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Bezeichnung der Erfindung: Ammonia production process
Title of invention:
Titre de l'invention :

Klassifikation / Classification / Classement : C 01 G 1/04

ENTSCHEIDUNG / DECISION
vom / of / du 19 May 1988

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent / Titulaire du brevet : Imperial Chemical Industries PLC

Einsprechender / Opponent / Opposant :
OI M W Kellogg Company
OII Exxon Corporation
OIII Linde AG

Stichwort / Headword / Référence : Ammonia process/ICI

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step - obvious combination of the prior art" "Person skilled in the art - team".

Leitsatz / Headnote / Sommaire

Case Number : T 57 /86 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1
of 19 May 1988

Appellant :
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Fourth party to the proceedings (Opponent III)	Linde AG Zentrale Patentabteilung D-8023 Höllriegelskreuth
Decision under appeal :	**Decision of Opposition Division of the European Patent Office of 24 October 1985 posted on 19 December 1985 revoking European patent No. 0 000 993 pursuant to Article 102(1) EPC.**

Composition of the Board :

Chairman : K.J.A. Jahn
Members : R.W. Andrews
R.L.J. Schulte

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 0 000 993 in respect of European patent application No. 78 300 276.9, filed on 10 August 1978 and claiming priority of 22 August 1977, 27 October 1977 and 28 October 1977 from three prior applications filed in the United Kingdom, was announced on 8 December 1982 (cf. Bulletin 82/49) on the basis of ten claims.

II. Notices of opposition were filed on 2 September 1983, 6 September 1983 and 8 September 1983 in which the revocation of the patent on the grounds that its subject-matter was not new and did not involve an inventive step was requested. It was also alleged by one of the Respondents that the patent did not disclose the invention in a manner sufficiently clear for it to be carried out by a skilled person. The oppositions were supported by, inter alia, the following documents:

(1) US-A-3 441 393
(2) Nitrogen, No. 102, pages 35 to 39, July/August 1976
(8) GB-A-1 156 002 and its equivalent
(10) US-A-3 442 613
(13) Hydrocarbon Processing, Volume 46, pages 197 to 202, 1967
(14) Nitrogen, No. 100, pages 71 to 75, March/April 1976 and
(17) GB-A-1 274 504.

III. By a decision of 24 October 1985, posted on 19 December 1985, the Opposition Division revoked the patent. The contested decision acknowledged that the subject-matter of the patent in suit was novel but considered it was obvious to combine the teachings of documents (10) and (2) to solve the technical problem as formulated by the Opposition Division of overcoming the disadvantages resulting from having a stoichiometric excess of nitrogen in the ammonia synthesis loop.

IV. A notice of appeal was lodged against this decision on 12 February 1986 with payment of the prescribed fee. A statement of grounds of appeal was filed on 14 April 1986.

In this statement and during the oral proceedings held on 19 May 1988 the Appellant argued that the claimed combination of process sequence and conditions represents a non-obvious selection of, inter alia, a limited degree of compression of the make-up gas and a hydrogen-equivalent to nitrogen molar ratio in the secondary reformer outlet gas of between 2.2 and 2.7 from the prior art. Thus, document (10) discloses the use of a large excess of air in the secondary reforming stage to reduce the fuel requirements in the primary reforming stage (hereinafter Purifier Process). This results in, for example, a hydrogen-equivalent to nitrogen molar ratio of 2.13 in the secondary reformer outlet gas. The excess of nitrogen together with the inerts in the make-up gas are removed in cryogenic purifier prior to compression to synthesis pressure. Although this process yields a high purity synthesis gas with greatly reduced purge requirements and consequently an increase in the partial pressures of the reactants in the synthesis loop it is necessary to expand all the make-up gas in the cryogenic unit with resulting pressure loss of, for example, 4.5 bar.

Document (2) teaches the use of a small excess of air in the secondary reforming stage such as to give a hydrogen-equivalent to nitrogen molar ratio of, for example, 2.84, to compensate for the nitrogen deficiency otherwise resulting from the hydrogen recovery and recycle from the purge of the ammonia synthesis loop (hereinafter Petrocarbon Process). This cryogenic process, which is applied to an ammonia-free purge, removes inerts from the purge gas and enables hydrogen and some nitrogen to be returned to the synthesis loop.

In the Appellant's view the skilled person would be deterred from combining the Purifier and Petrocarbon Processes since the advantage of low purge requirement of the Purifier Process would be lost, the hydrogen recovery unit would have to be much larger than taught by document (2) and an increase in compressive power would be required. The Appellant considered that this opinion is also supported by the fact that nobody considered modifying the Purifier and Petrocarbon Processes in the ten years between their disclosure in 1967 and the priority date of the disputed patent even though there was a great deal of activity in this technological field during this period and it also included the large increases in oil prices of the early seventies.

The Appellant also took the view that in the assessment of inventive step consideration should be given as to who is to be regarded as the skilled person. He argued that in the present case the skilled person would be a process engineer. Unlike the design engineer who would undertake the design of completely new ammonia production plants, a process engineer is only capable of making minor modifications to existing plants to cater for the requirements of prospective customers. Since the changes necessary to arrive at the present invention are not of a minor nature they would not have occurred to the notional skilled person as defined above.

V. Respondent I contended that the claimed combination of process sequence and conditions is merely an aggregation of the teaching of the prior art selected to optimise the economics of the whole process by adjusting the process conditions of the individual stages. This involved changing the nitrogen rejection step of the Purifier Process from before the ammonia reactor to after it. However this operational procedure is applied in the Petrocarbon Process. Furthermore the characterising features of Claim 1 can be derived from the teachings of document (2) and document (8) or its equivalent (10) and (13). Furthermore, in the Respondent's view the process variation according to Claim 1 (set D) is disclosed in Hydrocarbon Processing, pages 103 to 106, January 1976 (18).

Respondent II argued that the claimed invention is a result of the optimisation of a process derived by the permissible combination of the whole disclosure of documents (2) and (10). Although values for the hydrogen-equivalent to nitrogen molar ratio of the secondary reformer outlet in the range 2.2 to 2.7 are not explicitly disclosed in the prior art, no evidence has been provided by the Appellant to demonstrate any advantages for the claimed range. Therefore no distinction with regard any effects can be drawn between the claimed values of 2.2 and 2.7 and the known values of 2.13 and 2.84. Moreover the claimed process provides those benefits as would be expected by a combination of the processes of documents (2) and (10).

VI. The Appellant requested that the decision under appeal be set aside and the patent maintained on the basis of Claims 1 to 9 (Set C) filed on 2 April 1988 or Claims 1 to 8 (Set D) filed 2 April 1988. Claim 1 (Set C) reads as follows:

"An ammonia production process which comprises:

(a) primary catalytically reforming at superatmospheric pressure a hydrocarbon feedstock with steam in conditions of steam-to-carbon ratio, pressure, and temperature, to produce a primary reformed gas containing carbon oxides, hydrogen, and at least 10% v/v methane on a dry basis;

(b) secondary catalytically reforming the primary reformed gas by introducing air and bringing the mixture towards equilibrium, whereby to produce a gas stream containing nitrogen, carbon oxides, hydrogen, and a decreased quantity of methane, the quantity of air employed being such as to introduce an excess of nitrogen over that required for ammonia synthesis;

(c) converting carbon monoxide in the gas stream catalytically with steam to carbon dioxide and hydrogen;

(d) removing carbon oxides from the gas stream;

(e) removing non-reacting gases and said excess of nitrogen; and

(f) after increasing the pressure of the gas stream to ammonia synthesis pressure, and after said removal of carbon oxides, in a recycle loop synthesising ammonia from the hydrogen and nitrogen in the gas stream and separating ammonia from the reacted gas; characterised by:

(i) operating the primary reforming stage at a steam-to-carbon ratio in the range 2.5 to 3.5, an outlet pressure in the range 30 to 120 bar abs and an outlet temperature in the range 750°C to 850°C;

(ii) using such a quantity of air in the catalytic secondary reforming step that the secondary reformer outlet gas has a hydrogen-equivalent : nitrogen molar ratio of 2.2 to 2.7;

(iii) limiting the extent of said pressure increase by compressing the gas stream, while still including said excess of nitrogen, by not more than 100 bar prior to ammonia synthesis; and

(iv) effecting the removal of non-reacting gases and said excess of nitrogen after introduction of said gas stream to the synthesis loop by:

treating reacted gas, after said separation of ammonia therefrom, to separate a hydrogen-enriched stream and a hydrogen-depleted stream containing said non-reacting gases and excess of nitrogen,

discarding the hydrogen-depleted stream from the loop, and

returning the hydrogen-enriched stream to the ammonia synthesis.

Claim 1 (Set D) in accordance with the auxiliary request differs from the above claim only insofar as in step (f) of the precharacterising part of the claim the recycle loop is defined as comprising a synthesis catalyst, an ammonia separator and a circulator and in step (iv) of the characterising part the non-reacting gases and excess nitrogen are removed by treating part of the reacted gases leaving the circulator and the resulting hydrogen-enriched stream is returned to the inlet of said circulator.

Both Respondents requests that the appeal be dismissed.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. There are no formal objections under Article 123 EPC to any of the versions of the present claims since they are all adequately supported by the original disclosure and do not extend the scope of protection conferred. In view of the later findings no detailed substantiation of this is required.

3. The patent in suit relates to a process for the production of ammonia from a hydrocarbon feedstock comprising reacting a substantially stoichiometric mixture of hydrogen and nitrogen obtained by the following steps:

   (a) primary catalytically steam reforming the hydrocarbon feedstock;
   (b) reacting the primary reformed gas with such an amount of air as to introduce an excess of nitrogen over that required for ammonia synthesis;
   (c) catalytically converting carbon monoxide in the secondary reformed outlet gas to carbon dioxide;
   (d) removing carbon oxides from the resulting gas stream; and
   (e) removing non-reacting gases and said excess nitrogen.

   Such a process is the one referred to above as the Purifier Process. This process is described in document (10) and the corresponding British patent specification, document (8), and documents (13) and (14). However, a disadvantage of this prior art process was considered to lie in the fact that the excess nitrogen resulting from the use of the excess air in the secondary reforming stage together with the non-reacting gases are removed in a cryogenic purifier from the main stream of the make-up gas supplied to the synthesis loop. Thus, any failure of this cryogenic unit or its shut down for necessary maintenance results in the closure of the whole plant since no make-up gas can be supplied to the synthesis loop. Moreover, in view of the significant pressure drop across the cryogenic purifier, the amount of compression of the make-up gas prior to synthesis is greater than that required in the standard ammonia synthesis process such as that disclosed in document (1).

4. In the light of this prior art the technical problem underlying the disputed patent may be seen in providing a process for the production of ammonia from a hydrocarbon feedstock involving the use of excess air in the secondary reforming stage in which the disadvantages of this prior art process are overcome, especially to reduce the energy consumption.

According to the patent in suit this problem is essentially solved by selecting particular primary and secondary reforming conditions including the use of such an amount of air in the secondary reforming stage that the secondary reforming gas outlet has a hydrogen-equivalent to nitrogen molar ratio of 2.2 to 2.7, limiting the amount of post-reforming pressure increase to not more than 100 bar, removing the excess nitrogen and non-reacting gases from the loop as a purge and recovering and recycling hydrogen from the purge.

5. In the Purifier Process as disclosed in the above-mentioned documents excess air is used in the secondary reforming stage in order to reduce the fuel requirements in the

primary reforming stage (cf. (8), page 2, lines 61 to 71, (10) Col. 3, lines 61 to 69, (13) 2nd paragraph in the right-hand column of page 197 and (14) 4th paragraph in the left-hand column of page 71. Since the disclosure of these documents is directed to one and the same process, for the sake of succinctness only a single document will be referred to in the following).

In accordance with document (8) the amount of excess air used in the secondary reforming stage is sufficient to provide a stoichiometric excess of nitrogen from 2 to 150 mole percent (cf. page 2, line 114), with an optimum for the particular design of 53 mole percent (cf. page 2, lines 109 to 113). From the example of document (8) it has been calculated that the hydrogen-equivalent to nitrogen molar ratio in the secondary reformer outlet gas is 2.15, i.e. as exemplified and put into practice the Purifier Process uses a larger excess of air in the secondary reformer stage than the presently claimed process. However, it is clearly taught in document (8) that once the stoichiometric molar ratio of hydrogen to nitrogen of 3:1 is exceeded, the determining quantity of process air in the secondary reforming stage is dependent on various factors. Thus, the reduction in reforming energy has to be balanced against the increase in compression energy. The amount of air used in the secondary reforming stage influences the amount of methane, a diluent in the synthesis gas, in the effluent gas from the secondary reformer. The optimum amount of air is determined by striking an economic balance between those factors resulting from the use of excess air and other cost factors. Therefore, document (8) teaches that the exemplified value of 2.15 for the hydrogen-equivalent to nitrogen molar ratio in the secondary reformer outlet is to be considered as the optimum value for the process as exemplified and that any alterations to the process would entail an optimisation of the amount of excess air used in the secondary reforming stage.

5.1 A further process in which an amount of air is employed in the secondary reformer so as to introduce an excess of nitrogen over that required for ammonia synthesis is the Petrocarbon Process, disclosed in documents (2), (17) and (18). An excess of air is used in the secondary reforming stage to make-up for the nitrogen deficiency in the treated purge gas resulting from the recycling of the hydrogen recovered from the ammonia synthesis purge gas (cf. (2) 2nd complete paragraph in the right-hand column of page 38 in combination with Figure 2 on page 104 of (18)). From the Example in document (17) it has been calculated that the hydrogen-equivalent to nitrogen mole ratio in the secondary reformer outlet is 2.84, i.e. less air is used in the secondary reforming stage than in the claimed process.

Therefore, the claimed range of 2.2 to 2.7 for the hydrogen-equivalent to nitrogen molar ratio in the secondary reformer gas outlet falls between these two exemplified values but completely within the broad range disclosed in document (8) and it is considered to lie within the competence of the skilled person to optimise this parameter having regard to the other process steps, particularly in view of the encouragement to do so provided by document (8).

5.2 The conditions specified for operating the primary reforming stage are, in general, known. Thus, document (1), which relates to the conventional process for the production of ammonia from a hydrocarbon feedstock, discloses steam to carbon ratios of 2.5 and 3.5 (Col. 10, line 17), operating pressures of about 29 to 53 bar (400 to 700 psig) and effluent temperature, of 704-899°C (1300 to 1600°F; Col. 9, lines 61 to 72). It is also known in the art that methane conversion by catalytic steam reforming is favoured by high temperatures and low pressures and that in conventional ammonia plants the primary reforming stage is usually operated under conditions such that the methane concentration in outlet from this stage is in the region of 10%. However, since by using an excess of air in the secondary reforming stage a higher concentration of methane can be tolerated in the primary reformer outlet gas (cf. (14) 3rd complete paragraph in the right-hand column of page 72 and the paragraph bridging pages 72 and 73), the skilled person would realise that by using higher reforming pressure the total compression requirements would be reduced.

5.3 Furthermore it is also known in the art that although the conversion of hydrogen and nitrogen to ammonia is favoured by high pressures, for example above 150 bar, it is possible to operate at lower pressures if a lower rate of conversion to ammonia can be accepted. Therefore, a skilled person would immediately realise that, in order to limit the extent of the pressure increase of the make-up gas prior to ammonia synthesis to, for example, not more than 100 bar, a compromise must be reached between the contradictory requirements of low pressures for the primary reforming stage and high pressure for the ammonia synthesis. It must be considered to be part of the skilled person's routine duties to determine the optimum solution to this problem.

5.4 The Petrocarbon Process, which is described in documents (2), (17) and (18), comprises an ammonia production process in which hydrogen and some nitrogen are recovered from the ammonia synthesis purge gas and recycled to the synthesis loop. As illustrated in Figure 2 on page 104 of document (18), after removal of ammonia, some of the reacted gases are removed from the synthesis loop and treated in a cryogenic unit to yield a hydrogen-enriched stream comprising 90 to 91% hydrogen, less than 1% argon and methane and the balance nitrogen (cf (2) 2nd complete paragraph in the right-hand column of page 38) and a hydrogen depleted stream which is discarded from the loop and used as fuel gas. The Petrocarbon Process was designed for the addition to ammonia production processes as a means of increasing overall ammonia production efficiency by either increasing ammonia capacity or reducing feedstock consumption of the plant.

Although document (2) and (18) suggest the application of the Petrocarbon Process to conventional ammonia production processes such as that disclosed in document (1), nevertheless, a skilled person confronted with the problem underlying the patent in suit would realise that the combination of the Petrocarbon Process with the Purifier Process would overcome the disadvantage of the Purifier Process resulting from the siting of the cryogenic unit in the main stream of the make-up gas. By making this combination it would be clear to the skilled person that the advantage of the greatly reduced purge requirement afforded by the Purifier Process would be lost but the advantages of reduced fuel requirements in the primary reforming stage resulting from the use of an excess of air in the secondary reforming stage and of increased over-all ammonia synthesis efficiency from the recycling of recovered hydrogen would be retained.

The Appellant's arguments that the Petrocarbon and Purifier processes cannot be fairly combined is only valid if the skilled person were to make the combination solely on the basis of the process as exemplified in the relevant cited literature. Thus, the skilled person is not obliged to use such an amount of air in the secondary reforming stage that the hydrogen-equivalent to nitrogen molar ratio in the secondary reformer outlet gas is 2.15. He would, as a matter of course, optimise the amount of air used in the secondary reforming stage within the limits taught in document (8) for his particular design, which would also include the hydrogen recovery stage. In fact it is clearly taught in document (8) that this optimisation should be performed (cf. page 2, lines 95 to 109).

Furthermore, it would be obvious to the skilled person that the economics of the process would be further improved if the pressure increase between the reforming stage and ammonia synthesis stage were to be limited. It would lie within the competence of the skilled person to determine this limit for a process in which the reforming and ammonia synthesis stages are operated under conditions known *per se*.

5.5 An indication for the presence of an inventive step may be the lapse of time between the publication date of the relevant prior art and the priority date of a patent, particularly, if during this period of time an urgent need for improvement has been shown to exist. Although in the present case the large increases in oil prices in the early seventies may be considered to provide a stimulus for the development of processes with reduced hydrocarbon feedstock requirements, the lapse of ten years cannot be considered to be a long time in the field of bulk chemical production. It is a matter of common experience that the time required to design and construct a large chemical plant, for example a 1000 tonne per day ammonia production plant, amounts to several years. Moreover, in view of the large capital investment involved in such plants, provided the plants operate in a satisfactory manner there is no incentive to consider their modification.

5.6 With respect to the disputed patent the skilled person must be presumed to be a process engineer aware of the Purifier and Petrocarbon Processes and what was common general knowledge in the art of ammonia production plants. It is established jurisprudence that, in his search for the solution to his problem a skilled person may consult a skilled person in another technical field. Furthermore, the skilled person may even be thought of in terms of a group of persons forming a team. Therefore the Board cannot agree with the Appellant that a distinction should be drawn between the level of skill of a process engineer and that of a design engineer, since, if the problem underlying the disputed patent was such as to be outside the range of duties normally entrusted to a design engineer, the assessment of whether the proposed solution involves an inventive step must be based on the knowledge and ability of the appropriate skilled person, i.e. a design engineer.

5.7 In the Board's judgement it would have been obvious to this skilled person that the solution to the problem underlying the patent in suit of overcoming the disadvantages of the Purifier Process would lie in its modification in the light of his knowledge of the Petrocarbon Process and the optimisation of the operating conditions of the resulting process. Therefore, the subject-matter of Claim 1 in accordance with the main request does not involve an inventive step.

5.8 The additional features of Claim 1 in accordance with the auxiliary request requiring that the non-reacting gases and excess nitrogen are removed by treating part of the reacted gases leaving the circulator and that the hydrogen-enriched stream so obtained is returned to the inlet of said circulator do not render the subject-matter of this claim inventive.

In Figure 2 on page 104 of document (18) part of the reacted gases leaving the circulator in the synthesis loop is removed and treated in a cryogenic unit to recover a hydrogen-enriched stream which is returned to the inlet of the synthesis gas compressor. However, it would be immediately clear to the skilled person that, if the pressure of the hydrogen-enriched stream and the make-up gas is only slightly lower than the pressure in the synthesis loop, the necessary compression of this gas stream could be provided by the increase in pressure encountered in the synthesis loop circulator; thus obviating the need for an additional synthesis gas compressor.

5.9 Dependent Claims 2 to 9 and 2 to 8 in accordance with the main and auxiliary requests, relating to preferred embodiments of the processes of the respective main claims, do not contain any independent inventive features and are, therefore, unpatentable in the absence of a corresponding allowable main claim.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar

The Chairman