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**Datasheet for the decision
of 20 February 2009**

Case Number: T 0851/06 - 3.3.05

Application Number: 01101536.9

Publication Number: 1162170

IPC: C01B 3/38

Language of the proceedings: EN

Title of invention:

Method of manufacturing a synthesis gas to be employed for the synthesis of gasoline, kerosene and gas oil

Patentee:

MITSUBISHI HEAVY INDUSTRIES, LTD.

Opponent:

L'AIR LIQUIDE. S.A. A DIRECTOIRE ET CONSEIL DE SURVEILLANCE
POUR L'ETUDE ET L'EXPLOITATION DES PROCEDES GEORGES CLAUDE

Headword:

Synthesis gas/MITSUBISHI

Relevant legal provisions:

EPC Art. 123(3), 56
RPBA Art. 13(1) and 15(3)

Relevant legal provisions (EPC 1973):

-

Keyword:

"Amendments (allowable)"
"Inventive step (yes): reformulated problem derivable from description"

Decisions cited:

-

Catchword:

-



Case Number: T 0851/06 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 20 February 2009

Appellant:
(Opponent)
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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
19 April 2006 concerning maintenance of
European patent No. 1162170 in amended form.

Composition of the Board:

Chairman: G. Rath
Members: J.-M. Schwaller
S. Hoffmann

Summary of Facts and Submissions

I. This appeal was lodged by the opponent (hereinafter "the appellant") against the interlocutory decision of the opposition division to maintain the European patent 1 162 170 on the basis of the set of claims according to the auxiliary request filed during oral proceedings on 14 March 2006, independent claims 1 and 5 of which read as follows:

"1. A method of manufacturing a synthesis gas containing CO and H₂, which is suited for use in synthesizing gasoline, kerosene and gas oil by way of the Fisher-Tropsch reaction system; the method comprising the steps of:

feeding a steam-mixed natural gas to a reformer (10) which is provided with a combustion radiation portion (12) for burning a fuel, the reformer (10) being designed to be heated by the combustion radiation portion (12);

recovering carbon dioxide from a combustion exhaust gas generated at the combustion radiation portion (12); and

adding the carbon dioxide to the steam-mixed natural gas at a location on an upstream side of the reformer (10),

thereby allowing a reforming reaction to take place to obtain a synthesis gas comprising CO and H₂ at a molar ratio of H₂/CO = 1 to 2.5, which is suited for use in synthesizing gasoline, kerosene and gas oil,

characterized in that a preliminary reformer (36) is arranged at an upstream of the reformer (10), the natural gas to which steam has been added is supplied to the reformer (10) via the preliminary reformer (36), the carbon dioxide recovered from the combustion

exhaust is fed to a passageway connecting the reformer (10) and the preliminary reformer (36), and in that the step of adding the carbon dioxide to the steam-mixed natural gas is performed by arranging a moistening device (37) at a posterior stage of the reformer (10), introducing a synthesis gas from the reformer (10) into the moistening device (37), heating the moistening device (37) with waste heat of the synthesis gas, feeding natural gas and water to the moistening device (37), and adding steam to the natural gas in the moistening device (37).

5. A method of manufacturing a synthesis gas comprising CO and H₂, which is suited for use in synthesizing gasoline, kerosene and gas oil by way of the Fisher-Tropsch reaction system; the method comprising the steps of:

feeding a steam-mixed natural gas to a reformer (110) which is provided with a combustion radiation portion (112) for burning a fuel, the reformer (110) being designed to be heated by the combustion radiation portion (112);

recovering carbon dioxide from a combustion exhaust gas generated at the combustion radiation portion (112);

adding the carbon dioxide to the steam-mixed natural gas at a location on an upstream side of the reformer (110), thereby allowing a reforming reaction to take place; and

introducing a reformed gas from the reformer (110) into a partial oxidation furnace (133) simultaneous with an introduction of oxygen into the partial oxidation furnace (133), thereby allowing the reformed gas to react with the oxygen to obtain a synthesis gas comprising CO and H₂ at a molar ratio H₂/CO = 1 to 2.5,

which is suited for use in synthesizing gasoline, kerosene and gas oil, characterized in that a preliminary reformer (138) is arranged at an upstream of the reformer (110), the natural gas to which steam has been added is supplied to the reformer (110) via said preliminary reformer (138), the carbon dioxide recovered from the combustion exhaust is fed to a passageway connecting the reformer (110) and the preliminary reformer (138), and in that the step of adding the carbon dioxide to the steam-mixed natural gas is performed by arranging a moistening device (139) at a posterior stage of the reformer (110), introducing a synthesis gas from the reformer (110) into the moistening device (139), heating the moistening device (139) with waste heat of the synthesis gas, feeding natural gas and water to the moistening device (139), and adding steam to the natural gas in the moistening device (139)."

II. In the contested decision, which *inter alia* relied upon the following documents:

D2: "A new process to make oxo-feed", S. Teuner, Hydrocarbon Processing, July 1987, page 52,

D3: WO 00/09441,

D6: US 5 264 202,

the opposition division held that the subject-matter of independent claims 1 and 5 as maintained involved an inventive step in essence because the use of a moistening device in the process claimed solved the

problem of reducing the quantity of steam to be fed to the reformer.

Document D3, which required a saturator in its reforming process, did not disclose to contact the gas feed with steam but merely with heated water. Combining the teaching of D3 with that of documents D2 and D6 would thus not lead the skilled person to the subject-matter of said claim 1 or 5.

III. With its statement setting out the grounds of appeal, the appellant submitted two new documents:

D8: EP 0 970 939 A1,

D9: US 5 063 250,

and objected to the subject-matter of independent claims 1 and 5 as maintained as lacking an inventive step over a combination of documents D2, D6 and D8 (or D9). It also objected to the subject-matter of these claims as lacking clarity, arguing that the sentence beginning with "*the step of adding the carbon dioxide to the steam-mixed natural gas is performed by ...*" concerned the addition of CO₂ to the mix of steam and gas, while the features following this sentence dealt with the preparation of the mix of steam and natural gas.

IV. Under cover of the letter dated 30 April 2007, the patentee (hereinafter "the respondent") requested not to admit the documents D8 and D9 into the appeal proceedings and submitted an amended set of claims 1 to

9 in replacement to the claims maintained in the contested decision.

V. On 18 December 2008, the appellant objected that the amendment of the feature "the step of adding the carbon dioxide to the steam-mixed natural gas is performed by ..." in claims 1 and 5 into "the step of adding the steam to the natural gas is performed by ..." would be contrary to Article 123(3) EPC. It also reiterated its lack of inventive step objection against claims 1 and 5 and informed the board that it would not attend the scheduled oral proceedings.

VI. At the beginning of the oral proceedings, which took place on 20 February 2009, the respondent submitted that independent claim 5 filed with the set of claims dated 30 April 2007 had been insufficiently amended and that its subject-matter should be harmonized with that of claim 1. To this purpose, it filed an amended version of the set of claims dated 30 April 2007 wherein the term "steam-mixed" was deleted from claim 5.

Independent claims 1 and 5 of the sole request on file now read as follows:

*"1. A method of manufacturing a synthesis gas containing CO and H₂, which is suited for use in synthesizing gasoline, kerosene and gas oil by way of the Fisher-Tropsch reaction system; the method comprising the steps of:
feeding a steam-mixed natural gas to a reformer (10) which is provided with a combustion radiation portion (12) for burning a fuel, the reformer (10) being*

designed to be heated by the combustion radiation portion (12); recovering carbon dioxide from a combustion exhaust gas generated at the combustion radiation portion (12); and adding the carbon dioxide to the steam-mixed natural gas at a location on an upstream side of the reformer (10), thereby allowing a reforming reaction to take place to obtain a synthesis gas comprising CO and H₂ at a molar ratio of H₂/CO = 1 to 2.5, which is suited for use in synthesizing gasoline, kerosene and gas oil, characterized in that a preliminary reformer (36) is arranged at an upstream of the reformer (10), the natural gas to which steam has been added is supplied to the reformer (10) via the preliminary reformer (36), the carbon dioxide recovered from the combustion exhaust is fed to a passageway connecting the reformer (10) and the preliminary reformer (36), and in that the step of adding the steam to the natural gas is performed by arranging a moistening device (37) at a posterior stage of the reformer (10), introducing a synthesis gas from the reformer (10) into the moistening device (37), heating the moistening device (37) with waste heat of the synthesis gas, feeding natural gas and water to the moistening device (37), and adding steam to the natural gas in the moistening device (37).

5. A method of manufacturing a synthesis gas comprising CO and H₂, which is suited for use in synthesizing gasoline, kerosene and gas oil by way of the Fisher-Tropsch reaction system; the method comprising the steps of:

feeding a steam-mixed natural gas to a reformer (110) which is provided with a combustion radiation portion (112) for burning a fuel, the reformer (110) being designed to be heated by the combustion radiation portion (112);
recovering carbon dioxide from a combustion exhaust gas generated at the combustion radiation portion (112);
adding the carbon dioxide to the steam-mixed natural gas at a location on an upstream side of the reformer (110), thereby allowing a reforming reaction to take place; and
introducing a reformed gas from the reformer (110) into a partial oxidation furnace (133) simultaneous with an introduction of oxygen into the partial oxidation furnace (133), thereby allowing the reformed gas to react with the oxygen to obtain a synthesis gas comprising CO and H₂ at a molar ratio H₂/CO = 1 to 2.5, which is suited for use in synthesizing gasoline, kerosene and gas oil, characterized in that a preliminary reformer (138) is arranged at an upstream of the reformer (110), the natural gas to which steam has been added is supplied to the reformer (110) via said preliminary reformer (138), the carbon dioxide recovered from the combustion exhaust is fed to a passageway connecting the reformer (110) and the preliminary reformer (138), and in that the step of adding the steam to the ~~steam-mixed~~ natural gas is performed by arranging a moistening device (139) at a posterior stage of the reformer (110), introducing a synthesis gas from the reformer (110) into the moistening device (139), heating the moistening device (139) with waste heat of the synthesis gas, feeding natural gas and water to the moistening device (139),

and adding steam to the natural gas in the moistening device (139)."

The respondent no longer disputed the admissibility into the appeal procedure of the documents D8 and D9 filed with the grounds of appeal.

- VII. The appellant requested in writing that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims 1 to 9 filed during the oral proceedings.

Reasons for the Decision

1. *Amendments - Admissibility*

The present decision relies on the set of amended claims submitted during the oral proceedings before the board. This set differs from the one on which the contested decision was based only by the deletion in independent claim 5 of the term "steam-mixed" in the step of adding the steam.

The respondent stated that this amendment had to be made in order to harmonize the subject-matter of independent claim 5 with that of claim 1 amended and submitted under cover of the letter dated 30 April 2007.

The board observes that although the appellant was absent at the oral proceedings, it could not plead in

the present context that it was taken by surprise by the amendment to independent claim 5, because the amendment is identical to the one already made to claim 1 in response to a clarity objection raised prior to the oral proceedings and that the appellant had already commented in its letter of 18 December 2008 with respect to claim 1. Furthermore, potential comments concerning the present amendment to claim 5 would have been the same as those concerning the amendments carried out in claim 1. The board therefore admits the amendment to claim 5 under Articles 13(1) and 15(3) RPBA.

2. *Amendments - Allowability*

2.1 As indicated by the respondent, the basis in the application as filed for the amended claims 1 and 5 of the present request is as follows:

- claim 1: claims 1 and 4; page 42, lines 8 to 34;
- claim 5: claims 8 and 11; page 93, line 22 to page 94, line 10;

Dependent claims 2, 3, 4, 6, 7, 8, 9 have themselves a basis in claims 2, 3, 7, 9, 10, 14 and 15 of the application as filed, respectively.

The requirements of Article 123(2) EPC are therefore fulfilled.

2.2 Concerning appellant's objection under Article 123(3) EPC, the board observes that independent claims 1 and 5 as granted comprised inter alia the

features "feeding a steam-mixed natural gas to a reformer" and "adding the carbon dioxide to the steam-mixed natural gas at a location on an upstream side of the reformer", without however specifying how the steam was added to the natural gas. Accordingly, the scope of protection of the independent claims 1 and 5 as granted was very broad as regards the addition of steam, since their subject-matter was open to any possibility of adding the steam to the natural gas.

The amendment now carried out in independent claims 1 and 5, which specifies how the addition of steam is to be made, restricts the addition of steam to the features now specified in independent claims 1 and 5 and thus confers to these claims a **narrower** scope of protection in comparison to that conferred by claims 1 and 5 of the granted patent.

Accordingly, since the protection conferred has not been extended, it is concluded that the amendment in claims 1 and 5 is in conformity with the requirements of Article 123(3) EPC.

3. *Novelty*

The novelty of the amended claims has not been contested. The board is satisfied that the requirements of Article 54 EPC are met. So, further comments on this matter need not to be made.

4. *Inventive step*

4.1 The patent in suit relates to a method of manufacturing a synthesis gas to be employed for the synthesis of

gasoline, kerosene and gas oil by way of the Fisher-Tropsch reaction (paragraph [0001]). An object of the contested patent was to provide a method procuring CO₂ inside the synthesis gas-manufacturing plant, thereby enabling the synthesis gas to be cheaply manufactured without being restricted by the location of CO₂ gas source, such as an ammonia plant (paragraph [0011]).

- 4.2 A similar process being already known from D2, the parties acknowledged the latter document as representing the closest state of the art and, hence, the starting point for assessing inventive step.

D2 discloses a steam-hydrocarbon reforming process producing synthesis gas directly from the reformer step with the right ratio of hydrogen and carbon monoxide (H₂/CO), typically in a ratio of 1:1.

According to the process flow diagram as illustrated in Figure 1, a mixture of the feed (natural gas, LPG, methanol or related hydrocarbons) with steam and CO₂ enters the reformer where the mixture is converted to a product mixture of H₂, CO, CO₂ and H₂O in the presence of newly developed catalysts. Both the product mixture and hot flue gas from the reformer pass through separate waste heat boilers, coolers and CO₂ absorbers. Both absorbers use a recycled MEA (monoethanolamine) solution to absorb CO₂. The recovered CO₂ supplies the needs of the reformer feed. The desired H₂/CO ratio is obtained by selecting a proper ratio of CO₂, steam and hydrocarbon within the reformer feed. The overall investment cost for this process, which does not generate excessive hydrogen, is nearly 40% less than that for a common steam reformer.

- 4.3 The respondent stated that in the light of D2, the problem to be solved was to be seen in the provision of a method of manufacturing a synthesis gas wherein the steam reforming reaction was accelerated in a way that the reaction rate between hydrocarbon and steam within the natural gas could be maximized.

The board observed at the oral proceedings that there was no basis in the contested patent for this problem, and the respondent - by reference to paragraphs [0108] and [0109] of the contested patent - then reformulated the problem to be solved as being the provision of decreasing the quantities of fuel and steam fed to the reformer in order to manufacture a synthesis gas at lower costs.

- 4.4 As a solution to this problem, the patent in suit proposes the processes according to independent claims 1 and 5, which are in particular characterized by:

- i) a preliminary reformer being arranged upstream of the reformer, the natural gas to which steam has been added being supplied to the reformer via the preliminary reformer,
- ii) the carbon dioxide recovered from the combustion exhaust being fed to a passageway connecting the reformer and the preliminary reformer,
- iii) the step of adding the steam to the natural gas being performed by arranging a moistening device at a posterior stage of the reformer, introducing

a synthesis gas from the reformer into the moistening device, heating the moistening device with waste heat of the synthesis gas, feeding natural gas and water to the moistening device, and adding steam to the natural gas in the moistening device.

- 4.5 As regards the question whether the problem defined in item 4.3 *supra* has been solved, it can be seen from paragraphs [0108] to [0110] and [0233] to [0235] of the contested patent that the implementation of the features i) to iii) identified in item 4.4 above leads to a reduction of the fuel and steam quantities supplied to the reformer. The board is therefore satisfied that the technical problem underlying the patent in suit has been successfully solved by the proposed solution.
- 4.6 It remains to be decided whether the proposed solution to the above problem is obvious or not in view of the state of the art. The appellant argued in this respect that said solution would be suggested by the combined teachings of documents D6 and D8 (or D9).
- 4.6.1 D6 is directed to the field of reforming hydrocarbons to hydrogen and carbon monoxide-containing product gases (column 1, lines 9 to 11). In its first preferred embodiment (column 5, line 30 to column 6, line 13), D6 discloses an integrated process including a steam-methane reformer and a convective heat transfer, the natural gas feed being heated in a heat exchanger against hot reformat leaving the overall process as a product and steam being mixed with the feed gas stream to provide a steam to carbon ratio of 0.5 up to 3.5.

Carbon dioxide can be optionally added to the mixed feed in ratios of 0.1 to 2.5 carbon dioxide to carbon content. The heated feed stream is split into a stream feeding a conventional steam-methane reformer in conventional catalyst filled reformer tubes and a feed stream feeding a convective heat transfer reformer. The feed to the conventional steam-methane reformer is heated and reformed at 1500 to 1800°F (816 to 982°C) by essentially radiant heat provided by the combustion of fuel gas with air or oxygen-enriched gas. The hot steam-methane reformer reformat is directed to the convective heat transfer reformer to recover waste heat by convective heat transfer. The feed stream to the convective heat transfer reformer is optionally mixed with additional process steam to increase the steam to a carbon ratio to 3.0 up to 6.0. The feed stream to the convective heat transfer reformer is heated and reformed. The heat of reaction in the convective heat transfer reformer is provided by the waste heat recovered from the reformat leaving the conventional steam-methane reformer.

According to D6, column 6, lines 20 to 42, the above process can further integrate an adiabatic prereformer with the feed gas being taken through the adiabatic prereformer after being heated in the convective section coil of the primary reformer. The integration of such an adiabatic prereformer reactor upstream of both the conventional steam-methane reformer and prereforming convective heat transfer reformer reduces the duty and physical size of the conventional primary reformer by about 5% and the prereforming convective heat transfer reformer by about 2.5%. It also slightly lowers the overall fuel consumption.

From the above teaching, the board observes that even if the skilled person faced with the problem identified in item 4.3 *supra* might be prompted to integrate in the process known from D2 an adiabatic prereformer upstream the reformer with the feed gas being taken through the adiabatic prereformer after being heated in the convective section coil of the reformer with the hope that the integration of an adiabatic prereformer would lower the overall fuel consumption, the process obtained by such an integration however would still lack the features ii) and iii) identified under item 4.4 *supra*.

4.7 Concerning the other documents on which the appellant further relied upon in its objection of lack of inventive step, the board observes the following:

4.7.1 D8 (claim 1) discloses a process for producing methanol from hydrocarbon comprising the steps of:

- (a) reacting hydrocarbon with steam to generate synthesis gas containing hydrogen, carbon monoxide and carbon dioxide as main components,
 - (b) reacting the synthesis gas on a catalyst for methanol synthesis and recovering crude liquid methanol from the reacted gas, and
 - (c) distilling said crude methanol into refined methanol and waste water containing lower boiling point organic compounds, higher boiling point organic compounds and organic acid,
- wherein the hydrocarbon comes in contact with the waste water which has been neutralized with alkali metal salt or hydroxide in the step (c) so as to be humidified, and then comes in contact with condensed water separated from the synthesis gas obtained in the step

(a) so as to be further humidified, and is then supplied to the step (a).

In the passages specifically quoted by the appellant, on the one hand the above two-steps humidifying system is detailed (see paragraphs [0043] to [0045]) while on the other hand the problem to be solved is defined as being "to provide a process for humidifying hydrocarbon and reducing steam for the process, that is, reducing expensive boiler water by utilizing waste water obtained in a distillation step" (paragraph [0013]).

4.7.2 D9 (column 1, lines 9 to 14) relates to a process for the production of methanol from hydrocarbon so as to reduce an amount of effluent water from an apparatus for the production of methanol and to save and reduce boiler water.

In its claim 1, D9 discloses a process for the production of methanol from hydrocarbon, which comprises the steps of:

- (a) reacting hydrocarbon with steam to generate a synthesis gas composed of hydrogen, carbon monoxide and carbon dioxide as main components,
- (b) reacting the synthesis gas on a methanol synthesis catalyst to synthesize crude methanol,
- (c) feeding the crude methanol to a distillation column, without adding any alkaline compound to neutralize organic acids in the crude methanol, to distill the crude methanol and separate it into a purified methanol and effluent water containing low-boiling organic compounds, high-boiling organic compounds and organic acids, said distillation column composed of stainless steel having a composition of 18 to 20% by weight of Cr,

8 to 10.5% by weight of Ni and not more than 0.08% by weight of C in its portions contacting the low-boiling organic compounds, high-boiling organic compounds or organic acids,

(d) bringing the effluent water into contact with preheated gaseous hydrocarbon in an apparatus composed of stainless steel having a composition of 16 to 26% by weight of Cr, 6 to 14% by weight of Ni, 2 to 4% by weight of Mo and not more than 0.03% by weight of C in its portions contacting the low-boiling organic compounds, high-boiling organic compounds or organic acids to humidify the hydrocarbon, and

(e) feeding the humidified hydrocarbon for use in step (a), wherein step (d) contains a step of humidifying the hydrocarbon with the effluent water from step (c) by means of a heat exchanger humidifier using, as a heat source, heat of the synthesis gas generated in step (a), or a step of humidifying the hydrocarbon, pre-heated to 250° to 430°C, with the effluent water from step (c) by means of a heat-insulated humidifier, and then humidifying the hydrocarbon with water by means of a heat exchanger humidifier using, as a heat source, heat of the synthesis gas generated in step (a).

In the passage of D9 (column 7, lines 1 to 12) upon which the appellant specifically relied when arguing against the inventive step, the principle of the humidifying device using the heat of the synthesis gas for producing a mixture of hydrocarbons and steam is further explained.

- 4.7.3 The board notes that the possibility of adding carbon dioxide to the hydrocarbon-steam reaction mixture is envisaged both in D8 (paragraph [0019]) and in D9

(column 4, lines 11 to 17). However neither D8 nor D9 disclose that a preliminary reformer may be arranged upstream of the reformer, nor that carbon dioxide may be recovered from a combustion exhaust gas generated at a combustion radiation portion of the reformer, let alone do these documents disclose that the carbon dioxide recovered from the combustion exhaust be fed to a passageway connecting the reformer and the preliminary reformer.

4.7.4 Accordingly, even if the documents D8 and D9 might appear to suggest the use of a humidifying system with the aim of reducing steam or boiler water in the above processes, it is to be noted that none of the documents quoted by the appellant (D2, D6, D8, D9) discloses the feature that carbon dioxide recovered from the combustion exhaust is to be fed to a passageway connecting the reformer and the preliminary reformer, so that even if the skilled person faced with the problem identified in item 4.3 *supra* might be prompted to implement the humidifying system of D8 or D9 in the process known from D2 and/or D6, it would still not arrive at the subject-matter of present claims 1 or 5.

4.8 The remaining documents cited during the opposition phase were no longer relied upon by the appellant during the appeal proceedings. In the board's judgment, they also do not contain further information which would point towards the claimed solution of the problem stated above.

4.9 Accordingly, for the reasons indicated above, the subject-matter of independent claims 1 and 5 is not obvious to a person skilled in the art in view of the

cited prior art and, therefore, it involves an inventive step within the meaning of Article 56 EPC.

As claims 2 to 4 and 6 to 9 represent particular embodiments of the subject-matter of independent claims 1 and 5, they derive their patentability from claims 1 and 5 on which they depend.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside
2. The case is remitted to the department of first instance with the order that the patent be maintained on the basis of the set of new claims 1 to 9 filed during the oral proceedings, and a description, including the Figures, to be adapted.

The Registrar:

The Chairman:

C. Vodz

G. Rath