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Datasheet for the decision of 27 February 2015

Case Number: T 0016/11 - 3.3.05

Application Number: 01951910.7

Publication Number: 1316352

IPC: B01D53/64, B01D53/86, F23J15/00

Language of the proceedings: EN

Title of invention:
METHOD FOR TREATING MERCURY IN EXHAUST GAS AND EXHAUST GAS TREATING SYSTEM

Applicant:
MITSUBISHI HEAVY INDUSTRIES, LTD.

Headword:
Feedback- and feedforward control/MITSUBISHI

Relevant legal provisions:
EPC Art. 56

Keyword:
Inventive step - (no)

Decisions cited:

Catchword:

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Case Number: T 0016/11 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 27 February 2015

Appellant: (Applicant)	MITSUBISHI HEAVY INDUSTRIES, LTD. 5-1, Marunouchi 2-chome, Chiyoda-ku, Tokyo 100-8315 (JP)
Representative:	Henkel, Breuer & Partner Patentanwälte Maximiliansplatz 21 80333 München (DE)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 14 July 2010 refusing European patent application No. 01951910.7 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman	G. Raths
Members:	G. Glod
	O. Loizou

Summary of Facts and Submissions

I. The present appeal lies from the decision of the examining division to refuse European patent application No. EP 01 951 910.

II. The examining division found that the patent application did not meet the requirements of Articles 84, 83 and 56 EPC. The following documents were cited in the decision:

D1: EP-A-860 197
D2: DE-A-44 22 661
D3: DE-A-42 08 355
D4: EP-A-706 815
D5: US-A-5 435 980
D6: EP-A- 208 036

III. In its communication under Article 15(1) of the Rules of Procedure of the Boards of Appeal (RPBA), the Board expressed its preliminary non-binding opinion that the requests on file appeared inter alia not to meet the requirements of Articles 84 and 56 EPC.

IV. In reply to said communication, the applicant (appellant) submitted new requests and the following documents:

D7: US-A-4 418 541
D8: US-A-4 553 924
D9: Integrated dry NO_x/SO_x emissions control system: advanced retractable injection lance SNCR test report, DOE contract Number DE-FC22-91 PC90550, 1997, Section 3.

Claim 1 of the main and auxiliary request 1 is as

follows:

"*1. A method for the treatment of mercury present in exhaust gas of a boiler, wherein exhaust gas produced in the boiler and containing each of nitrogen oxides, sulfur oxides and mercury, after a chlorinating agent has been added thereto, is subjected to a reductive denitration treatment in the presence of a solid catalyst and then to wet desulfurization using an alkaline absorbing fluid,*
the method being ***characterized by*** *:*
detecting the boiler load;
measuring the mercury concentration in the exhaust gas after the wet desulfurization (A);
calculating a predicted value (Y) of the inlet mercury concentration before the reductive denitration treatment on the basis of the measured mercury concentration (A) and on the basis of the detected boiler load; and
controlling the feed rate of the chlorinating agent added prior to the reductive denitration treatment, according to the deviation of the predicted value (Y) of the inlet mercury concentration before the reductive denitration treatment from a reference inlet mercury concentration."

Claim 1 of the second auxiliary request includes in addition the following underlined wording:

"*1. A method for [...]*
and on the basis of the detected boiler load<u>, wherein the predicted value (Y) of the inlet mercury concentration before the reductive denitration treatment is calculated on the basis of the feed rate of the chlorinating agent and the degree of removal of</u>

mercury in the exhaust gas treatment system to which the method is applied; and
controlling the feed rate [...] from a reference inlet mercury concentration."

V. Oral proceedings took place on 27 February 2015.

VI. The arguments provided by the appellant and relevant to the present decision can be summarised as follows:

One could directly conclude from the remaining amount of mercury measured at the outlet the amount of mercury originally present at the inlet, assuming that the removal rates of mercury chloride and mercury(0) and the conversion/oxidation rate were known or could be predetermined in advance based on the configuration and operation of the processing equipment used in the treatment system.

The invention was based on the conclusion that, given that the parameters relating to the equipment could be predetermined, the presumed/predicted concentration of mercury at the inlet could be calculated if the concentration of mercury measured at the outlet and the boiler load were known.

In industrial processes, the actual or current boiler load was constantly monitored and was normally indicative of a percentage between 0 and 100% of the rated boiler load. The commonly applied control equipment in a power plant provided this information in the form of a boiler load signal that under normal operating conditions was proportional to the amount of electric power generated. This was known to the skilled person, who was an engineer with experience in the field of power plants and exhaust gas treatment.

The reference inlet mercury concentration was a predetermined value at the outlet of the boiler for different types of coal and boiler load.

The method of the invention could compensate variations occurring in the process of exhaust treatment, in the boiler load, or in the composition of the coal.

None of the cited prior-art documents appeared to teach that the boiler load signal should be considered when calculating a particular value of the mercury concentration at the various stages in the exhaust gas treatment to accommodate variations in a boiler load and the influence of these variations on the removal of mercury from the exhaust gas.

There was some delay in noting the variations, due to the residence/processing time in the exhaust gas treatment system when the mercury concentration was measured at the outlet.

By taking the boiler load signal into account, this delay could be avoided and the amount of chlorinating agent could be quickly adjusted.

D5 did not teach or suggest to adjust the amount of chloride based on both the type of fuel and the boiler load.

Since the amount of mercury emitted from a boiler did not depend only on the type of fuel but also on the boiler load, variations in the boiler load would change the mercury concentration in the exhaust gas leaving the boiler. Since the mercury detection device was located at the downstream end of the exhaust gas

treatment system, there was a delay in detecting variations in the mercury concentration in the exhaust gas leaving the boiler. This led to inaccurate dosing of chlorinating agent.

This could be avoided by additionally considering the boiler load when calculating the predicted values of the inlet mercury concentration, before the reductive denitration treatment, and of the outlet mercury concentration after the wet desulfurisation treatment.

By providing an arithmetic unit with information about the boiler load, the delay in detecting variations in the mercury concentration of the exhaust gas was minimised and the proper amount of chlorinating agent could be more quickly supplied to the exhaust gas.

D5 was completely silent as to the problem of variations in the load of a boiler causing variations in the mercury concentration of exhaust gas produced therein. D5 was a fast-responding system and did not concern the same setup as in D1.

D5 taught only a feedback control, but no feedforward control. None of the prior-art documents related to feedforward control.

The measurement of mercury before the removal of sulfur was not easy, so continuous monitoring of the input mercury concentration was cumbersome.

According to the invention, the chlorinating agent was added before the denitration catalyst.

In view of the feedback and feedforward control in the

method according to claim 1, the amount of chlorinating agent could be easily and reliably adjusted.

VII. Requests:

The appellant requests that the decision of the examining division be set aside and that a patent be granted on the basis of the claims of the main request or, alternatively, of auxiliary requests 1 or 2, all filed with its letter dated 27 January 2015.

Reasons for the Decision

Main request

1. Article 56 EPC

1.1 Invention

The invention concerns the removal of mercury from an exhaust gas comprising nitrogen oxides, sulfur oxides and mercury by the addition of a chlorinating agent.

1.2 Closest prior art

D1 can be considered as closest prior art since it discloses in claim 1 the removal of mercury from a combustion exhaust gas containing nitrogen oxides, sulfur oxides and mercury by adding a chlorinating agent.

In the embodiment according to figure 1 of D1, an ammonia injecting unit 2 for injecting ammonia supplied from an ammonia tank 3 into exhaust gas and an hydrogen chloride injecting unit 4 are installed in the passage between a boiler 1 and a reduction denitrating unit 5,

which means that the chlorinating agent is added before the exhaust gas reaches the catalyst used for the reduction denitration. Exhaust gas from the boiler 1 is introduced into the reduction denitrating unit 5. Ammonia and hydrogen chloride are injected into the exhaust gas. Nitrogen oxide reacts with ammonia and mercury (the metal) is oxidised to mercury chloride in the presence of hydrogen chloride in the reduction denitrating unit 5 at the same time. The exhaust gas flows through an air preheater 6 and a heat exchanger 7 to reach an electric precipitator 8 where dust is removed and then sulfur dioxide and mercury chloride in the exhaust gas are removed at the same time in a wet desulfurising unit 9 (D1: figure 1 and page 4, lines 15 to 21).

The exhaust gas of dust coal is used in the examples (D1: page 4, line 45).

1.3 Problem

According to the application in suit the problem was to reduce system-equipment corrosion and utility costs (see page 3, lines 15 to 21 of the application as filed).

1.4 Solution

As a solution to this problem the application proposes a method according to claim 1 characterised by detecting the boiler load, measuring the mercury concentration in the exhaust gas after the wet desulfurisation (A), calculating a predicted value (Y) of the inlet mercury concentration before the reductive denitration treatment on the basis of the measured mercury concentration (A) and on the basis of a

detected boiler load, and controlling the feed rate of the chlorinating agent added prior to the reductive denitration treatment, according to the deviation of the predicted value (Y) of the inlet mercury concentration before the reductive denitration treatment from a reference inlet mercury concentration.

1.5 Success of the solution

The board accepts that the problem has been solved, since it is generally known that controlling the feed rate of a chemical reactant in relation to the reaction conditions allows optimisation of the dosing of the reactant. In the present case, controlling the feed rate of the chlorinating agent makes it possible to avoid overdosage and underdosage.

The board is satisfied that there is no need to reformulate the technical problem in the light of document D1.

1.6 Obviousness

It remains to be decided whether the solution to the problem is obvious.

The board will address three points, in the following order:
(1) the claimed method
(2) the teaching of the prior art
(3) the common general knowledge of the skilled person
(4) before reaching its conclusion.

(1) The claimed method

The method according to claim 1 includes the measurement of the mercury concentration after desulfurisation. This value is taken to calculate a predicted value of the inlet mercury concentration on the basis of the known parameters relating to the equipment and in addition on the basis of the boiler load. If this value is not in accordance with the reference inlet mercury concentration, the dosage of the chlorinating agent was not correct and needs to be adjusted.

In other words, the adjustment of the dosage of the chlorinating agent depends on two variables:

(a) a feedback control: the measured mercury concentration after treatment,

(b) a feedforward control: the input into the boiler before the addition of the chlorinating agent.

The question is whether such a control mechanism is derivable from the cited prior art.

The board will assess whether D5 relates to
(a) a feedback control and
(b) a feedforward control,
and will then also consider possible
(c) differences between document D5 and the claimed method.

(2) The teaching of the prior art

D5 relates to flue gas purification and to the removal of mercury from flue gases originating from the combustion of coal (D5: column 1, lines 7 to 10). It is thus in the same field as D1.

In the process disclosed in D5 (illustrated in the drawing) a stream of flue gas from a pre-heater of a coal-fired boiler is introduced into a drying chamber 1 via a duct 4. An aqueous absorbent suspension is prepared in a mixing vessel 5 and by means of an atomiser wheel 6 atomised into the chamber 1. By contact with the hot flue gases in the chamber the water evaporates from the atomised suspension, whereby the temperature of the gas decreases substantially and at the same time acidic substances, mainly sulfur dioxide, in the flue gas react with the basic absorbent, producing a particulate material primarily comprising salts formed by said reaction, together with non-reacted absorbent. Some mercury present in the flue gas is also removed together with the particulate material. The amount of mercury removed together with particulate material is far less than desired if the chloride content of the gas is low (D5: column 5, lines 23 to 65).

Therefore, either a device 11 for measuring the chloride concentration in the flue gas is inserted into the duct 4, or alternatively **the chloride concentration in the flue gas is calculated or estimated on the basis of the chloride contents of the coal used in the boiler** (D5: column 5, line 66 to column 6, line 2).

If the chloride content is found to be below a certain value, hydrogen chloride and activated carbon are together introduced into the flue gas and, admixed with the latter, introduced into the spray drying absorption zone, thereby substantially increasing the mercury sorption taking place in this zone (D5: column 6, lines 3 to 23).

For the board, D5 relates to
(a) a feedback control and
(b) a feedforward control.

(a) feedback control

The amount of chloride introduced, and the amount of activated carbon possibly dosed, **may be adjusted also on the basis of the mercury content of the treated gas** measured by means of a device 18 arranged in the duct 9 (column 6, lines 44 to 46).

It is evident from D5 that the goal of the process is to add chloride in such an amount that mercury is efficiently removed (avoid underdosing of chloride). D5 like D1 discloses that the mercury can be removed together with sulfur oxides in the form of mercury chloride (see also claim 1 of D5).

D5 teaches that the amount of chloride, which is the chlorinating agent, is adjusted **based on the mercury concentration measured at the end of the treatment**, which is typical of a feedback control.

(b) feedforward control

Furthermore, the chloride dosing is also adjusted based on the calculated chloride concentration present in the flue gas. This calculation is based on the amount of chloride present in the coal and is a type of feedforward control, since it makes it possible to adjust the chloride concentration to be added to the exhaust gas on the basis of chloride already present in the fuel.

This **feedforward** does not help to control the feed-rate

of the chlorinating agent based on the mercury input, but **based on the chloride input.**

Both the feedforward and feedback controls ensure that the concentration of chloride (and activated carbon) is high enough to maximise the amount of mercury removed.

(c) Differences between D5 and the claimed method

However, D5 is silent about problems arising due to overdosing of hydrogen chloride. Therefore, the only difference between the control system of D5 and claim 1 is that the feedforward control is not used for calculating the mercury concentration and for subsequently controlling the feed-rate of the chlorinating agent based thereon. In other words, D5 does not teach to adapt the feed-rate of the chlorinating agent based on changes in the input.

So D5 generally teaches a feedback control and a feedforward control, but not with the purpose of avoiding overdosage of hydrogen chloride. However, it is self-evident that not only the chloride input depends on the type of coal, but also the mercury input.

(3) The common general knowledge

The skilled person (a process engineer with experience in the field of power plants and exhaust gas treatment) additionally knows that in industrial processes the actual or current boiler load is constantly monitored, which means that changes in the input are known. He is able to incorporate values obtained from this monitoring into control systems. It is also generally known to him that in a chemical process that includes

reactions between different reactants the dosing of the reactants needs to be optimised and adjusted to changes in order to avoid the disadvantages which otherwise arise (e.g. cost, side-reactions, waste).

Therefore, this very same skilled person will inevitably adjust the amount of hydrogen chloride (chlorinating agent) if the amount of mercury in the input changes.

Changes in the input are mainly due to changes in the type and amount of fuel. Factors influencing the output of mercury include the operating conditions of the system. If the operating conditions and type of fuel remain constant, the output is influenced only by the boiler load.

The skilled person whilst trying to avoid adverse effects due to incorrect dosing of the chlorinating agent knows that the feed rate of the chlorinating agent can be adjusted based not only on the measured mercury outlet concentration (feedback control) but also on the mercury input, which includes the type of fuel and the boiler load (feedforward control).

(4) Conclusion

The solution to the posed problem is considered obvious in view of the teaching of D5 and the skilled person's general knowledge, with the result that claim 1 of the main request lacks inventive step.

The main request must fail.

Auxiliary request 1

Claim 1 of this request is identical to claim 1 of the main request, so the conclusion reached for claim 1 of the main request also applies here.

Therefore, auxiliary request 1 must also fail.

Auxiliary request 2

Claim 1 of this request includes the additional feature that the inlet mercury concentration is calculated on the basis of the feed rate of the chlorinating agent (hydrogen chloride) and the degree of removal of mercury in the exhaust gas treatment system to which the method is applied.

This is considered a different way of calculating the predicted value Y that also requires some kind of preliminary calibration, since the degree of removal can only be known if the operation parameters of the plant and the tabulation of removal degrees for the specific type of fuel and system are known. Such a calculation has to be considered as being within the knowledge of the person skilled in the art.

It is generally known that a process can be controlled based on several parameters, and the skilled person would use the most relevant parameters when setting up a control system. The process of claim 1 is based on boiler load and measured mercury concentration in the exhaust gas after wet desulfurisation. However, it does not include all possible parameters, e.g. a feedforward control based on changes in the type of fuel.

Therefore, it is concluded that the controlling of the

feed-rate of the chlorinating agent is an evident process step that the skilled person would fine-tune as needed.

Hence, the subject-matter of claim 1 of this request does not involve an inventive step. Auxiliary request 2 must also fail.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

C. Vodz G. Raths

Decision electronically authenticated