(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
9 March 2017 (09.03.2017)

WIPO | PCT

(10) International Publication Number
WO 2017/037076 A1

(51) International Patent Classification:
*B01D 53/14* (2006.01) *B01D 53/50* (2006.01)
*B01D 53/18* (2006.01) *B01D 53/78* (2006.01)

(21) International Application Number:
PCT/EP2016/070439

(22) International Filing Date:
31 August 2016 (31.08.2016)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
15183213.6 31 August 2015 (31.08.2015) EP

(71) Applicant: **STEINMÜLLER ENGINEERING GMBH** [DE/DE]; Fabrikstrasse 5, 51643 Gummersbach (DE).

(72) Inventors: **RASCHE, Dorian**; c/o Steinmüller Engineering GmbH, Fabrikstrasse 5, Gummersbach 51643 (DE). **BINKOWSKI, Stefan**; c/o Steinmüller Engineering GmbH, Fabrikstrasse 5, 51643 Gummersbach (DE).

(74) Agent: **GODEMEYER BLUM LENZE PATENTANWÄLTE, PARTNERSCHAFT MBB**; An den Gärten 7, 51491 Overath (DE).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

**Declarations under Rule 4.17**:

— *as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii))*

*[Continued on next page]*

(54) Title: METHOD AND DEVICE FOR FLUE GAS DESULFURIZATION

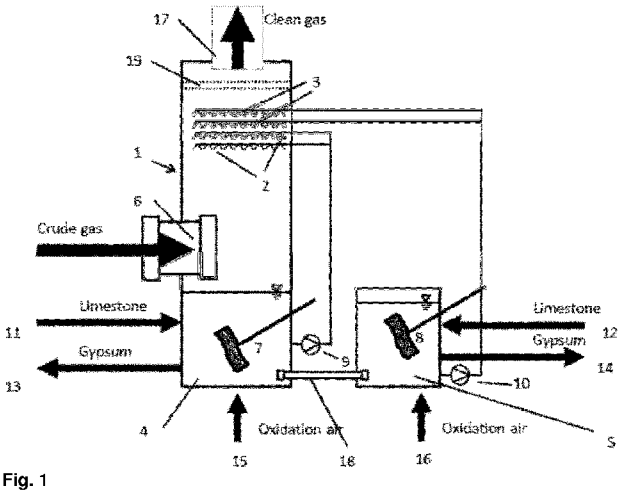


Fig. 1

(57) Abstract: The invention relates to a method for the treatment of a flue gas stream for reducing the $SO_X$ content in said flue gas by using absorbents based on suspensions that contain calcium compounds, wherein the flue gas initially streams through a first section and then through a second section of a flue gas scrubber, wherein the flue gas in the first section and in the following second section is sprayed with absorbent, wherein the pH of the absorbent sprayed in the first section is lower than the pH of the absorbent sprayed in the second section, and wherein the absorbent sprayed in the first and second section is collected together. Furthermore, the invention relates to a device for the treatment of a flue gas stream using absorbents based on suspensions that contain calcium compounds, wherein the device has a container, first and second spraying devices, located inside said container, for spraying said absorbents, wherein the second spraying devices are located above the first spraying devices, as well as an absorber sump and a second absorbent receptacle downstream of said absorber sump for receiving the absorbent, wherein, above the absorber sump and below the first spraying devices, a gas inlet for the flue gas stream is provided, wherein two absorbent loops are provided, wherein, in the first absorbent loop a first partial stream of the absorbent is conducted directly from the absorber sump to the first spraying device in order to be sprayed, and a second partial stream of the absorbent is conducted from the absorber sump to the second absorbent receptacle downstream of said absorber sump and from there to the second spraying devices, wherein the absorbent streams sprayed in the first and second spraying devices are conducted back into the absorber sump.

WO 2017/037076 A1

**Published**:

— *with international search report (Art. 21(3))*

# Method and Device for Flue Gas Desulfurization

The invention relates to methods for the treatment of a flue gas stream for reducing the $SO_x$ content in the flue gas by using absorbents based on suspensions that contain calcium compounds, as well as to a device for implementing said method.

## Technical Background

Such methods and devices are especially used for the desulfurization of exhaust gases which are generated in fossil-source fueled power plants for the generation of electricity or in garbage incineration plants. These exhaust gases or flue gases, respectively, contain a plurality of noxious substances such as $SO_x$. Considering environmental protection, it is necessary to reduce the emission of such substances into the atmosphere, making their separation from the exhaust gas essential.

Desulfurization of flue gas is carried out according to the following basic principle: unpurified flue gas is introduced into a flue gas scrubber and, in the scrubbing or absorption zone, respectively, sprinkled with a mixture of water and limestone, causing the sulfur dioxide being largely absorbed by chemical reactions. In the process, the gaseous sulfur dioxide initially is dissolved in the absorption liquid. By reacting sulfur dioxide with limestone, eventually calcium sulfite and $CO_2$ are produced. In the lower part of the scrubbing tower, in the absorber sump, the scrubbing or absorbent suspension, respectively, accumulates, loaded with calcium sulfite. By injecting air, a gypsum suspension is produced which can be suspended by agitators in order to avoid the formation of deposits in the absorber sump.

The wet desulfurization of flue gas from coal-fired power plants by using a limestone suspension is a method that can so far be carried out in a single-loop process or in a double-loop process.

In the single-loop process, the flue gas is usually conducted into the center of the absorber, streams through it in an upward direction and leaves the absorber in its upper section. The scrubbing suspension is conducted from the absorber sump to the spray levels by way of circulation pumps, sprayed via the nozzles inside the absorber, thus scrubbing the flue gas in counter flow. After the $SO_2$ has been absorbed, the drops fall back into the absorber sump, thus closing the (single) loop of the scrubbing suspension.

In the double-loop process, the absorption zone is divided into two sections that are normally arranged on top of each other. The flue gas consecutively streams through these sections in an upward direction. The scrubbing suspension is conducted in counter flow, but diverted in the center of the absorption zone, which creates two scrubbing suspension loops that are mostly independent of each other. The limestone suspension is (mainly) added to the upper loop, the gypsum is withdrawn from the lower loop. The absorber is controlled by transferring the suspension from the upper to the lower loop.

In DE 29 44 536 and DE 30 11 592 a method is described in which at least a part of the consumed absorbent of the second upper loop is collected and conducted back to an absorbent supply container via a collecting and recirculation device.

From DE 43 45 364 it is known that in such a double-loop scrubber the consumed absorbent of the second (upper) loop is conducted back into the absorbent container, where the reaction between sulfur oxide compounds and calcium compounds continues.

DE-A-196 01 193 relates to a method in which the absorbent is stored inside the scrubber within a container that is open at the top and inside of which the absorbent is at least partially collected after the scrubbing, wherein the liquid level in the container is higher than the liquid level in the sump of the scrubber.

In DE 10 2010 002 768 A1 different methods are described. Figure 2 relates to a device for a single-loop process. Figure 1 relates to a device for a double-loop process, wherein the absorbent sprayed in the upper section is collected by a collector unit that is integrated above the lower spraying device and then conducted back to the upper spraying device and, thus, remains (almost) completely separated from the lower loop. Figure 3 relates to a device for a double-loop process, wherein the absorbent sprayed in the upper section is collected together with the absorbent that is sprayed by the lower spraying device. The absorbent enters the absorber sump and is subsequently divided into two partial streams in order to supply the lower and the upper spraying devices directly.

The task of the present invention was to carry out the wet desulfurization of flue gases in a more efficient way and to achieve a better fine cleaning of the flue gas.

**Description of the Invention**

The technical task is solved by a method for the treatment of a flue gas stream in order to reduce the $SO_x$ content in the flue gas by using absorbents based on suspensions that contain calcium compounds, wherein the flue gas initially streams through a first section and then through a second section of a flue gas scrubber, wherein the flue gas in the first section and in the following second section is sprayed with absorbent, wherein, according to the invention, the pH of the absorbent sprayed in the first section is lower than the pH of the absorbent sprayed in the second section, and wherein the absorbent sprayed in the first and second section is collected together.

In a preferred embodiment the pH of the absorbent sprayed in the first section is from 4.5 to 5.9 and the pH of the absorbent sprayed in the second section is from 5.0 to 6.8.

Furthermore, a method is preferred in which in a first loop the flue gas in the flue gas scrubber is sprayed with an absorbent in a first section of the flue gas

stream, the absorbent then reaches an absorber sump, and a first partial stream of the absorbent is conducted directly from the absorber sump to the first section of the flue gas scrubber for being sprayed again, wherein the method contains a second absorbent loop wherein a second partial stream of the absorbent is conducted from the absorber sump into a second absorbent receptacle downstream of said absorber sump, and is subsequently sprayed in the second section of the flue gas stream, and wherein the absorbent returns to the absorber sump together with the absorbent that was sprayed in the first section.

In another preferred method the flue gas scrubber has spraying devices for spraying the absorbents as well as an absorber sump for receiving the absorbent, wherein the flue gas streams through the flue gas scrubber from bottom to top and is being sprayed with absorbent that is sprayed by first spraying devices and by second spraying devices, the absorbent then reaches the absorber sump, and a first partial stream of the absorbent is conducted directly from the absorber sump to the flue gas scrubber in order to be sprayed,
wherein a second partial stream of the absorbent is conducted from the absorber sump into a second absorbent receptacle downstream of said absorber sump, then sprayed inside the flue gas scrubber via the second spraying devices and subsequently returns to the absorber sump,
wherein the second spraying devices are located above the first spraying devices and, with regard to the flue gas stream, downstream of the first spraying devices.

Furthermore, the pH of the absorbent sprayed in the first spraying devices is lower than the pH of the absorbent sprayed in the second spraying devices. Thus, the pH of the absorbent sprayed in the first spraying devices is from 4.5 to 5.9 and the pH of the absorbent sprayed in the second spraying devices is from 5.0 to 6.8.

The pH of the absorbent in the absorber sump is lower than the pH of the absorbent in the second absorbent receptacle that is located downstream. Thus, the

pH of the absorbent in the absorber sump is from 4.5 to 5.9 and the pH of the absorbent in the second absorbent receptacle is from 5.0 to 6.8.

In another preferred embodiment, for the purpose of oxidation, air or oxygen is fed into the absorber sump and into the second absorbent receptacle downstream of said absorber sump.

Furthermore, it is preferred that fresh absorbent based on suspensions that contain calcium compounds is fed to the absorber sump continuously or discontinuously. It is particularly preferred when fresh absorbent based on suspensions that contain calcium compounds is fed continuously or discontinuously into the absorber sump and into the second absorbent receptacle downstream of said absorber sump.

In a very particularly preferred embodiment, fresh absorbent based on suspensions that contain calcium compounds is fed continuously or discontinuously into the absorber sump and into the second absorbent receptacle downstream of said absorber sump, as well as air or oxygen for the purpose of oxidation.

As already mentioned, according to the invention, the flue gas is sprayed with absorbent in the first section and in the following second section of the flue gas stream, wherein the pH of the absorbent sprayed in the first section is lower than the pH of the absorbent sprayed in the second section. The absorbent sprayed in the first and second section is received or collected together and enters the absorber sump together. This means that essentially the entire absorbent sprayed in the second section, i.e. at least 90 %, preferably at least 95 %, further preferred at least 98 %, even more preferred at least 99 %, of the absorbent sprayed and recycled in the second section is collected together with the absorbent sprayed in the first section and directly goes into the absorber sump.

The present invention also provides a device for the treatment of a flue gas stream using absorbents based on suspensions that contain calcium com-

pounds, wherein the device has a container, first and second spraying devices located inside the container for spraying the absorbents, wherein the second spraying devices are located above the first spraying devices, as well as an absorber sump and a second absorbent receptacle downstream of said absorber sump for receiving the absorbent, wherein above the absorber sump and below the first spraying devices a gas inlet for the flue gas stream is provided, wherein two absorbent loops are provided, wherein in the first absorbent loop a first partial stream of the absorbent is conducted directly from the absorber sump to the first spraying devices for being sprayed, and a second partial stream of the absorbent is conducted from the absorber sump to the second absorbent receptacle downstream of said absorber sump and from there to the second spraying devices, wherein the absorbent streams sprayed in the first and second spraying devices are conducted back into the absorber sump.

According to the present invention, the second absorbent receptacle, which is located downstream of the absorber sump, is a container that is separated from the absorber sump and which is in fluid connection with said absorber sump. In the simplest embodiment, the connection between the absorber sump and the downstream second absorbent receptacle is represented by a pipeline. The connection between the absorber sump and the downstream second absorbent receptacle may, in addition, be equipped with a pump, which conducts the absorbent to the downstream second absorbent receptacle.

In a preferred device, the absorber sump and the second absorbent receptacle downstream of said absorber sump have one or more supply lines for supplying air or oxygen for the purpose of oxidation.

In another preferred device, the absorber sump has one or more supply lines for supplying fresh absorbent based on suspensions that contain calcium compounds. Particularly preferred is a device in which the absorber sump and the second absorbent receptacle located downstream of said absorber sump each

have one or more supply lines for supplying fresh absorbent based on suspensions that contain calcium compounds.

In a preferred device, it is provided that the absorber sump and the second container are each equipped with a supply for oxidation air and a supply for fresh absorbent.

In another preferred device, a mist eliminator is provided above the second spraying device.

Preferably, in the device according to the present invention, there is particularly no separation between the first and the second spraying device, i.e. the first and second spraying device are located within the same area of the container, so that the absorbent sprayed in the first and second spraying device is collected together and enters the absorber sump together.

The new method for wet desulfurization of flue gas or the new device for carrying out said method, respectively, combines the advantages of the single-loop and the double-loop scrubber. By separating the absorber sump, the absorbent can be controlled at two different pH values. This means that in the first absorbent loop the pH is below and in the second loop the pH is above the usual pH of a single-loop scrubber, which allows for the primary tasks of the absorber to be accomplished considerably more efficient.

At a lower pH, dissolving of the limestone is considerably faster in the first absorber sump, so that in the first absorber sump an increased dissolution occurs. As a result, the carbonate content of the gypsum decreases and a considerably smaller reaction volume (sump volume) has to be provided for dissolving the limestone. In the second absorbent receptacle, which is located downstream of the (first) absorber sump, the pH is higher than in the (first) absorber sump. The higher pH in the downstream absorbent receptacle is already achieved by the subsequent dissolution of the substances (limestone) in the absorbent that is

coming from the absorber sump. Preferably, in the downstream absorbent receptacle, the pH can additionally actively be adjusted to a higher value as compared to the pH inside the first loop or the absorber sump, respectively. The higher pH in the downstream absorbent receptacle or in the second loop, respectively, causes the $SO_2$ absorption to be carried out considerably more efficient. The positive influence of the increased pH of the absorbent in the second loop is especially noticeable in the fine-cleaning section of the flue gas, for which reason the upper spray levels are supplied from the second absorbent receptacle.

A further advantage of the present invention is that it is not necessary to install a collection device for the scrubbing suspension or the absorbent in the absorber, respectively, so that the flue gas can stream through the absorber without hindrance. Thereby, pressure loss is decreased and a more even distribution of the flue gas across the cross section is possible. This also leads to an enhanced overall cleaning.

In the method according to the invention the flue gas is sprayed with absorbent in the first section and in the following second section. According to the invention, the pH of the absorbent sprayed in the first section is lower than the pH of the absorbent sprayed in the second section. The absorbent sprayed in the first and second sections is collected together and enters the absorber sump together. In contrast to the double-loop process described in the prior art, the absorbent which is sprayed in the second loop or sprayed by the second spraying devices, respectively, is, according to the invention, not collected and discharged separated from the absorbent which is sprayed in the first loop or sprayed by the first spraying devices, respectively. This means that, according to the present invention, the absorbent sprayed in the first and second section enters the absorber sump together and is mixed therein.

Advantageously and preferably, the two absorbent receptacles, the absorber sump and the downstream absorbent receptacle are provided with agitators.

By separating the absorber sump into two stirrer tanks that are connected in series, the total volume can be reduced considerably, maintaining the efficiency of the sump, so that the added volume of the two sumps is lower than the volume of a conventional single-loop scrubber. Despite this reduction of the sump volume, the effective dwell time of the substances that are important in terms of process engineering (limestone, gypsum) can be maintained and even systematically be controlled independently of each other.

By over-dimensioning the second absorbent receptacle, it is possible to omit the storage tank for an emergency draining of the power plant, which leads to further cost advantages. The storage tank for an emergency draining is seldom used and in case of an inspection it receives the content of the absorber to allow for re-using the absorbent when the facility will be started afterward. When employing the method of the present invention, in which two receptacles for the absorbent, i.e. the absorber sump and the downstream absorbent receptacle, are used, the storage tank for an emergency draining is not needed, since the second sump could remain filled by using a valve between the two sumps. Excess liquid might, if necessary, be disposed of.

Additionally, the above-described technology of a flue gas desulfurization plant (FDG plant) having two absorbent receptacles may be retrofitted in existing plants. If the absorber sump of an existing plant is dimensioned too small and an additional spray level is to be retrofitted, the total separation efficiency of the plant can additionally be increased by installing a second absorbent receptacle.

**Figure 1** shows a device for the treatment of a flue gas stream. The flue gas scrubber depicted in the figure has a container 1, in the base of which an absorber sump 4 is formed. Above the absorber sump 4, said container 1 is provided with an inlet 6 for a gas to be treated. In order to establish a first scrubbing stage, a first spraying device 2, schematically depicted in the figure, is provided spaced apart from the surface of the absorber sump 4. The absorbent from the absorber sump 4 is circulated using one or more pumps 9.

Above the first spraying device 2 a second spraying device 3 is located. The spraying devices 2, 3 may have one or more spray levels, wherein in the figure two spray levels for the first spraying device 2 and two spray levels for the second spraying device 3 are depicted. The second spraying device 3 is supplied with an absorbent which is circulated by way of the second absorbent receptacle 5, one or more pumps 10, the second spraying device 3, the absorber sump 4 and a connection line 18 to the second absorbent receptacle 5. Above the second spraying device 3, a mist eliminator 19 is located which separates the liquid that was carried away by the gas and reintroduces it to the process, i.e. the separated liquid enters the absorber sump 4. Accordingly, the gas is demisted.

In the embodiment shown in the figure, the absorber sump 4 and the second absorbent receptacle 5 are fed with fresh alkali-containing absorbent suspension via the supply lines 11, 12 when the desulfurization is carried out. The absorber sump 4, as well as the second absorbent receptacle 5, are provided with one or more agitators 7, 8.

For the generation of gypsum, the absorber sump 4 and the second absorbent receptacle 5 are additionally fed with oxidation air via the supply lines 15, 16. The gypsum-containing suspension is withdrawn from the absorber sump 4 and from the second absorbent receptacle 5 at position 13 and 14, respectively. The treated gas is withdrawn at the top of container 2 via a gas outlet connector 17.

**List of Reference Signs**

1 container
2 first spraying device
3 second spraying device
4 absorber sump
5 absorbent receptacle
6 inlet for flue gas
7 agitator (of the absorber sump)
8 agitator (of the absorbent receptacle)
9 pump (1st loop)
10 pump (2nd loop)
11 supply line for calcium-containing absorbent (to absorber sump)
12 supply line for calcium-containing absorbent (to absorbent receptacle)
13 withdrawal outlet for gypsum (from the absorber sump)
14 withdrawal outlet for gypsum (from the absorbent receptacle)
15 supply line for oxidation air (to absorber sump)
16 supply line for oxidation air (to absorbent receptacle)
17 gas outlet connector
18 connection line between absorber sump and absorbent receptacle
19 mist eliminator

## Claims

1. A method for the treatment of a flue gas stream for reducing the $SO_x$ content in said flue gas by using absorbents based on suspensions that contain calcium compounds, wherein the flue gas initially streams through a first section and then through a second section of a flue gas scrubber, wherein the flue gas is sprayed with absorbent in the first section and in the following second section, **characterized in that** the pH of the absorbent sprayed in the first section is lower than the pH of the absorbent sprayed in the second section, and that the absorbent sprayed in the first and second section is collected together.

2. The method according to claim 1, **characterized in that** the pH of the absorbent sprayed in the first section is from 4.5 to 5.9, and the pH of the absorbent sprayed in the second section is from 5.0 to 6.8.

3. The method according to claim 1 or 2, wherein, in a first loop, the flue gas in the flue gas scrubber is sprayed with absorbent in a first section of the flue gas stream, the absorbent then enters an absorber sump and a first partial stream of the absorbent is conducted directly from the absorber sump to the first section of the flue gas scrubber in order to be sprayed again, **characterized in that** said method contains a second absorbent loop, wherein a second partial stream of the absorbent is conducted from the absorber sump into a second absorbent receptacle downstream of said absorber sump, and subsequently sprayed in the second section of the flue gas stream, and the absorbent reenters the absorber sump together with the absorbent that was sprayed in the first section.

4. The method according to any one of claims 1 to 3, **characterized in that** the flue gas scrubber has spraying devices for spraying the absorbents, as well as an absorber sump for receiving the absorbent, wherein the flue gas streams through the flue gas scrubber from bottom to top and is sprayed

with absorbent that is sprayed by first spraying devices and by second spraying devices, the absorbent then enters the absorber sump and a first partial stream of the absorbent is conducted directly from the absorber sump into the flue gas scrubber in order to be sprayed,
wherein a second partial stream of the absorbent is conducted from the absorber sump into a second absorbent receptacle downstream of said absorber sump, then sprayed inside the flue gas scrubber using the second spraying devices and then reenters the absorber sump,
wherein the second spraying devices are located above the first spraying devices and, with regard to the flue gas stream, downstream of said first spraying devices.

5. The method according to claim 4, **characterized in that** the pH of the absorbent sprayed in the first spraying devices is lower than the pH of the absorbent that is sprayed in the second spraying devices.

6. The method according to claim 4 or 5, **characterized in that** the pH of the absorbent sprayed in the first spraying devices is from 4.5 to 5.9, and the pH of the absorbent sprayed in the second spraying devices is from 5.0 to 6.8.

7. The method according to any one of claims 3 to 6, **characterized in that** the pH of the absorbent in the absorber sump is lower than the pH of the absorbent in the downstream second absorbent receptacle.

8. The method according to any one of claims 3 to 7, **characterized in that** the pH of the absorbent in the absorber sump is from 4.5 to 5.9, and the pH of the absorbent in the second absorbent receptacle is from 5.0 to 6.8.

9. The method according to any one of claims 3 to 8, **characterized in that** air or oxygen is fed into the absorber sump and into the second absorbent receptacle downstream of said absorber sump for the purpose of oxidation.

10. The method according to any one of claims 3 to 9, **characterized in that** fresh absorbent based on suspensions that contain calcium compounds is fed continuously or discontinuously into the absorber sump.

11. The method according to any one of claims 3 to 9, **characterized in that** fresh absorbent based on suspensions that contain calcium compounds is fed continuously or discontinuously into the absorber sump and into the second absorbent receptacle downstream of said absorber sump.

12. The method according to any one of claims 3 to 8, **characterized in that** fresh absorbent based on suspensions that contain calcium compounds is fed continuously or discontinuously into the absorber sump and into the second absorbent receptacle downstream of said absorber sump, as well as air or oxygen for the purpose of oxidation.

13. A device for the treatment of a flue gas stream using absorbents based on suspensions that contain calcium compounds, wherein the device has a container, first and second spraying devices, located inside said container, for spraying said absorbents, wherein the second spraying devices are located above the first spraying devices, as well as an absorber sump and a second absorbent receptacle downstream of said absorber sump for receiving said absorbent, wherein a gas inlet for the flue gas stream is provided above the absorber sump and below the first spraying devices,
**characterized in that** two absorbent loops are provided, wherein, in the first absorbent loop, a first partial stream of the absorbent is conducted directly from the absorber sump to the first spraying devices in order to be sprayed, and a second partial stream of the absorbent is conducted from the absorber sump to the second absorbent receptacle downstream of said absorber sump and from there to the second spraying devices, wherein the absorbent streams sprayed in the first and second spraying devices are conducted back into the absorber sump.

14. The device according to claim 13, **characterized in that** the second absorbent receptacle, located downstream of the absorber sump, for receiving the absorbent is a container that is separated from the absorber sump and which is in fluid connection therewith, and wherein, preferably, the absorber sump and the second container are each provided with a supply for oxidation air and with a supply for fresh absorbent.

15. The device according to claim 13 or 14, **characterized in that** a mist eliminator is provided above the second spraying device.

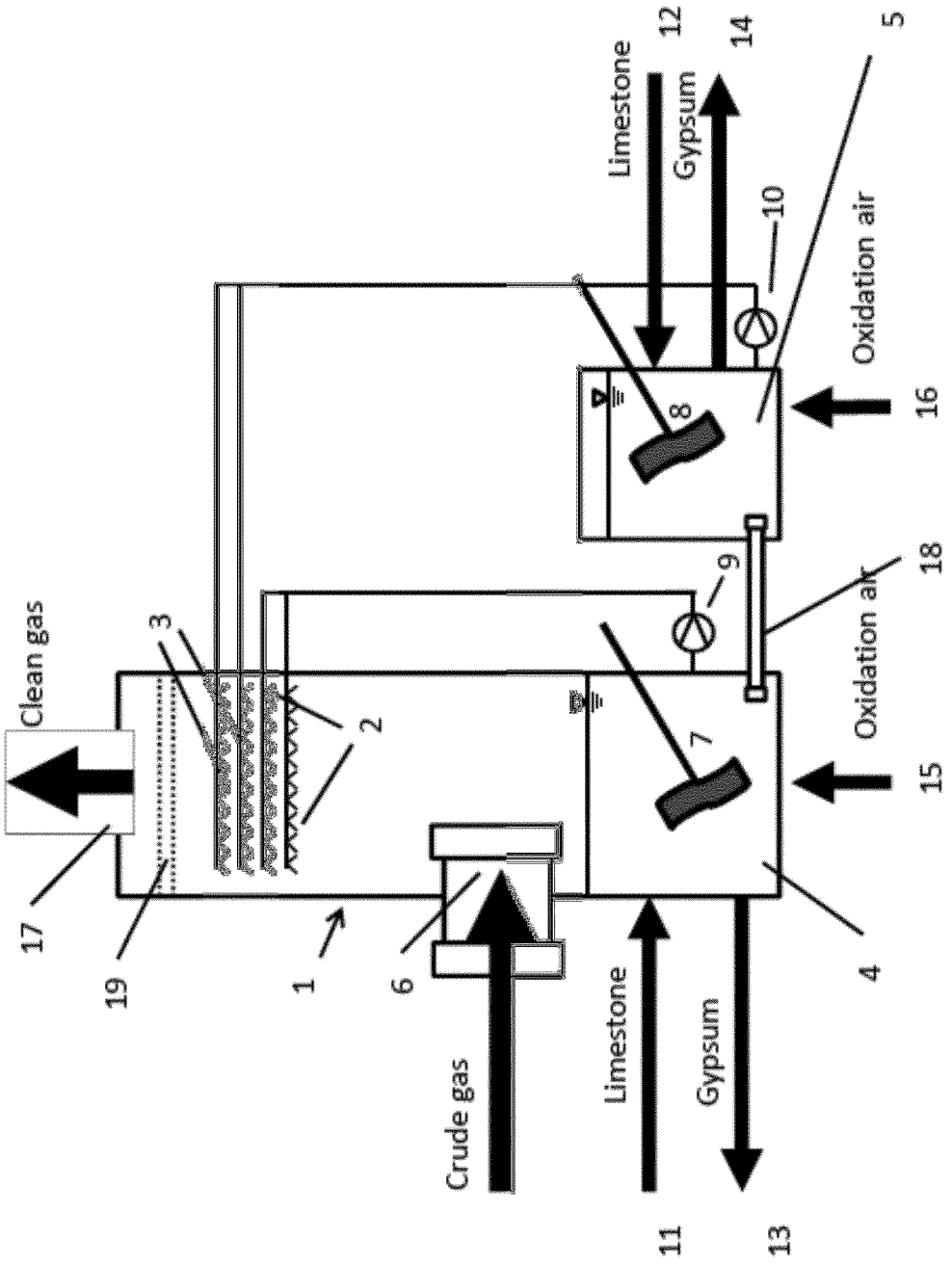
Clean gas
Crude gas
Limestone
Gypsum
Limestone
Gypsum
Oxidation air
Oxidation air
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Fig. 1

# INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2016/070439

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01D53/14 B01D53/18 B01D53/50 B01D53/78
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| X | US 4 305 909 A (WILLETT HOWARD P ET AL) 15 December 1981 (1981-12-15) | 1,3-5, 7-15 |
| Y | columns 1,4,8,9; figure 1 | 2,6,8 |
| | ----- | |
| X | US 5 512 072 A (LASLO DENNIS J [US]) 30 April 1996 (1996-04-30) column 6 | 1 |
| | ----- | |
| X | DE 23 13 264 A1 (OSAKA KOHAI CO) 20 September 1973 (1973-09-20) | 1,3-5,7, 9-15 |
| Y | figure 1 | 2,6,8 |
| | ----- | |
| X | DE 37 21 684 A1 (KRC UMWELTTECHNIK GMBH [DE]) 12 January 1989 (1989-01-12) | 1,2 |
| Y | claim 1; figure 1 | 2,6,8 |
| | ----- | |
| | -/-- | |

[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

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| European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. (+31-70) 340-2040,<br>Fax: (+31-70) 340-3016 | Bergt, Thomas |

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