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D E C I S I O N
of 12 November 1996

Case Number: T 0134/94 - 3.3.2

Application Number: 86306229.5

Publication Number: 0213840

IPC: C01B 3/16

Language of the proceedings: EN

Title of invention:
Hydrogen production

Patentee:
IMPERIAL CHEMICAL INDUSTRIES PLC

Opponent:
Metallgesellschaft AG
LINDE AKTIENGESELLSCHAFT

Headword:
Hydrogen production/ICI

Relevant legal provisions:
EPC Art. 56, 87(1)

Keyword:
"Priority - not acknowledged for claim broadened over
disclosure of priority document"
"Inventive step - no"

Decisions cited:
T 0073/88; T 0212/88; T 0131/92

Catchword:

-



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Boards of Appeal

Chambres de recours

Case Number: T 0134/94 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 12 November 1996

Appellant:
(Opponent)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 30 December 1993
rejecting the oppositions filed against European
patent No. 0 213 840 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: F. Antony
Members: G. J. Wassenaar
J. van Moer

Summary of Facts and Submissions

- I. European patent No. 0 213 840 was granted in response to European patent application No. 86 306 229.5, filed on 12 August 1986, claiming British priority of 30 August 1985. The mention of the grant was published in European Patent Bulletin 91/42 of 16 October 1991.
- II. Notices of Opposition were filed against the European patent by the Appellant (Opponent 01) on 2 July 1992 and the party as of right (Opponent 02) on 16 July 1992. Revocation of the patent was requested on the grounds of lack of inventive step and insufficient disclosure (Articles 100(a) and (b) EPC).

The opposition was supported, inter alia, by the following documents:

Ullmanns Encyklopädie der technischen Chemie, 4th Edition, Vol. 14, pages 422-424 (D2)
DE-B-1 542 632 (D4)
GB-A-1 267 603 (D5)
EP-A-0 081 948 (D6).

Later in the opposition proceedings the following documents were mentioned:

EP-A-0 157 480 (D7)
AT-A-286 926 (D8).

- III. By its decision announced at the oral proceedings held on 14 December 1993 the Opposition Division rejected the oppositions. During the oral proceedings the objection of insufficient disclosure was withdrawn. In the Grounds for the decision, issued on 30 December 1993, the Opposition Division held that the closest prior art was represented by D5, which like the patent

in suit disclosed a "low temperature" (LT) process for the production of hydrogen in a tubular shift reactor. The problem underlying the invention was considered to reduce the amount of catalyst for the same gas flow rate. This problem was solved by the selection of a relatively high reaction outlet temperature range of 230 to 280°C. It was concluded that, since there was no incentive in the prior art documents to select this range in order to reduce the required amount of catalyst, the claimed subject matter was inventive.

- IV. An appeal against this decision was lodged by the Appellant on 9 February 1994. The Statement of Grounds was filed on 22 April 1994.

In the Statement of Grounds the Appellant contested the reasons given by the Opposition Division and argued that granted claim 1 lacked an inventive step over D8, whose content, apart from a clerical error, corresponded to that of D5.

It was stressed that D8 disclosed that the reaction should be performed at temperatures of 150 to 290°C, so that an outlet temperature of 230 to 280°C could be expected. Attention was further drawn to D2, disclosing that for the low temperature shift reaction copper catalysts are suitable in a temperature range of 210 to 270°C (page 423 bottom of right hand column). With respect to the gas flow rate required by claim 1, it was argued that the range of 200 to 800 kmol per hour per m³ of catalyst (kmol/h/m³) was comprised by the range of 0.1 to 5 m/s for the linear flow velocity mentioned in D8, if applied to reactor conditions used in practice.

V. In his counter-statement to the Statement of Grounds the Respondent (Proprietor) agreed that the value of 14 mm for the tube diameter in Example 2 of D5 was an error and should read 40 mm. It was submitted that according to the patent in suit it was sufficient to use about 25 to 30 m³ of shift catalyst in a 1000 ton/day (t/d) ammonia plant, in contrast to the 145 m³ mentioned in D2. The wet gas flow rate in Example 2 of D5 was calculated to be 121 kmol/h/m³ on the assumption that the linear velocity of 2m/s quoted in Example 2 related to the dry gas linear velocity corrected to standard temperature and pressure (NTP). Applied to a 1000 t/d ammonia plant the amount of catalyst employed would be about 75 m³, i.e. about the same amount of conventional low temperature shift catalyst (LT) taught by D2. The problem posed by D5 and solved by the patent in suit was reducing the amount of catalyst required in the process of D5. It was argued that D5 had a strong preference for the use of temperatures in the range of 180 to 230°C and to operate at temperatures favourable for equilibrium. The equilibrium temperatures for Examples 1 and 2 of D5 were calculated to be 300 and 330°C, respectively. A closer approach to equilibrium could be achieved by a lower space velocity. It was submitted that, therefore, the skilled person was not led to use greater space velocities, i.e. less catalyst, as this would result in a product being even further away from equilibrium. There would be no suggestion in D5 that by using higher temperatures far less catalyst could be employed. It was further argued that, although D5 suggested that a certain range of linear gas velocities could be used, there was nothing in D5 to suggest that the linear gas velocity and the tube length could be varied independently and that a

carbon monoxide content below 1% could be achieved at high linear velocities in short tubes, and consequently there was no suggestion that such a conversion could possibly be achieved at the wet gas flow rates specified in present claim 1.

VI. With letter dated 8 October 1996 the Respondent filed a new set of claims as auxiliary request and requested that, in the event the Board would hold that the granted claims were not entitled to the claimed priority, the existing claims for the designated states DE, FR, GB, IT and NL be replaced by the set of claims filed according to the said auxiliary request. Claim 1 thereof reads as follows:

"1. A process for the production of a hydrogen containing gas stream comprising subjecting a raw gas containing steam, carbon dioxide, hydrogen, and carbon monoxide, and having a carbon monoxide content of at least 8% by volume on a dry basis, to the catalytic shift reaction at a pressure over 30 bar abs. in a single stage in the presence of a catalyst containing copper metal and at least one oxidic support material at an outlet temperature in the range 230 to 280°C, said bed being equipped with heat exchange tubes and/or plates with boiling water as a coolant on the cold side of said tubes and/or plates, whereby to reduce the carbon monoxide content to less than 1% by volume on a dry basis
characterised in that

- (a) the heat exchange tubes and/or plates provide 50 to 200m² of heat exchange surface contacted by the gas stream within the catalyst bed per m³ of catalyst, and
- (b) the gas flow rate through the catalyst is in the range 200 to 800 kmol per hour per m³ of catalyst."

VII. In the oral proceedings held on 12 November 1996, the Respondent maintained that the granted claims were entitled to the claimed priority date so that D7 was only relevant for novelty. In particular, it was argued that, although the granted main claim was indeed broadened in comparison to the priority document in that the pressure requirement of at least 30 bar was dropped and the range of the heat exchange surface was extended from 50 to 200 m² to 30 to 200 m², the invention remained the same within the meaning of Article 87(1) EPC. If a priority document destroyed the novelty of an application based thereon, as in this case, the priority should be acknowledged. In support of this argument reference was made to decisions T 73/88 (OJ EPO 1992, 557) and T 212/88 (OJ EPO 1992, 28). After discussion and deliberation the Board announced the intermediate decision that the claims as granted were not entitled to the claimed priority date. Thereupon the Respondent deleted those claims and made the set of claims according to the former auxiliary request his sole request.

The parties agreed that, in Example 2 of D5, the reference to a tube diameter of "14 mm" was a clerical error and should read "40 mm", so that there was no need to introduce D8 into the proceedings. Inventive step was then discussed on the basis of documents D2, D5 and D6 only.

VIII. The Appellant requested that the decision under appeal be set aside and the patent be revoked in its entirety.

The Respondent requested that the decision under appeal be set aside and that the patent be maintained on the basis of Claims 1 to 9 of 8 October 1996 for the contracting states DE, FR, GB, IT and NL (the designations for the contracting states AT, BE, CH/LI, LU and SE having been withdrawn).

Reasons for the Decision

1. The appeal is admissible.

2. *Allowability of amendments*

Amended Claim 1 differs from Claim 1 as granted for the remaining contracting states, and in turn based on Claim 1 as originally filed, by a more detailed specification of the reaction conditions. The added reaction conditions, viz. a pressure of at least 30 bar abs. and a heat transfer surface of at least 50 m² per m³ catalyst, are based on the description as originally filed (see page 2, lines 31 to 33 and page 3, lines 7 to 8) and do not extend the protection conferred by the patent as granted. Present claim 1 is therefore in accordance with Article 123 EPC. In fact, no objections in this respect were raised by the Appellant.

3. *Priority*

3.1 The subject matter of amended claim 1 is completely disclosed in the priority document (see particularly page 2, paragraph 1), as now agreed by all parties. The claimed priority date of 30 August 1985 can thus be acknowledged.

3.2 While the Respondent's former main request, directed to the claims as granted, is no longer maintained, the reasons for the Board's intermediate decision disallowing the claimed priority for Claim 1 thereof are given below:

3.2.1 According to the priority document, the process of the invention comprises reacting a raw gas containing steam, carbon dioxide, hydrogen and carbon monoxide in a single stage over a catalyst containing copper metal under specific conditions indicated as features (a) to (d); see page 2, lines 6 to 13. Feature (a) requires that the pressure is over 30 bar abs., and feature (c) requires that the heat exchange surface in the catalytic bed is at least 50 m² per m³ of catalyst. These requirements did not appear in granted claim 1 of the patent in suit, so that the scope of the invention as defined thereby comprises areas which according to the priority document did not belong to the invention. Thus at the priority date the technical problem underlying the invention was considered to be solved only if each of the conditions (a) to (d) was satisfied. According to granted claim 1 features (a) and (c) were no longer necessary for solving the problem underlying the invention. Thus the invention defined by granted claim 1 was not the same as the invention defined in the priority document, hence the requirement of Article 87(1) EPC, that the priority right shall be enjoyed only "in respect of the same invention", was not fulfilled.

3.2.2 The decisions of the technical Boards of Appeal cited by the Respondent do not relate to situations in which the patent protection is extended with respect to the disclosure of the priority document: In T 73/88 the priority right was acknowledged in a case where the claims under consideration contained additional features, not disclosed in the priority document, which limited the extent of protection (points 2.3 to 2.5 and headnotes I and II).

In T 212/88 the priority right was acknowledged in a case where in claims 1 and 2 of the patent in suit a slightly different definition of essentially the same product was used (points 4.5 and 4.6).

3.2.3 The intermediate decision given in the present case is consistent with decision T 131/92 (point 5; not published in OJ EPO) where the priority was disallowed in a case where the definition of a molecular weight range of a class of polymers in the claim was extended with respect to the range disclosed in the priority document.

3.2.4 In summary, the Respondent's position, that the priority claim should be acknowledged whenever the disclosure of the priority document would destroy the novelty of the claims of the later application or patent ("novelty test"), has no basis in either the EPC or in the EPO case law. Such a position would imply that essential features of an invention disclosed in a priority document could be omitted in a later application based thereon without loss of priority right. However, if an essential feature of an invention is so omitted, the invention is no longer the same, i.e. the requirement of Article 87(1) EPC is not fulfilled.

4. *Inventive step*

In view of the claimed priority being acknowledged for the sole set of claims now in the case, D7 is prior art in accordance with Article 54(3) EPC and is not to be considered here (Article 56 EPC).

4.1 As agreed by all parties, the closest prior art is D5. This document, referred to in the description of the patent in suit, discloses a method of producing hydrogen which comprises reacting a gas containing

carbon monoxide with water vapour at a temperature of 150 to 290°C and a pressure of 10 to 30 bar in the presence of a copper containing catalyst in an externally cooled tubular reactor (claims 1 and 8). In Example 2 (including its reference to Example 1), a process is disclosed wherein a tubular reactor is used with 14 tubes having a diameter of 40 mm and a length of 4000 mm filled with a copper oxide containing catalyst and surrounded by boiling water at 200°C and 15 atm. gauge. The gas to be treated contains 15% by volume of CO and 85% by volume of nitrogen and enters the reactor at a temperature of 190°C with a linear velocity of 2 m/s. The discharged gas contains less than 0.2% by volume of CO. A tube diameter of 40 mm implies that the heat exchange surface is 100 m²/m³ catalyst as agreed by the Respondent (page 7, line 2 of Respondent's letter of 22 August 1994). According to the Respondent the reaction conditions disclosed in Example 2 further imply that the gas flow rate is 121 kmol/h/m³ catalyst (page 3, penultimate paragraph of Respondent's same letter). This rate was calculated on the assumption that the linear gas velocity of 2 m/s relates to the dry gas velocity reduced to NTP. This assumption is consistent with the space velocity and the linear velocity given in Example 1. The linear velocity of 2.1 m/s mentioned in Example 1 is only in agreement with the indicated dry gas space velocity of 1500 per hour if what is meant is the linear **dry** gas velocity. If the value of 2 m/s in Example 2 were the actual linear wet gas velocity at the prevailing pressure and temperature, the gas flow rate expressed in kmol/h/m³ could not be calculated, since the gas temperature and pressure are not disclosed in Example 2. On the assumption that the gas temperature and pressure in said Example 2 were about the same as those given for the cooling water, i.e. 200°C and 15 atm. gauge, and the value of 2 m/s were the actual

linear wet gas velocity, the gas flow rate would be about 1000 kmol/h/m³, which would be unreasonably high. Therefore the Board accepts the Respondent's submission that the gas flow rate in Example 2 of D5 is substantially lower than required by claim 1 of the patent in suit. Since the gas flow rate is expressed per m³ of catalyst, a lower gas flow rate is tantamount to a correspondingly higher quantity of catalyst for treating the same amount of gas.

- 4.2 On the above basis the Board agrees with the Respondent that the technical problem underlying the patent in suit was to reduce the required amount of catalyst in a shift reactor where the carbon monoxide content of a raw gas having a carbon monoxide content of at least 8% by volume on a dry basis is reduced to less than 1% by volume.

According to claim 1 this problem is to be solved by performing the shift reaction at a pressure over 30 bar abs., at a reactor outlet temperature of 230 to 280°C, with a heat exchange surface of 50 to 200 m² per m³ of catalyst and a gas flow rate in the range of 200 to 800 kmol/h/m³.

According to the Example reported on page 5, lines 19 to 49, of the patent in suit, the CO content of about 12% (406/406+198+1631+23+1107+42) in a raw gas has been reduced to 0.5% (19/19+585+2018+23+1107+42) at a gas flow rate of 394 kmol/h/m³. According to the second Example (commencing page 5, line 50) the CO content of about 14% (450/450+154+1675+20+830+10) in a raw gas has been reduced to 0.56% (20/20+584+2105+20+830+10) at a gas flow rate of 303 kmol/h/m³. These flow rates are substantially higher than the flow rate calculated for

Example 2 of D5, which means that the amount of catalyst to treat the same amount of gas is indeed reduced. Therefore, the Board is satisfied that the claimed process actually solves the above mentioned technical problem.

4.3 It remains to be decided whether or not, for solving the above stated problem, it was obvious to apply the reaction conditions mentioned in claim 1.

4.3.1 In Example 2 of D5, heat exchange tubes with a diameter of 40 mm are used. As shown above, the heat exchange surface of such tubes is equal to $100 \text{ m}^2/\text{m}^3$ of catalyst; in other words feature (a) of claim 1 is already fulfilled by Example 2 of D5. The only relevant features of claim 1 not disclosed by D5 are a pressure above 30 bar abs. (see point 4.3.3 below), an outlet temperature in the range 230 to 280°C (see point 4.3.4 below) and a gas flow rate in the range 200 to 800 $\text{kmol}/\text{h}/\text{m}^3$ of catalyst (see point 4.3.2 below).

4.3.2 Because of the inverse proportional relationship between the amount of catalyst and the gas flow rate it is indispensable to increase the gas flow rate if the amount of catalyst is to be reduced. While the gas flow rate as such is not mentioned in D5, the linear flow velocity is, and so is its relationship to the degree of conversion. It is indicated that the upper limit of the flow velocity is determined by the desired degree of conversion and is normally about 5.0 m/s for complete conversion. In the manufacture of hydrogen for synthesis gas the flow velocity should not exceed 5 m/s (page 1, lines 71-78). According to Example 2 of D5, a CO conversion from an initial 15% to a final 0.2% content by volume is achieved at a linear velocity of 2 m/s. Considering the aforementioned relationship between degree of conversion and linear velocity it was obvious to a skilled person that using the same

apparatus as in said Example 2 the linear velocity could be increased substantially if a CO content of up to 1% by volume is tolerated as per claim 1 of the patent in suit. When wanting to reduce the amount of catalyst in a shift reactor according to Example 2 of D5 for the manufacture of hydrogen with a CO content of up to 1% by volume, the skilled person would, in view of the upper limit of 5 m/s for the manufacture of hydrogen for synthesis gas mentioned in D5, take into consideration linear velocities of up to 5 m/s. On the basis of the Respondent's assumptions mentioned under point 4.1 above, a linear velocity of 5 m/s in said reactor would correspond to $5/2 \cdot 121 \text{ kmol/h/m}^3 = 303 \text{ kmol/h/m}^3$. Thus the skilled person was aware that an increased gas flow rate was necessary to reduce the catalyst requirement, and that gas flow rates as claimed were achievable using conventional tube reactors under conventional shift reactor conditions, provided a CO content of up to 1% by volume was acceptable.

- 4.3.3 According to D5 the pressure in the reactor should preferably be between 10 and 30 bar (claim 8). Since the gas flow rate is expressed in kmol/h/m^3 , it is increased if the amount of gas is increased. Furthermore it is common general knowledge that in gas reactions the amount of gas passing through a reactor is proportional to the pressure of the gas. Thus, in order to reduce the required amount of catalyst, i.e. to increase the gas flow rate, it was obvious to increase the pressure. Pressures up to 40 bar are regarded as conventional in the art (cf. D4, column 3, lines 11-13). Therefore, in order to reduce the required amount of catalyst, it was obvious for the skilled person to use pressures over 30 bar abs.

4.3.4 An outlet temperature is not disclosed in Example 2 of D5. Disclosed is a temperature of 200°C for the cooling water, which implies that the outlet temperature is at least 200°C. While the outlet temperature in said example is probably below 230°C, the teaching of D5 is not limited to this example. According to the general teaching of D5, the reactor temperature may be up to 290°C, the range of 180 to 230°C being preferred. The reason for this preference is not explained but it is probably related to the nature of the catalyst. It is common general knowledge evidenced by D2 that the shift reaction using copper catalysts should be performed at temperatures between 210 and 270°C, the upper limit being dictated by a degeneration of the catalyst at higher temperatures (cf. D2, page 423, last paragraph to page 424, first paragraph). Allowing that the outlet temperature is generally 10 to 30°C lower than the maximum temperature in the reactor (patent specification, page 3, lines 36-39), a maximum temperature of at least 270°C as envisaged by both D2 and D5, implies an outlet temperature of more than 230°C. Looked at from another angle, as pointed out by the Respondent himself, it is common general knowledge that the lower the reaction temperature, the lower the rate of reaction, hence the more catalyst is required to effect a given rate of reaction. Conversely, a skilled person wanting to reduce the required amount of catalyst will perform the reaction at a temperature as high as other circumstances permit, the upper limit set by thermodynamical considerations being the equilibrium temperature to obtain the required CO reduction. As submitted by the Respondent the equilibrium temperature in Example 2 of D5 is 330°C; there was thus no thermodynamical reason not to increase the reaction temperature, and while there is no positive suggestion in D5 that by using higher temperatures than those said

to be preferred the space velocity or gas flow rate could be significantly increased, there is certainly no teaching in D5 which would deter a skilled person from using reaction temperatures over the whole range of 150 to 290°C disclosed as suitable.

- 4.4 In summary, no inventive feature or surprising result is apparent to the Board. The solution to the existing problem, viz. reduction in required shift catalyst, is the result of routine optimisation measures at the expense of reduced CO conversion and 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:

1. The decisions under appeal is set aside.
2. The patent is revoked.

The Registrar:

P. Martorana

The Chairman:

F. Antony