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(54) **Improved catalyst for the selective oxidation of hydrogen sulfide**

(57) The invention is directed to a catalyst bed for the selective oxidation of hydrogen sulfide, said catalyst bed comprising a first catalyst layer and a second catalyst layer, wherein the first catalyst layer comprises first catalyst particles that comprise a first support material and a first active material and the second catalyst layer comprises second catalyst particles that comprise a second support material and a second active material, wherein said first catalyst particles have a higher catalyst loading than said second catalyst particles. In further aspects, the invention is directed to a process for the selective oxidation of hydrogen sulfide, comprising passing a gas stream comprising hydrogen sulfide over the catalyst bed and to a plant for the selective oxidation of hydrogen sulfide comprising the catalyst bed.

P112947NL00

Title: Improved catalyst for the selective oxidation of hydrogen sulfide

The invention is in the field of selective oxidation of sulfur-containing compounds, in particular of hydrogen sulfide. The invention is in particular directed to processes and apparatuses suitable for oxidizing hydrogen sulfide entrained in tail gas streams of a sulfur recovery unit, such as a Claus plant.

Sulfur recovery units are employed to remove sulfur-containing compounds, in particular hydrogen sulfide (H_2S), from acid gas streams such as resulting from treatment of natural gas and to produce elemental sulfur. For gas streams comprising high amounts of hydrogen sulfide (*e.g.* 10 vol% or more), the Claus process is the most common desulfurization process. However, the conversion of H_2S in the Claus process is not complete and tail gas treatment processes are required to reduce the amount of H_2S in the tail gas of the Claus process before the tail gas can be discharged into the atmosphere.

Known methods to treat the tail gas of sulfur recovery units include the low-temperature Claus method, the reduction-absorption method and the selective oxidation method. The selective oxidation method is particularly suitable for the tail gas treatments since, in contrast to the Claus process, the reaction is thermodynamically complete meaning that a theoretically full conversion of H_2S to elemental sulfur is possible.

The selective oxidation of H_2S with oxygen over a catalyst to form elemental sulfur and water is reviewed by Zhang *et al.* (*ACS Catalysis* 2015, 5, 1053-1067). Moreover, the selective oxidation process and particularly suitable catalysts therefore are described in EP0409353 and EP0242920, which are incorporated herein in their entirety.

The selective oxidation of H_2S by oxygen to elemental sulfur is believed to proceed according the following reaction:

$$2\ H_2S + O_2 \rightarrow 2\ H_2O + \tfrac{1}{4}\ S_8 \qquad (1)$$

However, the selective oxidation to form elemental sulfur may be adversely affected by the occurrence of one or more of the following side reactions:

Over oxidation of H_2S: $2\ H_2S + 3\ O_2 \rightarrow 2\ H_2O + 2\ SO_2$ (2)

Oxidation of S_8: $1/8\ S_8 + O_2 \rightarrow 2H_2O + SO_2$ (3)

Nasato *et al.* (*Oil & Gas Journal*, 1994, 45-48) describe a catalytic system by loading the catalyst with alpha-alumina (α-Al) based and silica (Si) based catalysts in a layered manner. This allowed the reduction of the inlet temperature of the catalyst bed end and resulted in a higher elemental sulfur yield.

The formation of SO_2 is undesirable and the side reactions are preferably minimized to maximize the yield of obtained elemental sulfur.

The present inventors surprisingly found that by reducing the catalyst loading of the catalyst particles that catalyze the selective oxidation, a higher selectivity towards the formation of elemental sulfur from H_2S is obtained. However, a reduced catalyst loading generally results in a lower catalyst activity at an equal space velocity (*e.g.* gas hourly space velocity, GHSV, or weight hourly space velocity, WHSV) and thus in a lower yield of elemental sulfur. Accordingly, reducing the catalyst loading must generally be counterbalanced with an increase catalyst volume resulting in increased process costs and for instance a different design of the process plant. Alternatively the temperature of the catalyst bed can be increased to

enhance the reaction rate. A higher catalyst bed temperature however also promotes formation of SO_2.

The present inventors however surprisingly found that by providing a layered catalyst bed, wherein each layer comprises catalyst particles having a different catalyst loading, the advantage of the higher selectivity could be provided without having to resort to a lower space velocity.

Accordingly, an aspect of the present invention is directed to a catalyst bed for the selective oxidation of hydrogen sulfide, said catalyst bed comprising a first catalyst layer and a second catalyst layer. The first catalyst layer comprises first catalyst particles that comprise a first support material and a first active material and the second catalyst layer comprises second catalyst particles that comprise a second support material and a second active material, wherein said first catalyst particles have a higher catalyst loading than said second catalyst particles.

A further aspect of the present invention is a process for the selective oxidation of hydrogen sulfide, comprising passing a gas stream comprising hydrogen sulfide over the catalyst bed, wherein the gas stream first contacts the first catalyst layers and subsequently contacts the second catalyst layer. The gas stream generally comprises between 0.1 to 2.5 vol%, preferably between 0.5 and 1.5 vol% hydrogen sulfide.

The present invention results in an excellent selectivity towards and yield of elemental sulfur in the selective oxidation process. The yield of elemental sulfur is herein expressed as the degree of elemental sulfur production and is equal to the degree of H_2S conversion corrected for the formation of SO_2. Accordingly, the yield can be defined as

$$yield = \frac{[H_2S_{in}] - [H_2S_{out}] + [SO_{2,in}] - [SO_{2,out}]}{[H_2S_{in}]} \times 100\% \qquad (4)$$

The selectivity of the selective oxidation process is herein expressed as the fraction of converted hydrogen sulfide that is converted to elemental sulfur. Accordingly, the selectivity can be defined as

$$selectivity = \frac{[H_2S_{in}] - [H_2S_{out}] + [SO_{2,in}] - [SO_{2,out}]}{[H_2S_{in}] - [H_2S_{out}]} \times 100\% \qquad (5)$$

The activity of the selective oxidation process is herein expressed as the degree of hydrogen sulfide that is converted. Accordingly, the activity can be defined as

$$activity = \frac{[H_2S_{in}] - [H_2S_{out}]}{[H_2S_{in}]} \times 100\% \qquad (6)$$

Without wishing to be bound by theory, the present inventors believe that an improved sulfur yield can be obtained for each catalyst layer due to the exothermic nature of the selective oxidation reaction of hydrogen sulfide. Since the present selective oxidation of hydrogen sulfide is typically carried out in an adiabatic reactor and the oxidation reaction is exothermic, the temperature increases when the gas stream contacts the catalyst bed where the reaction takes place. The temperatures increases further when the gas stream passed further through the catalyst bed from one layer to the next layer. The inlet temperature of the catalyst bed is typically maintained above 150 °C, which generally results in a temperature of the catalyst bed up to between 160 and 300 °C.

In general, the yield of the selective oxidation is optimum at a certain temperature. At lower temperature, the catalyst activity decreases and at higher temperature, one or more of the above-described side reactions may become more pronounced (*i.e.* the selectivity decreases). Thus, in case the catalyst loading remains constant throughout the catalyst bed (as is the case in conventional catalyst beds), the increasing temperature

results in an increasingly lower local selectivity and elemental sulfur yield. The present inventors have solved this problem by providing a multilayered catalyst system wherein preferably the catalyst loading of each layer substantially corresponds to the optimum local temperature such that a substantially optimal local yield of elemental sulfur and thus a substantially optimal overall yield is obtained.

Although an improved yield is already obtained by providing two catalyst layers in the catalyst bed, it may be preferred that the catalyst bed of the present invention comprises one or more further catalyst layers, each catalyst layer comprising catalyst particles. In this particular embodiment, the catalyst loading of the catalyst particles of each catalyst layer is lower than of the catalyst particles of the preceding catalyst layer such that the catalyst loading of the catalyst particles decreases over the layers of the catalyst bed. If the layers are sufficiently thin, the catalyst loading of the catalyst particles may decrease gradually over the catalyst bed.

The temperature may of the catalyst be influenced by the amount of oxygen that is used for the oxidation of the hydrogen sulfide. Preferably, the gas stream further comprises oxygen and the molar ratio of oxygen to hydrogen sulfide is maintained at 0.5 to 1.5.

In general, any known catalyst for the selective oxidation of sulfur-containing compound may suffer from the side reactions as discussed above. Accordingly, the present invention may be applied to the selective oxidation, irrespective of the type of catalyst used. Particularly preferred catalyst beds and layers are those comprising active materials such as carbon and/or metal-oxide. Zhang *et al.* (*ACS Catalysis* 2015, 5, 1053-1067) describe a number of catalysts that are applicable in the selective oxidation and may thus also be applied for the present invention.

The catalysts described in EP0409353 and EP0242920 comprise iron compounds (*e.g.* iron oxide) or a mixture of iron compounds with other metal species as active material and these catalysts are most preferred. The

catalyst further preferably comprises a carrier material comprising silica, alumina or a combination thereof.

The different catalyst layers in the catalyst bed may each be based on different or on the same active materials. It is preferred that the first active material and the second active material comprises carbon or metal-oxide, more preferably iron-oxide.

The catalyst loading of a catalyst particle can be expressed by weight of the active material based on the total weight of the catalyst particle. In a preferred embodiment, in particular when the first active material comprises iron-oxide, the catalyst particles have a catalyst loading of 3-20 wt%, preferably 5-10 wt% first active material based on the total weight of the first catalyst particles.

Said second catalyst particles preferably have a catalyst loading of less than 5 wt%, preferably less than 3 wt% second active material based on the total weight of the second catalyst particles. This is also particularly preferred in case if the second active material comprises iron-oxide.

The catalyst particles with the desired catalyst loading can be obtained by standard preparation procedures such as impregnation (*e.g.* incipient wetness impregnation) of the support material.

The support material may be any support material, but particularly good results can be obtained when the first support material and/or the second support material comprises alpha-alumina or silica or silicon carbide or activated carbon or other supports that do not catalyze the Claus reaction, preferably silica.

The layers of the catalyst bed may further comprise inert particles which are typically based on the same support material as the catalyst particles in the catalyst bed. Interestingly, increasing the ratio inert particles versus catalytic particles typically decreases the activity of the catalyst bed, but does not result in a higher selectivity of the selective

oxidation reaction. This illustrates the unpredictable nature of the selective oxidation reaction and the surprising element of the present invention.

Without wishing to be bound by theory, the present inventors believe that the lower activity and the coupled higher selectivity as a result of a lower catalyst loading, may be connected to certain intraparticle transportation effects within the catalyst particles. The present invention was found to give particular good results when the first catalyst particles have a pore volume of 0.8-1.2 mm^3/g, preferably about 1 mm^3/g and the second catalyst particles have a pore volume of more than 1.2 mm^3/g. These pore volumes are particularly preferred for catalyst particles based on iron oxide as the active material and silica as the carrier material.

Aspects such as particle size distribution of the catalyst particles, average particle size of the catalyst particles and interaction of the active material with the support material can possibly influence the selectivity of the reaction. These aspects may change the intrinsic properties of the catalyst particle.

Another parameter that may be advantageously used to improve the elemental sulfur yield is the volumetric ratio of the different catalyst layers within the catalyst bed. As described herein-above, it is preferred that the catalyst loading of each layer substantially matches the local temperature. The temperature of the gas stream may not increase linear throughout the catalyst bed and/or the difference in catalyst loading may not be linear from one layer to the next layer. In general, it is preferred that the volume of the first catalyst layer is 15-50 vol%, preferably 20-30 vol% of the total catalyst bed volume.

The volume of the second catalyst layer is preferably 50-85 vol%, preferably 70-80 vol% of the total catalyst bed volume.

The catalyst bed of the present invention provides particular good results when it is provided in an adiabatic reactor. However, it may in principle be advantageously used in any reactor that is isolated such that a

temperature profile over the catalyst bed is obtained during the selective oxidation process (*i.e.* the reaction temperature increases in the direction of gas stream flow over the catalyst bed).

The present invention can be illustrated by the following examples.

Example 1

An inlet stream comprising 1.0 vol% H_2S, 0.07 vol% SO_2, 30 vol% H_2O, balance N_2 together with oxygen in a molar ratio of 2:1 oxygen to H_2S was passed over a bilayered catalyst bed comprising 25 v% of a first (top) layer of 5 wt% iron oxide on silica catalyst and 75 v% of a second (bottom) layer of 2 wt% iron oxide on silica catalyst. A space velocity of 2000 h^{-1} was set. The outlet stream was analyzed via a gas chromatograph to determine the conversion, selectivity and yield of sulfur.). The results are provided in Table 1.

Table 1

Inlet Temperature (°C)	Selectivity (%)	Yield of Sulfur (%)
195	97.79	45.73
200	97.94	59.29
210	96.79	86.17
220	96.80	95.26
225	96.14	95.90
230	96.06	96.26
235	96.12	96.03
240	95.85	95.75
245	93.63	93.54
250	92.47	92.40
260	88.56	88.46
270	80.28	80.15

Reference Example 1 - single-layered catalyst bed

An inlet gas stream comprising 1.0 vol% H_2S, 0.07 vol% SO_2, 30 vol% H_2O, balance N_2 to 100 vol%, was passed over a single-layered catalyst bed comprising 50 v% of 5 wt% iron oxide on silica catalyst blended with 50 v% of inert alumina together with oxygen in a molar ratio of 2:1 oxygen to H_2S. A space velocity of 2000 h^{-1} was used.

The outlet stream was analyzed via a gas chromatographer to determine the selectivity and yield of sulfur. The results are provided in Table 2.

Table 2

Inlet Temperature (°C)	Selectivity (%)	Yield of Sulfur (%)
220	96.67	84.51
225	96.77	86.88
230	96.62	93.10
235	95.96	94.53
240	95.95	95.45
245	96.04	95.51
250	94.87	94.71
260	92.85	92.72
270	88.57	88.40

Reference Example 2 - single-layered catalyst bed

Reference Example 1 was repeated, but the H_2S was passed over a single-layer catalyst bed comprising 50 v% of 2 wt% iron oxide on silica catalyst blended with 50 v% of inert alumina. The results are provided in Table 3.

Table 3

Inlet Temperature (°C)	Selectivity (%)	Yield (%)
225	97.65	64.47
230	97.30	78.46
235	97.68	87.79
240	97.26	93.89
250	96.54	95.83
260	95.81	95.60
270	93.91	93.77

Reference Example 3 - effect of catalyst dilution with inert material

Reference Example 1 was repeated, but the H_2S was passed over a single-layer catalyst bed comprising 100 v% of 5 wt% iron oxide on silica catalyst (thus no inert material was present). The results are provided in Table 4.

Table 4

Inlet Temperature (°C)	Selectivity (%)
220	96.86
225	96.34
230	96.28
240	95.74
250	94.28
260	91.83
270	85.95

The maximum selectivity which is obtained for the present example (undiluted 5 wt% iron-oxide on silica catalyst) is 96.86%. The data

indicate that the selectivity curve is similar for both Reference Examples 1 and 3.

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Conclusies

1. Katalysatorbed voor de selectieve oxidatie van waterstofsulfide, waarbij genoemd katalysatorbed een eerste katalysatorlaag en een tweede katalysatorlaag omvat, waarbij de eerste katalysatorlaag eerste katalysatordeeltjes omvat, die een eerste dragermateriaal en een eerste actief materiaal omvatten, en de tweede katalysatorlaag tweede katalysatordeeltjes omvat, die een tweede dragermateriaal en een tweede actief materiaal omvatten, waarbij genoemde eerste katalysatordeeltjes een grotere katalysatorbelading dan genoemde tweede katalysatordeeltjes hebben.

2. Katalysatorbed volgens conclusie 1, waarbij genoemde eerste katalysatordeeltjes een katalysatorbelading van 3 tot en met 20 gew.-%, bij voorkeur 5 tot en met 10 gew.-% eerste actief materiaal, gebaseerd op het totale gewicht van de eerste katalysatordeeltjes, hebben.

3. Katalysatorbed volgens een der voorafgaande conclusies, waarbij genoemde tweede katalysatordeeltjes een katalysatorbelading van minder dan 5 gew.-%, bij voorkeur minder dan 3 gew.-%, gebaseerd op het totale gewicht van de tweede katalysatordeeltjes, hebben.

4. Katalysatorbed volgens een der voorafgaande conclusies, waarbij het eerste dragermateriaal en/of het tweede dragermateriaal alfa-aluminiumoxide of siliciumdioxide of siliciumcarbide of geactiveerde koolstof of andere dragermaterialen, die de Claus-reactie niet katalyseren, bij voorkeur siliciumdioxide, omvatten/omvat.

5. Katalysatorbed volgens een der voorafgaande conclusies, waarbij het eerste actieve materiaal en/of het tweede actieve materiaal koolstof of metaaloxide, bij voorkeur ijzeroxide, omvatten/omvat.

6. Katalysatorbed volgens een der voorafgaande conclusies, waarbij het volume van de eerste katalysatorlaag 15 tot en met 50 vol.-%, bij voorkeur 20 tot en met 30 vol.-% van het totale katalysatorbedvolume bedraagt.

7. Katalysatorbed volgens een der voorafgaande conclusies, waarbij het volume van de tweede katalysatorlaag 50 tot en met 85 vol.-%, bij voorkeur 70 tot en met 80 vol.-% van het totale katalysatorbedvolume bedraagt.

8. Katalysatorbed volgens een der voorafgaande conclusies, waarbij de eerste en/of tweede katalysatordeeltjes een porievolume van 0,8 tot en met 1,2 ml/g hebben.

9. Katalysatorbed volgens een der voorafgaande conclusies, waarbij de eerste katalysatorlaag en/of de tweede katalysatorlaag voorts inerte deeltjes omvatten/omvat.

10. Katalysatorbed volgens een der voorafgaande conclusies, omvattende één of meer bijkomende katalysatorlagen, waarbij elke katalysatorlaag katalysatordeeltjes omvat, waarbij de katalysatorbelading van de katalysatordeeltjes van elke katalysatorlaag kleiner is dan van de katalysatordeeltjes van de voorafgaande katalysatorlaag, dusdanig dat de katalysatorbelading van de katalysatordeeltjes over de katalysatorlagen van

het katalysatorbed afneemt.

11. Werkwijze voor de selectieve oxidatie van waterstofsulfide, omvattende het over het katalysatorbed volgens conclusies 1-10 leiden van een gasstroom, waarbij de gasstroom eerst met de eerste katalysatorlagen in aanraking komt en vervolgens met de tweede katalysatorlaag in aanraking komt.

12. Werkwijze volgens conclusie 11, waarbij de inlaattemperatuur van het katalysatorbed boven 150 °C wordt gehandhaafd.

13. Werkwijze volgens een der conclusies 11 of 12, waarbij de gasstroom 0,1 tot en met 2,5 vol.-%, bij voorkeur tussen 0,5 en 1,5 vol.-% waterstofsulfide omvat.

14. Werkwijze volgens een der conclusies 11 tot en met 13, waarbij de gasstroom voorts zuurstof omvat en de molaire verhouding van zuurstof tot waterstofsulfide bij 0,5 tot en met 1,5 wordt gehandhaafd.

15. Installatie voor de selectieve oxidatie van waterstofsulfide, omvattende het katalysatorbed volgens een der conclusies 1 tot en met 10, waarbij genoemde installatie stroomafwaarts van een zwavelherwinningseenheid, in het bijzonder een Claus-installatie, is gelokaliseerd.

Title: Improved catalyst for the selective oxidation of hydrogen sulfide

Abstract

The invention is directed to a catalyst bed for the selective oxidation of hydrogen sulfide, said catalyst bed comprising a first catalyst layer and a second catalyst layer, wherein the first catalyst layer comprises first catalyst particles that comprise a first support material and a first active material and the second catalyst layer comprises second catalyst particles that comprise a second support material and a second active material, wherein said first catalyst particles have a higher catalyst loading than said second catalyst particles. In further aspects, the invention is directed to a process for the selective oxidation of hydrogen sulfide, comprising passing a gas stream comprising hydrogen sulfide over the catalyst bed and to a plant for the selective oxidation of hydrogen sulfide comprising the catalyst bed.

SAMENWERKINGSVERDRAG (PCT)

RAPPORT BETREFFENDE NIEUWHEIDSONDERZOEK VAN INTERNATIONAAL TYPE

IDENTIFICATIE VAN DE NATIONALE AANVRAGE	KENMERK VAN DE AANVRAGER OF VAN DE GEMACHTIGDE
	P112947NL00
Nederlands aanvraag nr. **2017533**	Indieningsdatum **26-09-2016**
	Ingeroepen voorrangsdatum
Aanvrager (Naam) **Jacobs Nederland B.V.**	
Datum van het verzoek voor een onderzoek van internationaal type **03-12-2016**	Door de Instantie voor Internationaal Onderzoek aan het verzoek voor een onderzoek van internationaal type toegekend nr. **SN67886**

I. CLASSIFICATIE VAN HET ONDERWERP (bij toepassing van verschillende classificaties, alle classificatiesymbolen opgeven)

Volgens de internationale classificatie (IPC)

B01J35/00;B01J21/08;B01J23/745;B01J35/10;B01D53/86

II. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK

Onderzochte minimumdocumentatie

Classificatiesysteem	Classificatiesymbolen
IPC	**B01J;B01D**

Onderzochte andere documentatie dan de minimum documentatie, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen

III.	☐	**GEEN ONDERZOEK MOGELIJK VOOR BEPAALDE CONCLUSIES**	(opmerkingen op aanvullingsblad)
IV.	☐	**GEBREK AAN EENHEID VAN UITVINDING**	(opmerkingen op aanvullingsblad)

ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2017533

A. CLASSIFICATIE VAN HET ONDERWERP
INV. B01J35/00 B01J21/08 B01J23/745 B01J35/10 B01D53/86
ADD.

Volgens de Internationale Classificatie van octrooien (IPC) of zowel volgens de nationale classificatie als volgens de IPC.

B. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK

Onderzochte minimum documentatie (classificatie gevolgd door classificatiesymbolen)
B01J B01D

Onderzochte andere documentatie dan de minimum documentatie, voor dergelijke documenten, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen

Tijdens het onderzoek geraadpleegde elektronische gegevensbestanden (naam van de gegevensbestanden en, waar uitvoerbaar, gebruikte trefwoorden)
EPO-Internal, WPI Data

C. VAN BELANG GEACHTE DOCUMENTEN

Categorie *	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
X	EP 1 442 781 A1 (JACOBS NEDERLAND BV [NL]; GASTEC NV [NL]) 4 augustus 2004 (2004-08-04) * alinea [0001]; conclusies; voorbeelden *	1-15
A	EP 1 355 720 A1 (CONOCOPHILLIPS CO [US]) 29 oktober 2003 (2003-10-29) * het gehele document *	1-15
A	US 6 800 261 B1 (BORSBOOM JOHANNES [NL] ET AL) 5 oktober 2004 (2004-10-05) * het gehele document *	1-15
A	US 2012/251436 A1 (ALKHAZOV TOFIK GASAN [US]) 4 oktober 2012 (2012-10-04) * het gehele document *	1-15

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[X] Verdere documenten worden vermeld in het vervolg van vak C.

[X] Leden van dezelfde octrooifamilie zijn vermeld in een bijlage

* Speciale categorieën van aangehaalde documenten

"A" niet tot de categorie X of Y behorende literatuur die de stand van de techniek beschrijft

"D" in de octrooiaanvrage vermeld

"E" eerdere octrooi(aanvrage), gepubliceerd op of na de indieningsdatum, waarin dezelfde uitvinding wordt beschreven

"L" om andere redenen vermelde literatuur

"O" niet-schriftelijke stand van de techniek

"P" tussen de voorrangsdatum en de indieningsdatum gepubliceerde literatuur

"T" na de indieningsdatum of de voorrangsdatum gepubliceerde literatuur die niet bezwarend is voor de octrooiaanvrage, maar wordt vermeld ter verheldering van de theorie of het principe dat ten grondslag ligt aan de uitvinding

"X" de conclusie wordt als niet nieuw of niet inventief beschouwd ten opzichte van deze literatuur

"Y" de conclusie wordt als niet inventief beschouwd ten opzichte van de combinatie van deze literatuur met andere geciteerde literatuur van dezelfde categorie, waarbij de combinatie voor de vakman voor de hand liggend wordt geacht

"&" lid van dezelfde octrooifamilie of overeenkomstige octrooipublicatie

Datum waarop het onderzoek naar de stand van de techniek van internationaal type werd voltooid	Verzenddatum van het rapport van het onderzoek naar de stand van de techniek van internationaal type
7 juni 2017	

Naam en adres van de instantie	De bevoegde ambtenaar
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	de Cauwer, Robby

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ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2017533

C.(Vervolg). VAN BELANG GEACHTE DOCUMENTEN

Categorie *	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
A	WO 2016/070805 A1 (YANG NAN [CN]) 12 mei 2016 (2016-05-12) -----	1-15
A	Elmo Nasato: "NEW CATALYST IMPROVES SULFUR RECOVERY AT CANADIAN PLANT - Oil & Gas Journal", oil and gas journal, 28 februari 1994 (1994-02-28), XP055378502, Gevonden op het Internet: URL:http://www.ogj.com/articles/print/volume-92/issue-9/in-this-issue/gas-processing/new-catalyst-improves-sulfur-recovery-at-canadian-plant.html [gevonden op 2017-06-02] * het gehele document * -----	1-15
A	US 6 652 827 B1 (BORSBOOM JOHANNES [NL] ET AL) 25 november 2003 (2003-11-25) * het gehele document * -----	1-15

ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Informatie over leden van dezelfde octrooifamilie

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2017533

In het rapport genoemd octrooigeschrift	Datum van publicatie	Overeenkomend(e) geschrift(en)	Datum van publicatie
EP 1442781	A1 04-08-2004	CA 2517090 A1	12-08-2004
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WRITTEN OPINION

File No.	Filing date *(day/month/year)*	Priority date *(day/month/year)*	Application No.
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Applicant
Jacobs Nederland B.V.

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 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 application
- ☒ Box No. VIII Certain observations on the application

	Examiner
	de Cauwer, Robby

Application number

WRITTEN OPINION

NL2017533

Box No. I Basis of this opinion

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application and necessary to the claimed invention, this opinion has been established on the basis of:

   a. type of material:

   ☐ a sequence listing

   ☐ table(s) related to the sequence listing

   b. format of material:

   ☐ on paper

   ☐ in electronic form

   c. time of filing/furnishing:

   ☐ contained in the application as filed.

   ☐ filed together with the application in electronic form.

   ☐ furnished subsequently for the purposes of search.

3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty	Yes:	Claims	
	No:	Claims	1-15
Inventive step	Yes:	Claims	
	No:	Claims	1-15
Industrial applicability	Yes:	Claims	1-15
	No:	Claims	

2. Citations and explanations

see separate sheet

WRITTEN OPINION

Box No. VIII Certain observations on the application

see separate sheet

Re Item VIII

Certain observations on the application

1 The claims are not clear.

1.1 Claim 1 relates to a catalyst bed comprising two undefined catalyst layers, which can actually be identical (see below). The wordings "catalyst layer", "carrier material" and "active material" are undefined and not suited to delimit the subject-matter of the claims over the prior art. It is further noted that there exists only a very limited amount of support (examples) in the application that could actually demonstrate a technical effect.

1.2 Claim 1 is further rendered unclear since carbon can be at the time support or active material (see claims 4 and 5). This further makes the reference to the loading with active material impossible to verify.

1.3 The reference to "catalyst loading" in claims 1 and 10 does not make sense, since before there is no reference to said "catalyst loading" and what it is supposed to mean. Said term is further rendered unclear in that it does not specify whether said loading is to be considered as an absolute or relative loading. In the latter case it should specify in relation to what said loading should be relative (mass, size, number, volume,...). It is further unclear how it is to be determined (which method), especially in the case where more than one active material might be present.

1.4 Claims 2 and 3 are unclear in that the active material is not defined, it is therefore also unclear to know as what the active material is to measured (element, (specific) oxide, compound) to determine the wt%. Claim 3 does not even specify what the wt% relates to, since the catalyst loading has not been defined. Since the catalyst loadings are not determinable, it is therefore possible that just one layer of catalyst might take away the novelty of the claims.

1.5 Claim 4 is not clear since the feature "other carrier materials that do not catalyze the Claus reaction" is not defined and therefore not suitable to delimit the subject-matter of the claims. Preferential features are just that, preferential, and in no way limiting.

1.6 Claims 6 and 7 are not limiting since it has not been defined whether the catalyst bed is fixed or fluidized. It is further unclear whether the different vol % are corresponding to the difference in catalyst loading in case the catalyst loading is to be interpreted as absolute. It is furthermore unclear how to

interpret said values when considering the subject-matter of claim 10 which states that more catalyst layers might be present, even if the total catalyst bed space has been filled up with the layer 1 and 2.

1.7 Claim 8 is not sufficiently defined in that it does not specify by which method the pore volume should be determined. It is further noted that said value has not been determined for the examples in the application. It is therefore not clear whether the examples fall under the scope of said claim. In any case it has not been demonstrated how said catalyst with said properties has been obtained and no effect has been shown that would correlate with said feature.

1.8 The term "inert" in claim 9 is vague and undefined, it is completely dependent on the reaction conditions, which materials can be considered inert or not.

1.9 The content of the gas stream in claim 11 should be indicated. Said claim does not make sense if no H2S is contained in the gas stream. The claim further refers to "first catalyst layers", whereby the previous claims only refer to on first layer. Furthermore, the claim should include all the features essential to the invention, at present this is merely a contacting step. It is clear to the skilled person that indications of temperatures, concentrations and gas velocities are essential for the method of the invention.

1.10 Claim 14 should indicate at what position in the bed said molar ratio is maintained.

Re Item V

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

2 Furthermore, the above-mentioned lack of clarity notwithstanding, the subject-matter of claims 1-15 is not new, and the criteria of patentability are therefore not met.

D1 (see cited references cited in the search report) discloses a catalyst bed for the selective oxidation of H2S, having 2 or more catalyst layers inherently having a different loading of active material, at least one of the catalyst layers containing iron oxide (5 wt%) on silica filling the 20 vol% of the catalyst bed. D1 further discloses the selective oxidation of H2S with said catalyst bed by contacting a gas stream containing 1,5 vol% H2S and 0,6 vol% SO2, 30 % H2O and 1,3 vol% CO at 230°C with the bed. D1 further discloses that said catalytic bed for the selective oxidation of H2S should be located downstream of a Claus installation.

It should be further observed that it is at present not clear what technical effect has been demonstrated by the examples. No technical effect can be attributed to the use of a multi-layered bed since the single layered beds of ref. example 2 and 3 attain much better values at comparable temperatures.