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D E C I S I O N
of 28 January 1997

Case Number: T 0561/93 - 3.3.2

Application Number: 86302121.8

Publication Number: 0195688

IPC: C01B 3/38

Language of the proceedings: EN

Title of invention:
Heat exchange steam-reforming process and reactor

Patentee:
Haldor Topsoe A/S

Opponent:
Linde Aktiengesellschaft, Wiesbaden

Headword:
-

Relevant legal provisions:
EPC Art. 52(1), 56

Keyword:
"Inventive step (no)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0561/93 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 28 January 1997

Appellant: (Opponent)
Linde Aktiengesellschaft, Wiesbaden
Zentrale Patentabteilung
D-82049 Höllriegelskreuth (DE)

Representative:
-

Respondent: (Proprietor of the patent)
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 12 May 1993 concerning maintenance of European patent No. 0 195 688 in amended form.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: M. M. Eberhard
J. H. Van Moer

Summary of Facts and Submissions

I. European patent No. 0 195 688 based on application No. 86 302 121.8 and claiming a priority date of 21 March 1985 was granted with effect from 3 October 1990 on the basis of eight claims.

II. The appellants (opponents) filed a notice of opposition requesting revocation of the patent on the grounds of lack of inventive step. Of the documents cited by the parties during the opposition procedure, only the following documents are relevant to the present decision:

DE-A-2 751 251 (hereinafter D7),

DE-A-1 667 573 (hereinafter D2),

EPRI Report EPRI AP-5319, August 1987.

III. In its interlocutory decision posted on 12 May 1993, the opposition division maintained the patent in an amended form. The opposition division held that the subject matter of the amended claims filed on 22 April 1993 met the requirements of patentability. According to the decision, the technical problem which had been solved with respect to the closest prior art D7 was the extension of the lifetime of the metallic components of the system. It appeared that when the heating of the second catalyst compartment was carried out solely by the hot flue gas, the maximum temperature at the end of the catalyst chamber was not as high as in the last part of the catalyst chamber of D7. The opposition division took the view that this effect was not obvious from the teaching of D7 and from the other cited documents.

IV. The appellants lodged an appeal against this decision on 15 June 1993. In the statement of grounds of appeal, the appellants contested that the claimed process involved an inventive step. They pointed out in particular that in the alternative of D7 where a shorter plug 46 was used, the heat exchange between the reformed gas leaving the outlet 36 of the reaction chamber and the process gas to be reformed in the said chamber was negligibly low. Therefore, in this alternative the upper portion 56 of the reaction chamber was heated only by the hot flue gas, while the lower portion 58 thereof was heated by both the moderately hot flue gas and the product stream.

V. The respondents submitted an amended set of claims on 24 March 1994 as well as a declaration by one of the inventors. This declaration included a computational comparison of the wall temperatures and lifetime of the second reactor tube for the co-current situation according to the claimed process and the counter-current situation of D7. On 19 December 1996, in reply to a communication from the board, the respondents filed an amended set of five claims, replacing the previous set of claims, as well as a further declaration by the same inventor. Oral proceedings were held on 28 January 1996. Claim 1 submitted on 19 December 1996 reads as follows:

"1. A process for the steam reforming of hydrocarbons in a reforming reactor by the passage of a feed stream comprising steam and one or more hydrocarbons as process gas under steam reforming conditions and under external supply of heat through a given volume of steam reforming catalyst, which catalyst is present in first and second separate catalyst compartments, to form a product stream rich in hydrogen, comprising the following successive steps:

(a) passing the process gas through the portion of the steam reforming catalyst in said first compartment containing 25-75% of said catalyst, heat needed for the endothermic reactions occurring in the process gas and for heating the process gas being supplied partly from a moderately hot flue gas as defined under (b) and partly from the product stream, and then

(b) passing the process gas partly reformed in step (a) through the steam reforming catalyst in said second compartment containing the remainder of said catalyst, the heat needed for the further endothermic reactions and heating of the process gas being supplied solely by a hot flue gas generated by combustion of a fluid fuel, the hot flue gas being thereby cooled to form the moderately hot flue gas employed in step (a)

wherein the moderately hot flue gas and the product stream, supplying the heat in step (a), are passing simultaneously and separately counter-currently in indirect exchange contact with the process gas passing step (a), whereas, the hot flue gas supplying the heat to the process gas in step (b) is passing co-currently in indirect heat exchange contact with the process gas passing step (b) to form the product stream."

VI. Regarding the issue of inventive step, the appellants put forward, *inter alia*, the following arguments:

The process of amended claim 1 did not involve an inventive step over the teaching of D7 and D2. It was known from D7 to heat the process gas in step (a) by passing the moderately hot flue gas and the product stream counter-currently to the process gas. Furthermore, the heating of the process gas in step (b) by the co-current flow of a hot flue gas was obvious in view of the general knowledge or of D2. In the conventional reformers used before the priority date, the necessary heat was supplied to the feed stream by a

co-current flow of hot flue gas. D7 disclosed for the first time performing the reforming process in two portions of the catalyst bed, wherein the process gas was heated in the lower portion ("pre-reformer") partly by a moderately hot flue gas and partly by the product stream, both flowing counter-currently to the process gas, while, in the upper portion ("fired-reformer"), the heat was supplied by a hot flue gas flowing counter-currently to the process gas. This led to high thermal efficiency and the apparatus was very compact. In D7 the hot flue gas was passed counter-currently to the process gas in the "fired-reformer" to achieve reactor compactness. If compactness was not required, then it would have been obvious to the skilled person to combine the co-current heating by the hot flue gas used in conventional reformers or disclosed in D2 with the counter-current heat transfer used in the "pre-reformer" section 58 of D7, since, on the one hand, D7 disclosed that the latter led to energy savings and, on the other hand, it was well known before the priority date that the problem of high maximum temperature on the tube wall could be solved by using co-current flow of the hot flue gas instead of counter-current flow.

VII. The respondents' arguments can be summarised as follows:

In the declaration filed on 19 December 1996, it was pointed out in particular that the apparatus of D7 had been commercially known for some time and that the problem encountered with such apparatus was the unstable heating effect of such a system leading to large fluctuations in the heat exposure of the metallic components, and therefore the unstable and unpredictable behaviour of those components. The problem underlying the present invention was to produce a much more stable and predictable heat flux leading to

high thermal efficiency whilst achieving a maximisation of component lifetime. It was not correct to characterise the starting point simplistically as the need to deal with high temperature exposure at the end of the catalyst chamber (point 36) in D7.

The respondents contested that co-current heating of the process gas with the hot flue gas was conventional in 1985 and pointed out that at that time the tubes received the necessary heat by radiation in conventional steam reformers. D2 did not represent the general knowledge. It was confirmed that the skilled person would have been aware before the priority date of the problems of unstable heating effect and difficult control of the maximum tube wall temperature arising with a counter-current flow of hot flue gas and would have known that the heating by means of a co-current flow did not have these drawbacks. The respondents contended that the appellants' submissions regarding the lack of inventive step in their letter dated 9 January 1995 were based on *ex post facto* analysis. The route to the present invention as suggested in this letter started from the wrongly oversimplified problem of high temperature at point 36 in Figure 1. It was complex and involved around six steps with an alternative choice at each step and with a jump in the logical chain of thought. D7 led away from the use of co-current heating by the hot flue gas, and clearly emphasised that heating by a counter-current flow of hot flue gas was critical. There was no teaching whatsoever leading to a combination of co-current flow with counter-current flow and there was no known way of making such a combination in a single steam reforming reactor before the priority date. Even if somehow a skilled person had decided to combine co- and counter-current flow in a single apparatus, he would still have had to come to the claimed specific combination of features, namely

two physically separated portions of catalyst; hot flue gas heating the second portion co-currently, with the remaining moderately hot flue gas heating the first portion counter-currently; and process gas heating the first portion counter-currently. There was clearly a huge jump involved in combining these features and no such combination was apparent from D7 and D2. In connection with D2, the respondents made reference to the passage at page 2, last sentence of the second paragraph, and pointed out that the process of D2 involved the use of a hot flue gas under pressure. The question was not whether the skilled person could have combined the co-current flow of hot flue gas with the counter-current flow but whether he would have done this.

VIII. The appellants requested that the decision under appeal be set aside and that the patent be revoked. The respondents requested that the decision under appeal be set aside and that the patent be maintained on the basis of the amended claims filed on 19 December 1996.

Reasons for the Decision

1. The appeal is admissible.

2. The amended claims are considered to meet the requirements of Article 123(2) and (3). In particular, claim 1 is based on the combination of claims 1 and 2 as filed with additional features disclosed in claim 6, at page 11 of the original description and in Figures 1 and 2. Apparatus claim 5 corresponds to a combination of the features recited in original claims 6, 7 and 8. The amendments do not broaden the scope of protection of the granted claims either.

3. None of the cited documents discloses a process or a reactor having the combination of features as defined in claims 1 and 5. Therefore the subject matter of these claims is new. This not being in dispute, there is no need to give further details.

4. Turning to the issue of inventive step, the board considers, in agreement with all the parties and the opposition division, that D7 represents the closest prior art.

This document discloses a process for the steam reforming of hydrocarbons in a reforming reactor by passing a feed stream comprising steam and a reformable hydrocarbon fuel as process gas under steam reforming conditions and under external supply of heat through a suitable catalyst bed to form a product stream rich in hydrogen. The reforming reactor comprises a lower portion 58 and an upper portion 56 containing the catalyst. The process gas is first passed through the catalyst in the lower portion 58 of the reactor, where it is heated partly by the moderately hot flue gas flowing counter-currently thereto through the annular flue gas passageway 64, and partly by the product stream passing through the annular regeneration chamber 48 counter-currently to the process gas. Then the process gas partly reformed in the lower portion 58 of the reactor further flows upward through the catalyst in the upper portion 56 of the reaction chamber, where it picks up the additional heat needed for the reaction in particular from the hot flue gas travelling counter-currently (see page 10, fourth paragraph; page 12, last paragraph; page 13, line 29 to page 15, line 13; Figures 1 and 2). When the plug 46 does not extend over the entire length of the reaction chamber, ie when a shorter plug is used, the process gas flowing through the upper portion 56 of the reaction chamber receives heat mainly from the hot flue

gas flowing counter-currently thereto (see page 12, lines 7 to 14). Half the length of the reactor used in the examples extends into the burner cavity. Conversion rates of 88% and 95% and overall reactor thermal efficiencies of 87% and 90% are achieved in examples 1 and 2 respectively.

4.1 As pointed out by the respondents and not contested by the appellants, the problem encountered with the process and apparatus of D7 is the unstable heating effect leading to large fluctuations in the heat exposure of the metallic components and thus in the maximum wall temperature, which in turn leads to the unpredictable behaviour, in particular as regards lifetime, of those components. Starting from D7 as the closest prior art, the problem underlying the patent in suit can be seen as the provision of a steam reforming process which makes it possible to maintain a high thermal efficiency whilst achieving a maximisation of the component lifetime.

The patent in suit proposes to solve this problem by a process comprising the combination of features as defined in the amended claim 1. This solution differs from the process of D7 in that (i) the hot flue gas supplying the heat to the process gas in step (b) is passing co-currently in indirect heat exchange contact with the process gas, (ii) the catalyst is present in two physically separated compartments, and (iii) the process gas is heated solely by the hot flue gas in step (b). In view, on the one hand, of the results given in the EPRI report of August 1987 as regards the stability of reformer temperatures, metal wall temperatures and overall thermal efficiency (see in particular pages S-14 and S-16, page 2-5, page 3-24

mentioning an efficiency of 86.8%, and page 4-1) and, on the other hand, of the computational comparison of the wall temperatures and lifetime submitted with the respondents' letter dated 22 March 1994, the board is satisfied that the technical problem has been solved by the claimed process.

4.2 According to both the appellants and the respondents, it was known to the skilled person in this field of chemical engineering before the priority date that heating the process gas by a counter-current flow of hot flue gas had drawbacks, namely the unstable heating effect and difficult control of the maximum wall temperatures, and that these disadvantages could be avoided by using a co-current flow of hot flue gas. In these circumstances, the skilled person would have inferred that the difficulties encountered with the process of D7 (see point 4.1 above) could be attributed to the use of a counter-current flow of hot flue gas for heating the process gas in particular in the relatively hot end of the reaction chamber. Therefore, in view of the teaching of D2, which discloses co-current heat transfer from the hot flue gas to the process gas, a skilled person aware of the advantages and drawbacks of counter-current and co-current heat transfer would have considered heating the process gas by a co-current flow of hot flue gas instead of a counter-current flow at least in the hot end (upper portion 56) of the reaction chamber of D7, since he would have expected that this substitution might solve the problem of unstable heating effect and difficult control of the maximum wall temperature.

However, taking into account (i) that the problem to be solved consists not only in maximising the lifetime of the metallic components but also in achieving a high thermal efficiency, and (ii) that D7 stresses the importance of using counter-current flows and narrow

annular heating gas passageways in the lower portion 58 of the reactor for maximising the heating rate and the reactor thermal efficiency (see page 8, line 17 to page 9, line 5 and page 14, line 24 to page 15, line 13), a skilled person would also have considered maintaining unchanged the counter-current heat transfer in the lower portion 58 of the reactor in order not to impair the reactor thermal efficiency. Therefore, a skilled person seeking a solution to the problem stated above would have contemplated replacing the counter-current flow of hot flue gas in the upper portion 56 of the reaction chamber by a co-current flow without changing the advantageous heat transfer by counter-current flow in the lower portion thereof.

4.3 As pointed out by the respondents, the cited prior art contains no information as to how the reaction chamber of D7 could be modified in order to achieve such a combination of co-current flow and counter-current flow of the hot flue gas in a reforming reactor with a single catalyst bed. However, a skilled person expecting that such a combination might solve the said technical problem would certainly not have been deterred from trying such a combination simply because a single catalyst bed was used in D7 or D2. He would have immediately realised that the physical separation of the upper portion 56 and the lower portion 58 of the catalyst chamber in D7 would have rendered such a combination possible. The board is not convinced that there were a great number of alternatives other than the physical separation of the catalyst bed into two compartments and that it would have involved an inventive skill to envisage a separation of the catalyst bed in view of D7 which already distinguishes between two different portions of the reaction chamber. It is noted in this context that the respondents did not mention other specific alternatives.

4.4 The respondents' arguments that D7 very strongly leads away from the use of co-current application of heat cannot be accepted by the board. According to the respondents, D7 at page 8, lines 19 to 21 and 24 to 27 very clearly teaches that only counter-current heating by flue gas must be utilised, and that the use of counter-current flow of flue gas is critical (see letter of 24 March 1994). However, D7 does not refer at page 8, lines 19 to 21 to the counter-current flow of flue gas as such, but to the counter-current flow of flue gas through a narrow annulus along the outer wall of the annular reaction chamber, and it is stated in this context in subsequent lines 24 to 27 that the use of counter-current flows and narrow annular heating gas passageways is critical to maximising heating rate and thermal efficiency. In the sentence bridging pages 8 and 9, it is again emphasised that the size of the annular passageways for the hot gases is critical to the heat transfer. It also derives from the statements at page 14, line 16 to page 15, line 13 that the counter-current flow of flue gas in the narrow passageway 64 over the lower portion 58 of the reactor is in particular critical for maximising heating rate and reactor thermal efficiency. The corresponding acceptable size of the flue gas passageway 64 is given at page 17. Therefore, the said passage at page 8 does not lead away from heating the process gas by a co-current flow of hot flue gas in the upper portion 56 of the reactor. The passage at page 15, line 31 to page 16, line 2 would also not have deterred the skilled person from combining co-current heating by the hot flue gas in the upper portion 56 of the reactor with counter-current heating in the lower portion thereof, since this passage relates to a prior art process wherein co-current heating is used along the whole length of the catalyst bed.

The respondents' arguments that several generally known options existed for lowering the maximum temperature at the outlet 36 of the reaction chamber of D7, such as lowering the temperature of the counter-current flow of flue gas, increasing the flow, using excess air or using recirculation, cannot change the findings in the preceding points, since this approach only takes into consideration the problem of too high a temperature at the outlet 36 without considering the problem of unstable heat flux, difficult control of the wall temperature and the need to maintain a high thermal efficiency as defined above.

The appellants referred to the passage in D2, at page 2, last sentence of the second paragraph, and emphasised that the process of D2 was performed using a flue gas under pressure. However, it is not clear to the board in what way this could have an influence on the outcome of the present decision as the wording of amended claim 1 does not exclude the use of a flue gas under pressure. Furthermore, the said passage concerns a prior art process in which the tubes are directly heated with the flame of the burners.

4.5 As regards the third feature distinguishing the claimed process from the process of D7, namely the heating of the process gas in step (b) "solely" by the hot flue gas, the respondents themselves acknowledged at the oral proceedings that this feature was of no significance for solving the technical problem stated above. Taking into account that in D7 the process gas in the upper portion 56 of the reaction chamber is mainly heated by the hot flue gas, in particular in the embodiment where a shorter plug 46 is used (see page 12, lines 7 to 14), a skilled person would obviously have inferred therefrom that the process gas might be heated "solely" by the hot flue gas in the said portion. This feature, which is considered to be

of no significance by the respondents, can only be regarded as an obvious alternative to the heating of the process gas "essentially" by the hot flue gas in that portion of the catalyst chamber.

4.6 It follows from the above that the claimed combination of features as defined in amended claim 1 does not meet the requirement of inventive step set out in Articles 52(1) and 56. For this reason the appellants' sole request cannot be allowed and the remaining claims of this request fall with claim 1.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: The Chairman:

P. Martorana P. A. M. Lançon