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Solvent and Method for Removal of an Acid Gas from a Fluid Stream

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SOLVENT AND METHOD FOR REMOVAL
OF AN ACID GAS FROM A FLUID STREAM

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Abstract:
A solvent for removal of an acid gas from a fluid stream includes a promoter amine with a pKa of between 6.5 and 10.5 and a tertiary amine with a pKa of between 8.5 and 10.5.

15 Claims, 5 Drawing Sheets
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FIG. 1
FIG. 2a

The graph shows the pH over time for different solutions.

- **MDEA (0.5M)**
- **10% SA**
- **MDEA (0.6M)**
- **15% SA**
- **20% SA**
- **10% APN**
- **15% APN**
- **20% APN**

The x-axis represents time in seconds, scaled by 10^8, and the y-axis represents pH from 8 to 11.5.
FIG. 3a

- ○ DMEA (0.2M)
- □ 2.5% SA+15% APN

pH

Time (x 10^3)

0  5  10  15  20  25
FIG. 3b

Bicarbonate (mM)

Time (x 10^6)

DMEA bicarb
SA/APN bicarb
1. SOLVENT AND METHOD FOR REMOVAL OF AN ACID GAS FROM A FLUID STREAM

TECHNICAL FIELD

This document relates to the removal of CO₂ and other acid gases from a fluid stream and more particularly to a solvent of promoter and tertiary amines used for separating an acid gas from that fluid stream.

BACKGROUND

The cleanup of acid gases or sour gas, such as CO₂, is a critical aspect of the petroleum and natural gas industries. CO₂ emissions are of concern due to their contribution to global climate change. Various methods are employed to capture CO₂ from sources such as coal and gas-fired power plants, natural gas processing, and industrial processes.

Primary (RNH₂) and secondary (R₂NH) amines are typically used in the direct chemical reaction with CO₂ to form a carbonate. However, this process can be energy-intensive and may require additional equipment to regenerate the amine solution.

The absorption process seeks to exploit the advantages of both systems without inheriting the limitations exhibited by either. More particularly, requiring 2 moles of amine to capture 1 mole of CO₂ directly; however, they show a lower than desirable capacity. It requires 2 moles of amine to capture 1 mole of CO₂ since the carbonate is then negatively charged another amine must absorb the proton formed. Furthermore, due to the high enthalpy of absorption from the formation of the carbonate the regeneration energy is high. Tertiary amines (R₃N) show a significant decrease in mass transfer rates due to the high volatility and low viscosity. The described process seeks to exploit the advantages of both systems without inheriting the limitations exhibited by either. More specifically, by promoting the CO₂ capture reaction rate the absorber will be smaller and reduce the process capital. The intrinsic lower energy of regeneration for the tertiary amine solvent minimizes operating costs.

While gas sweetening applications represent the most immediate opportunity for application of the described invention, post-combustion CO₂ capture could represent a large potential application of the described technology. The market driver for this application will be the regulation of CO₂ emissions due to concern about its environmental impact towards global climate change or a need for CO₂ for utilization purposes such as enhanced oil recovery (EOR).

Carbon dioxide capture and sequestration (CCS) from large stationary sources such as fossil fuel, combusting electricity generators represents one method to reduce the increase in atmospheric CO₂ levels. The challenges of post-combustion CO₂ capture include the fact that flue gas from utility boilers is at near-atmospheric pressure and the concentration of CO₂ in the flue gas is relatively low at 12-14%. Another technical hurdle is the energy requirements for the CO₂ capture/sequestration using MEA is estimated to reduce a PC plant’s output by about 30 percent, which equates to a very substantial 60-80% increase in the cost of electricity. The ability to capture and store CO₂ more efficiently will be highly valued by utilities.

In broad terms the described method seeks to add a promoter amine, in the form of a substituted primary and/or secondary amine to a tertiary amine to form a solvent. The promoters serve to increase the overall mass transfer of the acid gas into the absorption solvent. The promoters are designed to have particularly low volatility without contributing significant viscosity to the solution. The described promoters achieve this attribute without being an ionic compound which can negatively impact mass transfer. The low volatility and low viscosity are achieved by using alternate functional groups in addition to the amine functional group that reacts with the CO₂ molecule. Low volatility is important to reduce amine loss in the CO₂ capture process. This property is often achieved by using alcohol groups in addition to the amine group. However, the alcohol groups are hydrogen bond donors which add more solution viscosity due to intermolecular bonding.

SUMMARY

A solvent is provided for the removal of an acid gas such as carbon dioxide from a fluid stream such as a flue gas stream. The solvent comprises (a) a promoter amine selected from a group of amines consisting of primary amines with a pKa between 6.5 and 10.5, secondary amines with a pKa between 6.5 and 10.5 and mixtures thereof and (b) a tertiary amine with a pKa of between 8.5 and 10.5. More particularly the solvent includes 0.01-15.0 weight percent promoter amine, 10.0-65.0 weight percent tertiary amine and 34.44-89.99 weight percent water.

The promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter is N-methyltetrahydrothiophen-3-amine, 1,1-dioxide. In one embodiment the promoter is 2,2'-sulfonyldipropaneamine. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter is N-methyltetrahydrothiophen-3-amine, 1,1-dioxide. In one embodiment the promoter is 2,2'-sulfonyldipropaneamine. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA).

In one embodiment the promoter is 6-aminohexanenitrile. In one embodiment the promoter is 3-(methylamino )propanenitrile. In one embodiment the promoter is 3,3'-nyldiethanamine. In one embodiment the promoter is 2-aminoethyl methyl sulfone. In one embodiment the promoter is N,N-dimethyl-N-(3-aminopropyl)ethane-1,1-diamine. In one embodiment the promoter is N,N-dimethyl-N-(3-aminopropyl)ethane-1,1-diamine. In one embodiment the promoter is 3-N-sulfonylamine (SA). In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter amine includes at least one non-volatilizing functional group selected from a group consisting of —SO₂ —OP(OR)₃ —CN —OPR₃ —OR and —COOR and mixtures thereof where R —H or alkyl. In one embodiment the promoter amine is 3-N-sulfonylamine (SA).
embodiment the promoter is diethyl (3-aminopropyl)phosphonate. In one embodiment the promoter is diethyl (4-aminobutyl)phosphonate. In one embodiment the promoter is diethyl (5-aminopentyl)phosphonate. In one embodiment the promoter is diethyl (6-aminohexyl)phosphonate. In one embodiment the promoter is 2-(tert-butoxy)ethan-1-amine. In one embodiment the promoter is N,N,N,N'-tetraalkyl-1,8-naphthalenediamine, N,N,N',N'-tetraalkyl-1,6-hexanediamine, N,N,N',N'-tetraalkyl-1,5-pentanediamine, N,N,N',N'-tetraalkyl-1,4-butane diamine, N,N,N',_N'-tetraalkyl-1,3-propanediamine, N,N,N',N'-tetraalkyl-1,2-ethanediamine, N,N,N',N'-tetraakis(2-hydroxyethyl)ethylenediamine, N,N,N,N'-pentaalkylidiethylentetramine, N,N,N,N'-pentaalkylidipropylenetramine, N,N,-dialkylethylene diamine, N,N,N,N'-tetraalkylhexylamine, N,N,N,N'-tetraalkylblys (aminoethoxy)ether, N,N, _dimethyl-2(2-aminoethoxy)ethanol, where alkyl represents any methyl, ethyl, propyl, butyl isomer, and mixtures thereof. In one particularly useful embodiment the promoter amine comprises a mixture of 3-N-sulfonylamine (SA) and 3-aminopropionitrile (APN) while the tertiary amine is methyldiethanolamine (MDEA). More particularly the solvent comprises about 0.1 weight percent 3-N-sulfonylamine (SA), about 0.25 weight percent 3-aminopropionitrile (APN) and between about 2.0 and about 65.0 weight percent methyldiethanolamine (MDEA).

In accordance with an additional aspect, a method is provided for removing an acid gas from a fluid stream. That method comprises contacting the fluid stream with a solvent for the removal of an acid gas from the fluid stream. That solvent includes a promoter amine with a pKa of between about 6.5 and about 10.5 and a tertiary amine with a pKa of between about 8.5 and 10.5. More particularly the solvent comprises about 0.01-15.0 weight percent promoter amine, about 10.0-65.0 weight percent tertiary amine and about 34.44-89.99 weight percent water.

A solvent or solvent system for removal of an acid gas from a fluid stream comprises a promoter amine with a pKa of between 6.5 and about 10.5 and a tertiary amine with a pKa of between 8.5 and 10.5. More specifically the solvent includes about 0.01-15.0 weight percent promoter amine, about 10.0-65.0 weight percent tertiary amine and about 34.44-89.99 weight percent water.

Additional steps of the method include binding the acid gas in the fluid stream to the solvent to form an acid gas-solvent complex, separating the acid gas solvent complex from the fluid stream, releasing the acid gas from the complex, and circulating the acid gas-solvent complex back to the fluid stream.

In the following description there is shown and described preferred embodiments of the solvent method for removing an acid gas from a fluid stream such as, for example, CO₂ from flue gas. As it will be realized, these solvents and methods are capable of other different embodiments and their several details are capable of modification in various, obvious aspects. Accordingly, the drawings and descriptions will be regarded as illustrative in nature and not as restrictive.

**BRIEF DESCRIPTION OF THE DRAWINGS**

The accompanying drawings incorporated herein and forming a part of the specification, illustrate several aspects of the present invention and together with the description serve to explain certain principles of the invention. In the drawings:

**FIG. 1** is a schematic illustration of a process for removing acid gas from a fluid stream utilizing a solvent and thermal swing regeneration.

**FIG. 2a** is a graphical illustration of the pH-drop of a 0.5 M MDEA solution with various concentrations of solvent promoters.

**FIG. 2b** is a graphical illustration of bicarbonate formation via CO₂ hydration in 0.5 M MDEA and 0.5 M MDEA with 20% APN.

**FIG. 3a** is a graphical illustration of the pH-drop of a 0.2 M DMEA solution and 0.2 M DMEA solution with solvent promoters.

**FIG. 3b** is a graphical illustration of the bicarbonate formation via CO₂ hydration in 0.2 M DMEA and 0.2 M DMEA with a 2.5% and 15% APN mixture.

Reference will now be made in detail to the present preferred embodiment of the invention, examples of which are illustrated in the accompanying drawings.

**DETAILED DESCRIPTION**

A solvent or solvent system for removal of an acid gas from a fluid stream comprises a promoter amine with a pKa of between 6.5 and about 10.5 and a tertiary amine with a pKa of between 8.5 and 10.5. More specifically the solvent includes about 0.01-15.0 weight percent promoter amine, about 10.0-65.0 weight percent tertiary amine and about 34.44-89.99 weight percent water.

The primary amine useful in the solvent system includes a primary amine and/or a secondary amine with at least one non-volatilizing functional group selected from a group consisting of —SO₃ —OP(OR), —CN —OP₃, —OR and —COOR and mixtures thereof where R=—H or alkyl. For purposes of this document alkyl refers to branched and unbranched alkyl compounds with between 1 and 10 carbon atoms. In one embodiment the promoter amine is 3-N-sulfonylamine (SA). In one embodiment the promoter is N-methyltetrahydrothiophen-3-amine 1,1-dioxide. In one embodiment the promoter is 2,2'-sulfonyldiethyamine. In one embodiment the promoter is 3,3'-sulfonyldipropencamine. In one embodiment the promoter is 4,4'-sulfonyldibutanenitrile. In one embodiment the promoter is 2-aminomethyl sulfone. In one embodiment the promoter amine is 3-aminopropionitrile (APN). In one embodiment the promoter amine is 3-aminopropionitrile (APN). In one embodiment the promoter amine is 3-amino propionitrile (APN). In one embodiment the promoter amine is 3-amino propionitrile (APN). In one embodiment the promoter amine is 3-aminopropionitrile (APN).

In one embodiment the promoter amine is diethyl (2-aminomethyl)phosphonate. In one embodiment the promoter is diethyl [2-(methylamino)ethyl]phosphonate. In one embodiment the promoter is diethyl (3-aminopropyl)phosphonate. In one embodiment the promoter amine is diethyl (4-aminobutyl)phosphonate. In one embodiment the promoter amine is diethyl (5-aminopentyl)phosphonate. In one embodiment the promoter is diethyl (6-aminohexyl)phosphonate. In one embodiment the promoter is 2-(tert-butoxy)ethan-1-amine. In one embodiment the promoter is N-methyl-2-{[2-(methyl-2-propanyl)oxy]ethanamine. In one embodiment the promoter amine is a mixture of two or three amines selected from a group consisting of 3-N-sulfonylamine (SA), 3-aminopropionitrile (APN) and diethyl 2-aminoethane phosphonate (EtP₂).
The method includes separating the acid gas solvent complex to reduce the potential for atmospheric emissions from the method includes recycling the regenerated solvent for con­ the step of binding the acid gas in the fluid stream to the solvent and related method for removal of an acid gas representative of the solvent and method it should be appre­ nolamine (DMEA) for examples, with pKa ranging between 6.5 and 10.5. Functional Vapor pressure

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<th>Functional group</th>
<th>Example compound</th>
<th>Vapor pressure (Pa*</th>
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<tr>
<td>HO</td>
<td>Monoethanolamine (MEA)</td>
<td>55</td>
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<tr>
<td>SO2</td>
<td>Sulfonylamine (SA)</td>
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</tr>
<tr>
<td>CN</td>
<td>3-aminopropionitrile</td>
<td>89</td>
</tr>
<tr>
<td>OP(OH)3</td>
<td>Aminoethyl-dimethoxyphosphine oxide</td>
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*Vapor pressure predicted using ACD/I-Lab at 25° C.

Addition of 3-N-sulfonylamine (SA), 3-aminopropionitrile (APN), or a combination of the two to a tertiary amine solution (0.2 M-0.5 M) resulted in an up to 300% increase in relative rate of CO2 hydration (FIG. 2). As shown in FIG. 2a, the relative rate of CO2 hydration increases with increased amounts of solvent promoters added. However, it was observed that different promoters, based on pKa, show different maximum levels of promotion. For example, SA shows no increase in promotion above ~3.5 mol % of tertiary amine concentration, while APN maintains its promoter effects up to at least 20 mol % based on tertiary amine concentration. The effectiveness of the solvent promoter was judged on its ability to increase the relative rate of bicarbonate formation from CO2 and H2O (FIG. 2b). It was observed that addition of 20 mol % APN to a 0.5M MDEA solution gives significant enhancement of the relative rate of bicarbonate formation. The rate enhancement and increased bicarbonate formation is not a factor of the summation of the two amines acting independently (data not shown). If the amount of bicarbonate formed by APN independently is added to the amount of bicarbonate formed by MDEA independently then the total amount of bicarbonate is less than that of when the two are combined in one solution. This strongly suggests that APN is acting as a promoter for carbon capture by MDEA.

The solvent promoters show promising results in MDEA solutions. In order to understand the scope of these promoters, various solvents were tested. It was observed that the different promoters showed a varying amount of activity in other solvents. For example, the optimal mixture used for the best enhancement in MDEA (2.5% SA and 15% APN) showed a ~150% increase in relative rate in DMAE solutions (FIG. 3). While some solvents showed more significant results than others, it shows the versatility of using less basic primary amines as promoters for more basic tertiary amines.

The foregoing has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the embodiments to the precise form disclosed. Obvious modifications and variations are possible in light of the above teachings. For example, while certain promoter amines and tertiary amines are identified above as being useful in the present solvent and method, these are to be considered illustrative of possible promoter and tertiary amines that may be used rather than limiting. Similarly, while this document references CO2 capture from a flue gas, the method is suited for other applications where acid gas capture from a fluid stream is desired. All such modifications...
What is claimed:
1. A solvent for removal of an acid gas from a fluid stream, comprising:
   a promoter amine comprising a mixture of N-sulfonylamine (SA) and 3-aminopropionitrile (APN), wherein SA comprises 3-5 mol% and APN comprises 1-20 mol%; and
   a tertiary amine with a pKa of between 8.5 and 10.5.
2. The solvent of claim 1 including 0.01 to 15.0 weight percent promoter amine, 10.0 to 65.0 weight percent tertiary amine and 34.99 to 89.99 weight percent water.
3. The solvent of claim 1, wherein said tertiary amine is selected from a group of tertiary amines consisting of methyldiethanolamine (MDEA), triethanolamine (TEA), N,N,N',N'-tetraalkylethanolamine, N,N,N',N'-tetraalkyl-1,8-naphthalenediamine, N,N-N',N'-tertionalyl-1,8-naphthalenediamine, N,N-N',N'-tertionalyl-1,6-hexanediame, N,N,N',N'-tertionalyl-1,5-pentanediame, N,N,N',N'-tertionalyl-1,4-butanediame, N,N,N',N'-tertionalyl-1,3-propanediame, N,N,N',N'-tertionalyl-1,2-ethanediame, N,N,N',N'-tetrakis (2-hydroxyethyl) ethylenediamine, N,N,N',N'-pentaklydiethylenetriame, N,N,N',N',N'-pentaalkyldipropyaminetraime, N,N,-dialkylecloxyleyhamine, N,N,N',N'-tertionalylbis (aminoethyl)ether, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethylethylenetriame, and mixtures thereof.
4. The solvent of claim 1, wherein said tertiary amine is selected from a group of tertiary amines consisting of methyldiethanolamine (MDEA), triethanolamine (TEA), dimethylethanolamine (DMEA), N,N,N',N'-tetraalkylethanolamine, N,N,N',N'-tetraalkyl-1,8-naphthalenediamine, N,N,N',N'-tetraalkyl-1,6-hexanediame, N,N,N',N'-tetraalkyl-1,5-pentanediame, N,N,N',N'-tetraalkyl-1,4-butanediame, N,N,N',N'-tetraalkyl-1,3-propanediame, N,N,N',N'-tetraalkyl-1,2-ethanediame, N,N,N',N'-tetrakis (2-hydroxyethyl) ethylenediamine, N,N,N',N'-pentaklydiethylenetriame, N,N,N',N',N'-pentaalkyldipropyaminetraime, N,N,-dialkylecloxyleyhamine, N,N,N',N'-tertionalylbis (aminoethyl)ether, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethylethylenetriame, and mixtures thereof.
5. The solvent of claim 1, wherein said tertiary amine is methyldiethanolamine (MDEA).
6. The solvent of claim 5, wherein said solvent comprises about 0.1 weight percent N-sulfonylamine (SA), about 0.25 weight percent aminopropionitrile (APN) and between about 2.0 and about 65.0 weight percent methyldiethanolamine and a remainder is water.
7. The solvent of claim 1, wherein said acid gas is carbon dioxide.
8. A method of removing an acid gas from a fluid stream, comprising:
   contacting said fluid stream with a solvent for removal of said acid gas from said fluid stream, and said solvent including:
   (a) a promoter amine comprising a mixture of N-sulfonylamine and 3-aminopropionitrile, wherein N-sulfonylamine comprises 3-5 mol% and 3-aminopropionitrile comprises 1-20 mol%; and
   (b) a tertiary amine with a pKa of between 8.5 and 10.5.
9. The method of claim 8, including providing said solvent with 0.01 to 15.0 weight percent promoter amine, 10.0 to 65.0 weight percent tertiary amine and 34.99 to 89.99 weight percent water.
10. The method of claim 8, including using a tertiary amine selected from a group of tertiary amines consisting of methyldiethanolamine (MDEA), triethanolamine (TEA), dimethylethanolamine (DMEA), N,N,-dimethylbenzylamine, diethylmonoethanolamine, dipropylmonoethanolamine, N,N,N',N'-tetramethyldiethylethlenetriame, N,N,N',N'-tetramethylethylenetriame, N,N,N',N'-tetramethyldiethylethlenetriame, N,N,N',N'-tetramethylethylenetriame, N,N,N',N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, N,N,N',N'-pentamethyldiethylethlenetriame, N,N,N',N'-pentamethylethylenetriame, and mixtures thereof.
11. The method of claim 8, including using methyldiethanolamine (MDEA) as said tertiary amine.
12. The method of claim 8, further including binding acid gas in said fluid stream to said solvent to form an acid gas-solvent complex.
13. The method of claim 12, including separating said acid gas solvent complex from said fluid stream.
14. The method of claim 13, including regenerating said solvent by releasing said acid gas.
15. The method of claim 14, including recycling said regenerated solvent for contacting said fluid stream.