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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Production of alkylene oxides and catalysts therefor

Klassifikation / Classification / Classement : B01J 23/50

ENTSCHEIDUNG / DECISION

vom / of / du 10 October 1990

Anmelder / Applicant / Demandeur : Imperial Chemical Industries PLC

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence : Cylindrical catalysts/ICI

EPÜ / EPC / CBE Art. 56

Schlagwort / Keyword / Mot clé : "Reassessment of the state of the art - Redefinition of the problem"
"Inventive step (affirmed) - non-obvious combination of properties"

Leitsatz / Headnote / Sommaire

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Case Number : T 189/89 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 10 October 1990

Appellant : Imperial Chemical Industries PLC
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Decision under appeal : Decision of Examining Division 033 of the European Patent Office dated 3 October 1988 refusing European patent application No. 85 307 070.4 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : P. Lançon
Members : M. Eberhard
F. Benussi

Summary of Facts and Submissions

I. European patent application 85 307 070.4 (publication No. 0 179 584) was refused by a decision of the Examining Division. The decision was based on Claims 1 to 7 filed on 12 August 1988. Independent Claim 1 reads as follows:

"1. A catalyst for the production of an alkylene oxide, by contacting the corresponding olefine and oxygen with the catalyst, which comprises silver deposited on a porous heat resisting support, in which the catalyst is in the form of substantially cylindrical pellets in which the ratio of the length to the diameter is in the range 2:1 to 1:2, pierced by 3 to 15 substantially longitudinal holes passing completely through the pellets, the total volume of the holes within the pellet being 5 to 25% of the volume of the pellets including the holes, the disposition of the holes across the cross section of the pellet facilitating access of gas to the substance of the pellet."

Independent Claim 5 relates to a process of producing an alkylene oxide which is carried out in the presence of this catalyst.

II. The ground for refusal was that the subject-matter of Claim 1 did not involve an inventive step in the light of the disclosure in DE-A-1 920 976, document (1), and in US-A-2 408 164, document (2). It was held that in view of the teaching about the catalyst shape in (1), the skilled person was directly invited to give special consideration to the influence of the catalyst shape in the alkylene oxide production. Therefore he would have considered (2) which dealt, under a broad aspect, with the shaping of catalysts in order to render them more efficient. According to the decision, the explicit reference to the

increased contact surface in this document was a clear advice for the skilled person to try the multiholed cylinders when his task was to improve the catalyst performance. In the Examining Division's opinion the selection of the size ratio and of the number of holes mentioned in Claim 1 was obvious in view of example 1 and Figures 1H and 1M of (2). Designing the holes so that the total volume thereof is 5-25% of the volume of the pellets was regarded as an obvious result of ordinary design. It was further held that the process of Claim 5 lacked inventive step since it was known per se from EP-A-3 642, document (3), and differed therefrom only in the shape of the catalyst which was itself obvious.

III. The Applicant lodged an appeal against this decision with payment of the prescribed fee. A Statement of Grounds was filed in due time.

IV. In a Board's communication it was questioned about a document illustrating the prior art referred to in general terms at page 1 of the application and about the silver content of the tested catalysts. The Appellant's attention was further drawn to FR-A-1 137 622, document (4). In reply thereto the Appellant cited new documents, in particular US-A-4 471 071, document (5). Oral proceedings were held on 10 October 1990.

V. At the oral proceedings, the Appellant agreed that document (5) represented the closest prior art. He pointed out that the catalysts of (5) were tested without supplying nitric oxide to the process contrary to the tests effected in the refused application. However the comparative examples thereof showed that the improvement of 0.8% in selectivity was attributable to the change of shape since all the other conditions remained unchanged. As regards the silver content of the tested catalysts, the

Appellant submitted porosity measurements of the one hole and seven hole support and calculations of surfaces in these supports. He further stressed that the claimed catalyst exhibited a better selectivity and a slightly higher activity than the known rings (i.e. hollow cylinders) without any adverse consequence on the other properties such as resistance to abrasion, resistance to breakage, pressure drop and packing density. According to the Appellant, the prior art shape had been already designed to expose the optimum contact surface to the gases and in the invention the question of better access of the gases to the material was considered.

In his written submissions and during oral proceedings the Appellant contended amongst others that the technological teaching of (1) was of so vague a nature that the skilled person could not determine from it on what principles he could proceed to devise other shapes. In view of (1) the skilled person would not have expected any advantage in selectivity by changing the catalyst shape. As regards (2) the Appellant argued that its disclosure did not relate to oxidation processes or ethylene oxide production and was very general. He pointed out that the whole paragraph at column 9, lines 60 to 68, was concerned with increasing contact surface, but roughened or corrugated surfaces as well as thin walled pellets or multiholed pellets and even all the pellets forms shown in Figure 1 were proposed. In the Appellant's view the disclosure did not point to any advantage of the multiholed cylinders over the other alternatives. Under these circumstances, the skilled person would not have been encouraged to effect experiments with such supports at the commercial scale. As regards document (4) it was contended that its teaching was not very clear.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims submitted on 12 August 1988.

Reasons for the Decision

1. The appeal is admissible.

2. There are no objections under Article 123(2) to the amended claims since they are supported by Claims 1 to 4 and 6 to 8 as originally filed.

3. The refused patent application relates to a catalyst for the production of an alkylene oxide, in particular ethylene oxide, by contacting the corresponding olefine and oxygen with the catalyst. This latter comprises silver deposited on a porous heat resisting support, for example an alpha-alumina support. Such catalysts are already known, in particular from document (5) in which they are preferably in the form of rings, i.e. in the form of hollow cylindrical pellets (cf. column 1, lines 5 to 10; column 5, lines 23 to 52 and examples). This document, which was cited by the Appellant in reply to the Board's question concerning identification of the prior art referred to in the application, is considered to be the closest state of the art (see decision T 248/85, OJ EPO 1986, 261).

3.1 In the light of this closest prior art, the technical problem stated in the application must be defined more precisely and can be seen in providing a catalyst which exhibits an improved selectivity and a slightly improved activity without increasing the pressure drop in the catalyst bed.

According to Claim 1, this problem is solved by a catalyst in the form of substantially cylindrical pellets pierced by 3 to 15 longitudinal holes passing completely therethrough, the total volume of the holes being 5 to 25% of the volume of the pellets. The pellets have a ratio of length to diameter in the range of 2:1 to 1:2 and the disposition of the holes is such that it facilitates access of gas to the substance of the pellets.

3.2 The comparative test reported in the application is effected with a ring shaped catalyst, i.e. the single hole pellets, containing the same promoter as the seven hole catalyst according to the invention. Both catalysts are prepared and tested under identical conditions the only significant difference therebetween lying in their shape. The comparative test shows an improvement of 0.8% in selectivity and a slight improvement in activity with the claimed catalyst and no difference in pressure drop. From the porosity measurements and from the calculations of surfaces submitted during the oral proceedings, it can be concluded that even if the seven hole catalyst retains a higher amount of silver complex in the absence of shaking, the possible differences in Ag content has no significant influence on the outcome of the comparison. Although the comparative test was not carried out with a silver catalyst prepared as described in (5) but with a catalyst differing from that of the invention only by its shape, i.e. by the claimed distinguishing feature, it demonstrates more clearly that the improvement is attributable to the change of shape. Under these circumstances the Board is satisfied that the technical problem is plausibly solved by the multiholed catalysts (see in this context T 35/85 of 16 December 1986, not published in the OJ EPO).

4. After examination of the cited documents, the Board has come to the conclusion that none of them discloses a catalyst for the production of an alkylene oxide, comprising silver deposited on a porous heat resisting support and having the form of multiholed cylinders as defined in Claim 1. Since the issue of novelty has not been raised by the Examining Division, it is not necessary to consider this matter in further detail.

5. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the teaching of the cited documents.

5.1 Document (5) itself teaches that the support material is advantageously used in the form of granules, spheres, rings, pellets or the like (cf. column 5, lines 47 to 51). In all the examples the catalyst has a ring shape. This document deals with the problem of improving the selectivity, the activity and the life of silver catalysts and lowering the dependence of the selectivity on the ethylene conversion. However this improvement is obtained by the selection of a particular amine mixture for the impregnation solution and this document does not suggest either that the shape of the catalyst is critical or that other shapes can improve its selectivity or its activity.

5.2 Document (1) also relates to the partial oxidation of ethylene in the presence of a silver supported catalyst. It teaches the use of a support material with pore dimensions falling within a specific range in order to avoid the necessity of employing a halogenated inhibitor. The resulting catalyst makes possible operation at lower temperatures and/or improvement in productivity (see Claim 1; page 1, first paragraph and whole page 4). This document further discloses in connection with the shape of the support that it may take almost any geometrical

configuration; cylindrical, spheroical and spherical pellets being appropriate from the standpoint of simplicity (cf. page 7, last paragraph). Spherical pellets are used in the example. According to page 8, first paragraph, it is well known that technological factors such as ability to pack uniformly, mechanical strength, pressure drop and stability may influence the choice of configuration and accordingly it can be necessary to use complicated configurations, for example saddles or rings. Therefore, the skilled person is aware that other shapes may be designed to improve the technological factors listed above, however he would not be able to infer from this teaching that the "complicated" configurations referred to in (1) favourably affect the selectivity of the catalyst. The optimum combination of ethylene oxide selectivity and catalyst activity mentioned in the first paragraph of page 7 is not correlated with the shape of the catalyst but with its porosity and its specific pore dimensions. Moreover, the improved selectivity and activity shown in table II of (1) do not result from a change of shape but from different pore diameter ranges and different average pore diameters. Therefore, in the Board's opinion, the skilled person confronted with the problem of improving the selectivity and activity of a ring shaped silver catalyst without impairing the pressure drop would not consider the shape of the catalyst as particularly important and crucial for the selectivity improvement in view of the teaching of (1).

5.3 As emphasised by the Appellant, document (2) does not address either oxidation processes in general or ethylene oxide processes in particular. It relates to catalyst preparation, more precisely to a process for shaping catalysts into suitable forms in order to render them more efficient for use in the various purposes to which they are applied (cf. column 1, lines 1 to 5). Although the

claims are directed to the preparation of Friedel-Crafts catalysts, the description is very general and the process may be applied to any solid material or mixture of materials used in catalysis. Alumina and silica are cited among others (cf. column 5, line 64 to column 6, line 6; column 11 last paragraph). In the Board's opinion the skilled person seeking a solution to the problem stated above would indeed first look for suggestions in the field of alkylene oxide catalysts, but, in the absence of useful suggestions therein, it is quite reasonable to expect him to refer to the field of catalysts or catalyst preparation in general and to take into consideration documents dealing in a broad aspect with improvement in efficiency of catalysts, for example of catalysts containing alumina, such as document (2).

Document (2) discloses a process of forming catalytic masses into rigid pellets which comprises shaping the mass of catalyst particles under pressure while they are heated to a temperature substantially below their normal melting point such as to cause bonding at their points of contact without substantial reduction in the original inter-particle voids (see column 1, lines 1 to 12 and column 4, lines 45-73). With respect to the shape of the pellets it is in particular referred to Fig. 1 which shows eighteen various shapes including spheres, rings, plates, cylinders, hollow cylinders, multiholed cylinders, hollow cylinders with corrugated outer walls etc. Other possible shapes of greater sizes than pellets (i.e. "massive shapes") such as saddles, discs, rings etc. are further mentioned in the description. According to (2), the various proposed forms are designed to promote the efficiency of the catalyst in two factors especially, namely the obtention of a maximum contact surface per weight or volume unit of catalyst and the necessity of avoiding channelling (see column 6, lines 25-46). However

as pointed out in this passage, the choice of any given form or combination of forms is determined by the conditions to be met in the use of the catalyst and document (2) does not give any information as to which shape is more suitable when selectivity must be improved without impairing pressure drop.

In connection with the desired high contact surface, document (2) indeed teaches that the surface of the pellets may be advantageously roughened or corrugated to increase their contact surface and, to the same purpose, it proposes to provide cylinders with a multiplicity of longitudinal holes therethrough. In the same context it also refers to any shape shown on Fig. 1 in particular to thin walled cylindrical shapes (cf. column 9, lines 60-67). Therefore, according to (2) a maximum contact surface may be achieved with a lot of different shapes, however this document does not point to any advantage of the multiholed cylinder over other shapes with a similarly high contact surface or with a higher surface than that of the cylinder, such as the hollow cylinder, the various possible shapes with roughened or corrugated outer walls, or other shapes shown on Fig. 1.

Although attention is focussed in particular to Friedel-Crafts catalysts, no information is given about the influence of the catalyst shape upon its selectivity in this particular reaction, let alone in oxidation reactions and in the partial oxidation of ethylene to ethylene oxide. As regards pressure drop document (2) recommends to use regular geometrical shapes to obtain low pressure drop in the catalyst bed (cf. column 6, lines 46-49), but no emphasis is put on a particular form as being more advantageous than the other proposed shapes in this respect. In the examples, catalysts having the form of hollow cylinders with corrugated outer walls, of

perforated cylinders or of "rounded diamonds" are illustrated. However, even in the examples, neither their catalytic performances (selectivity, activity, conversion, reaction temperature) nor the pressure drop in the catalyst bed are indicated.

Thus in the Board's opinion document (2) does not contain any indication from which the skilled person could infer or expect that the multiholed cylinders are more advantageous than hollow cylindrical pellets with respect to catalyst selectivity and simultaneously do not increase the pressure drop. Under these circumstances this document would not have provided the skilled person with any incentive to replace the support of the known ethylene oxide catalyst in the form of hollow cylindrical pellets by a support having the shape of multiholed cylindrical pellets illustrated in (2), in order to solve the technical problem stated above.

5.4 The influence of the catalyst shape upon the performance of the ethylene oxide catalyst is discussed in document (4). This document teaches to use spheres with a rugged or corrugated surface instead of a support with irregular shapes and dimensions in order to decrease the pressure drop. Comparison of the selectivity and conversion datas given in the table at page 4 shows that the spheres lead to a slightly better yield (selectivity x conversion) than the catalyst with irregular shapes although this latter has a much greater outer surface. This disclosure, which is more recent than that of (2), would not enable the skilled person to predict that among the various shapes disclosed in (2), the multiholed cylinder would solve the present technical problem.

5.5 Document (3) was cited in the decision under appeal only in connection with the features of the dependent claims. The Board agrees with the Examining Division that this document is not relevant to the subject-matter of Claim 1 since the catalyst is used in form of crushed and sieved particles.

5.6 Therefore, for the reasons given above, the Board considers that the replacement of the ring-shaped support of the known ethylene oxide catalyst by multiholed cylindrical pellets as defined in Claim 1 in order to improve the catalyst selectivity and activity without impairing the pressure drop is not obvious in the light of the cited prior art. Thus the subject-matter of Claim 1 involves an inventive step.

6. The process of producing an alkylene oxide according to Claim 5 is known per se from document (3) however as it is carried out in the presence of the inventive catalyst, its patentability derives from that of the catalyst.

Claims 1 and 5 being allowable, the same applies to the dependent Claims 2-4 and 6-7 which represents preferred embodiments thereof and whose patentability is supported by that of the main claims.

Order

For these reasons, it is decided that:

1. **The decision under appeal is set aside.**

2. **The case is remitted to the Examining Division with the order to grant a patent on the basis of the claims submitted on 12 August 1988 with a description including the indication of the relevant background art.**

The Registrar: The Chairman:

M. Beer P. Lançon