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DECISION
of 26 July 2001

Case Number: T 0623/96 - 3.3.5

Application Number: 92100746.4

Publication Number: 0496295

IPC: B01D 53/34

Language of the proceedings: EN

Title of invention:
High-Performance flue-gas desulfurization process and apparatus

Applicant:
MITSUBISHI JUKOGYO KABUSHIKI KAISHA

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Main request and auxiliary requests 1A to 2A: inventive step (no); obvious combination of prior art teaching with expected effects"
"Auxiliary requests 3A to 4A: inventive step (no); routine optimisation of process parameters"

Decisions cited:
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Catchword:

-

Case Number: T 0623/96 - 3.3.5

DECISION
of the Technical Board of Appeal 3.3.5
of 26 July 2001

Appellant:
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 14 February 1996 refusing European patent application No. 92 100 746.4 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: R. K. Spangenberg
Members: A.-T. Liu
J. H. Van Moer

Summary of Facts and Submissions

I. The appeal is from a decision of the examining division refusing the European patent application No. 92 100 746.4.

II. The impugned decision was based on claims 1 to 6 as amended by letter of 8 August 1995. Claim 1, which was maintained on appeal as basis for the main request, read as follows:

"A flue-gas desulfurization process in accordance with a wet lime/gypsum method by which sulfur oxides are removed from a combustion exhaust gas, said process comprising the steps of bringing the combustion exhaust gas into contact with a gypsum slurry containing 1 wt. % or less of calcium carbonate in a first absorption tower, and then bringing said combustion exhaust gas into contact with a slurry containing 1 wt. % or more of calcium carbonate in a second absorption tower, wherein the first and second slurries remain in contact with the combustion exhaust gas in tanks provided in bottom portions of the first and second absorption towers."

III. The examining division made reference to the following documents in its decision:

D1	US-A-4 040 803
D2	FR-A-2 534 150

IV. Starting from D2 as representing the closest prior art teaching, it was held that the technical problem to be solved was to obtain a high grade of desulfurisation and a gypsum by-product of high purity and to avoid the formation of scale in the desulfurisation apparatus. Essentially, the examining division was of the opinion that the technical problem as stated was by itself obvious and the solution proposed in claim 1 was arrived at by routine optimisation.

V. Oral proceedings before the Board of Appeal were held on 13 September 2000, during which the appellant was handed over the following documents:

D2'	DE-C-3 236 905
D3	Zement-Kalk-Gips, Nr.6/1982 (35. Jahrgang), pages 313 to 317.

D3 was acknowledged in D2' which was the German patent specification upon which the priority of the French patent application D2 was based. The Board had cited D3 as reference for the common general knowledge concerning the chemical reactions involved in the desulfurisation process and their significance in the discussion of the pH value of the absorbent slurry at the respective desulfurisation stage.

VI. At the appellant's request, the oral proceedings were concluded with the decision to continue the proceedings in writing. By letter of 25 July 2001, the appellant submitted inter alia amended claims as basis for auxiliary requests numbered 1A to 3A.

VII. Further oral proceedings were held before the same Board of Appeal on 26 July 2001, at which a new claim 1 was filed to serve as basis for the auxiliary request 4A.

VIII. Claim 1 of the auxiliary request 1A, essentially based on claim 1 of the main request, incorporated at the end of the text of the latter the additional stipulation:

"so as to obtain a purity of the by-product gypsum of 95% or more and a concentration of SO_x in the desulfurized gas of 10 ppm or less."

Claim 1 of the auxiliary request 2A differed from claim 1 of the auxiliary request 1A in that it stipulated that the concentration of SO_x to be obtained in the desulfurised gas be:

"10 ppm or less, particularly 1 ppm or less", instead of "10 ppm or less".

Claim 1 of the auxiliary request 3A differed from claim 1 of the main request in that the claimed process was further characterised by:

"the combustion exhaust gas being desulfurized in the first adsorption tower to such extent that the SO_x concentration is decreased from about 1,000 ppm to about several tens of ppm, so as to obtain a purity of the by-product gypsum of 95% or more and a concentration of SO_x in the desulfurized gas exiting the second absorption tower of 10 ppm or less."

Claim 1 of auxiliary request 4A differed from claim 1 of the auxiliary request 3A in the stipulation of:

"a concentration of SO_x in the desulfurized gas exiting the second absorption tower of 1 ppm or less."

IX. The appellant's arguments, submitted in writing and orally, may be summarised as follows:

- D1 and not D2 (or D2') should be considered to represent the closest prior art teaching.

- The main problem to be solved with respect of D1 was the provision of a highly efficient desulfurisation process. This was achieved by using a high amount of calcium carbonate as absorbent, which in turn would result in the additional problem of scaling.

- The problem of scaling was solved in D1 by keeping the pH at a value less than 7. Since it was common knowledge that a higher calcium carbonate concentration would lead to an undesirable increase of the pH, thus augmenting the risk of scale formation, the skilled person would not have modified the process of D1 by increasing the concentration of calcium carbonate, at least not without additional measures.

- The additional measure for solving the problem of scaling resided in maintaining a permanent contact of the gas to be desulfurised with the calcium carbonate slurry.

- The skilled person did not have any incentive for combining with D1 the teaching of D2 (or D2') which only accidentally disclosed the permanent contact of gas and slurry.

- The additional functional feature in claim 1 of auxiliary request 1A was to be interpreted as a clarification and not as a further limitation with respect to the combination of features of claim 1 of the main request.

- The limit of 1 ppm SO_x in claim 1 of auxiliary request 2A was optional.

- The skilled person knew how to achieve the aims set by the additional functional features in the respective claim 1 of auxiliary requests 3A and 4A.

X. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of claims 1 to 6 as amended by letter of 8 February 1995 or, in the alternative, on the basis of, in the given order, auxiliary requests 1A to 3A filed with the letter dated 25 July 2001 and auxiliary Request 4A submitted at the oral proceedings of 26 July 2001.

Reasons for the Decision

Main request

1. Inventive step

1.1 Claim 1 is directed to a process for the desulfurisation of flue-gas by bringing the gas into contact in a first absorption tower with a gypsum slurry containing 1 wt. % or less of calcium carbonate, then in a second absorption tower with a slurry containing 1 wt. % or more of calcium carbonate. In the process, the gas to be treated remains in contact with carbonate slurries in tanks provided in the bottom portion of the respective absorption tower. According to the process, a combustion gas can be desulfurised to such an extent that an SO_x concentration in the gas is 10 ppm or less and the by-product gypsum can also be obtained with high purity (see description of patent application, page 3, lines 10 to 14).

1.2 The Board can accept the appellant's view that the closest prior art is represented by D1 which also relates to a method of wet flue gas desulfurisation using calcium carbonate (also called "lime") as absorbent. Essentially, the absorption is also conducted here in a "former stage" and a "latter stage" absorbing device (corresponding to the first and second absorption tower of the present process, respectively). The flue gas is passed successively in the "former" then in the "latter" stage, whilst the lime absorbent slurry is circulated in the latter stage and then in the former stage (column 4, lines 54 to 62). Each stage is operated in such a manner that the circulating liquid is maintained at a prescribed lime concentration and the liquid discharged from the former stage for gypsum recovery is substantially free of unreacted lime (column 1, lines 47 to 52 and column 3, lines 14 to 19). It is undisputed that the process of D1 provides a high rate of flue gas desulfurisation and a gypsum by-product of high purity (see also column 3, Table 1).

1.3 The appellant has submitted that the claimed process leads to a highly pure gypsum and a desulfurisation efficiency which is improved with respect to D1. However, there are no data on file demonstrating that, compared to D1, the present process would always lead to a purer gypsum at a given desulfurisation level or alternatively, a higher desulfurisation efficiency for the same gypsum purity.

The Board also fails to accept the appellant's assertion that, with regard to D1, the technical problem to be solved was to avoid the formation of scaling. As is explicitly indicated in D1, no formation or deposition of scale was observed within the absorbing devices (see column 3, lines 47 to 48).

Consequently, the Board can see that, with respect to D1, the problem to be solved by the present application is the provision of a further process with the same effects.

1.4 The solution proposed in claim 1 is a process which is essentially distinguished from that of D1:

(i) in the respective concentration of the calcium carbonate slurries in the absorption towers and

(ii) in that these calcium carbonate slurries remain in contact with the flue gas in tanks provided in bottom portions of the first and second absorption towers.

1.5 The question is whether the modification as proposed in claim 1 is obvious in view of the available prior art.

1.5.1 Re.: characterising feature (i)
Concentration of the calcium carbonate slurries

For the two embodiments whose data are listed in Table 1, the process of D1 involves mixing lime material with water to form a slurry, supplying the slurry to a circulation tank and eventually feeding this slurry, with a starting concentration of 6 wt%, to the second ("latter") absorption tower. After reaction with the flue gas within the second absorption tower, the lime slurry is returned to the circulation tank, to be recycled within the same second absorption tower. During this period, the major part of the lime in the absorbing liquid is being used up. Part of this absorbing liquid is withdrawn from the circulation system of the second tower and fed into another circulation tank for use in the first ("former") absorption stage (see column 2, line 50 to column 3, line 8). The slurry fed to the first tower is thus the absorbing liquid which has gone through treatment in the second tower. It is undisputed that the slurry concentration at this point is 0.035 or 0.032 $CaCO_3$ mol/l, respectively, corresponding to a concentration of approximately 0.3% by weight (column 3, Table 1).

From the disclosure of D1, the Board can thus derive the concentration of the carbonate slurries as they are fed into the absorption towers. The Board, however, agrees with the appellant that, whilst the slurry circulated in the first absorption stage always has a carbonate concentration below 1 wt%, there is no clear disclosure in D1 that the slurry in circulation in the second stage has at the same moment a concentration of at least 1 wt%.

It is however explicitly explained in D1 and not refuted by the appellant, the skilled person knows that a considerably large amount of lime in excess of the stoichiometrical equivalent is necessary for obtaining a high absorption rate of sulfur contained in the flue gas (see column 1, lines 28 to 38). As a consequence, the Board holds that the optimisation of the lime slurry concentration to be applied to the absorption of sulfur, be it as a function of the sulfur loading of the flue gas to be treated or of the targeted concentration of sulfur in the treated flue gas, belongs to the routine tasks of the notional skilled person, not requiring inventive activity.

1.5.2 Re.: characterising feature (ii)
Constant contact of flue gas with calcium carbonate slurries

As is illustrated in the drawings, the circulation tanks (6) and (9) for the lime slurries in D1 are separate from the absorbing towers (2) and (3) (see Figures 1, 4 and 5). D1 thus does not disclose that the absorbent slurries are kept in permanent contact with the flue gas. According to the present application, the circulation tanks are provided, in contrast, directly within the absorption towers, such that the lime slurries remain in contact with the flue gas during the course of the process, as stipulated in claim 1. It is undisputed that such process modification is known from D2' (Figure 1, column 4, lines 25 to 54 and column 5, lines 10 to 17). Thus, when looking for an alternative to the process of D1, the proposed modification is a priori one of the possibilities the skilled person **could** consider.

1.5.3 Re.: combination of features of claim 1

The appellant has submitted that the combination of the characterising features (i) and (ii) is essential in that the use of a highly concentrated lime slurry as in claim 1 results in scaling, which is prevented by permanently contacting the slurry and the gas. The Board observes however, that, following from D1, the scaling problem is known in the field of flue gas desulfurisation. In order to avoid this, it is already taught in D1 that care must be taken to maintain the pH value of the absorbing liquid between 6.5 and 7.0 (column 4, lines 20 to 28).

Although D2' does not indicate the process parameters, it refers to D3 in its introductory part as the process it sets out to improve, the improvement over D3 being the heat exchange which is not of concern for the present discussion. However, it is clear from the reference that the process conditions in the absorption towers of D2 are the same as those of D3, the only difference being that the absorption stages are side by side in the earlier case whilst it is on top of each other in the latter case (see D2', column 2, lines 1 to 9 and lines 22 to 30). The Board therefore considers that the disclosure of D3 is fully incorporated in the disclosure of D2' as far as the chemical reactions which take place during these absorption stages are concerned. It is thus known that, at the contact with the 10% lime slurry in the top stage of D3 (thus in the second tower of D2'), hydrogen carbonate is formed in the slurry due to the excess of lime, acting as a natural buffer keeping the pH of the slurry between 6 and 7 (D3, page 316, subparagraph 3.3). This is no longer in dispute (see applicant's letter dated 13 February 2001, page 1, paragraph 2).

In summary, it is common knowledge that, where the flue gas to be treated is kept in contact with a highly concentrated lime slurry (for example a 10% slurry as in D2'/D3), the pH value of this slurry is buffered between 6 and 7. Thus, contrary to the appellant's assertion, D2' does not "accidentally" disclose the permanent contact of gas and slurry, without the connection between the pH and that contact being known. In the Board's judgment, the skilled person therefore not only **could** consider but **would** particularly resort to this specific teaching with the aim to avoid scaling, since the absorbing slurry is then automatically maintained within the pH range as recommended in D1.

The appellant has not submitted further arguments with respect to the combination of features in claim 1. Nor is it plausible that the claimed combination leads to any unexpected effect.

1.6 The Board does not agree with the appellant that there is no incentive for the skilled person to combine the teachings of D1 and D2'.

1.6.1 Although D2' primarily concerns the problem of reheating the treated flue gas, it generally relates to a process for desulfurising flue gas to obtain gypsum as a by-product (column 2, lines 1 to 4). Furthermore, it teaches the use of a 10% lime slurry as absorbing liquid in the second absorption stage. Thus, when considering the use of a concentrated lime absorbent slurry, the skilled person would naturally look into D2' for the measures applied therein.

1.6.2 The appellant has argued that the pH range of the second absorption stage in D2' is between 5 and 8, which would not make D2' a suitable candidate for combining with D1.

As is noted above, D2' only gives cursory indications as to the reaction conditions in the absorption towers and refers instead to D3 (see point 1.5.3). In that document, it is clearly indicated that the pH value of the slurry concerned is buffered at 6 to 7, thus within the error margins of the pH range as recommended in D1.

1.7 As a corollary of the above, the subject-matter of claim 1 lacks an inventive step in view of D1 in combination with D2'.

Auxiliary request 1A

2. Claim 1 of this request differs from claim 1 of the main request in that it further incorporates the stipulation that the process is intended "to obtain a purity of the by-product gypsum of 95% or more and a concentration of SO_x in the desulfurized gas of 10 ppm or less."

2.1 As is submitted by the appellant, the additional feature is not a functional feature which further limits the scope of the claim. Rather, the results indicated are automatically obtained when the process is conducted within the boundaries as defined by the preceding features of the claim.

2.2 In view of the applicant's explanation, the Board concludes that the subject-matter of claim 1 of this request is the same as that of claim 1 of the main request.

2.3 For the same reasons as elaborated above, the claimed process therefore lacks an inventive step with regard to D1 in combination with D2'.

Auxiliary request 2A

3. Compared to claim 1 of the auxiliary request 1A, the present claim 1 contains the additional feature that the concentration of SO_x in the desulfurised gas is "particularly 1 ppm or less."

3.1 The appellant has not indicated and the Board cannot find a basis for the stipulation that the concentration of SO_x in the desulfurised gas can be **less than 1 ppm**. In the original description, it is only indicated that "the exhaust gas ... is desulfurized to an SO_x concentration of 10 ppm or less, even **to 1 ppm** in particular, in the second absorption tower" (see page 5, line 23 to page 6, line 3). It is therefore questionable whether claim 1 as amended meets the requirements of Article 123(2) EPC.

3.2 Notwithstanding the above remark, the Board holds that the process as claimed also lacks an inventive step.

Considering that the additional feature is only optional, as is expressly confirmed by the appellant, the scope of present claim 1 remains the same as that of claim 1 of the main request. As a result, the reasoning and finding for claim 1 of the main request and that of the auxiliary request 1A equally apply to present claim 1.

Auxiliary request 3A

4. Compared to the main request, the process of present claim 1 is further limited by the stipulation of "the combustion exhaust gas being desulfurized in the first adsorption tower to such extent that the SO_x concentration is decreased from about 1,000 ppm to about several tens of ppm".

4.1 With respect to D1 as closest prior art teaching, the Board can see the problem to be solved in the provision of a further process of desulfurisation, for the same reasons as explained in point 1.3 above.

4.2 Compared to the process of claim 1 of the main request, the present process is thus further distinguished from that of D1 by the stipulation according to which the absorption of sulfur is mainly carried out in the first absorption tower and not in the second tower as in D1 (column 4, lines 66 to 68).

4.3 It is common ground that the present application does not explicitly disclose how the desired effect, namely that the absorption of sulfur be mainly carried out in the first absorption tower, is to be achieved. As is explained by the appellant, however, the functional feature concerned only implies that the lime absorbent slurry for use in the first tower must have a lower limit of concentration, which can be determined by the skilled person using his common general knowledge.

The appellant's submission is congruous with the teaching of D1 which discloses that the amount of lime in the absorbent slurry is responsible for the absorption of sulfur from the flue gas. On the other hand, it is warned in D1 that it is disadvantageous to have unreacted lime in the slurry after the treatment since it has to be removed before gypsum can be recovered from the absorbent slurry. By ensuring that the spent slurry removed from the first absorption tower is free of unreacted lime, gypsum can be obtained directly without previous treatments (see D1, column 1, lines 28 to 46 and column 3, lines 5 to 21). Needless to say, any unreacted gypsum which is still contained in the spent liquid will affect the purity of the gypsum obtained therefrom.

It is thus undisputed that, when gypsum is directly produced from the spent slurry of the first absorption tower, a compromise has to be made between the purity of gypsum and the rate of sulfur removal, the determining factor being the lime concentration of the absorbent slurry. In consequence, the Board cannot see an inventive activity in the selection of a lower limit of lime concentration with the aim to ensure a particular level of desulfurisation, which selection being, in agreement with the appellant's submission, the direct result of routine optimisation tasks of the notional skilled person (see also point 1.5.1 above).

4.4 The appellant has not convinced the Board that the combination of the new functional feature with the other features of the claim leads to an unexpected result. Specifically, it has not been proved that the claimed process leads to a more efficient desulfurisation (see point 1.3 above). In consequence, the Board holds that the additional feature does not, either by itself or in combination with the remaining features of the claim, involve an inventive step. The finding for claim 1 of the main request therefore applies to claim 1 of the present request.

Auxiliary request 4A

5. Compared with claim 1 of the preceding request, the process of present claim 1 is further limited by the stipulation that the desulfurised gas exiting the second absorption tower must have an SO_x concentration of 1 ppm or less.

5.1 As is already observed in point 3.1, the stipulation that the concentration of SO_x in the desulfurised gas must be less than 1 ppm does not seem to have a basis in the application documents as filed. The same question arises here therefore as to the conformity of the amended claim 1 with the requirements of Article 123(2) EPC.

5.2 Notwithstanding the above objection, the Board holds that this additional feature cannot contribute to an inventive step.

It is common ground that the targeted sulfur concentration is by itself not inventive. Quite apart from the fact that such target normally depends on external requirements set by the authorities, a desulfurisation to an SO_x concentration of 1 ppm is not even unusual in the art (see present description, page 10, lines 1 to 11).

As is on the other hand submitted by the applicant, the additional functional feature is to be construed as imposing a lime concentration in the second tower such as to achieve the set sulfur concentration in the exiting gas. Such an optimisation of the absorbent concentration as a function of a targeted sulfur absorption belongs, however, to the routine tasks of the notional skilled person (see also points 1.5.1 and 4.3 above).

Consequently, the present request must fail, following the same logic as applied to the preceding request(s).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

G. Rauh R. Spangenberg