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INTERNATIONAL SEARCHING AUTHORITY

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PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing
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Applicant's or agent's file reference see form PCT/ISA/220		FOR FURTHER ACTION See paragraph 2 below
International application No. PCT/EP2017/072648	International filing date (day/month/year) 08.09.2017	Priority date (day/month/year) 15.09.2016
International Patent Classification (IPC) or both national classification and IPC INV. C10G21/27 C10G29/20 C10L3/10		
Applicant SCHÜLKE & MAYR GMBH		

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

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Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:
 - ☒ the international application in the language in which it was filed.
 - ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).
2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))
3. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:
 - a. ☐ forming part of the international application as filed:
 - ☐ in the form of an Annex C/ST.25 text file.
 - ☐ on paper or in the form of an image file.
 - b. ☐ furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.
 - c. ☐ furnished subsequent to the international filing date for the purposes of international search only:
 - ☐ in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).
 - ☐ on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).
4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
5. Additional comments:

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	<u>1-16</u>
	No: Claims	
Inventive step (IS)	Yes: Claims	<u>1-16</u>
	No: Claims	
Industrial applicability (IA)	Yes: Claims	<u>1-16</u>
	No: Claims	

2. Citations and explanations

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1 Reference is made to the following documents:

- D1 US 5 128 049 A (GATLIN LARRY W [US]) 7 July 1992 (1992-07-07)
- D2 WO 2008/124404 A1 (MI LLC [US]) 16 October 2008 (2008-10-16)
- D3 WO 90/07467 A1 (QUAKER CHEM CORP [US]) 12 July 1990 (1990-07-12)
- D4 WO 98/02501 A1 (BAKER HUGHES INC [US]) 22 January 1998 (1998-01-22)

2 Document D1 is regarded as being the prior art closest to the subject-matter of claim 1, and discloses the use of a composition that comprises:

a1) a hydrogen sulfide-scavenging agent in the form of a N,N',N'' tris(2-hydroxyethyl) hexahydrotriazine which is regarded as condensation product of an ethanolamine and formaldehyde (see col. 2, lines 13-15; col. 2, lines 26-27

a2) diluent which can be selected from monoethylene glycol (see col. 3, line 40)

wherein component a) is present in an amount from 10 to 30% by weight (see col. 3, line 44) and component b) is present in an amount from 70 to 90% by weight (see col. 3, lines 44-45) , in each case based on the weight of the composition,

in the removal of hydrogen sulfide compounds from process streams (see col. 3, lines 36-38; col. 1, lines 6-8) .

The subject-matter of claim 1 therefore differs from this known use in that the composition used in claim 1 for the removal of sulphur compounds from process streams contains a condensation products of 1-aminopropan-2-ol and formaldehyde .

The subject-matter of claim 1 is therefore new (Article 33(2) PCT).

The problem to be solved by the present invention may be regarded as to provide a composition that increases the removal of sulphur compounds from process streams .

The solution to this problem proposed in claim 1 of the present application is considered as involving an inventive step (Article 33(3) PCT) for the following reasons:

Document D1 taken alone or in combination with any of the other documents cited in the international search report does suggest that the use of a composition comprising the combination of one or more condensation products of 1-aminopropan-2-ol and formaldehyde (component a)) with a monoethylene glycol (component b)) wherein component a) is present in an amount from 5 to 70% by weight of the composition and component b) is present in an amount from 30 to 95% by weight of the composition , leads to an increased removal of sulphur compounds from process streams.

3 Claim 15 is directed at a corresponding method for removing sulphur compounds from a process stream using the same composition and as such is also new and inventive.

4 Claim 16 is directed at the use of a composition as mentioned in claim 1 wherein the weight ratio between component a) and b) is specified and as such is also new and inventive.

5 Claims 2-14 are dependent on claim 1 and as such also meet the requirements of the PCT with respect to novelty and inventive step.

Re Item VIII

Certain observations on the international application

Although claims 1 and 16 have been drafted as separate independent claims in the same category, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought and/or in respect of the terminology used for the features of that subject-matter. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.



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amendments (Rule 48.2(h))

(54) Title: USE OF COMPOSITIONS HAVING A CONTENT OF CONDENSATION PRODUCT OF 1-AMINOPROPAN-2-OL AND FORMALDEHYDE IN THE REMOVAL OF SULPHUR COMPOUNDS FROM PROCESS STREAMS

(57) Abstract: The invention relates to the use of a composition that comprises a) one or more condensation products of 1-amino-propan-2-ol and formaldehyde and b) monoethylene glycol, wherein component a) is present in an amount from 5 to 70% by weight and component b) is present in an amount from 30 to 95% by weight, in each case based on the weight of the composition, in the removal of sulphur compounds from process streams.



**Use of compositions having a content of condensation product
of 1-aminopropan-2-ol and formaldehyde in the removal
of sulphur compounds from process streams**

The present invention relates to the use of a composition that comprises a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde and b) monoethylene glycol, in the removal of sulphur compounds from process streams. In addition, the invention relates to a corresponding method for removing one or more sulphur compounds from a process stream.

H₂S is an unpleasantly smelling and toxic gas that is a hazard to health to a great extent and furthermore leads in industrial systems to serious corrosion phenomena. The legislator has therefore enacted strict requirements for lowering the H₂S content in gaseous and liquid streams.

WO90/07467 A1 discloses condensation products of formaldehyde and ethanolamine (such as 3,3'-methylenebisoxazolidine and N,N',N''-tris(2-hydroxyethyl)hexahydrotriazine) and use thereof in gaseous and liquid hydrocarbon streams, in order to decrease therein the amount of hydrogen sulphide and organic sulphides. In addition, it is indicated that, in the case of hydrocarbon streams that contain water, glycols can be added in order at the same time to reduce the water content. An exemplary and preferred glycol is triethylene glycol.

WO2016/100224 A2 discloses compositions for removing sulphur compounds from moist or dry gaseous streams. The compositions, in addition to at least one compound for capturing the sulphur compound, further contain at least one hygroscopic agent. Preferred compounds for capturing the sulphur compounds are triazines of monoethanolamine, methylamine or methoxypropylamine. An exemplary hygroscopic agent is glycerol.

Grotan® OX (i.e. 3,3'-methylenebis[5-methyloxazolidine]) is an anhydrous condensation product of formaldehyde and 1-

aminopropan-2-ol (monoisopropanolamine, MIPA) in the molar ratio 3:2, (Schülke & Mayr GmbH, Norderstedt, Federal Republic of Germany). The product Grotan® WS contains about 80% by weight of the formaldehyde donor compound α,α',α'' -trimethyl-1,3,5-triazine-1,3,5-(2H,4H,6H)triethanol (N,N',N''-tris(2 hydroxypropyl)hexahydrotriazine, hereinafter TTT). TTT is produced by condensation of 1-aminopropan-2-ol with formaldehyde (in the molar ratio 1:1). Grotan® OX and Grotan® WS show good efficacy in the chemical neutralization of H₂S.

The problem addressed by the present invention is to provide compositions that remove sulphur compounds from process streams, more precisely also from those process streams that preferably contain little or no water, including liquid and gaseous process streams (such as, in particular, hydrocarbon process streams). H₂S trapping compositions having improved efficiency and advantages in application which additionally are also more economical are sought.

It has now surprisingly been found that this problem is solved by using, in the removal of sulphur compounds from process streams

- a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde and
- b) monoethylene glycol,

wherein the weight ratio used of a):b) is in the range from 5:95 to 70:30. The components a) and b) are preferably used according to the invention as a composition comprising a) and b), wherein component a) is present in an amount from 5 to 70% by weight and component b) is present in an amount from 30 to 95% by weight, in each case based on the weight of the composition.

The invention is based, inter alia, on the fact that it has been found that the efficiency of condensation products of 1-aminopropan-2-ol and formaldehyde in the removal of sulphur

compounds from process streams may be significantly increased by addition of monoethylene glycol. In addition, it is advantageous that, in the compositions according to the invention, the proportion of component b) does not impair those product properties such as handleability and flashpoint. Even in the case of the condensation product N,N',N''-tris(2-hydroxypropyl)hexahydrotriazine, that already acts well when used alone, there is an increase in activity when monoethylene glycol is added. Although, by addition of propylene glycol, a certain improvement in activity may be achieved, this improvement, however, is markedly less than compared with the effect with monoethylene glycol.

In a first aspect, the invention therefore relates to the use of a composition that comprises

- a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde and
- b) monoethylene glycol,

wherein component a) is present in an amount from 5 to 70% by weight and component b) is present in an amount from 30 to 95% by weight, in each case based on the weight of the composition,

in the removal of sulphur compounds from process streams.

The component a) is a condensation product of 1-aminopropan-2-ol and formaldehyde, or a mixture of such condensation products. Preferably, the condensation product is produced by reacting 1-aminopropan-2-ol and formaldehyde in the molar ratio 1:0.8 to 1:1.8, preferably in the molar ratio 1:0.9 to 1:1.6.

In a first preferred embodiment, the condensation product is 3,3'-methylenebis[5-methyloxazolidine], i.e. the condensation product of formaldehyde and 1-aminopropan-2-ol in the molar ratio 3:2. In this case it is preferred that the component a) is

3,3'-methylenebis[5-methyloxazolidine]. In this first preferred embodiment, preferably component a) is present in an amount from 20 to 60% by weight, and component b) is present in an amount from 40 to 80% by weight.

5

Particularly preferably, in the first preferred embodiment, component a) is present in an amount from 30 to 50% by weight and component b) is present in an amount from 50 to 70% by weight, wherein, in particular, component a) is present in an amount of about 40% by weight and component b) is present in an amount of about 60% by weight.

This first preferred embodiment in which component a) comprises 3,3'-methylenebis[5-methyloxazolidine] and preferably, component a) is 3,3'-methylenebis[5-methyl-oxazolidine], wherein, particularly preferably, component a) is present in an amount from 30 to 50% by weight and component b) is present in an amount from 50 to 70%, is therefore particularly advantageous because the surprising effect in lowering the content of sulphur compounds already results after a short exposure time and the reinforcement of the effect by the content of monoethylene glycol is particularly pronounced (see Examples 6 to 8 and, in particular, Example 7).

In this first preferred embodiment, it is further preferred that the composition comprises less than 10% by weight of water, preferably less than 5% by weight of water, in particular less than 2% by weight of water, such as less than 1% by weight of water.

Particularly, preference is therefore given to the use of a composition that consists of a) about 40% by weight of 3,3'-methylenebis[5-methyloxazolidine] and b) about 60% by weight of monoethylene glycol.

In a second preferred embodiment, the condensation product is N,N',N''-tris(2-hydroxypropyl)hexahydrotriazine, i.e. the

condensation product of formaldehyde and 1-aminopropan-2-ol in the molar ratio 3:3 (1:1), wherein component a) is preferably N,N',N''-tris(2-hydroxypropyl)hexahydrotriazine. In this second preferred embodiment, the composition preferably comprises 2 to 25% by weight of water, more preferably 5 to 20% by weight of water, in particular 10 to 15% by weight of water, such as 12% by weight of water.

In addition, in this second preferred embodiment, it is preferred that component a) is present in an amount from 35 to 60% by weight and component b) is present in an amount from 30 to 50% by weight, wherein preferably component a) is present in an amount from 40 to 55% by weight and component b) is present in an amount from 35 to 45% by weight, wherein, in particular component a) is present in an amount of about 48% by weight and component b) is present in an amount of about 40% by weight.

In all embodiments of the invention it is advantageous when the composition does not comprise a dialkylhydroxylamine of the formula $RR'NOH$, wherein R and R' independently of one another are selected from linear, branched and cyclic C_1 to C_{10} alkyl groups.

It is in addition advantageous when the composition does not contain an additive selected from urea, urea derivatives, amino acids, guanidine and guanidine derivatives.

The process stream is preferably selected from liquid and gaseous process streams.

Preferably, the process stream contains at most 40% by weight of water, more preferably at most 35% by weight of water, particularly preferably at most 30% by weight of water, such as at most 25% by weight of water, or at most 20% by weight, or at most 15% by weight, for instance at most 10% by weight, or at most 5% by weight, for example at most 1% by weight, of water.

Fields of use are, inter alia, biogas plants, the transport of petroleum and natural gas, the processing, storage and transport of fossil energy carriers and the removal of H₂S liberated by sulphate-reducing bacteria under anaerobic conditions. Examples of H₂S-containing streams or products are petroleum, crude oil, mineral oil, heating oil, diesel fuel, bitumen, distillation residues, drilling liquid and wastewater. Particular preference is given to the use according to the invention of the compositions in process streams that are hydrocarbon streams. Preferably, the process stream is a liquid or gaseous hydrocarbon stream.

The sulphur compound, for the removal of which the described composition is used, is selected from hydrogen sulphide, inorganic and organic sulphides, mercaptans and mercaptides, wherein the composition is preferably used in the removal of hydrogen sulphide from process streams.

In a second aspect, the invention relates to a method for removing one or more sulphur compounds from a process stream, in which the process stream that contains the sulphur compound or sulphur compounds is contacted with a composition that comprises

- a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde and
- b) monoethylene glycol,

wherein component a) is present in an amount from 15 to 70% by weight and component b) is present in an amount from 30 to 85% by weight, in each case based on the weight of the composition.

According to a third aspect of the present invention, it is possible to add components a) and b) to the process stream separately in the said weight ratio. Correspondingly, the invention also relates to the use of a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde, and b)

monoethylene glycol, wherein the weight ratio used of a):b) is in the range from 5:95 to 70:30, in the removal of sulphur compounds from process streams.

According to the invention, it is in any case preferred to use, according to the first and second aspects, compositions which comprise the components a) and b). These compositions have a low viscosity and offer advantages with respect to cost savings, economic efficiency, cold stability and flow behaviour.

The advantages of the invention result, in particular, from the following examples. Percentages, unless stated otherwise, relate to the weight.

Examples

1. Condensation products used

Grotan® OX:

The reaction product of isopropanolamine and paraformaldehyde (91%) is formed in the molar ratio 2:3. In this case 3,3'-methylenebis(5-methyloxazolidine) is formed. The reaction water and the water from the paraformaldehyde are distilled off.

Grotan® WS:

The reaction product is formed from isopropanolamine and paraformaldehyde (91%) in the molar ratio 3:3 (1:1). This forms α,α',α'' -trimethyl-1,3,5-triazine-1,3,5-(2H,4H,6H)-triethanol (N,N',N''-tris(2-hydroxypropyl)hexahydrotriazine, TTT). The reaction water and the water from the paraformaldehyde remain in the product. The TTT content is about 80% by weight.

2. Method: Determination of the sulphide concentration (based on IP 570, determination of hydrogen sulphide in mineral oils)

- Action of the differing sulphur capturing agents on the sample at various temperatures and for differing times

- Dilution of the sample with alkylbenzene in order to arrive at the linear working range of the analysis system
- Injection of the sample under investigation (including sulphur capturing agent) into the analysis system
- 5 - Addition of acid (2M H_3PO_4 in water) and optionally heating the analytical sample in the analysis system
- Quantitative expulsion of the resultant hydrogen sulphide in the analysis system by means of air and transfer of the hydrogen sulphide to an electrochemical measuring electrode
- 10 in the analysis apparatus
- The hydrogen sulphide generates, at the electrochemical measurement electrode, a measurement signal that is proportional to the respective hydrogen sulphide amount
- By means of evaluation software, the resultant peak area
- 15 (composed of measured signal intensity against time) is determined and converted to a sulphide content, on the basis of a calibration line

3. Study of various formaldehyde condensation products, glycols and mixtures thereof

In order to show the advantages of the compositions used according to the invention, a test was made as to what action various compositions have on the removal of H_2S in C_{10-13} alkyl benzene (sulphide concentration 200 ppm). The samples were

25 heated for this purpose to 50°C and the compositions were used in an amount in each case of 1000 ppm, wherein in the table hereinafter, for the mixtures of condensation product and glycol (monoethylene glycol and propylene glycol) in each case the weight ratio is stated. The results in the case of percentage

30 reduction of H_2S content after exposure for 2 or 24 hours are given in Tab. 1.

Tab. 1

No.	Composition	2 h	24 h
1*	Monoethylene glycol	2.3%	5.8%
2*	Grotan® OX	7.9%	17.8%
3*	Grotan® OX + Monoethylene glycol (9:1)	9.7%	24.5%
4*	Grotan® OX + Monoethylene glycol (8:2)	18.7%	-
5	Grotan® OX + Monoethylene glycol (6:4)	21.9%	82.6%
6	Grotan® OX + Monoethylene glycol (1:1)	23.3%	86.1%
7	Grotan® OX + Monoethylene glycol (4:6)	25.4%	88.0%
8	Grotan® OX + Monoethylene glycol (3:7)	21.5%	93.6%
9	Grotan® OX + Monoethylene glycol (2:8)	22.3%	97.5%
10	Grotan® OX + Monoethylene glycol (1:9)	14.6%	99.4%
11*	Grotan® OX + Propylene glycol (4:6)	13.8%	34.0%
12*	Grotan® WS	14.0%	72.4%
13	Grotan® WS + Monoethylene glycol (6:4)	25.0%	88.9%

* Comparison

- Experiment not carried out

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The results verify that monoethylene glycol and the condensation products of 1-aminopropan-2-ol and formaldehyde (Grotan® OX and Grotan® WS) in each case alone reduce the H₂S content only insufficiently, and more precisely not only for the exposure time of 2 h, but also for the exposure time of 24 h (comparative experiments 1, 2 and 12). The same applies to compositions

consisting of Grotan® OX and monoethylene glycol in the weight ratios 9:1 and 8:2 (comparative experiments 3 and 4), that is to say having a small fraction of monoethylene glycol, and also to compositions consisting of Grotan® OX and propylene glycol (comparative experiment 11).

In contrast, the compositions according to the invention consisting of i) condensation product of 1-aminopropan-2-ol and formaldehyde (Grotan® OX or Grotan® WS) and ii) monoethylene glycol surprisingly act considerably better when the content of monoethylene glycol is higher than 20% by weight (experiments 5 to 10 and 13). In the case of compositions according to the invention consisting of i) Grotan® OX and ii) monoethylene glycol, for an exposure time of only 2 h, it is shown that a composition consisting of Grotan® OX and monoethylene glycol in the weight ratio 4:6 (experiment 7) already acts well (reducing the H₂S content by 25.4%), and for an exposure time of 24 h, the reduction is even 88%. In the case of the markedly longer exposure time of 24 h, in contrast, a mixture consisting of Grotan® OX and monoethylene glycol in the weight ratio 1:9 (experiment 10) acts excellently (reduction by 99.4%).

4. Particularly preferred composition

For production of the particularly preferred composition consisting of a) 40% by weight of 3,3'-methylenebis-[5-methyloxazolidine] and b) 60% by weight of monoethylene glycol, monoethylene glycol is charged and Grotan® OX is added. After brief stirring, a clear, colourless solution is obtained having the following properties (Tab. 2).

Tab. 2

Appearance	clear, colourless solution
Hazen colour number	11
Density at 20°C, g/ml	1.108
Refraction at 20°C	1.457
Cleavable HCHO, %	17.9

The storage stability of this composition was studied by storing samples for 1 month at 25°C or 40°C in PE bottles (Tab. 3):

Tab. 3

After 1 month of storage at	25°C	40°C
Appearance	clear, slightly yellowish	light yellow
Hazen colour number	29	70
Density at 20°C, g/ml	1.108	1.108
Refraction at 20°C	1.457	1.457
Cleavable HCHO, %	17.7	17.8
Reduction of H ₂ S content, 2 h, %	26.0	27.9
Reduction of H ₂ S content, 24 h, %	87.5	87.4

5

The results in Tab. 3 verify that this particularly preferred composition has not lost the activity in the reduction of H₂S content even after storage for one month at an elevated temperature of 40°C.

10

Claims

1. Use of a composition that comprises

- 5 a) one or more condensation products of 1-aminopropan-2-ol
 and formaldehyde and
 b) monoethylene glycol,

10 wherein component a) is present in an amount from 5 to 70%
by weight and component b) is present in an amount from 30
to 95% by weight, in each case based on the weight of the
composition,

15 in the removal of sulphur compounds from process streams.

2. Use according to Claim 1, **characterized in that** the
condensation product is produced by reacting
1-aminopropan-2-ol and formaldehyde in the molar ratio
1:0.8 to 1:1.8, preferably in the molar ratio 1:0.9 to
20 1:1.6.

3. Use according to Claim 2, **characterized in that** the
component a) is 3,3'-methylenebis[5-methyloxazolidine].

25 4. Use according to Claim 3, **characterized in that** component
a) is present in an amount from 20 to 60% by weight, and
component b) is present in an amount from 40 to 80% by
weight,

30 wherein preferably component a) is present in an amount
from 30 to 50% by weight, and component b) is present in an
amount from 50 to 70% by weight,

35 wherein, in particular component a) is present in an amount
of about 40% by weight and component b) is present in an
amount of about 60% by weight.

5. Use according to Claim 4, **characterized in that** the composition comprises less than 10% by weight of water, preferably less than 5% by weight of water, in particular less than 2% by weight of water, such as less than 1% by weight of water.

6. Use according to Claim 2, **characterized in that** component a) is N,N',N''-tris(2-hydroxypropyl)hexahydrotriazine.

7. Use according to Claim 6, **characterized in that** the composition comprises 2 to 25% by weight of water, preferably 5 to 20% by weight of water, in particular 10 to 15% by weight of water, such as 12% by weight of water.

8. Use according to Claim 6 or Claim 7, **characterized in that** component a) is present in an amount from 35 to 60% by weight and component b) is present in an amount from 30 to 50% by weight,

wherein preferably component a) is present in an amount from 40 to 55% by weight and component b) is present in an amount from 35 to 45% by weight,

wherein, in particular component a) is present in an amount of about 48% by weight and component b) is present in an amount of about 40% by weight.

9. Use according to one of the preceding claims, **characterized in that** the composition does not comprise a dialkylhydroxylamine of the formula $RR'NOH$, wherein R and R' independently of one another are selected from linear, branched and cyclic C_1 to C_{10} alkyl groups.

10. Use according to one of the preceding claims, **characterized in that** the composition does not comprise an

additive selected from urea, urea derivatives, amino acids, guanidine and guanidine derivatives.

11. Use according to one of the preceding claims,
5 **characterized in that** the process stream is selected from liquid and gaseous process streams.

12. Use according to one of the preceding claims,
10 **characterized in that** the process stream contains at most 40% by weight of water, preferably at most 35% by weight of water, particularly preferably at most 30% by weight of water, such as at most 25% by weight of water, or at most 20% by weight, or at most 15% by weight, for instance at most 10% by weight, or at most 5% by weight, for example at
15 most 1% by weight, of water.

13. Use according to one of the preceding claims,
characterized in that the process stream is a hydrocarbon stream.

14. Use according to one of the preceding claims,
characterized in that the sulphur compound is selected from hydrogen sulphide, inorganic and organic sulphides, mercaptans and mercaptides,

25 wherein the composition is preferably used in the removal of hydrogen sulphide from process streams.

15. Method for removing one or more sulphur compounds from
30 a process stream, in which the process stream that contains the sulphur compound or sulphur compounds is contacted with a composition that comprises

a) one or more condensation products of 1-aminopropan-2-ol and formaldehyde and

35 b) monoethylene glycol,

wherein component a) is present in an amount from 15 to 70% by weight and component b) is present in an amount from 30 to 85% by weight, in each case based on the weight of the composition.

5

16. Use of

a) one or more condensation products of
1-aminopropan-2-ol and formaldehyde

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and

b) monoethylene glycol,

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wherein the weight ratio used of a):b) is in the range from 5:95 to 70:30,

in the removal of sulphur compounds from process streams.

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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/072648

A. CLASSIFICATION OF SUBJECT MATTER
INV. C10G21/27 C10G29/20 C10L3/10
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)
C10G C10L 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.
A	US 5 128 049 A (GATLIN LARRY W [US]) 7 July 1992 (1992-07-07) column 3, lines 40,44	1-16
A	----- WO 2008/124404 A1 (MI LLC [US]) 16 October 2008 (2008-10-16) page 9, line 4 page 8, line 5 page 9, lines 26-28 page 6, lines 30-31 page 4, paragraph 17 - page 5, line 9	1-16
A	----- WO 90/07467 A1 (QUAKER CHEM CORP [US]) 12 July 1990 (1990-07-12) page 10, lines 3-5, 11-12 page 17, lines 9-11 page 17, lines 13-17 page 18, line 5 ----- -/-	1-16



Further documents are listed in the continuation of Box C.



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

"E" earlier application or patent but published on or after the international filing date

"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)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

22 December 2017

Date of mailing of the international search report

08/01/2018

Name and mailing address of the ISA/

European Patent Office, P.B. 5818 Patentlaan 2
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Authorized officer

Hilgenga, Klaas

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/072648

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 98/02501 A1 (BAKER HUGHES INC [US]) 22 January 1998 (1998-01-22) page 4, line 8 - page 5, line 8 page 6, line 1 - line 8 -----	1-16

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2017/072648

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5128049	A	07-07-1992	NONE

WO 2008124404	A1	16-10-2008	BR PI0809479 A2 09-09-2014
		CA 2682595 A1 16-10-2008	
		EA 200970914 A1 30-04-2010	
		EP 2155363 A1 24-02-2010	
		US 2010163255 A1 01-07-2010	
		WO 2008124404 A1 16-10-2008	

WO 9007467	A1	12-07-1990	AT 126092 T 15-08-1995
		AU 619375 B2 23-01-1992	
		DE 68923793 D1 14-09-1995	
		DE 68923793 T2 04-04-1996	
		EP 0408700 A1 23-01-1991	
		MX 172932 B 24-01-1994	
		NO 903477 A 07-08-1990	
		RU 2080909 C1 10-06-1997	
		WO 9007467 A1 12-07-1990	

WO 9802501	A1	22-01-1998	AU 719046 B2 04-05-2000
		CA 2231659 A1 22-01-1998	
		DK 0882112 T3 13-01-2003	
		EP 0882112 A1 09-12-1998	
		NO 981090 A 11-05-1998	
		US 6117310 A 12-09-2000	
		US 6339153 B1 15-01-2002	
		WO 9802501 A1 22-01-1998	

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter I of the Patent Cooperation Treaty)

(PCT Rule 44bis)

Applicant's or agent's file reference 2016P00518WO	FOR FURTHER ACTION	See item 4 below
International application No. PCT/EP2017/072648	International filing date (<i>day/month/year</i>) 08 September 2017 (08.09.2017)	Priority date (<i>day/month/year</i>) 15 September 2016 (15.09.2016)
International Patent Classification (8th edition unless older edition indicated) See relevant information in Form PCT/ISA/237		
Applicant SCHÜLKE & MAYR GMBH		

1.	This international preliminary report on patentability (Chapter I) is issued by the International Bureau on behalf of the International Searching Authority under Rule 44 bis.1(a).																								
2.	This REPORT consists of a total of 6 sheets, including this cover sheet.																								
In the attached sheets, any reference to the written opinion of the International Searching Authority should be read as a reference to the international preliminary report on patentability (Chapter I) instead.																									
3.	This report contains indications relating to the following items: <table style="width: 100%;"> <tr> <td style="width: 5%; text-align: center;">☒</td> <td style="width: 25%;">Box No. I</td> <td style="width: 70%;">Basis of the report</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. II</td> <td>Priority</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. III</td> <td>Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. IV</td> <td>Lack of unity of invention</td> </tr> <tr> <td style="text-align: center;">☒</td> <td>Box No. V</td> <td>Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. VI</td> <td>Certain documents cited</td> </tr> <tr> <td style="text-align: center;">☐</td> <td>Box No. VII</td> <td>Certain defects in the international application</td> </tr> <tr> <td style="text-align: center;">☒</td> <td>Box No. VIII</td> <td>Certain observations on the international application</td> </tr> </table>	☒	Box No. I	Basis of the report	☐	Box No. II	Priority	☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability	☐	Box No. IV	Lack of unity of invention	☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement	☐	Box No. VI	Certain documents cited	☐	Box No. VII	Certain defects in the international application	☒	Box No. VIII	Certain observations on the international application
☒	Box No. I	Basis of the report																							
☐	Box No. II	Priority																							
☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability																							
☐	Box No. IV	Lack of unity of invention																							
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement																							
☐	Box No. VI	Certain documents cited																							
☐	Box No. VII	Certain defects in the international application																							
☒	Box No. VIII	Certain observations on the international application																							
4.	The International Bureau will communicate this report to designated Offices in accordance with Rules 44bis.3(c) and 93bis.1 but not, except where the applicant makes an express request under Article 23(2), before the expiration of 30 months from the priority date (Rule 44bis .2).																								

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No. +41 22 338 82 70	Date of issuance of this report 19 March 2019 (19.03.2019) Authorized officer Nora Lindner e-mail: pct.team5@wipo.int
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PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:
see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference see form PCT/ISA/220		FOR FURTHER ACTION See paragraph 2 below
International application No. PCT/EP2017/072648	International filing date (day/month/year) 08.09.2017	Priority date (day/month/year) 15.09.2016
International Patent Classification (IPC) or both national classification and IPC INV. C10G21/27 C10G29/20 C10L3/10		
Applicant SCHÜLKE & MAYR GMBH		

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:  European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Fax: +31 70 340 - 3016	Date of completion of this opinion see form PCT/ISA/210	Authorized Officer Hilgenga, Klaas Telephone No. +31 70 340-0	
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Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:
 - ☒ the international application in the language in which it was filed.
 - ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).
2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))
3. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:
 - a. ☐ forming part of the international application as filed:
 - ☐ in the form of an Annex C/ST.25 text file.
 - ☐ on paper or in the form of an image file.
 - b. ☐ furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.
 - c. ☐ furnished subsequent to the international filing date for the purposes of international search only:
 - ☐ in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).
 - ☐ on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).
4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
5. Additional comments:

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	<u>1-16</u>
	No: Claims	
Inventive step (IS)	Yes: Claims	<u>1-16</u>
	No: Claims	
Industrial applicability (IA)	Yes: Claims	<u>1-16</u>
	No: Claims	

2. Citations and explanations

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1 Reference is made to the following documents:

- D1 US 5 128 049 A (GATLIN LARRY W [US]) 7 July 1992 (1992-07-07)
- D2 WO 2008/124404 A1 (MI LLC [US]) 16 October 2008 (2008-10-16)
- D3 WO 90/07467 A1 (QUAKER CHEM CORP [US]) 12 July 1990 (1990-07-12)
- D4 WO 98/02501 A1 (BAKER HUGHES INC [US]) 22 January 1998 (1998-01-22)

2 Document D1 is regarded as being the prior art closest to the subject-matter of claim 1, and discloses the use of a composition that comprises:

a1) a hydrogen sulfide-scavenging agent in the form of a N,N',N'' tris(2-hydroxyethyl) hexahydrotriazine which is regarded as condensation product of an ethanolamine and formaldehyde (see col. 2, lines 13-15; col. 2, lines 26-27

a2) diluent which can be selected from monoethylene glycol (see col. 3, line 40)

wherein component a) is present in an amount from 10 to 30% by weight (see col. 3, line 44) and component b) is present in an amount from 70 to 90% by weight (see col. 3, lines 44-45) , in each case based on the weight of the composition,

in the removal of hydrogen sulfide compounds from process streams (see col. 3, lines 36-38; col. 1, lines 6-8) .

The subject-matter of claim 1 therefore differs from this known use in that the composition used in claim 1 for the removal of sulphur compounds from process streams contains a condensation products of 1-aminopropan-2-ol and formaldehyde .

The subject-matter of claim 1 is therefore new (Article 33(2) PCT).

The problem to be solved by the present invention may be regarded as to provide a composition that increases the removal of sulphur compounds from process streams .

The solution to this problem proposed in claim 1 of the present application is considered as involving an inventive step (Article 33(3) PCT) for the following reasons:

Document D1 taken alone or in combination with any of the other documents cited in the international search report does suggest that the use of a composition comprising the combination of one or more condensation products of 1-aminopropan-2-ol and formaldehyde (component a)) with a monoethylene glycol (component b)) wherein component a) is present in an amount from 5 to 70% by weight of the composition and component b) is present in an amount from 30 to 95% by weight of the composition , leads to an increased removal of sulphur compounds from process streams.

3 Claim 15 is directed at a corresponding method for removing sulphur compounds from a process stream using the same composition and as such is also new and inventive.

4 Claim 16 is directed at the use of a composition as mentioned in claim 1 wherein the weight ratio between component a) and b) is specified and as such is also new and inventive.

5 Claims 2-14 are dependent on claim 1 and as such also meet the requirements of the PCT with respect to novelty and inventive step.

Re Item VIII

Certain observations on the international application

Although claims 1 and 16 have been drafted as separate independent claims in the same category, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought and/or in respect of the terminology used for the features of that subject-matter. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.