, it is used for the production of ethylene oxide in the presence of a halogenated inhibitor in opposition to the catalyst of document (2). Therefore, document (9) is considered by the Board to represent the closest prior art.

5.1 According to the patent in suit, the catalysts of documents (8) and (9) are not fully satisfactory in selectivity (cf. page 2, lines 35-49). Starting from (9) as closest prior art, the problem underlying the patent in suit can be seen in providing a silver catalyst for the production of ethylene oxide in the presence of a halogenated inhibitor, which catalyst exhibits an improved selectivity to ethylene oxide.

It is proposed to solve this problem by using a carrier in the shape of Intalox saddles or Berl saddles having the dimensions recited in Claim 1. Furthermore, the carrier has a specific pore volume in the range of 0.06 to 1.0 cm^3/g. The pore volume, which is not mentioned in the closest prior art, was however not alleged to be responsible for the improvement in selectivity.

It is indicated in the patent in suit that the claimed catalysts suffer only minimal pressure loss in the

catalyst bed. However, as pointed out by the Appellant in the appeal statement, the comparative examples of the patent in suit show that the pressure loss obtained with a carrier in the form of Berl saddles or Intalox saddles is about the same as the pressure loss of a catalyst comprising Raschig rings (cf. examples 4 and 5 compared to control example 8 or example 1 compared to control example 2). Accordingly, an improvement in the pressure loss cannot be taken into consideration for the definition of the problem.

5.2 The question arises whether the problem defined above has been solved by the features recited in Claim 1. The Appellant has not contested that a slight improvement in selectivity to ethylene oxide is achieved with the catalysts of the invention over those of the prior art. However, the Appellant has argued that it could not be derived from these comparative examples, in particular from example 1 and control example 2 on the one hand and from examples 4 and 5 and control example 8 on the other hand, that this slight improvement was attributable to the different shape of the carrier since not only the shape has been changed but also other parameters having an influence on the selectivity.

The catalysts of table 2 were prepared by the method of the closest prior art. The carriers of examples 4 and 5, i.e. Intalox saddles and Berl saddles exhibit the same pore volume, BET surface area and ratio of apparent surface area/apparent volume as the Raschig rings of the comparative control example 8, however their packed specific gravities are higher. The silver contents of the corresponding catalysts have accordingly been adapted so that the three tested catalysts contain similar amounts of silver in the packed bed. These catalysts were prepared by the same method and under the same operating conditions except for the silver oxalate

concentration in the solution to be impregnated, which was slightly lower in the examples 4 and 5 than in the control example 8, and for the temperature of the final treatment at high temperature (550-950°C) in nitrogen. The latter was performed at 630°C in examples 4 and 5 and at 640°C in control example 8.

Although different concentrations of the silver salt in the impregnation solution may have an influence on the distribution of the silver on the surface of the carrier and therefore on the selectivity, as pointed out by the Appellant, the differences of concentration in the compared examples are not so great that it could be *prima facie* concluded that they affect the selectivity. Likewise, the difference of 10°C between temperatures lying in the order of 630°C is relatively small so that no *prima facie* conclusion can be drawn on its possible influence upon the selectivity. It is indeed derivable from document (9) that the final treatment at a temperature between 550-950°C improves the selectivity to ethylene oxide in comparison with a catalyst which was not submitted to this heat treatment, however it does not follow therefrom that the difference of 10°C in the temperature of this heat treatment would be responsible for the improvement in selectivity. In this context it is further observed that the catalysts of examples 9 and 10 of table 3 (Intalox saddles and Berl saddles) exhibit an improved selectivity over the control example 13 (Raschig rings) although a heat treatment at high temperature (630 or 640°C) was not performed.

In connection with the different packed specific gravities, it should be noted that if the Intalox saddles have the same packed specific gravity as the Raschig rings, the improvement in selectivity over the catalyst in the form of Raschig rings is still achieved:

cf. table 1, example 1 and control example 2. The pore volume in control example 2 is indeed lower than in example 1 (0.27 cm^2/g instead of 0.32), however the Appellant has not shown that this lower pore volume would impair the selectivity of the catalyst of control example 2, which on the other hand exhibits a greater ratio of apparent surface area/apparent volume.

As regards the different reaction temperatures which the Appellant has made the basis of an objection at the oral proceedings, it is observed that these temperatures are those which give the **same** ethylene **conversion** of 13%. No evidence has been brought to show that the lower selectivity of the control catalyst 8 results from the difference of 7°C in the reaction temperatures (239°C for control example 8 and 232°C for example 5) and not from the different shapes.

The Appellant's assumption that the slight improvement in selectivity results from parameters other than the different shapes of the carrier is not in agreement with the statement in the patent in suit that the replacement of the Raschig rings by Intalox saddles or Berl saddles leads to a catalyst exhibiting a heretofore unattainable high selectivity. Furthermore, as indicated above, the Appellant has brought no evidence that the discussed other differences are so great that they are responsible for the improvement in selectivity. In such a situation where the Opponent makes an assertion which is contrary to the teaching of the patent in suit without bringing any evidence to substantiate this assertion, which is contested by the Patentee, the Board has to decide in favour of the party not having the burden of proof, i.e. in the present case the Respondent (cf. T 219/83, OJ EPO, 1986). The Appellant's argument that parameters which might affect the selectivity have been changed in addition to the shape of the carrier is certainly not

sufficient to reverse the burden of proof. In these circumstances, the Board concludes that in the absence of evidence to the contrary, it is credible in view of the examples and control examples of the patent in suit and of the statements at page 3, lines 26 to 31, and page 4, lines 2 to 9, that the technical problem has been solved by the shape and dimensions of the carrier defined in Claim 1.

5.3 Neither document (9) nor document (8), which are both concerned with the problem of improving the selectivity of the silver catalyst to ethylene oxide, suggests that this selectivity might be improved by a catalyst in the shape of saddles.

5.4 Document (2) aims at providing a silver catalyst for the partial oxidation of ethylene to ethylene oxide, which avoids the necessity of using a halogenated inhibitor and permits improved productivity and/or a lower operating temperature. This is achieved by selecting a carrier having an apparent porosity greater than 30% and pore diameters as well as an average pore diameter which fall within well defined ranges (cf. pages 4 and 5, Claim 1). According to (2) these features give the optimum combination of ethylene oxide selectivity and catalyst activity (see page 7, first paragraph and page 15, table II). Thus, the improvement in selectivity is not attributed to the shape of the support but to the particular pore dimensions of the carrier.

Document (2) further teaches that the carrier may take almost any geometrical configuration, the configuration being suitably cylindrical, spheroidal or spherical from the standpoint of simplicity (cf. page 7, last paragraph). Spherical pellets are used in the examples. According to page 8, first paragraph, engineering factors such as the ability to pack uniformly, the

mechanical strength, the pressure drop and the stability may influence the choice of the configuration. Accordingly the use of more complicated shapes, such as saddles or rings, may be necessary. This teaching does not suggest that the selectivity of the catalyst might be improved by using these more complicated configurations, let alone that saddles might lead to an improved selectivity over rings. Therefore, the skilled person would not be prompted in view of document (2) to replace the rings by saddles in the catalyst of document (9) in order to improve the selectivity to ethylene oxide.

5.5 Documents (3) and (3a) relate to packing materials which are used in particular for distillation. Different kinds of packing materials such as spheres, Raschig rings, Pall rings, Berl saddles, Intalox saddles, Torus saddles, Super saddles, Novolax saddles are disclosed, as well as the dimensions in which they are available. According to (3a) Berl saddles exhibit the most perfect mathematical configuration with the correspondingly favourable effect on the performance in the distillation and rectification of mixture of compounds with close boiling ranges. Intalox saddles are said to have a lower resistance to passage than the Berl saddles (cf. page 1199). These packing materials can be used in the field of catalysis and catalysts can be deposited thereon (cf. (3) page 2231). In document (4) which exclusively concerns distillation, there are also references to Raschig rings, Pall rings, Berl saddles, Intalox saddles and Torus saddles.

These documents do not contain a reference to the catalytic production of ethylene oxide nor do they deal with the problem of improving the selectivity of the silver catalysts used in the said reaction. Therefore, they would be of no assistance to the skilled person

confronted with the problem of how to improve the selectivity of the catalysts of (9), even in combination with the teaching of document (2), since the latter indeed mentions the possible use of saddles or rings instead of spherical pellets but not for improving the selectivity to ethylene oxide.

5.6 The Appellant alleged for the first time at the oral proceedings that a decrease of the pressure drop results in a shorter contact time of the gases with the catalyst and thus in an increase of the selectivity. The Respondent's representative pointed out that this relationship was not derivable from the cited documents. The Board also has doubts as regards the alleged predictable effect on the selectivity since in the case of the decrease in pressure drop being due to a different geometrical configuration, in particular a more sophisticated shape such as saddles, not only the gas velocity is changed but also other parameters which depend upon the shape of the carrier and may have an influence on the selectivity. Furthermore, the Appellant did not consider the effect on the conversion. It should be noted in this respect that the improvement of selectivity is achieved at the same ethylene conversion in the examples of the patent in suit. In these circumstances and taking into account that the Opponent's allegation was not supported by any evidence, the Board has to decide in favour of the Patentee on whom the burden of proof does not rest.

5.7 Document (5) relates to the influence of the catalyst shape and catalyst arrangement on the conditions governing the reaction in tubular reactors. Reference is made to reactions such as the production of phthalic anhydride, ethylene oxide, maleic anhydride or methanol in fixed-bed tubular reactors. The part of this article submitted (i.e. the introduction) contains only general

considerations. There is disclosed that an increase of the flow velocity improves the dissipation of heat produced by the exothermic reaction, that a reduction of the pressure drop in the bed would be of interest in order to make possible an increase of the flow velocity without raising the energy costs and that, consequently, catalyst shapes and arrangements which are more favourable to the flow have been discussed from time to time (cf. page 2, first paragraph). Although the reactor-bed heat transfer is a factor which may affect the selectivity, the latter is not discussed at all in (5) let alone other factors which depend upon the carrier shape and may also influence the selectivity. Therefore, the general teaching of (5) considered in combination with documents (2), (3), (3a) and (4) would not give the skilled person an incentive to replace the rings of the catalyst of (9) by Berl saddles or Intalox saddles as defined in Claim 1 in order to improve the selectivity to ethylene oxide.

The two remaining documents (6) and (7) which, like (5), were cited after the opposition period are less relevant than the preceding documents. They only disclose spheres, cylinders or rings as catalyst shapes and give their diameter as well as the reactor diameter. Selectively is only mentioned in (7) in connection with the use of a chlorinated inhibitor (cf. page 223). Therefore the teaching of these documents cannot change the preceding findings.

5.8 It follows from the above that it was not obvious to arrive at the claimed catalyst in view of the cited prior art. Therefore, the subject-matter of Claim 1 is considered to meet the requirements of inventive step set out in Article 52(1) and 56 EPC.

6. The methods for the preparation of this catalyst as defined in Claims 7 and 8 derive their patentability from that of the product claim. Claim 1 being allowable, the same applies to the dependent Claims 2 to 6 whose patentability is supported by that of Claim 1.

Order

For these reasons, it is decided that:

1. The appeal is dismissed.

2. The request for correction under Rule 88 EPC is granted.

3. The decision under appeal is set aside and the case is remitted to the first instance with the order to maintain the patent on the basis of the claims submitted on 4 December 1989 with Claim 1 amended by correction of "ehtylene" in line 2 to "ethylene", of "siler" in line 3 to "silver" and of "cc" in line 9 to "cm^3", and a description to be adapted.

The Registrar: The Chairman:

P. Martorana P.A.M. Lançon

H.B.
M.P.