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D E C I S I O N
of 3 December 1996

Case Number: T 0369/94 - 3.3.2

Application Number: 89201070.3

Publication Number: 0339748

IPC: B01J 8/02

Language of the proceedings: EN

Title of invention:
Process for the preparation of ethylene oxide

Patentee:
SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

Opponent:
O1) LINDE AKTIENGESELLSCHAFT
O2) BASF Aktiengesellschaft, Ludwigshafen

Headword:
Ethylene oxide/SHELL

Relevant legal provisions:
EPC Art. 56, 114(2)

Keyword:
"Inventive step - no"
"Late filed document - admitted"

Decisions cited:
-

Catchword:
-

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Case Number: T 0369/94 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 3 December 1996

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 16 February 1994 revoking European patent No. 0 339 748 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: G. J. Wassenaar
J. Van Moer

Summary of Facts and Submissions

I. European patent No. 0 339 748 was granted with 9 claims in response to European patent application No. 89 201 070.3. Claim 1 of the patent in suit reads as follows:

"1. Process for the preparation of ethylene oxide by reacting ethylene with molecular oxygen in the presence of a silver-containing catalyst, which comprises passing a gas mixture of ethylene and oxygen through a reaction zone containing the catalyst particles, while removing heat from the reaction zone by a cooling medium which flows through the reaction zone via one or more helical patterns, each pattern containing one or more helices."

II. Notices of opposition were filed by the Respondents (O1 and O2). Revocation of the patent in its entirety was requested on the grounds of lack of inventive step and insufficient disclosure (Articles 54, 56, 83, 100(a) and 100(b) EPC).

Amongst others, the following documents remaining relevant for the present decision were cited:

Linde-Berichte aus Technik und Wissenschaft, 58 (1986), pages 5 to 8 (D2)

EP-A-0 082 609 (D4)

GB-A-2 046 618 (D6)

Kirk Othmer's Encyclopedia of Chemical Technology, 3rd ed. (1980), pages 439 to 454 (D8)

Chemical Economy & Engineering Review, vol. 15(4), 1983, pages 14 to 16 (D11)

Chem. Ing. Tech. vol. 58(3), (1986), pages 212 to 215 (D15).

III. The Opposition Division revoked the patent. The decision was taken on the basis of the set of claims as granted.

They considered that claim 1 lacked an inventive step over D6 in combination with D15.

IV. The Appellant (Patentee) lodged an appeal against this decision.

In the statement of the grounds of appeal, the appellant contested the arguments of the Opposition Division and argued that the reactor according to D6 was inadequate for the production of ethylene oxide and therefore could not be regarded as the most relevant state of the art. He considered that D8 represented the most relevant state of the art. He submitted that the statement at the end of D15, that the Linde reactor is also suitable for partial oxidation reactions, did not imply that the reactor was suitable for the partial oxidation of ethylene to ethylene oxide. The envisaged partial oxidation reaction might have been the partial oxidation of methanol to formaldehyde. In this context reference was made to a new document:

Kirk-Othmer's Encyclopedia of Chemical Technology, 3rd Ed., vol. 11 (1980), pages 236 to 242 (D16),

which described the partial oxidation reactions of methanol to formaldehyde.

With respect to the Linde reactor, he submitted that D2 gave a more detailed description than D15. D2 contained no incentive for the person skilled in the art of ethylene oxide production to choose this reactor in preference to the long-known reactor of D8.

V. The Respondents submitted that the expression "partial oxidation" in D15 also related to partial oxidation reactions such as the partial oxidation of ethylene to ethylene oxide. The heat density in kW/m^3 in a reactor for the production of ethylene oxide was calculated as being lower than for the production of formaldehyde, so that a reactor suitable for the production of formaldehyde would also be suitable for the production of ethylene oxide. The arguments were supported by references to new documents:

Ullmann's Enzyklopädie der technischen Chemie, 4th ed., vol. 17 (1979), pages 483 to 514 (D17) and,

Chauvel et al., Hydrocarbon Processing 52 (9), 1973, pages 179 to 184 (D18).

VI. In reply thereto, the Appellant stressed that a prejudice existed against departing from external heat exchange in the production of ethylene oxide because of the risk of a runaway reaction and explosion. The admissibility of the late filed document D15 in the proceedings was questioned with the argument that this document was not exceptionally relevant.

VII. Oral proceedings were held on 3 December 1996. The parties maintained their requests. With respect to inventive step special attention was drawn to documents (2), (4), (6), (8), (11), (15) and (17).

The Appellant maintained that the skilled person would not use one of the reactors disclosed in D4 or D6 for the preparation of ethylene oxide because of the danger of a runaway reaction so that the closest prior art was represented by D8. The parties agreed that, independently of the choice of the starting document, the problem underlying the invention was in all cases how to scale up the process for the preparation of ethylene oxide by the catalytic oxidation of ethylene.

VIII. The Appellant requested that the decision under appeal be set aside and the patent be maintained.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. *Admissibility of late filed documents*

D15 was filed after the 9 months opposition period. However, it was filed in reply to the Appellant's submission in the opposition procedure that D2 only related to equilibrium reactions such as the methanol synthesis. It is thus questionable whether D15 should be considered as evidence not filed in due time within the meaning of Article 114(2) EPC. Even if one were to consider D15 as late filed evidence, the Opposition Division acted correctly by admitting D15 in the proceedings for the following reasons. Apart from exceptional circumstances, the essential criterion for accepting late filed evidence is its relevance for the decision. In the present case the Opposition Division based its decision to a large extent on D15. It is thus evident that in the opinion of the Opposition Division

D15 was very relevant. Under these circumstances the Board is of the opinion that the Opposition Division had not only the right but also the duty to admit this document into the proceedings. The Board is also of the opinion that D15 provides some additional information over D2 which is not without relevance for the decision. The Board therefore considers that D15 has been correctly introduced into the proceedings under Article 114(2) EPC.

3. *Inventive step*

3.1 The document representing the closest prior art is generally the document related to the same technical field and having more essential features in common with the invention under consideration than any other prior art document. The only prior art document relating to the preparation of ethylene oxide and disclosing, all the features of present claim 1, except the helical pattern flow of the cooling medium, is D4.

The Appellant's argument that D4 should be disregarded because the reactor disclosed therein has several drawbacks which makes the reaction inherently unstable and thus unsuitable for the ethylene oxide production cannot be followed. In D4 it is explicitly disclosed that the reactor is very suitable for carrying out strongly exothermic processes, for example the oxidation of ethylene to ethylene oxide by contacting ethylene with oxygen in the presence of a catalyst which is normally a supported silver catalyst (page 3, lines 29 to 34). In fact, this oxidation reaction is the only explicitly disclosed reaction in D4. The Appellant has not provided any evidence that the alleged drawbacks are of such a nature that it would be impossible to carry out the process according to D4. The argument that the reactor of D4 contains dead volumes wherein reactive gas is in contact with the

catalyst without sufficient cooling which might trigger a runaway reaction is not convincing. As pointed out by the Respondents, the entrance of the reactive gas in the dead volumes can be easily prevented by introducing an inert gas in the dead volumes and the contact of any reactive gas in the dead volumes with the catalyst can be prevented or at least greatly reduced by providing an inert packing at the top and the bottom of the catalyst bed as explained in D4, page 4, lines 34 to 36. The argument that the gas flow in the reactor of D4 is not a plug flow but gives rise to back-mixing is not convincing either, since in the reactor used according to the patent in suit the gas flow is also not a plug flow and back-mixing is not prevented either. The circumstance that, to the knowledge of the Appellant, the process of D4 has not been performed in practice does not mean that it cannot be performed. The state of the art to be taken into consideration for inventive step is not and cannot be limited to what has actually been performed. In fact, it is often the difficulties encountered by performing the instructions in the state of the art which define the technical problems to be solved. Without convincing evidence to the contrary, the Board is, therefore, of the opinion that D4 is an enabling disclosure representing the closest prior art.

3.2 According to the patent in suit there is nowadays an increasing demand for larger capacity equipment, but the upscaling of classical reactors of the externally cooled multitube type such as disclosed in D8 causes cooling problems. The Board accepts that temperature control is a general problem in upscaling reactors since in indirect cooling systems the cooling capacity increases with the cooling surface (to the square)

whereas the heat development increases with the volume (third power). According to the Respondents, cooling problems also arise with the upscaling of internally cooled reactors of the type as disclosed in D4 and D6.

Accordingly, the problem underlying the invention can be seen in increasing the reactor capacity of an isothermal ethylene oxide reactor.

According to claim 1 this problem is solved by cooling the reactor with a medium which flows through the reaction zone via one or more helical patterns, each containing one or more helices.

Reactors having such a flow pattern are known in the art. According to D2, a report with the heading "Linde Isothermreaktor für die Methanolsynthese", disclosing such a reactor, the use of cooling tubes bent in a helical pattern allows a higher range for the ratio between the cooling surface and reactor volume and increases the capacity with respect to reactors with straight cooling tubes (point 2, under "Reaktorgeometrie"). The Board is therefore satisfied that the claimed solution actually solves the said problem.

3.3 It remains therefore to be decided if it was obvious for the person skilled in the art to substitute the internally cooled reactor of D4 for the production of ethylene oxide with the equally internally cooled known reactor with helically bent cooling tubes (hereinafter referred to as the Linde reactor) in order to increase the reactor capacity.

The skilled person encountering cooling problems in upscaling the reactor of D4 will first look for solutions in the field of ethylene oxide reactors. The choice in this field is very limited, probably only the

reactor types disclosed in D6 and D8, which suffer from the same kind of cooling problems. D6 is cited in D4. In fact, the reactor disclosed in D4 is a further development of the reactor of D6; see D4, page 1, lines 4 to 13. The skilled person is taught in D6 that internally cooled reactors suitable for the production of ethylene oxide are also suitable for other chemical exothermic reactions such as the synthesis of methanol, methane and formaldehyde (page 8, lines 73 to 114). The skilled person trying to improve the reactor capacity will also take into consideration available reactors used in other exothermal reactions. The skilled person will certainly look for the more recent developments in the field of isothermal reactors and should be aware of the Linde reactor and the publications D2 and D15 both relating thereto. The skilled person knows from D2 (point 2) that by using cooling tubes in a helical pattern the cooling capacity can be increased and the heat exchange on a large technical scale can be improved. D2 also teaches that the reactor, developed for the synthesis of methanol, can also be used for other exothermal catalytic reactions such as for the preparation of higher alcohols or methane. The skilled person learns from D15 that the Linde reactor can also be used for partial oxidation reactions (point 3, last sentence). The present reaction of ethylene with molecular oxygen to ethylene oxide is a partial oxidation reaction. Whether or not the present reaction was intended to be included by the expression "partial oxidation" in D15, this indication will encourage the skilled person to use the Linde reactor for the synthesis of ethylene oxide too. In view of the positive indication in D2 that the capacity can be increased by using the Linde reactor and the general character of the cooling problems in isothermal reactors for exothermic reactions it must be

considered obvious to the skilled person to use the Linde reactor, one of the most recent developments in reactor technology, to solve the above mentioned problem.

3.4 The Appellant's main argument against obviousness is the existence of a prejudice against using an internally cooled reactor for the synthesis of ethylene oxide because of the risk of a runaway reaction typical for this reaction which is unavoidably also accompanied by a complete oxidation to carbon dioxide. The Board accepts that such a prejudice might have existed before the publication of documents D4 and D6. At the latest after the publication of D4, in which the only explicitly mentioned use of the reactor is for the oxidation of ethylene to ethylene oxide, this prejudice was removed. After the publication of D4 there was no obstacle for the skilled person to use the Linde reactor with its known advantages of large production and cooling capacity for solving the upscaling problem in the process for the oxidation of ethylene to ethylene oxide according to D4.

3.5 The Appellant's argument that D15, while mentioning "partial oxidation", does not point to the partial oxidation of ethylene to ethylene oxide but rather to the partial oxidation of methanol to formaldehyde is not convincing. D15 discloses in a very general statement that the reactors with helically bent cooling tubes can also be used for reactions which are not determined by equilibrium reactions such as hydrations or partial oxidations. This statement comprises most, if not all, of the large scale synthesis reactions and reinforces the general statement at the beginning of the article that the newly developed reactors are for

use in exothermal catalytic processes. There is no indication that some partial oxidation reactions were to be excluded therefrom. The Appellant's reference to D16, relating to the synthesis of formaldehyde is, therefore, of no relevance to this decision.

3.6 The Appellant's further argument that pressure and temperature conditions in the synthesis of methanol are very different from those in the synthesis of ethylene oxide so that it was not obvious to use a methanol reactor for solving a problem in the synthesis of ethylene oxide is not convincing either. In the first place, the reaction conditions are not fundamentally different; viz. 50 to 150 bar and 200 to 220°C for the methanol synthesis (D2) and 5 to 50 bar and 200 to 300°C for the ethylene oxide synthesis (D4, page 4, lines 2 to 6). Besides, it is known from D15 that the Linde reactor can be used in a methanising plant at temperatures from 300 to 650°C (page 215, paragraph 3.2). Thus it was evident to the skilled person that the Linde reactor could cope with the relatively moderate reaction conditions required for the ethylene oxide synthesis. Furthermore it is general knowledge that the same type of reactor can be used for many different reactions; e.g. in D6, amongst many others, the methanol synthesis and the oxidation reaction for preparing ethylene oxide are both mentioned as exothermic reactions which can be performed in the same kind of reactor (page 8, lines 73 to 114). Moreover, the skilled person will not use exactly the same reactor for the present reaction as used for the methanol synthesis but, sticking to the principle of internal cooling by helically bent tubes, will adapt it to the specific requirements of the ethylene oxide synthesis.

3.7 In summary, the claimed solution of the technical problem as defined above in point 3.2 results from applying recent developments in reactor technology for the preparation of a known compound with a known catalyst under known reaction conditions, which required no more than ordinary technical skill, without involving an inventive step in the meaning of Article 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana P. A. M. Lançon