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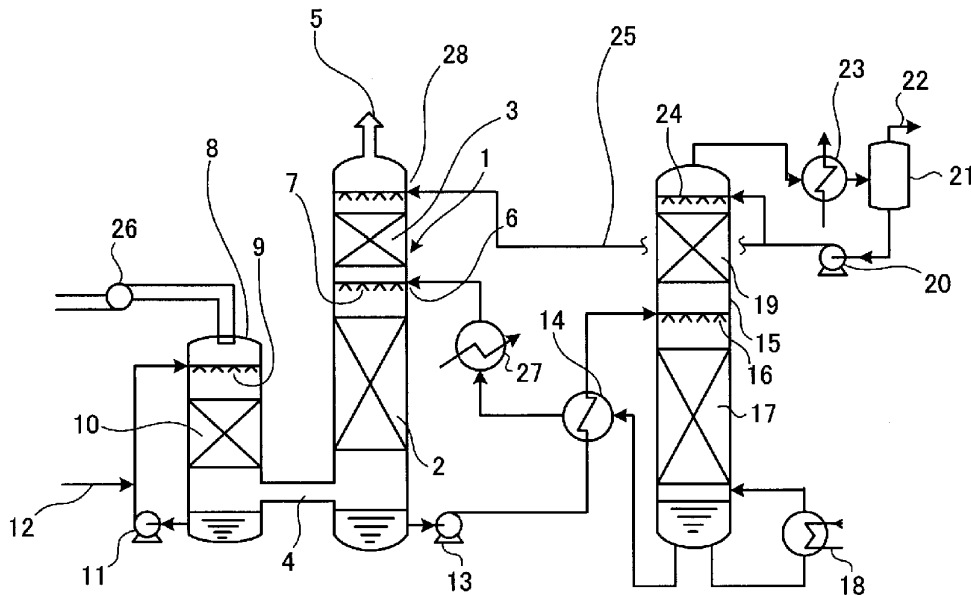
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[続葉有]

(54) Title: ABSORBING SOLUTION, AND METHOD AND APPARATUS FOR THE REMOVAL OF CO<sub>2</sub> OR H<sub>2</sub>S OR BOTH

(54) 発明の名称: 吸収液、CO<sub>2</sub>又はH<sub>2</sub>S又はその双方の除去方法及び装置



(57) Abstract: An absorbing solution capable of absorbing CO<sub>2</sub> or H<sub>2</sub>S or both contained in a gas, which is obtained by adding a tertiary monoamine preferably in an amount of 1 to 20 wt% to a secondary amine composite absorbent such as a mixture of a secondary monoamine with a secondary diamine. The tertiary monoamine inhibits the secondary amines contained in the solution from deterioration by oxygen contained in a gas or the like to bring about reduction in absorption loss, less lowering in performance and cost reduction. The absorbing solution is suitably usable in apparatus for the removal of CO<sub>2</sub> or H<sub>2</sub>S or both.

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(57) 要約: 本発明にかかる吸収液は、ガス中のCO<sub>2</sub>又はH<sub>2</sub>S又はその双方を吸収する吸収液であって、例えば2級モノアミンと2級ジアミンとの混合物等の2級アミン系複合吸収剤に対して、3級モノアミンを好ましくは1~20重量%添加してなるものであり、これにより、ガス中の酸素等による吸収液アミンの劣化を抑制することができ、この結果、吸収損失の低減、性能の低下及びコスト低減を図ることができる。この吸収液はCO<sub>2</sub>又はH<sub>2</sub>S又はその双方の除去装置に用いて好適である。

## DESCRIPTION

ABSORBING SOLUTION, METHOD AND APPARATUS FOR ABSORBING  
CO<sub>2</sub> OR H<sub>2</sub>S OR BOTH

5

## TECHNICAL FIELD

[0001] The present invention relates to an absorbing solution that removes at least one of carbon dioxide (CO<sub>2</sub>) and hydrogen sulfide (H<sub>2</sub>S) or both of CO<sub>2</sub> and H<sub>2</sub>S contained  
10 in gas, and an apparatus and a method that removes at least one of CO<sub>2</sub> and H<sub>2</sub>S or both of CO<sub>2</sub> and H<sub>2</sub>S by using the absorbing solution.

## BACKGROUND OF THE INVENTION

15 [0002] Various methods have been proposed for collecting and removing acidic gases such as CO<sub>2</sub> that are often contained in gases (treatment object gases), for example in industrial gases manufactured in chemical plants, such as a natural gas and synthetic gas and flue gases.

20 In the case of flue gases, a method of bringing a flue gas comprising CO<sub>2</sub> into contact with an alkanolamine solution or the like to remove and collect CO<sub>2</sub>, and a method of storing CO<sub>2</sub> without emitting CO<sub>2</sub> to the atmosphere have been actively studied.

25 [0003] As the alkanolamine, it is possible to use monoethanolamine (MEA), 2-methylaminoethanol, 2-ethylaminoethanol, 2-propylaminoethanol, n-butylaminoethanol, 2-(isopropylamino)ethanol, and 3-ethylaminoethanol.

30 [0004] For example, an absorbing-solution formed of a mixture of secondary amine or a mixture of secondary amine and tertiary amine is disclosed in a United States Patent specification. It is proposed that this mixed absorbing-solution is an advantageous absorbing-solution because an

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absorbing ability and regeneration energy are substantially improved in the mixed absorbing solution compared with an MEA single absorbing solution (Patent Document 1).

[0005] When a monoethanolamine (MEA) absorbing solution  
5 is used, there is a problem in that degradation in the absorbing-solution severely progresses because of oxygen or the like in gas.

A method has therefore also been proposed for stabilizing an absorbing-solution by adding, for example,  
10 trialkanolamine or methyldiethanolamine (MDEA) to the absorbing-solution (Patent Document 2 and Patent Document 3).

[0006] Patent Document 1: United States Patent No.  
5,618,506 specification

15 Patent Document 2: United States Patent No.  
3,535,260 specification

Patent Document 3: United States Patent No.  
4,840,777 specification

20 [0007] However, these patent documents are primarily concerned with the absorbing ability and the like of the absorbing-solution formed from the mixture of secondary amine. In particular, the documents do not disclose a method of preventing degradation in the absorbing-solution  
25 due to oxygen or the like present in the gas. Furthermore, control of degradation in the absorbing-solution still remains a problem to solve.

[0008] Taking the problem into consideration, the present invention seeks to provide an absorbing-solution which can  
30 minimize or prevent self-degradation due to oxygen or the like that is present in gas, and a method and an apparatus for removing any one of CO<sub>2</sub> and H<sub>2</sub>S or both.

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## SUMMARY OF THE INVENTION

[0009] The present invention provides an absorbing-solution that absorbs at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the absorbing-solution

5 comprising:

secondary-amine composite absorbent; and

tertiary monoamine that is added to the secondary-amine composite absorbent;

wherein the secondary-amine composite absorbent is a mixture  
10 of secondary monoamine and secondary diamine, and wherein 1 to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

[0011] In one embodiment, the secondary monoamine is a compound that is selected from at least one kind of 2-

15 methylaminoethanol, 2-ethylaminoethanol, 2-n-propylaminoethanol, 2-n-butylaminoethanol, 2-n-pentylaminoethanol, 2-isopropylaminoethanol, 2-sec-butylaminoethanol, and 2-isobutylaminoethanol, and the secondary diamine is a compound that is selected from at  
20 least one kind of piperazine, 2-methylpiperazine, 2,3-dimethylpiperazine, 2,5-dimethylpiperazine, N,N'-dimethylethanediamine, N,N'-dimethylpropanediamine, N,N'-diethylethylenediamine, N,N'-diethylpropanediamine, N,N'-diisopropylethylenediamine, and N,N'-ditertiary-  
25 butylethanediamine.

[0013] In a further embodiment, 2.5 to 100 weight percent of the secondary diamine is added to the secondary monoamine.

[0014] The present invention also provides a removing  
30 apparatus that removes at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the removing apparatus comprising:

an absorber that receives the gas, causes the gas to come in contact with an absorbing-solution whereby at least

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one of CO<sub>2</sub> and H<sub>2</sub>S or both is absorbed by the absorbing-solution; and

a regenerator that regenerates the absorbing-solution that absorbed at least one of CO<sub>2</sub> and H<sub>2</sub>S or both by removing  
5 at least one of CO<sub>2</sub> and H<sub>2</sub>S or both from the absorbing-solution, and supplies the absorbing-solution that is removed of at least one of CO<sub>2</sub> and H<sub>2</sub>S or both to the absorber, wherein the absorbing-solution includes secondary-amine composite absorbent, and tertiary monoamine that is  
10 added to the secondary-amine composite absorbent;  
wherein the secondary-amine composite absorbent is a mixture of secondary monoamine and secondary diamine, and wherein 1 to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

15 [0015] The present invention further provides a method for a removing apparatus to remove at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the method comprising:

causing the gas to come in contact with an absorbing-solution whereby at least one of CO<sub>2</sub> and H<sub>2</sub>S or both is  
20 absorbed by the absorbing-solution; and

regenerating the absorbing-solution that absorbed at least one of CO<sub>2</sub> and H<sub>2</sub>S or both by removing any one of CO<sub>2</sub> and H<sub>2</sub>S or both from the absorbing-solution, wherein the causing includes causing the gas to come in contact with an  
25 absorbing-solution that is regenerated at the regenerating step, wherein the absorbing-solution includes secondary-amine composite absorbent, and tertiary monoamine that is added to the secondary-amine composite absorbent;  
wherein the secondary-amine composite absorbent is a mixture  
30 of secondary monoamine and secondary diamine, and wherein 1 to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

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[0016] According to an embodiment of the present invention, it is possible to reduce a loss of an absorbing-solution due to degradation in amine used as the absorbing-solution, to minimize or prevent deterioration of performance, and to reduce a cost.

#### BRIEF DESCRIPTION OF DRAWINGS

[0017] The present invention is also illustrated by way of example only with reference to the accompanying drawing in which Fig. 1 is a diagram for explaining an example of a process that can be adopted in the present invention.

#### EXPLANATIONS OF LETTERS OR NUMERALS

[0018] In Figure 1:  
1 Decarbonator  
15 Absorbing-solution/regenerator

#### DETAILED DESCRIPTION OF THE INVENTION

[0019] Exemplary embodiment of the present invention is explained in detail below with reference to the drawings. The present invention is not limited by the embodiment and an example. Components in the embodiment and the example include components that those skilled in the art can easily anticipate, or include components that are substantially identical with the components that those skilled in the art can easily anticipate.

[0020] An absorbing solution according to an embodiment of the present invention is an absorbing solution that absorbs at least one of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  or both of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  in gas. The absorbing solution is formed by adding tertiary monoamine to a secondary-amine composite absorbent. Consequently, it is possible to control degradation in amine in the absorbing solution due to oxygen or the like in gas.

[0021] The secondary-amine composite absorbent is a

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mixture of secondary monoamine and secondary diamine.

[0022] The secondary monoamine may be an amine compound represented by Formula (1) below.



5 In the formula,  $R^1$  represents a lower alkyl group with a hydrogen or carbon number 1 to 4 and  $R^2$  represents a hydrogen or methyl group.

[0023] Specifically, examples of the secondary monoamine include a compound selected from at least one kind of 2-  
10 methylaminoethanol, 2-ethylaminoethanol, 2-n-propylaminoethanol, 2-n-butylaminoethanol, 2-n-pentylaminoethanol, 2-isopropylaminoethanol, 2-sec-butylaminoethanol, and 2-isobutylaminoethanol. However, the present invention is not limited to this.

15 [0024] Examples of the secondary diamine include a compound selected from at least one kind of piperazine, 2-methylpiperazine, 2,3-dimethylpiperazine, 2,5-dimethylpiperazine, N,N'-dimethylethanediamine, N,N'-dimethylpropanediamine, N,N'-diethylethylenediamine, N,N'-  
20 diethylpropanediamine, N,N'-diisopropylethylenediamine, and N,N'-ditertiary-butylethanediamine. However, the present invention is not limited to this.

[0025] The tertiary monoamine may be an amine compound indicated by Formula (2) below.



$R^3$  is a lower alkyl group with a carbon number 1 to 4,  $R^4$  is a lower alkyl group or hydroxyethyl group with a carbon number 1 to 4, and  $R^5$  is a lower alkyl group with a carbon number 2 to 4.

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As the tertiary monoamine indicated by Formula (2), it is desirable to use, for example, N-methyldiethanolamine (MDEA), N-ethyldiethanolamine, N-butyldiethanolamine, 2-dimethylaminoethanol, 2-diethylaminoethanol, 2-di-n-butylaminoethanol, N-ethyl-N-methylethanolamine, 3-dimethylamino-1-propanol, 2-dimethylamino-2-methyl-1-propanol, or 4-dimethylamino-1-butanol. However, the present invention is not limited to this.

[0026] The percentage of addition of tertiary monoamine to the secondary-amine composite absorbent is 1 to 20 weight percent.

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[0027] This is because, as indicated by Table 1 below, when the percentage exceeds 20 weight percent, a fall in an absorption capacity undesirably increases.

On the other hand, when the percentage is lower than 1  
5 weight percent, undesirably, it is impossible to eliminate an influence of contaminant.

[0028] Table 1

Table 1 (50°C, 10 mol% dry CO<sub>2</sub> condition)

Tertiary amine concentration with respect to a mixture of secondary monoamine and secondary diamine (weight %)	Absorption capacity ratio
0	1 (reference)
10	0.99
20	0.96
30	0.93

[0029] A percentage of addition of secondary diamine to  
10 secondary monoamine is not specifically limited. However, it is desirable to add 2.5 to 100 weight % of secondary diamine.

[0030] A process that can be adopted in the method of  
removing CO<sub>2</sub> or H<sub>2</sub>S in a flue gas or both of CO<sub>2</sub> and H<sub>2</sub>S  
15 according to the present invention is not specifically limited. An example of the process is explained with reference to Fig. 1.

In Fig. 1, only main equipment is shown and additional  
equipment is not shown. In Fig. 1, reference numeral 1  
20 denotes a decarbonator; 2, a lower filling unit; 3, an upper filling unit or a tray; 4, a decarbonator flue-gas supply port; 5, a decarbonator flue-gas exhaust port; 6, an absorbing-solution supply port; 7, nozzles; 8, a flue gas cooler provided when needed; 9, nozzles; 10, a filling  
25 unit; 11, a humidifying-coolant circulation pump; 12, a

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makeup-water supply line; 13, an absorbing-solution discharge pump for an absorbing-solution in which CO<sub>2</sub> is absorbed; 14, a heat exchanger; 15, an absorbing-solution regenerator; 16, nozzles; 17, a lower filling unit; 18, a reboiler; 19, an upper filling unit; 20, a reflux water pump; 21, a CO<sub>2</sub> separator; 22, a collected CO<sub>2</sub> exhaust line; 23, a regenerator reflex cooler; 24, nozzles; 25, a regenerator reflux-water supply line; 26, a flue-gas supply blower; 27, a cooler, and 28, a regenerator reflux-water supply port.

[0031] In Fig. 1, a flue gas is squeezed into the flue gas cooler 8 by the flue-gas supply blower [translator's comment: reference numeral 26 should be affixed]. The flue gas comes into contact with a humidifying coolant from the nozzles 9 in the filling unit 10 and is humidified and cooled and led to the decarbonator 1 through the decarbonator flue-gas supply port 4. The humidifying coolant coming into contact with the flue gas accumulates in a lower part of the flue gas cooler 8 and is circulated to the nozzles 9 by the pump 11 and used. Since the humidifying coolant is gradually lost by humidifying and cooling the flue gas, the humidifying coolant is filled by the makeup-water supply line 12.

[0032] The flue gas squeezed into the decarbonator 1 is brought into counter-contact with an absorbing-solution of a fixed concentration supplied from the nozzles 7 in the lower filling unit 2. CO<sub>2</sub> in the decarbonated flue gas is absorbed and removed by the absorbing-solution and the decarbonated flue gas flows to the upper filling unit 3. The absorbing-solution supplied to the decarbonator 1 absorbs CO<sub>2</sub>. Temperature of the absorbing-solution usually rises to be higher than temperature in the absorbing-solution supply port 6 because of reaction heat due to the absorption. The absorbing-solution is sent to the heat exchanger 14 by the

absorbing-solution discharge pump 13 for the absorbing-solution in which CO<sub>2</sub> is absorbed. The absorbing-solution is heated and led to the absorbing-solution regenerator 5. It is possible to perform temperature adjustment for the  
5 absorbing-solution regenerated using the heat exchanger 14 or the cooler 27 provided between the heat exchanger 14 and the absorbing-solution supply port 6 as required.

[0033] In the absorbing-solution regenerator 15, the absorbing-solution is regenerated in the lower filling unit  
10 17 according to heating by the reboiler 18, cooled by the heat exchanger 14, and returned to the decarbonator 1. In an upper part of the absorbing-solution regenerator 15, CO<sub>2</sub> separated from the absorbing-solution comes into contact with a reflux water supplied from the nozzles 24 in the  
15 upper filling unit 19 and cooled by the regenerator reflux cooler 23. Water vapor accompanying CO<sub>2</sub> is separated from the condensed reflux water by the CO<sub>2</sub> separator 21 and led to a CO<sub>2</sub> collection process from the collected CO<sub>2</sub> exhaust line 22. The reflux water is partially refluxed to the  
20 regenerator by the reflux water pump 20 and partially supplied to the regenerator reflux-water supply port 28 of the decarbonator 1 through the regenerator reflux-water supply line 25. Since a small quantity of absorbing-solution is contained in this regenerated reflux water, the  
25 absorbing-solution comes into contact with exhaust gas in the upper filling unit 3 of the decarbonator 1 and contributes to removal of a small quantity of CO<sub>2</sub> contained in the exhaust gas.

### 30 Example

[0034] A non-limiting example according to the present invention is explained.

In the example, temperature was set to 60°C and oxygen concentration in gas was set to 20 mol%.

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As a compounding ratio, concentration of tertiary amine with respect to a mixture of secondary monoamine and a secondary diamine was set to 2 weight percent.

In this example, secondary monoamine was used and a piperazine compound was used as secondary diamine to form a secondary-amine composite absorbent. 2 weight percent of methyldiethanolamine (MDEA) was added to the secondary-amine composite absorbent as tertiary monoamine. Thereafter, a predetermined quantity of water was added to the secondary-amine composite absorbent to form a CO<sub>2</sub> absorbing-solution. Concentration of a decomposition product (a vapor-like basic compound) in the CO<sub>2</sub> absorbing-solution obtained was 8 ppm.

[0035] On the other hand, concentration of a decomposition product (a vapor-like basic compound) in a CO<sub>2</sub> absorbing-solution formed of a secondary monoamine and a piperazine compound, which was a comparative example in which 2 weight percent of methyldiethanolamine (MDEA) was not added to the secondary-amine composite absorbent as tertiary monoamine, was 15 ppm

[0036] Thus, it was found that, when tertiary monoamine was added to the secondary-amine composite absorbent, it is possible to control degradation due to oxygen in exhaust gas.

[0037] As described above, the absorbing-solution according to the present invention is suitably used in a facility that removes CO<sub>2</sub> or H<sub>2</sub>S in a flue gas or both of CO<sub>2</sub> and H<sub>2</sub>S, in which a reduction in a loss of an absorbing-solution due to degradation in absorbing-solution amine, prevention of malfunction, and a reduction in cost

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can be realized.

The reference in this specification to any prior publication (or information derived from it), or to any matter which is known, is not, and should not be taken as an acknowledgment or admission or any form of suggestion that  
5 that prior publication (or information derived from it) or known matter forms part of the common general knowledge in the field of endeavour to which this specification relates.

Throughout this specification and the claims which  
10 follow, unless the context requires otherwise, the word "comprise", and variations such as "comprises" and "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of  
15 integers or steps.

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**THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:**

1. An absorbing-solution that absorbs at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the absorbing-  
5 solution comprising:

secondary-amine composite absorbent; and

tertiary monoamine that is added to the secondary-amine composite absorbent;

wherein the secondary-amine composite absorbent is a mixture  
10 of secondary monoamine and secondary diamine, and wherein 1 to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

2. The absorbing-solution according to claim 1, wherein  
15 the secondary monoamine is a compound that is selected from at least one of 2-methylaminoethanol, 2-ethylaminoethanol, 2-n-propylaminoethanol, 2-n-butylaminoethanol, 2-n-pentylaminoethanol, 2-isopropylaminoethanol, 2-sec-butylaminoethanol, and 2-  
20 isobutylaminoethanol, and

the secondary diamine is a compound that is selected from at least one of piperazine, 2-methylpiperazine, 2,3-dimethylpiperazine, 2,5-dimethylpiperazine, N,N'-dimethylethanediamine, N,N'-dimethylpropanediamine, N,N'-  
25 diethylethylenediamine, N,N'-diethylpropanediamine, N,N'-diisopropylethylenediamine, and N,N'-ditertiary-butylethanediamine.

3. The absorbing-solution according to claim 1 or 2,  
30 wherein 2.5 to 100 weight percent of the secondary diamine is added to the secondary monoamine.

4. A removing apparatus that removes at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the removing

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apparatus comprising:

an absorber that receives the gas, causes the gas to come in contact with an absorbing-solution whereby at least one of CO<sub>2</sub> and H<sub>2</sub>S or both is absorbed by the absorbing-solution; and

5 a regenerator that regenerates the absorbing-solution that absorbed at least one of CO<sub>2</sub> and H<sub>2</sub>S or both by removing at least one of CO<sub>2</sub> and H<sub>2</sub>S or both from the absorbing-solution, and supplies the absorbing-solution that is removed of at least one of CO<sub>2</sub> and H<sub>2</sub>S or both to the absorber, wherein the absorbing-solution includes secondary-amine composite absorbent, and tertiary monoamine that is added to the secondary-amine composite absorbent; wherein the secondary-amine composite absorbent is a mixture of secondary monoamine and secondary diamine, and wherein 1 to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

5. A method for a removing apparatus to remove at least one of CO<sub>2</sub> and H<sub>2</sub>S or both that are present in a gas, the method comprising:

causing the gas to come in contact with an absorbing-solution whereby at least one of CO<sub>2</sub> and H<sub>2</sub>S or both is absorbed by the absorbing-solution; and

25 regenerating the absorbing-solution that absorbed at least one of CO<sub>2</sub> and H<sub>2</sub>S or both by removing any one of CO<sub>2</sub> and H<sub>2</sub>S or both from the absorbing-solution, wherein the causing includes causing the gas to come in contact with an absorbing-solution that is regenerated at the regenerating step, wherein the absorbing-solution includes secondary-amine composite absorbent, and tertiary monoamine that is added to the secondary-amine composite absorbent; wherein the secondary-amine composite absorbent is a mixture of secondary monoamine and secondary diamine, and wherein 1

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to 20 weight percent of the tertiary monoamine is added to the secondary-amine composite absorbent.

6. An absorbing-solution according to claim 1  
5 substantially as hereinbefore described.

7. A removing apparatus according to claim 4 or a method for a removing apparatus according to claim 5 substantially as hereinbefore described.

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FIG.1

