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**D E C I S I O N**  
of 27 October 1999

**Case Number:** T 0860/95 - 3.3.5

**Application Number:** 88106910.8

**Publication Number:** 0339119

**IPC:** B01J 23/94

**Language of the proceedings:** EN

**Title of invention:**  
Method of reactivating catalyst

**Patentee:**  
Nippon Shokubai Kagaku Kogyo Co., Ltd.

**Opponent:**  
Rhone-Poulenc Chimie

**Headword:**  
-

**Relevant legal provisions:**  
EPC Art. 84, 56

**Keyword:**  
"Clarity of a relative expression"  
"Inventive step (yes) not obvious to try"  
"Closest state of the art - identification without the benefit  
of hindsight"

**Decisions cited:**  
T 0860/93, G 0010/91

**Catchword:**  
-



Case Number: T 0860/95 - 3.3.5

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.5  
of 27 October 1999

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

**Representative:** Kraus, Walter, Dr.  
Patentanwälte Kraus, Weisert & Partner  
Thomas-Wimmer-Ring 15  
80539 München (DE)

**Respondent:** Rhone-Poulenc Chimie  
(Opponent) 25, Quai Paul-Doumer  
92408 Courbevoie Cedex (FR)

**Representative:** Le Pennec, Magali  
Rhone-Poulenc Rorer SA  
Direction des Brevets  
20 Avenue Raymond Aron  
92165 Antony Cédex (FR)

**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 18 September 1995  
revoking European patent No. 0 339 119 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** R. K. Spangenberg  
**Members:** M. M. Eberhard  
M. B. Günzel

## Summary of Facts and Submissions

- I. European patent No. 0 339 119 based on application No. 88 106 910.8 was granted on the basis of one claim concerning a method of reactivating a catalyst. The respondent (opponent) filed a notice of opposition requesting revocation of the patent on the ground of lack of inventive step over the disclosure of EP-A-0 000 663 (hereinafter D1).
- II. The opposition division revoked the patent on the ground of lack of inventive step. According to the decision, the method of claim 1 as granted differed from the regeneration process of D1 only by the heating time being 5-24 hours instead of 1 hour. The technical problem to be solved with respect to D1 was to achieve the revival of the original properties of the catalyst and to maintain them for a long period of time. In view of D1 the skilled person faced with the said problem would have carried out the regeneration for a longer period of time since it was common general knowledge that the greater the time of contact between reagents, the greater the probability of the reaction being completed.
- III. The appellant (patentee) lodged an appeal against this decision and submitted additional comparative tests on 23 January 1996. Both parties relied upon EP-A-0 169 449 (hereinafter D3) at the appeal stage. Amended claims were filed in reply to a communication from the board. Oral proceedings were held on 27 October 1999. At the oral proceedings the appellant submitted two amended claims, one as the main request and the other one as the first auxiliary request as well as an amended description adapted to the claim of the main request. The claim according to the main request reads as follows:

"A method of reactivating a catalyst having reduced activity as a result of having been used for a long period of time in producing an unsaturated aldehyde as a main product by catalytic vapor-phase oxidation reaction of propylene, isobutylene or tertiary butanol, said catalyst being a catalytic oxide comprising molybdenum, bismuth and iron with a Mo:Fe atomic ratio of 12:at least 0.1; which method comprises heat-treating the used catalyst at a temperature of 300 to 380°C in the presence of a flowing oxidizing gas containing at least 5% by volume of molecular oxygen and at least 0.1% by volume of steam, the treating time being 5 to 24 hours."

IV. The appellant's arguments can be summarised as follows:

The expression "for a long period of time" incorporated into the claim met the requirements of clarity set out in Article 84 EPC. The skilled person was familiar with the fact that catalysts which stood continuous use for at least 4000 hours without showing a decrease in activity, were usually employed for the industrial production of unsaturated aldehydes. It was clear to the skilled person that this expression meant several thousands of hours when used in the context of the patent in suit. In D3 the same expression was used, thus showing that it was familiar to the skilled person.

D1, contrary to D3, was not concerned with the technical problem underlying the claimed process. Therefore, D3 should be considered as the closest prior art. The technical problem vis-à-vis D3 was to provide an improved method for reactivating a catalyst having reduced activity as a result of its use for a long period of time in the catalytic reaction concerned. This improvement could be inferred from the test report of 27 February 1991. The respondent's doubts that the

problem was not solved at a space velocity of less than  $100 \text{ h}^{-1}$  was based only on assumptions and not supported by evidence. Neither D1 nor D3 pointed to the claimed solution. In D1, the purpose of the treatment with oxygen and steam was to reoxidise the catalyst constituents which had been reduced whereas the claimed treatment aimed at increasing the amount of iron molybdate. In D3, the catalyst deterioration after a long period of use was caused by the sublimation of Mo from the surface of the catalyst and, thus, greatly differed from that of the present invention. If the redox test disclosed in D1 had been a routine method for regenerating a catalyst deactivated as a result of a long period of use, it would have been mentioned in D3 since D1 was published five years before the priority date of D3.

Regarding the objection of insufficiency of disclosure raised by the respondent during the appeal proceedings (see below), the appellant indicated with reference to decision G 10/91 that he did not agree to fresh grounds of opposition being introduced into the proceedings.

V. The respondent put forward the following arguments in writing:

The appellant's comparative tests of 23 January 1996 had been performed at a temperature of  $330^\circ\text{C}$  although an average temperature of  $350^\circ\text{C}$  was used in D1. The appellant had chosen for these tests the sole temperature at which the difference in the period of time showed a positive effect. No difference would have been obtained in the upper part of the temperature range and, at the lower limit of  $300^\circ\text{C}$ , the regeneration effect would have been non-existent for a treatment of 5 hours. D3 confirmed that the greater the heating temperature, the shorter the period of time for obtaining a certain degree of restoration. The skilled

person knew how to adapt the regeneration temperature and period of time to the desired degree of regeneration, to the degree of deterioration of the catalyst and to the technology used. The claimed process lacked an inventive step over the teaching of D1 and D3. The invention was not sufficiently disclosed for it to be carried out by the skilled person in the temperature range of 300-320°C since the exemplified temperatures represented only 56% of the claimed range.

At the oral proceedings the appellant further argued that the expression "for a long period of time" introduced into the amended claims was not precise. As this time depended on the circumstances of the reaction, in particular the steam content and space velocity of the feed and the temperature, this expression covered very disparate values such as 4000 or 12000 hours. D1 represented the closest prior art since it concerned the same reaction and disclosed the same oxygen/steam ratio and temperature as in the claimed regeneration process. It further showed that regeneration was excellent in examples 20 and 21. The amended claim did not include the space velocity at which regeneration was performed although this parameter had an influence on the regeneration and narrower conditions were stated on page 4 of the patent in suit. The comparative tests of 23 January 1996 had been carried out at a space velocity of 1300 h<sup>-1</sup>, ie under the sole conditions that gave an improvement. The surprising improvement had not been shown for the whole range of the claimed process. D1 placed no particular emphasis on the heating step or on the presence of steam. Heating in air, which anyway inevitably contained water, was considered in D1 as a routine regeneration method, the skilled person would automatically have thought about. The skilled person would routinely have contemplated treating the deactivated catalyst for a longer time if he had wanted

to achieve an improvement. In the regeneration process of D3 the atmosphere also inevitably contained the water present in air and D3 further disclosed the possibility of adding steam. It was then simply a matter of adjusting the relative amounts of oxygen and steam. Furthermore, example 4 of the patent in suit showed that the temperature was not an essential feature.

- VI. The appellant requested that the decision of the opposition division be set aside and that the patent be maintained on the basis of the amended claim according to the main request submitted in the oral proceedings. As an auxiliary request, the appellant requested that the patent be maintained on the basis of auxiliary request I filed in the oral proceedings. The respondent requested that the appeal be dismissed.

### **Reasons for the Decision**

1. The appeal is admissible.
  
2. The respondent raised an objection of insufficiency of disclosure under Article 100(b) EPC for the first time at the appeal stage. This objection is, therefore, a fresh ground of opposition within the meaning of G 10/91 (opinion of the Enlarged Board of Appeal, OJ EPO, 1993, 420). According to this decision, fresh grounds of opposition may be considered in appeal proceedings only with the approval of the patentee. As the patentee did not agree to this fresh ground of opposition being introduced into the proceedings, the respondent's objection concerning insufficiency of disclosure is not taken into consideration by the board.

3. The amended claim of the main request meets the requirements of Article 123(2) and (3) EPC. It is disclosed in the application as filed that the reactivation method of the present invention is applied to multi-component Mo.Bi.Fe type oxide catalysts which were degraded as a result of having been used **for a long period of time** in the reactions concerned: see page 3, lines 24 to 34; page 6, lines 20 to 23. By introduction of this additional feature into the granted claim, the scope of protection has been restricted.
  
4. Turning to the respondent's objection concerning the lack of clarity of the expression "for a long period of time", the board observes that the use of a relative term in a claim may be accepted where the skilled person is able to understand the meaning of this term in a given context (see T 860/93, OJ EPO 1995, 47). In the present case, the claim concerns a method of reactivation of a catalyst, the activity of which has been reduced as a result of its use "for a long period of time" in the production of an unsaturated aldehyde by catalytic vapour phase oxidation of propylene, isobutylene or tertiary butanol. As stated in the claim, the catalysts used for said reactions are oxide compositions comprising Mo, Bi and Fe with a Mo:Fe atomic ratio of 12:at least 0.1. These catalysts are known catalysts which are used for the industrial production of unsaturated aldehydes and the operating conditions generally used in the said reactions are also well-known to the skilled person. The skilled person is also aware of the fact that the performance of these catalysts is gradually reduced when they are used for a long period of time and that the decrease of activity depends on the conditions used for the reaction, in particular on the composition and space velocity of the feed. As pointed out by the appellant and not disputed by the respondent, it is also well-



known to the skilled person in this technical field that these catalysts were used on an industrial scale, in continuous operation, in general for at least several thousands of hours before being reactivated or replaced with a fresh catalyst. In the examples of the patent in suit, the known catalysts to which the claimed reactivation method is applied were used for 12 000 hours in the vapour-phase oxidation of isobutylene to methacrolein or 16 000 hours in the vapour-phase oxidation of propylene to acrolein, under the reaction conditions stated in these examples. It is evident that under different reaction conditions, and with other known Mo.Bi.Fe-containing multi-oxide catalysts, this time may be shorter or longer. The fact that this period of time might vary in a broad range including for example 4000 hours in the case of relatively severe conditions or 16 000 hours as in example 6 of the patent in suit (ie four times higher), does not mean that this expression is not sufficiently clear to enable the skilled person to understand the meaning of the claim. It is essential in the present case to bear in mind that the contested expression relates to the well-known use of known catalysts in well-known reactions and not to the new method of regeneration itself. It follows from the above that the skilled person would understand in the given context what is meant by the expression "for a long period of time". Therefore, the claim according to the main request meets the requirement of Article 84 EPC.

5. The reactivation method according to the claim of the main request is novel over the teaching of the cited prior art. This was not in dispute so that further considerations in this respect are not necessary.
6. The board considers, in agreement with the appellant, that D3 represents the closest prior art. The respondent's arguments that D1, and not D3, was the

closest prior art cannot be accepted by the board for the following reasons. D1 is not directed to a process of regeneration of a deteriorated catalyst but to the preparation of unsaturated aldehydes and acids by the vapour-phase oxidation of propylene or isobutylene with molecular oxygen using specific multi-oxide catalysts having an improved redox stability. D1 does not deal with the technical problem underlying the patent in suit, ie the problem of reactivation of Bi.Mo.Fe based multi-oxide catalysts which have been deactivated as a result of their use for a long period of time in producing the said unsaturated aldehydes. The problem addressed in D1 is to provide a catalyst having a high redox stability when used in the said reactions (see page 1, lines 12 to 29). The reoxidation step of the redox test disclosed on page 6 is performed at temperatures and with amounts of air and steam lying within the claimed ranges. However, taking into account that D1, contrary to D3, does not deal with the problem of the patent in suit, the selection of D1 as the closest prior art mainly on the basis of the similarities between the conditions used in the redox test and those used in the claimed reactivation method amounts, in the board's judgment, to an inadmissible ex post facto analysis of the relevant state of the art.

- 6.1 D3 discloses a process for regenerating a Mo.Bi.Fe-containing multi-oxide catalyst, the catalyst performance of which was deteriorated as a result of its use for a long period of time (for example several years) in vapour-phase oxidation of propylene or isobutylene into acrolein or methacrolein respectively. The regeneration method comprises heating the deteriorated catalyst in an atmosphere consisting essentially of air at a temperature of 380-540°C, preferably 390-530°C, so as to attain at least partial restoration of the catalytic performance thereof. The heating is desirably carried out until maximum

restoration is attained (see claim 1; page 4, lines 29 to 36; page 5, lines 16 to 23; page 7, lines 24 to 33; page 8, lines 3 to 7; page 10, lines 13 to 17). It can be inferred from the data in the examples of D3 that complete regeneration was not achieved, since the conversion and yield reported for the regenerated catalyst were in fact obtained at reaction temperatures which are higher than in the case of the fresh catalyst. It is also pointed out in the patent in suit that the regeneration method of D3 does not lead to a complete restoration of the catalyst activity (see page 2, lines 30 to 40, the analysis of US-A-4 604 370 which is the US equivalent of D3).

With respect to D3, the technical problem underlying the claimed process can be seen in the provision of a method for reactivating a Mo.Bi.Fe based multi-oxide catalyst having a reduced activity as a result of its use for a long period of time in the production of unsaturated aldehydes, which method makes it possible to achieve complete regeneration of the catalyst and to maintain the restored catalytic performance for a long period of time.

It is proposed that this problem be solved by heat treating the deactivated catalyst under the conditions defined in the claim. The claimed solution differs from the regeneration treatment of D3 by the **combination** of a relatively short treatment time of 5-24 hours with relatively low temperatures, i.e. between 300 and 380°C, the amount of steam in the oxidising atmosphere being of at least 0.1 vol.%. In view of the results reported in the examples of the patent in suit, it is credible that the problem indicated above has actually been solved by the method as defined in the claim of the main request.

6.2 Although the respondent's arguments about the comparative examples filed on 23 January 1996 concern the comparison of the claimed process with that of D1 and not with the regeneration process of D3, they raised the question whether or not complete regeneration would be achieved throughout the whole ranges of temperature, time and space velocity defined in the claim.

The board first observes that the temperature range of 300 to 380°C is claimed **in combination** with a treatment time of 5 to 24 hours. The fact that at 300°C a treatment period of 5 hours might not be sufficient to obtain complete regeneration does not, in the board's view, necessarily mean that the problem is not solved in the whole range of temperatures. As pointed out by the respondent himself, the skilled person is aware of the fact that lower temperatures generally require longer periods of time for obtaining the same degree of regeneration, and vice versa. Therefore, even if complete regeneration were not attained at 300°C by using a treatment period of 5 hours, it might be achieved with a longer period of up to 24 hours. Conversely the minimum treatment period of 5 hours might be suitable for higher temperatures of the claimed range. As shown by the appellant complete regeneration is for example obtained by a treatment at 330°C for 5 hours. In these circumstances and further considering that the respondent, who has the burden of proof with respect to this issue, has provided no evidence in support of his assumption, the board is not convinced that the problem indicated above is not solved for the whole range of temperatures and periods of time as claimed.

Regarding the influence of the space velocity on the degree of regeneration, it is stated on page 4, lines 39 to 41, of the patent in suit that "the degraded catalyst can be easily reactivated at a relatively low temperature of 300 to 500°C using a gas containing at least 5% by volume of molecular oxygen and at least 0.1% by volume of steam at a space velocity of at least 100 h<sup>-1</sup>." The board cannot infer from this statement that only a partial regeneration would be obtained at space velocities of  $\leq 100$  h<sup>-1</sup> since these conditions are those which permit achievement of the desired result **easily** and there is no information in the patent in suit from which it could be derived that a space velocity of at least 100 h<sup>-1</sup> is necessary to obtain complete regeneration. Therefore, in the absence of evidence to support this contested statement the board cannot accept it.

- 6.3 D3 discloses that an atmosphere consisting essentially of air at a temperature of 380 to 540°C is used for the catalyst regeneration, which means that the atmosphere may further contain a "small amount" of carbon dioxide, steam, nitrogen gas or the like. The said "small amount" of non-oxidising components is determined taking into account that the molybdenum should preferably be in a "high-order oxide state" since the rate of diffusion of Mo is increased as the ionic radius is decreased (see page 3, lines 24 to 26; page 6, line 30 to page 7, line 4). Although D3 does not exclude the presence of a small amount of steam, this document contains no information from which the skilled person could have inferred that the presence of steam might have a beneficial effect on the degree of restoration of the catalyst.

According to D3 the regeneration is performed at a temperature of from 380°C to 540°C, preferably from 390°C to 530°C. The heating period is varied depending upon the degree of deterioration and the desired degree of restoration of the catalyst. Generally, low temperature heating requires a long period of time and vice versa. At a temperature of less than 500°C, the heating time is at least 12 hours, preferably at least 24 hours, more preferably from a number of days to over ten days. In the examples, the deactivated catalyst is treated in air at 540°C for 3 hours, 470°C for 3 days, 410°C for 11 days, 410°C for 30 days, and 450°C for 7 days respectively. However, as already indicated above, the catalytic performance is not fully restored. In comparative example 2 of D3 where the deactivated catalyst is treated at 350°C for 10 days under flowing air, the degree of regeneration is far lower than in the examples illustrating the invention of D3, according to which the regeneration temperature must lie within the range 380 to 540°C, preferably 390 to 530°C. Neither this teaching nor the remaining disclosure in D3 suggests that performing the regeneration at temperatures between 300 and 380°C for 5 to 24 hours under a flowing oxidising atmosphere containing at least 0.1 vol.% steam in addition to the oxygen would have made it possible to achieve complete regeneration of the deactivated catalyst.

6.4 As already indicated above D1 does not relate to a process of regeneration of a catalyst which has been deactivated as a result of its use for a long period of time in the production of unsaturated aldehydes and acids by catalytic oxidation of propylene or isobutylene. The objective of D1 is to provide Mo.Bi.Fe multi-oxide catalysts which exhibit an improved redox stability in order to avoid a decrease of catalytic activity resulting from variations of the amount of oxygen in the feed with respect to the desired value. This

objective is achieved by using catalysts having the formula indicated in the claims of D1 for the said production (see page 1 line 14 to page 2, line 24; page 4, lines 18 to 30). In order to illustrate the excellent redox stability of the catalysts, the latter are tested by means of a redox test in which propylene is oxidised to acrolein at a temperature of 400°C for 2 hours using a feed with a relatively low oxygen content and the resulting reduced catalyst is reoxidised by feeding a mixture of air and steam with an air/steam ratio of 2.3/4 to the catalyst, at a temperature of 320, 350 or 380°C for 1 hour (see page 6, lines 11 to 35 and Table I). The results reported in Table 1 show that the performance of the catalysts is not fully restored after the reoxidation step of this redox test in the case of 22 catalysts out of 24. The initial performance is completely restored only for the two Mg containing catalysts of examples 20 and 21 which were reoxidised at 380°C and had a relatively low initial performance compared to the other catalysts. D1 contains no further information from which the skilled person could have inferred that the reoxidation step of this **redox test**, which in most cases does not lead to complete regeneration and involves relatively low temperatures and a short period of time compared to those used in D3, would have been suitable to achieve complete regeneration of catalysts which have been deactivated in a different way, i.e. by use for a long period of time in the normal production of unsaturated aldehydes. Furthermore, the skilled person knew from D3 that the said deactivation after a long period of use was due to Mo sublimation from the surface of the catalyst and that at heating temperatures of less than 380°C a sufficient diffusion of Mo from the interior of the catalyst particles to their surface cannot be achieved, temperatures of 390 to 530°C being preferred for the regeneration (see D3, page 7, lines 24 to 28, and page 6, lines 14 to 23). Therefore, in view of the

teaching of both D1 and D3, he would not have been encouraged to try the reoxidation treatment of the redox test disclosed in D1 in order to solve the problem of complete regeneration stated in point 6.1 above.

6.5 The respondent further argued that it could be deduced from example 4 of the patent in suit that the temperature was not an essential feature. This argument cannot be accepted by the board. Example 4 shows that a catalyst which has been deactivated as a result of its use for 12 000 hours in the production of methacrolein from isobutylene can be completely regenerated by treatment at a temperature of 410°C for 5 hours in a flowing gas composed of 2 vol.% of steam, 19 vol.% of oxygen, the remainder consisting mainly of nitrogen. The catalytic performance of the regenerated catalyst can be maintained for a long period of time. The conditions used in this example correspond to those stated in the claim except for the temperature which is higher than 380°C. Therefore, it can be derived from example 4, read in the context of the patent in suit, that the technical problem stated above can also be solved by treating the catalyst at a temperature of 410°C, ie a temperature lying within the broader temperature range of 300 to 500°C disclosed on page 4 of the description but higher than the claimed upper limit of 380°C. However, it cannot be inferred therefrom that the temperature is not an essential feature, let alone that the combination of the operating conditions indicated in the amended claim lacks an inventive step over the teaching of D1 and D3.

6.6 The other documents cited during the opposition proceedings were not relied upon at the appeal stage. They do not contain any additional information which could hint at the claimed solution, when considered alone or in combination with the teaching of D1 and D3.



6.7 It follows from the above that the subject-matter of the amended claim according to the main request also meets the requirement of inventive step set out in Articles 52(1) and 56 EPC.

**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 with the claim and description according to the main request filed in the oral proceedings.

The Registrar:

The Chairman:



S. Hue



R. Spangenberg

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