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D E C I S I O N
of 2 February 2001

Case Number: T 1018/97 - 3.3.5

Application Number: 91108734.4

Publication Number: 0460512

IPC: B01D 71/02

Language of the proceedings: EN

Title of invention:

Membrane formed of crystalline molecular sieve material

Patentee:

MOBIL OIL CORPORATION

Opponent:

Exxon Chemical Patents Inc.

Headword:

Molecular sieve/MOBIL

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step"

"Main and first auxiliary request - no, obvious alternative"

"Second auxiliary request - yes, non-obvious use of the product"

Decisions cited:

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Catchword:

-



Case Number: T 1018/97 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 2 February 2001

Appellant: MOBIL OIL CORPORATION
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Respondent: Exxon Chemical Patents Inc.
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Representative: White, Nicholas John, Dr.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 12 September 1997
revoking European patent No. 0 460 512 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: G. J. Wassenaar
M. B. Günzel

Summary of Facts and Submissions

- I. The appeal is from the decision of the Opposition Division to revoke European patent No. 0 460 512, which was granted in response to European patent application No. 91 108 734.4.
- II. The basis for the contested decision was a main request and five auxiliary requests filed by the patent proprietor during opposition proceedings. Claim 1 of the main request was directed to a microporous, self-supporting membrane consisting of a continuous array of ZSM-5 zeolite crystals. The main request and auxiliary requests 1 and 2 were rejected on the ground that the subject-matter of claims 1 thereof lacked an inventive step in view of

D1: DE-A-3 827 049 and

D3: EP-A-0 135 069.

The Opposition Division essentially argued that D1 disclosed a self-supporting membrane consisting of layers of zeolite crystals and that it was obvious to make such a membrane of ZSM-5 type zeolites, which were known in the art as components in separation membranes as evidenced by D3. With respect to the process for making the membrane (auxiliary request 2), which differed from the process disclosed in D1 only in the use of a non-porous carrier, it was argued that it was self-evident to use a non-porous carrier if the carrier was removed by a solvent. Moreover the use of a non-porous carrier was already contemplated in D3.

The auxiliary requests 3 and 4 were rejected on the

ground that the amended claims comprised subject-matter which extended beyond the content of the application as originally filed. Although the subject-matter of the claim according to auxiliary request 5 was considered to involve an inventive step, the request was rejected because the appellant did not adapt the description to the amended claim within the time limit set. An inventive step was acknowledged based on the finding that none of the cited documents disclosed the use of zeolitic membranes for the separation of the gas mixtures mentioned in the claim of auxiliary request 5 and that there was no suggestion to use ZSM-5 type membranes for that purpose.

III. With the statement of the grounds of appeal, the appellant submitted a new main request and argued that the Opposition Division's reasons refuting inventive step were not well-founded. The appellant's final requests, submitted during oral proceedings, which were held on 2 February 2001, comprised four sets of claims.

Claim 1 of the main request reads as follows:

"Use of a non-composited, microporous, self-supporting membrane consisting of a continuous array of crystalline molecular sieve material consisting of a siliceous zeolite of low or zero activity containing only trace amounts of two- or three- valent metals or none at all for separating gaseous or liquid mixtures."

Claim 1 of the first auxiliary request reads as follows:

"A method of preparing a non-composited, microporous, self-supporting membrane consisting of a continuous

array of crystalline molecular sieve material which comprises preparing a reaction mixture capable of forming a crystalline molecular sieve material, forming the mixture into a thin, cohesive, continuous membrane under crystallization conditions by allowing crystallization of the mixture in a continuous layer on a non-porous forming surface, removing the continuous layer on the forming surface from the mixture, rinsing and drying the layer and removing it from the forming surface to obtain a non-composited membrane."

The claim of the second auxiliary request reads as follows:

"Use of a non-composited, microporous, self-supporting membrane consisting of a continuous array of crystalline molecular sieve material consisting of a siliceous zeolite of low or zero activity containing only trace amounts of two- or three- valent metals or none at all for separating gaseous or liquid mixtures, wherein the mixture comprises oxygen and nitrogen, hydrogen and carbon monoxide, or a linear paraffin and a branched paraffin."

In the claim of the third auxiliary request, which substantially corresponded to the claim found allowable by the opposition division, the linear paraffin was limited to hexane and the branched paraffin to 2,2-dimethyl butane.

IV. The appellant's submissions with respect to the inventive character of the subject-matter of these claims can be summarized as follows:

D1 represented the closest prior art. It disclosed

composite membranes comprising zeolitic material on a porous carrier. Although there is also a disclosure that the porous carrier can be removed by dissolution, there is no explicit disclosure of a self-supporting membrane. With the self-supporting membranes according to the patent in suit separation characteristics could be improved. D1 only disclosed the use of a **porous** carrier for preparing the membrane. Only a low silica zeolite was prepared and tested, which led away from the use of the claimed high silica zeolites. D3 did not disclose or suggest the use of a self-supporting membrane. Neither of the cited prior art documents disclosed or suggested separating one of the mixtures mentioned in the claim of auxiliary request II.

- V. During the appeal proceedings the respondent (opponent) withdrew its opposition.

- VI. The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims of the main request filed during oral proceedings. As auxiliary requests, the appellant requested that the patent be maintained on the basis of any of auxiliary requests I to III filed during oral proceedings, taken in their numerical order.

Reasons for the Decision

- 1. *Main request*
 - 1.1 D1 relates to zeolitic molecular sieves for the separation of fluids. It discloses the formation of a composite membrane consisting of continuous crystalline

zeolite layers on a porous carrier, whereby the pores are so small that they are bridged by the zeolite crystals (page 2, lines 36 to 49 and Figure 3). It is further indicated that one can use a soluble carrier which after the formation of the zeolite layers can be removed by a solvent (page 2, lines 50 to 51 and claim 11). Although self-supporting zeolitic membranes are not explicitly mentioned in D1, the specifically disclosed alternative process disclosed in D1, according to which the carrier is removed by solving the carrier, inevitably results in the formation of a self-supporting zeolitic membrane. Thus the use of a non-composited, microporous, self-supporting membrane consisting of a continuous array of crystalline zeolite material for separating gaseous or liquid mixtures was contemplated in D1.

- 1.2 The appellant's submission that the membranes used according to claim 1 have improved separation characteristics has not been supported by any experimental evidence. It is also unlikely that the present high silica zeolite membranes have improved separation characteristics for any mixture since the performance of a membrane is dependent upon the pore size of the membrane and the effective diameter of the molecules to be separated. Thus even if the selectivity of a membrane according to present claim 1, was better than that of the membrane disclosed in D1 for one mixture, it is likely that there are other mixtures for which the membrane according to the examples of D1, having a different pore size, perform better. Under these circumstances the Board can only accept as the problem underlying the invention the provision of a further zeolitic membrane to be used for separating fluids. The patent in suit proposes solving this

technical problem by the use of a self-supporting membrane consisting of a high silica zeolite. It is indisputable and supported by Example 1 of the patent specification that with the membrane according to claim 1 gas mixtures can be separated. The Board is, therefore, satisfied that the membrane used according to claim 1 actually solves the above-mentioned problem. It remains to be decided whether the claimed solution was obvious to a person skilled in the art.

1.3 Supported high silica zeolitic membranes for the separation of fluid mixtures are known in the art. D3 discloses for instance the use of a composite membrane having a surface layer of ZSM-5 type zeolite for use in the molecular sieving of xylene isomers (page 8, line 19 to page 9, line 14). Specifically disclosed in this respect is silicalite, a ZSM-5 type siliceous zeolite containing only trace amounts of aluminium (three-valent metal), which, according to the definition given in the patent in suit (page 5, lines 40 to 42), has low or zero catalytic activity. The forming of an unsupported zeolitic membrane according to D1 is not limited to any specific zeolite. The skilled person trying to solve the above mentioned problem would, therefore, consider any available zeolite known to be capable of forming a membrane; thus also silicalite.

1.4 The appellant's submission that D1 specifically discloses only a low silica zeolite and therefore leads away from the invention cannot be accepted. The Na-A zeolite mentioned in D1 is used for the separation of hydrogen from a mixture with nitrogen (Example 2). The Board agrees that D1 does not point to the use of high silica zeolites, such as silicalite, for separating a

mixture of small molecules. Present claim 1 is, however, not limited to such a use. For the separation of a mixture comprising larger molecules, D1 does not discourage the skilled person from preparing an unsupported silicalite membrane. The clear incentive provided by D3 to use high silica ZSM-5 type zeolites for separating a mixture of larger molecules, such as xylene isomers, is thus not put in doubt by any technical information derivable from D1. The Board, therefore, holds that the use of the self-supported membrane according to claim 1 was obvious to a person skilled in the art for solving the above mentioned problem. Thus, the subject-matter of claim 1 lacks an inventive step within the meaning of Article 56 EPC.

2. *Auxiliary request I*

The method for preparing a self-supporting membrane according to auxiliary request I differs from the method disclosed in D1 only in the explicit requirement that the crystallisation of the zeolite takes place on a **non-porous** forming surface. Explicitly disclosed in D1 is the use of a **porous** forming surface. In a supported membrane the fluid to be treated must be able to penetrate the support. It is thus self-evident to use a porous forming surface if a supported membrane is prepared according to the main objective mentioned in D1. For the alternative embodiment mentioned in D1, whereby the carrier is removed by a solvent, the porosity of the carrier is not relevant. This is, in the Board's judgement and in accordance with the opponent's submission during the opposition proceedings (see the decision under appeal, reasons 6.3, third paragraph), immediately clear to a skilled reader of D1. For a skilled person who wants to execute said

alternative embodiment it is thus obvious that in that case he is not bound to a porous carrier and that a non-porous forming surface can also be used. The alleged advantages of using a non-porous forming surface mentioned by the appellant, such as a more homogeneous membrane layer and reduction in thickness variation, have not been supported by experimental evidence. Nevertheless, the Board can accept, on the basis of common general knowledge, that these advantages were really obtained. On the same basis, however, the Board must conclude that a skilled person would have expected them. In fact, therefore, they provide an incentive to use a non-porous polymer film as a forming surface if the polymer film is to be removed by treating it with a solvent as disclosed in D1 rather than indicating the presence of an inventive step. The Board, therefore, holds that the method according to auxiliary request I is an obvious modification of the process according to D1 and thus lacks an inventive step.

3. *Auxiliary request II*

3.1 The amendments made in the claim according to auxiliary request II are based on the application as originally filed (paragraph bridging pages 11 and 12 in combination with Example 1) and do not extend the protection conferred. The subject-matter of the claim according to auxiliary request II, therefore, fulfils the requirements of Articles 123(2) and (3) EPC.

3.2 The subject-matter of the claim according to auxiliary request II is essentially in conformity with the content of the claim according to auxiliary request 5 of the contested decision. The subject-matter of the

claim according to said auxiliary request 5 was regarded as involving an inventive step for the reasons summarized under point II above. During the appeal proceedings the opposition was withdrawn and no arguments or new evidence were put forward which would induce the Board to deviate from the findings in the contested decision in this respect. Under these circumstances the Board accepts that the subject-matter of the claim according to auxiliary request II involves an inventive step.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent with the claim of auxiliary request II filed during the oral proceedings and a description to be adapted.

The Registrar:

The Chairman:

S. Hue

R. Spangenberg