

The Commissioner of Patents  
PO Box 200  
WODEN ACT 2606

F B RICE & CO  
SYDNEY NSW  
Speed Dial 511

69 4 3 5 2

PATENTS

Our Ref: 71794

AUSTRALIA

*Patents Act 1990*

PATENT REQUEST: STANDARD PATENT

We, CHIYODA CORPORATION being the person(s) identified below as the Applicant, request the grant of a standard patent to the person identified below as the Nominated Person, for an invention described in the accompanying complete specification.

Full application details follow.

Applicant and Nominated Person: CHIYODA CORPORATION

Address: 12-1, Tsurumichuo 2-chome, Tsurumi-ku,  
Yokohama-shi, Kanagawa-ken Japan

Invention Title: Process for the desulfurization of sulfurous acid  
gas-containing waste gas

Name(s) of Actual Inventor(s): Haruo NISHINO; Yoshio OGAWA; Kazushige  
KAWAMURA

Address for Service in Australia: F B RICE & CO  
28A Montague St  
BALMAIN NSW 2041

Attorney Code: RI

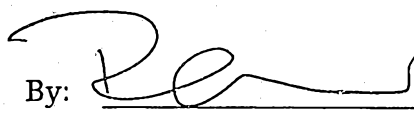
BASIC CONVENTION APPLICATION(S) DETAILS

| <u>Application No</u> | <u>Country</u> | <u>Country Code</u> | <u>Date of Application</u> |
|-----------------------|----------------|---------------------|----------------------------|
| 7-155119              | Japan          | JP                  | 30 May 1995                |

Drawing number recommended to accompany the abstract: Fig 1

Dated this twenty-eighth day of May 1996

CHIYODA CORPORATION

By: 

PAUL WHENMAN

Registered Patent Attorney

S066280 280596

NEW/PCW/waw/e28

AUSTRALIA  
Patents Act 1990

# NOTICE OF ENTITLEMENT

I (We) Masaaki Kashiwara  
authorised by CHIYODA CORPORATION  
of 12-1, Tsurumichuo, 2-chome, Tsurumi-ku, Yokohama-shi, Kanagawa-ken,  
Japan

the applicant in respect of an application for a patent for an invention entitled  
Process for the desulfurization of sulfurous acid gas-  
containing waste gas  
filed under Australian Application No. \_\_\_\_\_, state the following:

**PART 1 - Must be completed for all applications.**

The person(s) nominated for the grant of the patent

☐ is (are) the actual inventor(s)

~~or~~

☒ has, for the following reasons, gained entitlement from the actual inventor(s)

The nominated person is a person who would if a patent were  
granted upon an application made by the actual inventor(s)  
be entitled to have the patent assigned to it

**PART 2 - Must be completed if the application is a Convention application.**

The person(s) nominated for the grant of the patent is (are):

☒ the applicant(s) of the basic application(s) listed on the patent request form

~~or~~

☐ entitled to rely on the basic application(s) listed on the patent request form by reason of the following:

The basic application(s) listed on the request form is (are) the first application(s) made in a Convention  
country in respect of the invention.

**PART 3 - Must be completed if the application was made under the PCT and claims priority.**

The person(s) nominated for the grant of the patent is (are):

☐ the applicant(s) of the application(s) listed in the declaration under Article 8 of the PCT

or

☐ entitled to rely on the application(s) listed in the declaration under Article 8 of the PCT by reason  
of the following:

The basic application(s) listed in the declaration made under Article 8 of the PCT is (are) the first  
application(s) made in a Convention country in respect of the invention.

Dated this 22 day of May 19 96

Signed Masaaki Kashiwara Status President

Signatory's Name Masaaki Kashiwara



AU9654534

(12) PATENT ABRIDGMENT (11) Document No. AU-B-54534/96  
(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 694352

- (54) Title  
PROCESS FOR THE DESULFURIZATION OF SULFUROUS ACID GAS-CONTAINING WASTE GAS
- International Patent Classification(s)  
(51)<sup>6</sup> B01D 053/18
- (21) Application No. : 54534/96 (22) Application Date : 28.05.96
- (30) Priority Data
- (31) Number (32) Date (33) Country  
7-155119 30.05.95 JP JAPAN
- (43) Publication Date : 12.12.96
- (44) Publication Date of Accepted Application : 16.07.98
- (71) Applicant(s)  
CHIYODA CORPORATION
- (72) Inventor(s)  
HARUO NISHINO; YOSHIO OGAWA; KAZUSHIGE KAWAMURA
- (74) Attorney or Agent  
F B RICE & CO , 605 Darling Street, BALMAIN NSW 2041
- (56) Prior Art Documents  
US 5120518  
EP 0396375  
US 4368060
- (57) Claim

1. A process for the desulfurization of a sulfurous acid gas-containing waste gas, wherein said waste gas is blown into a pool of a stirred absorbing liquid, contained in a reaction vessel and having a liquid level, through a plurality of gas dispersing pipes each vertically extending downward from a partition plate into said absorbing liquid and each having a side peripheral wall provided with a multiplicity of gas injection holes at a lower portion thereof, so that said waste gas is desulfurized by contact with said absorbing liquid and the resulting desulfurized gas is passed to an upper space defined between said partition plate and said liquid level of said absorbing liquid, characterized

in that said gas injection holes of each of said gas dispersing pipes are substantially horizontally aligned;

in that each adjacent two gas injection holes of each of said gas dispersing pipes are spaced apart from each other such that, when each of said two gas injection holes is regarded as a circle having the same area as the area thereof, the distance P between the centroids of said

(11) AU-B-54534/96  
(10) 694352

-2-

adjacent two gas injection holes satisfies the following condition:

$$1.15 \leq P/D \leq 6$$

wherein D is a diameter of one of said two circles which is smaller than the other;

in that the maximum velocity  $V_{\max}$  of said waste gas passing through each of said gas injection holes is controlled so that the following conditions are satisfied::

$$Y \geq 4.5S$$

$$Y \leq 24S$$

$$0.05 \leq Y \leq 1.0$$

$$0.005 \leq S \leq 0.06$$

wherein Y represents a pressure of said waste gas required

for carrying out the desulfurization and S represents a value obtained by dividing the dynamic pressure of said waste gas injected through said gas injection hole at said maximum velocity  $V_{\max}$  by the density of said absorbing liquid;

in that said gas dispersing pipes are arranged such that the distance  $L_I$  between nearest two dispersing pipes satisfies the following condition:

$$1.5 \leq L_I/S \leq 10.0$$

wherein S is as defined above; and

in that said gas injection holes of each of said gas dispersing pipes are located so that the average distance  $L_{II}$  between the liquid level of said absorbing liquid in the state where no gases are injected thereinto and the center of each of said gas injection holes satisfies the following condition:

$$2 \leq L_{II}/S \leq 20$$

wherein S is as defined above.

# AUSTRALIA

## Patents Act 1990

CHIYODA CORPORATION

ORIGINAL

COMPLETE SPECIFICATION  
STANDARD PATENT

*Invention Title:*

*Process for the desulfurization of sulfurous acid gas-containing waste  
gas*

The following statement is a full description of this invention including the best method of performing it known to us:-

### Background of the Invention

This invention relates to a process for the desulfurization of a sulfurous acid gas-containing waste gas by contact with an absorbing liquid.

5 A desulfurization process is known wherein a waste gas containing sulfurous acid gas (sulfur dioxide) is blown into a pool of an absorbing liquid contained in a reaction vessel through a plurality of gas dispersing pipes (sparger pipes) each of which vertically extends downward from a partition plate into the absorbing liquid and has a side peripheral wall provided with a multiplicity of gas injection holes at a lower portion thereof, so that the waste gas is desulfurized by contact with the absorbing liquid and wherein the resulting desulfurized gas is passed to an upper space defined between the partition plate and the liquid level of the absorbing liquid and is discharged from the reaction vessel (JP-B-3-70532 and JP-A-3-72913).

10 The known process, however, has problems that a relatively high costs are required for operating the process and for constructing the apparatus therefor and that the process is not able to be performed in a stable manner for a long period of time.

### Summary of the Invention

15 It is, therefore, the prime object of the present invention to provide a process which can desulfurize a sulfurous acid gas-containing waste gas at a low operation cost in a stable manner.

20 In accomplishing the foregoing object, there is provided in accordance with the present invention a process for the desulfurization of a sulfurous acid gas-containing waste gas, wherein said waste gas is blown into a pool of a stirred absorbing liquid, contained in a reaction vessel and having a liquid level, through a plurality of gas dispersing pipes each vertically extending downward from a partition

25

30

35

plate into said absorbing liquid and each having a side peripheral wall provided with a multiplicity of gas injection holes at a lower portion thereof, so that said waste gas is desulfurized by contact with said absorbing liquid and the resulting desulfurized gas is passed to an upper space defined between said partition plate and said liquid level of said absorbing liquid, characterized

in that said gas injection holes of each of said gas dispersing pipes are substantially horizontally aligned;

in that each adjacent two gas injection holes of each of said gas dispersing pipes are spaced apart from each other such that, when each of said two gas injection holes is regarded as a circle having the same area as the area thereof, the distance P between the centroids of said adjacent two gas injection holes satisfies the following condition:

$$1.15 \leq P/D \leq 6$$

wherein D is a diameter of one of said two circles which is smaller than the other;

in that the maximum velocity  $V_{\max}$  of said waste gas passing through each of said gas injection holes is controlled so that the following conditions are satisfied::

$$Y \geq 4.5S$$

$$Y \leq 24S$$

$$0.05 \leq Y \leq 1.0$$

$$0.005 \leq S \leq 0.06$$

wherein Y represents a pressure of said waste gas required for carrying out the desulfurization and S represents a value obtained by dividing the dynamic pressure of said waste gas injected through said gas injection hole at said maximum velocity  $V_{\max}$  by the density of said absorbing liquid;

in that said gas dispersing pipes are arranged such that the minimum distance  $L_I$  between two adjacent dispersing pipes satisfies the following condition:

$$1.5 \leq L_I/S \leq 10.0$$

wherein S is as defined above; and

in that said gas injection holes of each of said gas dispersing pipes are located so that the average distance  $L_{II}$  between the liquid level of said absorbing liquid in the state where no gases are injected thereinto and the center of each of said gas injection holes satisfies the following condition:

$$2 \leq L_{II}/S \leq 20$$

wherein S is as defined above.

#### Brief Description of the Drawings

Other objects, features and advantages of the present invention will become apparent from the detailed description of the preferred embodiments of the invention which follows, when considered in light of the accompanying drawings in which:

Fig. 1 is an elevational, cross-sectional view diagrammatically showing one embodiment of a desulfurization apparatus suitable for carrying out the process of the present invention;

Fig. 2 is a developed, elevational view schematically showing one embodiment of the gas injection hole arrangement of a sparger pipe;

Fig. 3 is a view similar to Fig. 2 showing another embodiment of the gas injection hole arrangement of a sparger pipe;

Fig. 4 is an elevational, cross-sectional view, similar to Fig. 1, diagrammatically showing another embodiment of a desulfurization apparatus suitable for carrying out the process of the present invention; and

Fig. 5 is a graph showing a relationship between the pressure of the waste gas required for desulfurization and the S value.



Detailed Description of the Preferred  
Embodiments of the Invention

Referring first to Fig. 1, designated generally as 1 is a desulfurization apparatus suitable for carrying out the process of the present invention. The apparatus 1 has a housing member 2 within which first and second partition plate members 3 and 14 are disposed for dividing the inside space thereof into a first, lowermost chamber 4, a second, intermediate chamber 5 and a third, uppermost chamber 15. Each of the partition plate members 3 and 14 may be a horizontal plate or a plate inclined gradually or stepwise. An inclined plate is generally used as the partition plate member 14.

The first chamber 4 contains a pool of an absorbing liquid L such that an upper space B is defined above the liquid level LS of the absorbing liquid L.

A gas inlet port 6 is provided in the second chamber 5 for introducing the waste gas to be treated into the second chamber 5. A plurality of gas dispersing pipes (sparger pipes) 7 are secured to the partition plate 3 and vertically extending downward into the first chamber 4 such that the waste gas introduced into the second chamber 5 is blown into the absorbing liquid L through the sparger pipes 7. Each of the sparger pipes 7 has a side peripheral wall provided with a multiplicity of horizontally aligned gas injection holes 8 at a lower portion thereof.

Designated as W is a liquid level of the absorbing liquid L in the state where no gas is fed to the sparger pipes 7. The gas injection holes 8 are located below the liquid level W. Thus, the waste gas fed to the sparger pipes 7 is injected into the absorbing liquid L through the gas injection holes 8 so that a mixed gas-liquid phase layer A is formed on the surface of the absorbing liquid. The sulfurous acid gas contained in the waste gas is absorbed in the absorbing liquid L in this gas-liquid phase layer A. The

waste gas desulfurized by contact with the absorbing liquid L then flows into the upper space B above the liquid level LS. The absorbing liquid L may be an aqueous gypsum slurry containing an absorbent such as a calcium compound, e.g. lime stone or slaked lime.

One or more riser pipes 16 are provided for connecting the first and third chambers 4 and 15. The third chamber 15 has a gas discharge port 9 at an upper portion thereof. Thus, the desulfurized gas in the upper space B flows upward as well as in the horizontal direction. During the flow of the desulfurized gas in the upper space B, a greater part of the mist and solid particles contained therein are separated therefrom by gravity and by collision with the sparger pipes 7. The desulfurized gas from which such liquid and solid particles have been separated is passed through the riser pipes 16 to the third chamber 15. The upward gas flow is thus converted into a horizontal gas flow and is discharged from the third chamber 15 through the outlet port 9.

During the passage of the desulfurized gas through the third chamber 15, the entraining liquid and solid particles are separated and collected on the partition plate 14. A washing liquid, such as a gypsum-containing aqueous slurry, the absorbing liquid from which gypsum is separated, water or sea water, is supplied from a conduit 17 to the third chamber 15 to remove the deposits on the partition plate 14 and is discharged through a discharge conduit 18.

Preferably, the average ascending velocity of the desulfurized gas in the upper space B is 0.5-5 m/s, more preferably 0.7-4 m/s, from the standpoint of economy and efficiency of the separation of mist and solid particles. The average ascending velocity herein is based on the horizontal sectional area of the upper space B excluding the sectional areas of the sparger pipes 7 and the like structures which do not provide passages for the desulfurized

gas. The average horizontal velocity of the desulfurized gas in the upper space B is preferably 8 m/s or less, more preferably 6 m/s or less for reasons of the formation of stable mixed gas-liquid phase layer A. The average horizontal velocity is based on the vertical sectional area of the upper space B at a position adjacent to the riser pipe 16.

The velocity of the desulfurized gas flowing upward through the riser pipes 16 is preferably 6-20 m/s, more preferably 8-15 m/s for reasons of effective separation of the mist and solid particles and economy.

The desulfurized gas introduced into the third chamber 15 impinges against the top wall of thereof and is directed horizontally. Thus, the entraining liquid and solid particles are separated in the third chamber 15 by the impingement and by gravity. The average horizontal velocity of the desulfurized gas in the third chamber is preferably 10 m/s or less, more preferably 8 m/s or less for reasons of effective separation of these particles. The average horizontal velocity is based on the vertical sectional area of the third chamber at a location horizontally spaced apart a distance of 2 m from the outlet port 9.

Each of the sparger pipes 7 may have any desired sectional shape such as a circular shape, a polygonal (triangle, square or hexagonal) shape or a rectangular (trough) shape. The gas injection holes 8 formed on the side peripheral wall of each sparger pipe 7 may have any desired shape such as a circle, triangle, rectangular, hexagonal, slit or star-like shape. If desired, the injection holes 8 may be arranged into two or more arrays, as shown in Fig. 3. It is preferred that the equivalent inside diameter  $D_p$  of the sparger pipes 7 satisfy the following condition:

$2D_H \leq D_p \leq 12D_H$ , more preferably  $3D_H \leq D_p \leq 10D_H$ , where  $D_H$  represents the equivalent diameter of the gas injection hole 8.

Generally, the equivalent inside diameter  $D_p$  is 25-300 mm, preferably 50-300 mm. The equivalent diameter  $D_H$  of the injection hole 8 is generally 3-100 mm, preferably 5-50 mm.

The equivalent diameters  $D_p$  and  $D_H$  are as defined below:

$$D_p = 4S_p/L_p$$

wherein  $S_p$  represents the horizontal sectional area of the inside of the sparger pipe 7 at a location at which the gas injection holes 8 are provided and  $L_p$  represents the inside peripheral length of the gas sparger pipe 7 at the same location as above, and

$$D_H = 4S_H/L_H$$

wherein  $S_H$  represents the area of the gas injection hole 8 and  $L_H$  represents the inside peripheral length of the gas injection hole 8.

The lower open end of each of the sparger pipes 7 may be in any desired shape and may be, for example, horizontal, slanted, notched or waved.

The average axial distance  $L_{ax}$  between the center (centroid of geometry) of the gas injection hole and the lower end of the sparger pipe 7 is preferably so adjusted that almost no waste gas is passed through the lower open end of the sparger pipe or, in other words, that the liquid level of the absorbing liquid  $L$  always exists in the sparger pipe 7. This can be achieved by adjusting the distance  $L_{ax}$  to  $3S$  to  $8S$ , more preferably  $4S$  to  $7S$ , wherein  $S$  represents a value obtained by dividing the dynamic pressure of the waste gas injected through the gas injection hole at the maximum velocity  $V_{max}$  by the density of the absorbing liquid  $L$ .

One preferred sparger pipe 7 is a plastic cylinder having an inside diameter of 25-300 mm and provided with a plurality of equally spaced apart round holes having a diameter of 5-100 mm.

The riser pipe 16 may have any desired sectional shape such as a circular, square or rectangular shape.

It is important that each adjacent two gas injection holes 8 of each of the gas dispersing pipes 7 should be spaced apart from each other such a distance that, when each of the two gas injection holes 8 is regarded as a circle having the same area as the area of the hole 8, the distance P between the centroids (center of gravity of the geometry) of the adjacent two gas injection holes satisfies the following condition:

$1.15 \leq P/D \leq 6$ , preferably  $1.2 \leq P/D \leq 5$ , wherein D is a diameter of one of the two circles which is smaller than the other. Figs. 2 and 3 depict examples of the gas injection hole arrangement.

When the P/D ratio is smaller than 1.15, the desulfurization rate is considerably lowered since the waste gas flows injected through separate injection holes are apt to be combined. Namely, the jet flows from adjacent injection holes interfere with each other so that the mixed gas-liquid phase layer A (froth phase layer) becomes unstable. A P/D ratio of below 1.15 is also disadvantageous in fabrication and maintenance of the sparger pipe 7. On the other hand, too large a P/D ratio in excess of 6 causes lowering of the volume efficiency so that it is disadvantageously necessary to use a large-sized apparatus.

It is also important that the maximum velocity  $V_{max}$  of the waste gas passing through each of the gas injection holes 8 is controlled so that the following conditions (1)-(4) are satisfied:

- (1)  $Y \geq 4.5S$ , preferably  $Y \geq 6.5S$
- (2)  $Y \leq 24S$ , preferably  $Y \leq 22S$
- (3)  $0.05 \leq Y \leq 1.0$
- (4)  $0.005 \leq S \leq 0.06$

wherein Y represents a pressure of the waste gas required for carrying out the desulfurization and S represents a value obtained by dividing the dynamic pressure of the waste gas injected through the gas injection hole at the maximum

velocity  $V_{\max}$  by the density of the absorbing liquid.

The pressure of the waste gas required for the desulfurization (pressure in terms of absorbing liquid column; unit: m) is defined by:

5 
$$Y = T + L_{II}$$

wherein T represents a value obtained by dividing the pressure loss (unit:  $\text{kg/m}^2$ ) of the waste gas passing through the gas injection hole 8 by the density  $\rho_{II}$  (unit:  $\text{kg/m}^3$ ) of the absorbing liquid L and  $L_{II}$  represents an average distance  
10 between the centroid of the gas injection hole 8 and the liquid level W of the absorbing liquid in the state where no gas is injected into the sparger pipe 7. In other words, the pressure Y is a value obtained by dividing the pressure  
15 (unit:  $\text{kg/m}^2$ ) required for the waste gas fed to the sparger pipe 7 to pass through the gas injection hole 8 to the upper space B by the density  $\rho_{II}$  (unit:  $\text{kg/m}^3$ ) of the absorbing liquid L.

Practically, the value T is in the range of between 2.5S and 4S (where S is as defined above) and depends upon  
20 the shape of the gas injection hole 8 and flow rate of the waste gas. Since, in the present invention,  $L_{II}/S$  is between 2 and 20, preferably between 4-18, as described hereinafter, the pressure Y is expressed as follows:

$$\begin{aligned} Y &= T + L_{II} \\ &= (2.5 \text{ to } 4)S + (2 \text{ to } 20)S \\ &= (4.5 \text{ to } 24)S \end{aligned}$$

The maximum velocity  $V_{\max}$  and the S value have the following relationship:

30 
$$\begin{aligned} S &= (\text{dynamic pressure at the maximum velocity } V_{\max}) / \\ &\quad (\text{density } \rho_{II} \text{ of the absorbing liquid}) \\ &= (\rho_I \times V_{\max} \times V_{\max} / 2G) / \rho_{II} \\ &= V_{\max}^2 \times \rho_I / 2G\rho_{II} \end{aligned}$$

wherein  $\rho_I$  represents the density ( $\text{kg/m}^3$ ) of the waste gas,  $\rho_{II}$  represents the density ( $\text{kg/m}^3$ ) of the absorbing liquid and G  
35 represents the gravitational acceleration ( $9.8 \text{ m/s}^2$ ).

Fig. 5 shows a relationship between the S-value and the pressure Y at various desulfurization rates Z. The term "desulfurization rate Z" used herein is defined as follows:

$$Z = (1 - Q_{out}/Q_{in}) \times 100 (\%)$$

5 wherein  $Q_{out}$  represents the flow rate of sulfurous acid gas contained in the purified gas discharged through the outlet port 9 and  $Q_{in}$  represents the flow rate of sulfurous acid gas contained in the waste gas introduced through the inlet port 6. As seen in Fig. 5, there exists a minimum value in the pressure Y in a given desulfurization rate Z. It is preferred that the S value be selected so that Y is minimum at the intended desulfurization rate. For example, when a desulfurization rate Z of 90 % is intended, the S value of about 0.017 m is preferably used. When a desulfurization rate Z of 70 % is intended, the S value is preferably about 0.009 m. When the desulfurization is carried out while alternately shifting the intended desulfurization rate between 99 % and 70 %, the S value is preferably set at 0.035 m which provides the minimum pressure Y at the 99 % desulfurization rate and which satisfies the above conditions (1)-(4) at the 70 % desulfurization rate. Once the S value is determined, the maximum velocity  $V_{max}$  is determined according to the above-described formula:

$$S = V_{max}^2 \times \rho_I / 2Gp_{II}.$$

25 The number of the sparger pipes 7 and the total area of the openings of the gas injection holes 8 in each of the sparger pipes 7 are then determined on the basis of the maximum velocity  $V_{max}$ .

30 The arrangement of the sparger pipes 7 connected to the partition plate 3 is not specifically limited as long as the distance  $L_I$  between two adjacent sparger pipes 7 satisfies the following condition:

$$1.5 \leq L_I/S \leq 10.0, \text{ preferably } 2 \leq L_I/S \leq 8,$$

35 wherein S is as defined above. The distance  $L_I$  is the minimum distance from the outer periphery of one sparger pipe

7 to the outer periphery of that sparger pipe 7 which is located nearest of all. When the  $L_I/S$  ratio is smaller than 1.5, the desulfurization rate is considerably lowered since the jet flows from the two sparger pipes 7 interfere with each other so that the mixed gas-liquid phase layer A becomes unstable. On the other hand, too large a  $L_I/S$  ratio in excess of 10 causes lowering of the volume efficiency so that it is necessary to use a large-sized apparatus.

The distance  $L_I$  is generally 0.05-0.6 m, preferably 0.075-0.45 m, and is so selected as to meet with the requirement for the above  $L_I/S$  ratio. For reasons of an increased amount of the waste gas treated per unit area of the partition plate 3, it is preferred that the distance  $L_I$  be as small as possible. The  $S$  value is determined according to the equation described above. In this connection, the maximum velocity  $V_{max}$  is in the range of 8-35 m/s, the density  $\rho_I$  of the waste gas is 0.91-1.2 kg/m<sup>3</sup>, and the density  $\rho_{II}$  of the absorbing liquid is 1,000-1,300 kg/m<sup>3</sup>. For reasons of the reduction of operation costs (desulfurization costs) of the desulfurization apparatus, the  $S$  value is desired to be as small as possible, although, from the standpoint of construction costs, too small an  $S$  value is undesirable. By reducing the velocity of the waste gas passing through the gas injection holes, namely by increasing the equivalent diameter  $D_H$  of the gas injection holes or by increasing the number of the gas injection holes, the  $S$  value can be made small. As described previously, the equivalent diameter  $D_H$  of the injection hole 8 is generally 3-100 mm.

It is also important that the gas injection holes 8 of each of the sparger pipes 7 are located so that the average distance  $L_{II}$  between the liquid level  $W$  of the absorbing liquid  $L$  in the state where no gases are injected into the sparger pipe 7 and the center of each of the gas injection holes 8 satisfies the following condition:

$$2 \leq L_{II}/S \leq 20,$$



preferably  $4 \leq L_{II}/S \leq 18$ , more preferably  $6 \leq L_{II}/S \leq 16$ , wherein S is as defined above.

When the  $L_{II}/S$  ratio is less than 2, the waste gas fails to be sufficiently contacted with the absorbing liquid L so that the desulfurization efficiency is lowered. When the  $L_{II}/S$  ratio exceeds 20, the bubbles of the waste gas are combined and grow in size during the passage through the absorbing liquid L so that the efficiency in the liquid-gas contact is lowered. The depth  $L_{II}$  is generally 0.05-0.9 m, preferably 0.075-0.75 m.

When the S value is large or when the depth  $L_{II}$  is large, the pressure Y of the waste gas is high and the desulfurization rate increases. However, the operation costs which depend upon the pressure Y increase as the pressure Y increases. When the  $L_{II}/S$  ratio is maintained in the above-described range, it is possible to maintain the pressure Y of the waste gas supplied to the sparger pipes in a low level. Thus, it is possible to save the energy required for the desulfurization and to reduce the desulfurization costs.

By adjusting the depth  $L_{II}$  so as to meet with the above-described conditions  $4.5S \leq Y \leq 24S$  (Fig. 5) and  $2 \leq L_{II}/S \leq 20$ , the pressure Y required for the desulfurization can be made small at any intended desulfurization rate Z. The curves shown in Fig. 5 are examples in which only the intended desulfurization rate Z is varied while maintaining other parameters, such as the inside diameter of the sparger pipes, the flow rate of the waste gas per one sparger pipe, the pH of the absorbing liquid and the concentration of the sulfurous acid gas in the waste gas, constant. The shape and the position of each curve vary with these parameters.

As described previously, the S value should be  $0.005 \leq S \leq 0.06$ . The suitable S value, however, varies depending upon the intended desulfurization rate Z, as seen in Fig. 5. When the desulfurization apparatus is operated under various operation conditions, it is advisable to set

the S value at a high value so that the desulfurization can be performed with a low energy consumption.

5 The  $L_{II}/S$  ratio is an important parameter for controlling the performance of the desulfurization apparatus and provides an effective means for performing the desulfurization at an intended desulfurization rate Z with minimum operation costs. The depth  $L_{II}$  can be changed by changing the liquid level W. By controlling the amount of the absorbing liquid L in the reactor or by controlling the amount of the oxidizing gas, such as air, fed through a line 10 to the pool of the absorbing liquid L, the liquid level W may be changed to provide a suitable depth  $L_{II}$ .

15 In order to efficiently perform the desulfurization, it is necessary to stir the absorbing liquid L by one or more stirrers 10. The stirrer 10 may be composed of a rotating shaft 10' extending vertically or obliquely into the chamber 4 and one or more blades or propellers secured to the tip of the rotating shaft 10' for rotation therewith. In this case, it is preferred that the stirring of the absorbing liquid is carried out with one or more stirrers operated at a total driving power of 0.05-0.2 kW, more preferably 0.08-0.15 kW, per 1 m<sup>3</sup> of the absorbing liquid, for reasons of obtaining particularly stable desulfurization rate.

25 Preferably, the stirring is performed so that there is formed a main recirculating flow (shown by the arrow R in Fig. 1) in the stirred absorbing liquid L. The main flow is accompanied with randomly occurring flows. In Fig. 1, the reference numeral 11 designates an absorbing agent feeding conduit having an injection nozzle from which the absorbing agent is injected into the main recirculating flow R. The absorbing agent is rapidly diffused into the absorbing liquid L and is swiftly supplied to the gas-liquid mixed phase layer A. If desired, the absorbing agent may be fed through a plurality of conduits 11. The absorbing agent can be

30

35

supplied in the main recirculating flow R at a position upstream or downstream of the blade of the stirrer 10.

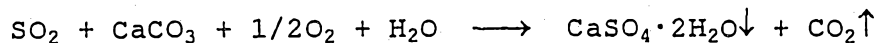
5 The absorbing agent injection nozzle generally has a diameter of 20-100 mm, preferably 25-75 mm. Preferably, a plurality of the nozzles are used for rapidly uniformly dispersing the absorbing agent into the absorbing liquid L and for preventing local increase of pH and deposition of scales on the walls of the sparger pipes. One nozzle is preferably used per 20-500 m<sup>3</sup>, more preferably 30-300 m<sup>3</sup>, of the absorbing liquid L. The absorbing agent is injected in an amount of 0.5-20 kg mol/hour, preferably 1-10 kg mol/hour, per one nozzle.

15 A part of the absorbing liquid L is discharged through a line 13 from the chamber 4 to maintain the content of gypsum in the absorbing liquid L below a predetermined level. If desired, part of the discharged liquid may be treated for the removal of the gypsum, mixed with the absorbing agent and recycled to the first chamber 4 through the line 11. The amount of the absorbing agent incorporated into the recycling absorbing liquid is preferably such that the molar ratio MG/MA of the gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O) contained therein to the absorbing agent is in the range of 0.1-20, more preferably 1-10, for reasons of preventing a local increase of pH in the region adjacent to the gas injection holes 8. Namely, the precipitation of fine gypsum crystals or fine CaCO<sub>3</sub> crystals is suppressed. Further, even when such fine crystals are formed, they grow into large crystal particles so that the clogging of the gas injection holes 8 or scaling of the wall of the sparger pipes 7 can be prevented.

30 If desired, a part of the absorbing liquid L may be recycled to and sprayed in the chamber 5 to cool and wash the waste gas introduced thereinto.

35 The above-described oxidized gas supplied through the line 12 is preferably injected into the main

recirculating flow R at a position downstream of the blade of the stirrer 10. In the gas-liquid mixed phase layer A, the following reaction occurs to fix the sulfurous acid gas contained in the waste gas as gypsum:



To improve the desulfurization rate Z, it is necessary that the above reaction efficiently proceeds in the gas-liquid mixed phase layer A. It is preferred that the oxidizing gas be introduced into the absorbing liquid L in such an amount that the molar ratio of the oxygen in the oxidizing gas to the sulfurous acid gas in the waste gas is 0.5-6, more preferably 1-5.

Fig. 4 depicts another embodiment of the desulfurization apparatus in which similar component parts have been designated by the same reference numerals. In this embodiment, the inside space of a housing member 2 is divided by one partition plate member 3 into a first, lower chamber 4 and a second, upper chamber 5. The first chamber 4 contains a pool of an absorbing liquid L such that an upper space B is defined above the liquid level LS of the absorbing liquid L. A waste gas to be treated is introduced through a gas inlet port 6 provided in the second chamber 5 and injected into a pool of the absorbing liquid L through a plurality of sparger pipes 7 secured to the partition plate 3 and a multiplicity of horizontally aligned gas injection holes 8 formed at a lower portion of each of the sparger pipes 7.

The waste gas desulfurized by contact with the absorbing liquid L then flows into the upper space B above the liquid level LS. Preferably, the average ascending velocity of the desulfurized gas in the upper space B is 0.5-5 m/s, more preferably 0.7-4 m/s, while the average horizontal velocity of the desulfurized gas in the upper space B is preferably 8 m/s or less, more preferably 6 m/s or less. During the flow of the desulfurized gas in the upper space B, a greater part of the mist and solid particles

contained therein are separated therefrom by gravity and by collision with the sparger pipes 7. The desulfurized gas from which such liquid and solid particles have been separated is discharged from the outlet port 9.

5           The following example will further illustrate the present invention.

#### Example

10           A waste gas containing 1,000 ppm of sulfurous acid gas was treated in accordance with the process of the present invention under the following conditions:

Reactor: 13 m x 13 m x 10 m (height)

Maximum flow rate of waste gas: 1,000,000 m<sup>3</sup>/hour

15           Range of flow rate of waste gas: 50-100 % (evenly operated)

Desulfurization rate Z: 90%

Density of waste gas  $\rho_I$ : 1.1 kg/m<sup>3</sup>

Density of absorbing liquid  $\rho_{II}$ : 1,100 kg/m<sup>3</sup>

Sparger pipe (cross-section: circular)

20           Diameter  $D_S$ : 0.15 m

Distance between adjacent sparger pipes  $L_I$ : 0.15 m

Number: 1,390

Gas injection hole (circular)

25           Diameter  $D_H$ : 0.03 m

Number: 12

Distance between adjacent holes P: 0.0393 m

Average distance  $L_{II}$ : 0.2 m

Maximum velocity  $V_{max}$ : 24.2 m/s

S value: 0.03

30           Pressure Y: about 0.28 m

The desulfurization treatment was found to be carried out with minimum costs including installation and construction costs and operation costs.

35           The invention may be embodied in other specific forms without departing from the spirit or essential

characteristics thereof. The present embodiments are  
therefore to be considered in all respects as illustrative  
and not restrictive, the scope of the invention being  
indicated by the appended claims rather than by the foregoing  
description, and all the changes which come within the  
meaning and range of equivalency of the claims are therefore  
intended to be embraced therein.

5

10

15

20

25

30

35

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:-

1. A process for the desulfurization of a sulfurous acid gas-containing waste gas, wherein said waste gas is blown into a pool of a stirred absorbing liquid, contained in a reaction vessel and having a liquid level, through a plurality of gas dispersing pipes each vertically extending downward from a partition plate into said absorbing liquid and each having a side peripheral wall provided with a multiplicity of gas injection holes at a lower portion thereof, so that said waste gas is desulfurized by contact with said absorbing liquid and the resulting desulfurized gas is passed to an upper space defined between said partition plate and said liquid level of said absorbing liquid, characterized

in that said gas injection holes of each of said gas dispersing pipes are substantially horizontally aligned;

in that each adjacent two gas injection holes of each of said gas dispersing pipes are spaced apart from each other such that, when each of said two gas injection holes is regarded as a circle having the same area as the area thereof, the distance P between the centroids of said adjacent two gas injection holes satisfies the following condition:

$$1.15 \leq P/D \leq 6$$

wherein D is a diameter of one of said two circles which is smaller than the other;

in that the maximum velocity  $V_{\max}$  of said waste gas passing through each of said gas injection holes is controlled so that the following conditions are satisfied::

$$Y \geq 4.5S$$

$$Y \leq 24S$$

$$0.05 \leq Y \leq 1.0$$

$$0.005 \leq S \leq 0.06$$

wherein Y represents a pressure of said waste gas required

for carrying out the desulfurization and S represents a value obtained by dividing the dynamic pressure of said waste gas injected through said gas injection hole at said maximum velocity  $V_{\max}$  by the density of said absorbing liquid;

5 in that said gas dispersing pipes are arranged such that the distance  $L_I$  between nearest two dispersing pipes satisfies the following condition:

$$1.5 \leq L_I/S \leq 10.0$$

wherein S is as defined above; and

10 in that said gas injection holes of each of said gas dispersing pipes are located so that the average distance  $L_{II}$  between the liquid level of said absorbing liquid in the state where no gases are injected thereinto and the center of each of said gas injection holes satisfies the following condition:

$$2 \leq L_{II}/S \leq 20$$

wherein S is as defined above.

20 2. A process as claimed in claim 1, wherein said range S satisfies the following conditions:

$$Y \geq 6.5S$$

$$Y \leq 22S$$

$$0.05 \leq Y \leq 1.0$$

$$0.005 \leq S \leq 0.06$$

25 wherein Y and S are as defined above.

30 3. A process as claimed in claim 1, wherein said minimum distance  $L_I$  is in the range of 0.05-0.6 m and said average distance  $L_{II}$  is in the range of 0.05-0.9 m.

4. A process as claimed in claim 1, wherein the amount of said absorbing liquid in said reaction vessel is controlled so that the ratio  $L_{II}/S$  is in the range of 2 to 20.

35 5. A process as claimed in claim 1, wherein air is



blown into said absorbing liquid in an amount so that the ratio  $L_{II}/S$  is in the range of 2 to 20.

5 6. A process as claimed in claim 1, wherein said stirring of said absorbing liquid is carried out with one or more stirrers operated at a total driving power of 0.05-0.2 kW per 1 m<sup>3</sup> of said absorbing liquid.

10 7. A process as claimed in claim 1, wherein the average ascending velocity of said desulfurized waste gas in said upper space is 0.5-5 m/s and the average horizontal velocity of said desulfurized waste gas in said upper space is 8 m/s or less.

15 8. A process as claimed in claim 1, wherein each of said gas dispersing pipes has an equivalent diameter of 25-300 mm and each of said gas injection holes has an equivalent diameter of 3-100 mm.

20 9. A process as claimed in claim 1, wherein said desulfurized waste gas in said upper space is introduced through at least one riser pipe into a chamber defined in an upper portion of said reaction vessel and is discharged from said reaction vessel through a discharge port provided in said chamber.

25 10. A process as claimed in claim 9, wherein the average horizontal velocity of said desulfurized waste in said chamber is 10 m/s or less.

30 DATED THIS 28 DAY OF MAY 1996

CHIYODA CORPORATION

Patent Attorneys for the

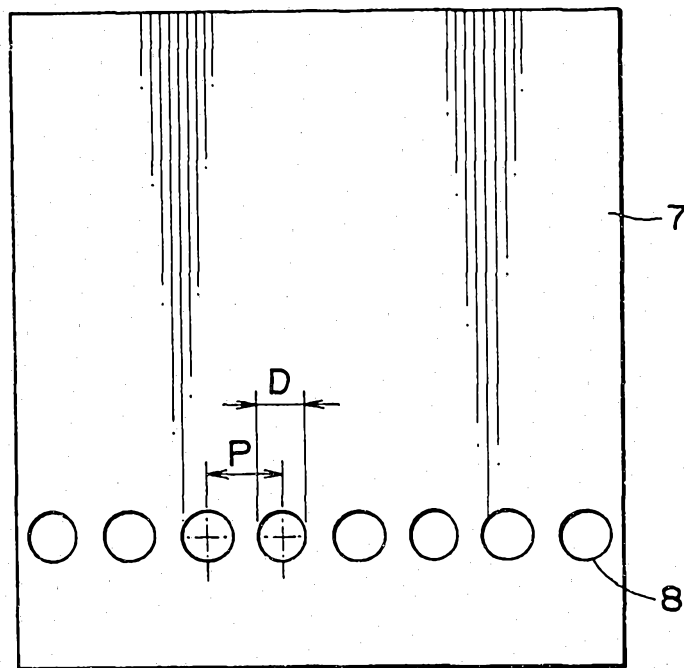
Applicant:-

F.B.RICE & CO.



2/5

FIG. 2



3/5

FIG. 3

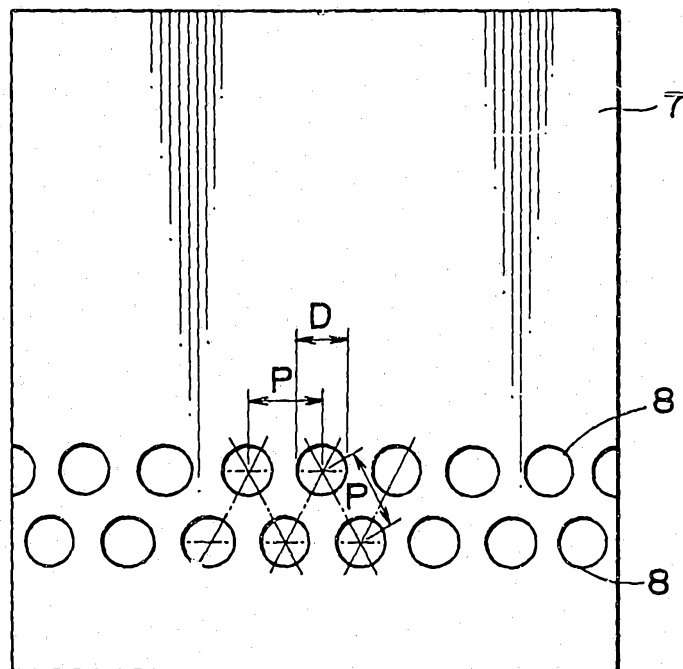


FIG. 4

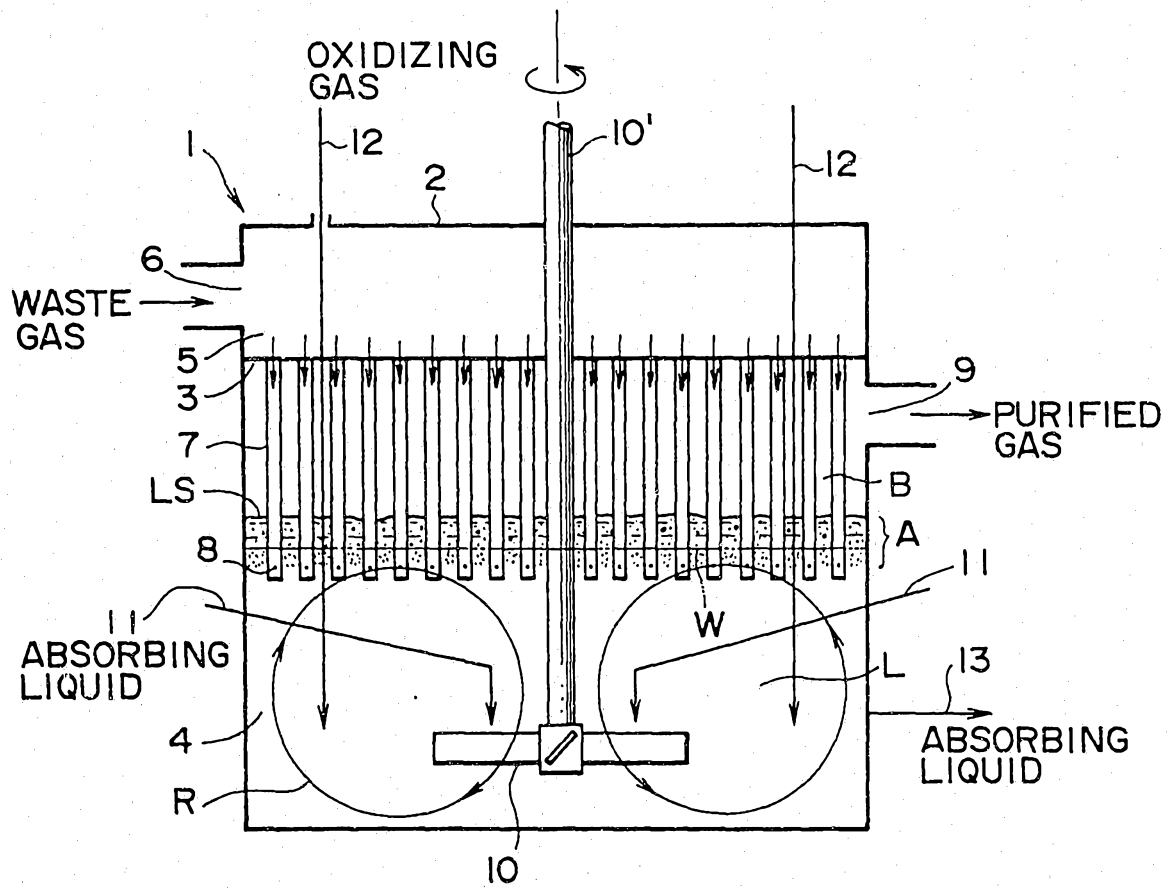


FIG. 5

