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**D E C I S I O N**  
**of 8 October 2001**

**Case Number:** T 0575/97 - 3.3.5

**Application Number:** 93610018.9

**Publication Number:** 0561730

**IPC:** B01D 53/34

**Language of the proceedings:** EN

**Title of invention:**  
Flue gas desulfurization process

**Applicant:**  
MITSUBISHI JUKOGYO KABUSHIKI KAISHA

**Opponent:**

-

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 56, 123(2)

**Keyword:**

"Main request and auxiliary request II: Inventive step (no):  
obvious alternative process"

"Auxiliary request I: Added subject-matter (yes)"

**Decisions cited:**

T 0201/83

**Catchword:**



Case Number: T 0575/97 - 3.3.5

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.5**  
**of 8 October 2001**

**Appellant:** MITSUBISHI JUKOGYO KABUSHIKI KAISHA  
5-1, Marunouchi 2-chome  
Chiyoda-ku  
Tokyo (JP)

**Representative:** Christiansen, Ejvind  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 20 December 1996  
refusing European patent application  
No. 93 610 018 pursuant to Article 97(1) EPC.

**Composition of the Board:**

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

## Summary of Facts and Submissions

I. The appeal is from the decision of the examining division refusing the European patent application No. 93 610 018.9 on the basis of claims 1 to 9 as filed with the letter of 16 June 1995.

II. The examining division held that the subject-matter of claim 1 did not involve an inventive step in view of the prior art document DE-A-3 713 143 (D1).

III. With the statement of the grounds of appeal, the appellant filed two new sets of claims, termed primary and secondary claims. Reference was also made to the following documents:

D1': US-A-4 802 966

D2: EP-A-0 405 619

D3: US-A-4 578 257.

D1' corresponded to the German patent application D1 cited in the decision by the examining division.

IV. In a communication dated 16 November 2000, the Board gave reasons as to why, in their preliminary view, it was unlikely that the appeal could succeed on the basis of those primary or secondary claims.

V. By letter of 30 May 2001, the applicant filed a new set of primary claims to replace the primary set of claims filed with the statement of the grounds of appeal, and a new set termed tertiary claims.

VI. In the annex to the summons to attend oral proceedings, the Board explained why the new submissions and the

accompanying arguments were not appropriate for meeting the objections raised in the previous communication.

VII. The primary set consisted of a newly amended independent process claim 1 and claims 2 to 10 dependent thereon. Claim 1 read as follows:

"A flue gas desulfurization process comprising:

- (a) contacting the gas (2) to be desulfurized with an aqueous adsorbent (sic) solution (3) for desulfurization of sulfur oxides in an absorption column (1); said absorption solution comprising:  
  
a calcium compound for providing calcium ions to react with said sulfur oxides;
- (b) absorbing sulfur oxides from the gas (2) into the absorbent solution (3);
- (c) reacting the absorbed sulfur oxides in the absorbent solution (3) with calcium ions from said calcium compound and oxidizing the reaction product with oxygen to form a plaster therein;
- (d) withdrawing a portion (7) of the plaster-containing adsorbent (sic) solution (3) from the adsorption (sic) column (1) for separating out the formed plaster (9) and for forming a filtrate (10, 12);
- (e) electrolyzing a portion (12) of the filtrate in an electro dialyzer (14) comprising diluting chambers and concentration chambers located between cation-exchange membranes (C) and anion-exchange

membranes (A) alternately arranged between a positive and a negative electrode for recovering of a diluted solution (16); and

(f) recycling the diluted solution (16) to the absorption column (1);

CHARACTERIZED in

that the absorption solution further comprises an auxiliary absorption agent for improving the removal of sulfur oxides; said agent being an organic carboxylic acid consisting of a straight chained hydrocarbon having 1 to 4 carbon atoms and having carboxylic groups at opposite ends of said hydrocarbon;

that said filtrate comprises water-soluble salts made up of calcium ions, magnesium ions and chloride ions; said calcium ions and magnesium ions being major components of said filtrate; and

that the anion-exchange membranes (A) are univalent-selective so that the auxiliary absorption agent in the form of a bivalent organic carboxylate ion (CA) migrating in the direction toward the positive electrode is retained in the diluted solution (16) of the diluting chambers between said membranes (A,C), so that the auxiliary agent is recycled in the diluted solution for further absorption."

VIII. The secondary set of claims consisted of an independent process claim 1 and claims 2 to 9 dependent thereon. Claim 1 of this set differed from claim 1 of the primary set in its characterising part which read:

"CHARACTERIZED in

that the absorption solution further comprises an auxiliary absorption agent for improving the removal of sulfur oxides; said agent being an organic carboxylic acid consisting of a straight chained hydrocarbon having 1 to 4 carbon atoms and having carboxylic groups at opposite ends of said hydrocarbon; and said agent having a concentration of about 5 mol/l;

that said filtrate comprises water-soluble salts made up of calcium ions, magnesium ions and chloride ions; and

that the anion-exchange membranes (A) are univalent-selective so that the auxiliary absorption agent in the form of a bivalent organic carboxylate ion (CA) migrating in the direction toward the positive electrode is retained in the diluted solution (16) of the diluting chambers between said membranes (A,C)."

- IX. The tertiary set consisted of 9 claims corresponding to claims 1 to 9 of the primary set.
- X. By letter of 1 October 2001, the applicant informed the Board that he did not intend to participate in the oral proceedings as scheduled.
- XI. Pursuant to Rule 71(2) EPC, oral proceedings were held on 8 October 2001 in the absence of the appellant.
- XII. The appellant's arguments, submitted in writing, may be summarised as follows:

- The invention as claimed was directed to a flue

gas desulfurisation process using carboxylic acid as auxiliary absorption agent. The problem to be solved was to avoid pollution caused by the release of this agent into the environment.

- The solution proposed with the claimed process was a method comprising subjecting the spent absorbent liquid to electrodialysis in such way as to retain the carboxylic acid for recycling.
- The closest prior art was not represented by D1' which did not address the same problems and disclosed the electrodialysis of a different type of absorbent liquid.
- D3 was proof that the use of a sulfosuccinate as auxiliary absorption agent as in D1' would not pose a pollution problem.
- In view of D1', the skilled person would not be prompted to realise that a three-chambered electrodialyser, and even less a two-chambered electrodialyser could be used to retain carboxylate in the treated liquid.
- The skilled person would consider document D2 as starting point for the claimed invention.
- The present process involved an inventive step with regard to D2 which neither disclosed the use of an auxiliary absorbing agent nor its recovery from the waste liquor.

XIII. The appellant's main request was that a patent be granted on the basis of claims 1 to 10 of the primary

set of claims filed by letter of 30 May 2001.  
Auxiliarily, a patent was to be granted on the basis of claims 1 to 9 of the secondary set of claims (auxiliary request I), filed with the statement of the grounds of appeal or on the basis of claims 1 to 9 of the tertiary set of claims filed by letter of 30 May 2001 (auxiliary request II).

## **Reasons for the Decision**

### *Main request*

#### 1. *Inventive step*

##### 1.1 Claim 1

Claim 1 is directed to a flue gas desulfurisation process, defined in the preamble essentially by the following process steps:

- (a) contacting the flue gas with a calcium containing aqueous absorbent in an absorption column,
- (b) absorbing the sulfur oxides from flue gas,
- (c) reacting the sulfur oxides with the calcium ions in the absorbent solution to form a plaster,
- (d) separating a filtrate from the plaster-containing absorbent solution,
- (e) electrolysing the filtrate in a electrolysers comprising diluting chambers and concentration



chambers located between cation-exchange membranes and anion-exchange membranes alternately arranged between a positive and a negative electrode for recovering a dilute solution, and

- (f) recycling the diluted solution to the absorption column.

The characterising features recited in claim 1 are essentially as follows:

- (i) the absorbent solution further comprises, as auxiliary absorption agent, a carboxylic acid consisting of a straight chained hydrocarbon having 1 to 4 carbon atoms and having carboxylic groups at opposite ends;
- (ii) the filtrate contains calcium ions and magnesium ions as major components, and chloride ions;
- (iii) the anion-exchange membranes are univalent-selective so that the carboxylate ion is retained in the diluted solution of the diluting chamber and recycled.

As is already stated in the patent application and later confirmed by the appellant's submissions, the aim of the claimed process is to avoid the entrainment of the carboxylic acid with the waste water (see published patent application, page 3, lines 33 to 55).

#### 1.2 Closest prior art document

The problem of loss of the carboxylate used as auxiliary absorption agent in a process for flue gas

desulfurization is addressed in D1' (column 1, lines 31 to 48 and column 2, lines 3 to 10). In contrast, as is recognised by the appellant, D2 does not disclose the use of an auxiliary absorbing agent in such processes and thus, necessarily does not address the problem of its loss. The Board therefore concurs with the examining division that the closest prior art to the process of claim 1 is that disclosed in D1'.

1.3 Technical problem with regard to D1'

The appellant has not submitted that the present process provides an improvement over that of D1'. Neither is the Board able to establish any such improvement on the basis of the available data. The technical problem to be solved is therefore held to reside in the provision of a further process as an alternative to that of D1'.

1.4 Solution proposed in claim 1

It is undisputed that D1' relates to a process for flue gas desulfurisation comprising features (a) to (d) and (f) as indicated in points VII and 1.1 above. In this prior art process, the (carboxylate containing) filtrate is treated in an electro-dialyser comprising units of three dialysing chambers as follows:

- (1) an intermediate chamber M located between two sheets of membrane  $A_1$  and  $A_2$  selectively permeable to monovalent anions,
- (2) a concentrating chamber C located between the above-mentioned membrane  $A_2$  selectively permeable to monovalent anions and a cation-exchange

membrane K and

- (3) a desalting chamber located between the cation-exchange membrane K and a sheet of membrane A<sub>1</sub> selectively permeable to monovalent anions of the neighbouring unit of three dialysing chambers

(column 3, lines 16 to 35; column 4, lines 9 to 21 and lines 52 to 64, and Figure).

In the process of claim 1, the filtrate is also electrolysed in an electrolysing chamber comprising diluting chambers and concentration chambers located between cation-exchange and univalent selective anion-exchange membranes. The claimed process is, however, essentially distinguished from that D1' in that these **different membranes are alternately arranged between a positive and a negative electrode** (emphasis added) (see feature (e) in points VII and 1.1 above). The apparatus involved will be referred to below as a two-chamber electrolysing chamber.

At this point, the Board notes that, although features (i) to (iii) are recited in the characterising portion of claim 1 (see points VII and 1.1 above), they do not distinguish the process of claim 1 from that of D1'. The reasons therefor will be given below (see points 1.8 and 1.9).

- 1.5 The Board accepts that, according to the process of claim 1, the auxiliary absorbent agent is effectively retained in the treated solution. The only question is therefore whether, in view of the available prior art, the skilled person would consider the stipulated process when seeking an alternative to the process of

D1'.

1.6 In the prior art process, when the absorbing liquid is treated in the three-chamber electrolysers, the organic substances present as anion in that liquid scarcely shift into the concentration chamber with the chloride ions (see D1': Example 2, Table 2; Example 3, Table 4; column 10, lines 15 to 18). In fact, D1' also discloses that, even when a two-chamber electrolyser is used, the concentration of the organic anion in the concentrated liquid is a factor 10 lower than that in the liquid to be treated (see Comparative example, Table 3). Thus, the skilled person can scarcely ignore the finding that the univalent selective anion-exchange membranes, which are permeable to the chloride ions, prevent most of the organic substances from passing through. It is also clear that the concentrated liquid is to be discarded while the treated ("diluted") liquid is kept for recycling (see column 4, line 65 to column 5, line 5; column 6, lines 32 to 44 and Figure). With the aim of retaining the organic substances in the recycle liquid and avoiding its discharge with the waste liquor, D1' thus offers the choice of using either a three-chamber electrolyser or a two-chamber electrolyser.

The Board does not ignore the fact that, in order to avoid the additional risk of scaling, D1' prefers treating the absorbent liquid in a three-chamber electrolyser (D1', paragraph bridging columns 9 and 10). When looking for an alternative to this preferred embodiment, the skilled person, however, would most naturally try to use the two-chamber electrolyser already suggested in D1', especially where the risk of scaling is either acceptable or is not to be expected

as in the present case. In such a case, it also appears obvious to use repeating units as for the three-chamber electrolysers. The Board therefore holds that, in the present case, the deliberate choice of using a less efficient electrolysers with repeating two-chamber units in lieu of the apparatus with repeating three-chamber units preferred by the prior art is merely a matter of choice which depends on the circumstances and not a matter of inventivity. The modification as proposed in claim 1 therefore lacks an inventive step (Article 56 EPC).

1.7 The appellant has asserted that D1' neither relates to the retention of an auxiliary absorption agent nor does it mention a pollution problem due to that auxiliary agent.

1.7.1 It is true that D1' does not mention the expressions "auxiliary absorption agent" or "pollution problem". However, it indicates that the carboxylic acid is being used in the desulfurisation process for improving the SO<sub>2</sub>-absorbability, thus clearly as an auxiliary absorbing agent (column 6, lines 65 to 68). Furthermore, it expressly advises against a method wherein added carboxylic acid is discharged as carboxylate with the waste liquor (see column 2, lines 3 to 11). The Board in this case holds that D1' in fact indirectly addresses the problem of pollution which would be encountered if the auxiliary absorbing agent were discharged with the waste liquor.

1.7.2 The appellant has observed that D3 proposes to use a sulfosuccinate salt in order to avoid pollution problems relating to the use of carboxylic acids such as adipic acid (see D3, column 3, lines 8 to 27). He

has gone on to conclude that the pollution problem would not arise for the process of D1' in which sulfosuccinate is used as auxiliary absorption agent.

However, D3 specifically concerns the pollution problem caused by the decomposition of carboxylic acids used as auxiliary absorbent agent in an off-gas desulfurisation process. In the Board's view, what is suggested in D3 is that sulfosuccinate should be preferred over adipic acid or other compounds which decompose to by-products with offensive odours (see column 2, line 61 to column 3, line 36). Thus, the pollution problem tackled in D3 is not the same as the problem of loss of the auxiliary absorbent agent which is discharged with the absorbing liquid.

On the other hand, D1' discloses the use as auxiliary absorbent agent of carboxylic acids in general, with particular preference for sulfosuccinic acid or adipic acid (column 6, lines 63 to 64). As is already outlined above, the problem of retaining this organic agent - be it sulfosuccinic acid or adipic acid - in the recycle liquid is discussed in D1' (see point 1.2 above) and the solution as proposed in claim 1 is obvious in view of the same document (see point 1.6).

- 1.8 The Board is also unable to follow the appellant's allegation that nothing in D1' indicates or suggests that the organic ions are retained in the desalting chamber and that the present process is therefore distinguished from D1' by the stipulation that the auxiliary agent be recycled in the diluted solution for further absorption (features (i) and (iii), see points VII and 1.1 above). Indeed, D1' clearly indicates that the carboxylic anion which is used as

auxiliary absorbing agent scarcely shifts into the concentrated liquid with the chloride ions but remains in the desalted solution (see column 6, lines 32 to 39 and column 10, lines 15 to 19). Thus, the auxiliary absorbing agent is necessarily recycled with this desalted (or "diluted") solution.

The above analysis is not in contradiction with the appellant's statement with respect to the disclosure of D1', according to which "it is accepted that a three-chambered electrodialyser may retain an auxiliary absorption agent".

- 1.9 The appellant has contended that flue gas desulfurisation processes using calcium compounds as absorbent are classified according to the main ionic components of the absorbent solution. The present application would concern a system wherein calcium ions and magnesium ions are major components (feature (ii), see points VII and 1.1 above). The skilled person therefore would not start from the process of D1' which deals with special problems arising from a large excess of magnesium ions in the absorbent liquid.

The Board, however, cannot accept the above argument since there is no doubt that calcium ions and magnesium ions are also comprised in the filtrate solutions of D1' (see column 7, lines 10 to 15; column 8, lines 5 to 10 and column 9, lines 15 to 20). Furthermore, it is expressly indicated in D1' that the disclosed process can be applied to remove chloride ions from the filtrate **without being limited by the kinds and amount of cations dissolved in the liquid saturated with gypsum** (emphasis added), see D1', column 9, line 67 to column 10, line 4. In addition, were the constitution

of the solution to be electrodialysed different from that of D1', the skilled person would look all the more for an alternative electro dialysis process to that of D1'. For the same reasons as set out in point 1.6 above, he would then arrive at the process as stipulated in claim 1 by an obvious route.

2. Even if the Board had followed the appellant's arguments and accepted D2 as closest prior art document, it would not have reached another conclusion with respect to inventive step.
  - 2.1 As is correctly submitted by the appellant, D2 discloses a process according to the preamble of claim 1, namely a process for treating chlorine-containing effluent discharged from a flue gas desulfurisation process by electro dialysis in a two-chamber electro dialyser.
  - 2.2 With respect to D2, the appellant has argued that the problem to be solved resides in the provision of a flue gas desulfurisation process such that an organic carboxylate acid, which is added to the absorbent solution to improve its desulfurisation efficiency, is retained in the absorbent solution.
  - 2.3 The technical problem as stated above is solved by the provision of a process essentially characterised in that:
    - (i) the absorbent solution further comprises a carboxylic acid and
    - (ii) the anion-exchange membranes are univalent-selective.



2.4 Starting from D2, the Board holds that, should the skilled person envisage improving the desulfurisation efficiency of the process according to D2, he would consider the teaching of D1' disclosing a desulfurisation process in which a carboxylic acid is added to the liquid used for absorbing SO<sub>2</sub> with the aim to improve the absorbability of the latter (see D1', column 1, lines 32 to 38). By thus doing, he would also avoid the loss of the carboxylic acid for the same reasons and in the same manner as suggested in D1' (see also discussion in point 1.6 above). As a result, the skilled person would be led to the process of claim 1 by an obvious and straightforward combination of D2 with D1'.

*Auxiliary request I*

The appellant has submitted that the newly added feature stipulating the concentration of the auxiliary agent is based on the original description page 10, lines 17-18.

The passage indicated relates to the only embodiment illustrating the claimed invention, wherein the carboxylic acid concentration of 5.0 mmol/l is one among a number of specified conditions. The description, however, does not discuss the interdependence of the quantitative values given. In the Board's judgment, it is not plausible, and certainly not proved, that the stipulated carboxylic acid concentration can be chosen by the skilled person, without being pinned down to the other process conditions, for example the quality (SO<sub>2</sub>-loading) and the quantity (flow rate) of the inlet gas. The Board therefore holds that the carboxylic acid concentration

is only originally disclosed in the particular context of the given example. By singling out this parameter and stipulating its combination with other process conditions in the more general context of claim 1, the resulting amendment constitutes subject-matter which extends beyond the content of the application as filed. Claim 1 thus contravenes the requirements of Article 123(2) EPC.

In the case cited by the appellant, an amendment was requested to change the lower limit of a preferred range of values to a specific value taken from an example. After an analysis of the working and comparative examples, the Board came to the conclusion that the amendment represented a reduction of a range to a value already envisaged within the document (see point 8 of the decision T 201/83, published in OJ EPO 1984, 481). The situation in the cited case law is thus different and the decision in question is therefore not relevant for deciding the present case.

#### *Auxiliary request II*

Since claim 1 of this request is the same as claim 1 of the main request, the finding of lack of inventive step in point 1.6 above also applies here.

#### **Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

C. Eickhoff

R. Spangenberg