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# A comparison between Claus and THIOPAQ sulfur recovery techniques in natural gas plants

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## Abstract

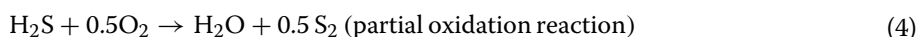
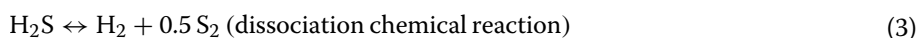
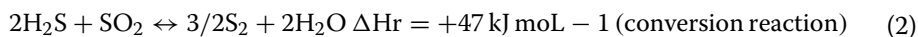
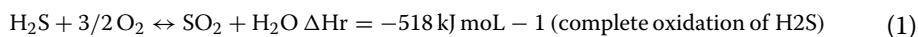
A sulfur recovery process is one of the most important processes in the oil and gas industry to get rid of hydrogen sulfide ( $H_2S$ ) which is produced from the acid gas removal process of sour natural gas to convert it into sweet natural gas. Actual data from a gas field is used to obtain a realistic comparison between two sulfur recovery techniques, through which researchers and/or manufacturers can obtain information that will help them choose the most appropriate and cheapest method. A total feed acid gas flow rate of 5.1844 MMSCFD with an  $H_2S$  concentration of 24.62% by mole percent was produced from amine acid gas removal units. Claus sulfur recovery technique is a traditional chemical process that uses thermal and catalytic reactors. Therefore, an acid gas enrichment unit is applied to increase the  $H_2S$  concentration to approximately 50% mole to provide reliable and flexible operation in the thermal and catalytic reactors. Moreover, a tail gas treatment unit is applied to increase the overall conversion efficiency to 99.90% with the Claus technique instead of 95.08% without it to achieve high sulfur recovery and reliable operation through the conversion of carbonyl sulfide (COS) and mercaptans. Studies on the safety and simplicity of the Claus technique revealed many important hazards and a large number of transmitters (379) and control loops (128) in one Claus train. THIOPAQ sulfur recovery as a new technology is a biological desulfurization process that uses a natural mixture of sulfide-oxidizing bacteria. It is also a unique  $H_2S$  removal process with an efficiency of 99.999%. In addition, studies on the safety and simplicity of the THIOPAQ technique have shown that the hazards, the number of transmitters (74), and the number of control loops (29) of a one THIOPAQ train are lower. The THIOPAQ technique showed higher efficiency, was safer, simpler, and had lower CAPEX and OPEX. This study was conducted using Aspen HYSYS V11 and actual data.

**Keywords:** Sulfur recovery, Claus, THIOPAQ, Sulfide-oxidizing bacteria, Desulfurization of sour natural gas

## Introduction

Acid gas removal technologies used in oil and gas fields only remove the acid gas from the sour natural gas to convert it into sweet gas, such as amine acid gas removal systems, and membrane acid gas removal systems [1–5]. However, the acid gas is still in the system and therefore needs to be removed. Work has been done on the design and development of sulfur recovery plants to get rid of the acid gas ( $H_2S$ ) by converting the  $H_2S$  into

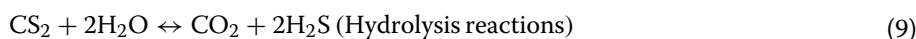
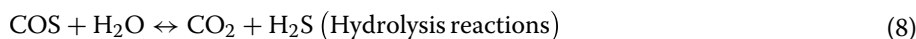
elemental sulfur, which can be used as a useful product in various fields. In general, Bio-sulfur and Claus sulfur are widely used in industries such as pharmaceuticals, fertilizers, and fungicides [6–8]. In this paper, a study is conducted between two technologies, one traditional and one newly developed, to determine the new developments that have been added and their effectiveness compared to old traditional methods under certain conditions. The study is also considered as a guide for researchers under different conditions. The oldest and best known technique for sulfur recovery is the Claus technique, which is mainly based on the combustion of  $\text{H}_2\text{S}$  with air to form sulfur and water vapor [6, 8–10]. There are other techniques for sulfur recovery, such as the THIOPAQ technique, which depends on sulfide-oxidizing bacteria that are considered the main catalyst for the conversion of  $\text{H}_2\text{S}$  to sulfur (bio-sulfur) in the presence of air (without combustion) [4, 5, 9, 11, 12]. It is therefore necessary to make a comparison between these techniques in order to determine which is better and cheaper, and to determine the proportional conditions for each of them. The Claus technique as a traditional process is a chemical process that mainly involves the conversion of  $\text{H}_2\text{S}$  into S in two stages. The first stage of the process is a thermal stage in which 1/3 of the  $\text{H}_2\text{S}$  is burned with air (by a controlled ratio of air to acid gas) to convert it to sulfur dioxide ( $\text{SO}_2$ ), which then reacts with the other 2/3 of the  $\text{H}_2\text{S}$  (unburned) and /or dissociation and partial oxidation reactions among the following main reactions at 1100 °C in the furnace to convert  $\text{H}_2\text{S}$  to sulfur:



There are also side reactions in the thermal stage that lead to the formation of COS and  $\text{CS}_2$  based on the concentration of  $\text{CO}_2$  and hydrocarbons in the acid gas:



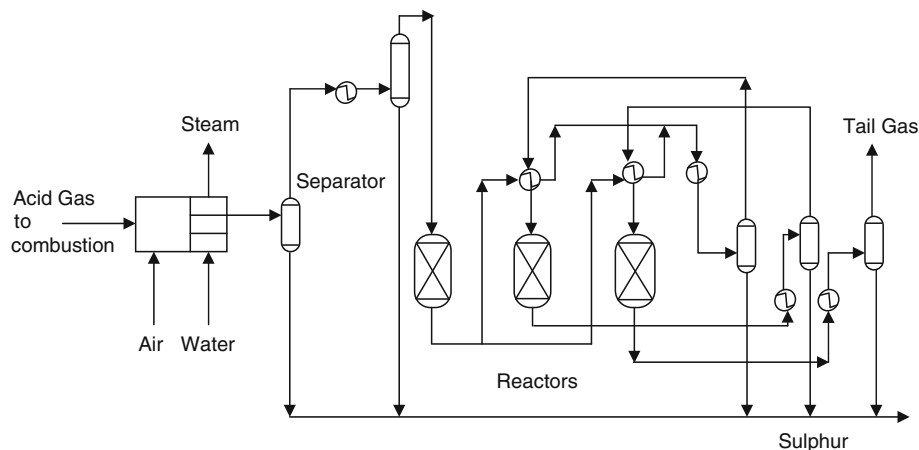
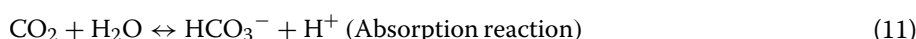
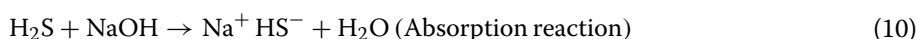
The conversion efficiency of the thermal stage reaches about 67%. The gas leaving the furnace cooled in a waste heat boiler and condenser to separate the elemental sulfur ( $\text{S}_2$ ) formed in the thermal reactor [13–18]. The second stage is the catalytic stage, which consists of two reactors. The first reactor mainly used for hydrolysis of thermal stage by-products (COS and  $\text{CS}_2$ ) into  $\text{H}_2\text{S}$  (90% titanium catalyst and 10% alumina catalyst), and the second reactor mainly used for Claus reaction (alumina catalyst). The residual gas from the thermal stage is reheated using a bypass hotline stream from the waste heat boiler (before the condenser) for purpose of catalytic activation and the flow enters the first catalytic reactor where COS and  $\text{CS}_2$  are converted into  $\text{H}_2\text{S}$  and then enters the second catalytic reactor where the sulfur formed. Hydrolysis and Claus reactions occur at temperatures around 240 °C to 320 °C [17, 19–21]:



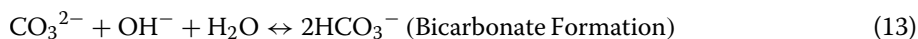
To achieve a more complete conversion of  $\text{H}_2\text{S}$ , multiple catalytic reactors are provided. There is a condenser behind each reactor to condense and separate the formed sulfur vapor, as shown in Fig. 1. The conversion efficiencies of the two stages (thermal and catalytic) range from 93 to 95% for two catalytic reactors and up to 97% for three catalytic reactors [17–19]. The exhaust gases are discharged to the combustion system or delivered to the tail gas treatment system. Tail gas treatment unit can increase the efficiency to 99.5–99.9% [17–19], but the cost of tail gas treatment is high, usually twice the cost of a Claus unit [17, 22].

The THIOPAQ O&G process that represents a cost-effective alternative for the traditional Claus process, which is a biochemical process that mainly depends (for converting  $\text{H}_2\text{S}$  to S) on a mixed population of haloalkaliphilic sulfide oxidizing bacteria (microorganisms' catalyst) which oxidize the  $\text{H}_2\text{S}$  biologically to bio-sulfur [4, 9, 11, 24]. Biological desulfurization (THIOPAQ) originally marketed by Paques to be used solely for the treatment of biogas and the first unit for biogas desulfurization was built in 1991–1993 [25–28]. Biological desulfurization is also used at high and low pressure in the natural gas process and oil refinery so it is known as THIOPAQ O&G. In addition, the first commercial THIOPAQ O&G unit was built to treat natural gas in Canada in 2002 [9, 11, 25].

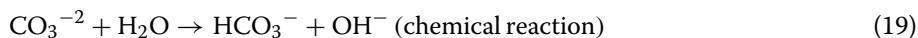
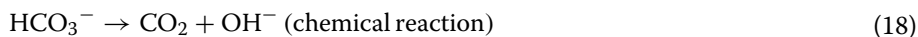
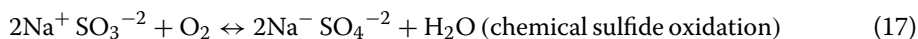
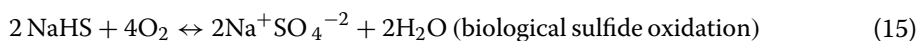
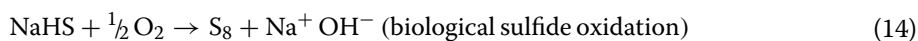
Generally, the THIOPAQ consists of three main sections. Absorption, biological reactions and sulfur separation sections [4, 9, 11, 26–29]. First section (absorption section): In this section, the acidic feed gas first comes into contact with the lean solution (alkaline solution) in the absorber. This solution absorbs  $\text{H}_2\text{S}$  and some  $\text{CO}_2$ , and forms sodium sulfide, carbonate and bicarbonate (rich solution) by the following reaction:



**Fig. 1** Schematic flow diagram of the basic Claus process [23]



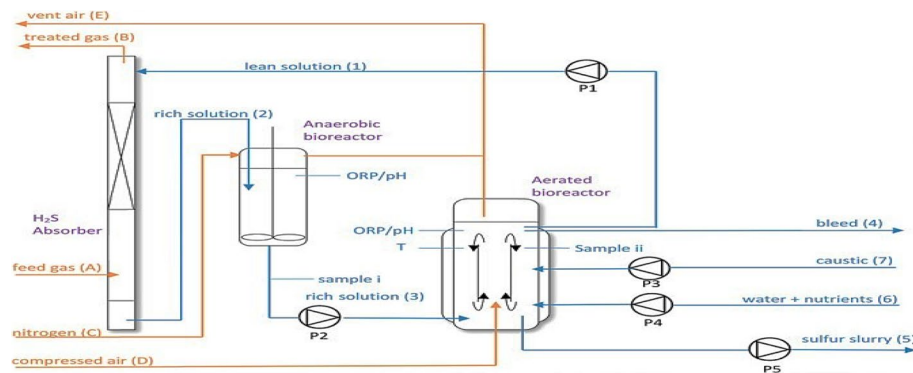
Sweet gas ( $\text{CO}_2$ , HC gas) exits from the absorption tower. The rich solution must be fed directly into the flash vessel to reduce the high pressure if found and flush out any hydrocarbon gases soluble in the solution. In the second section (bio-reaction section): The rich solution flows into a flash tank or directly into a bioreactor operating at atmospheric pressure and ambient temperature. This contains microorganisms (sulfide-oxidizing bacteria) that oxidize sulfide ( $\text{HS}^-$ ) to elemental sulfur, resulting in the rich solution into a lean solution. Sulfate and Thiosulfate Formation (by-products) occur. Carbonate and bicarbonate decomposition, resulting in the release of  $\text{CO}_2$  into the air through the vent line:



The bio-reaction section was developed to increase the selectivity of sulfur production. In the old version of THIOPAQ there is only one aerated bioreactor with a sulfur production selectivity of 75.6%, so in the new version of THIOPAQ two bioreactors, one anaerobic bioreactor (first) and one aerated bioreactor (second), increase the selectivity of sulfur production to 96.6%, thus reducing by-products and increasing the purity of the sulfur produced [11].

In the third section (sulfur separation section): the solution recirculated over the absorber and a part of the solution is routed to a decanter centrifuge where the elemental sulfur granules are separated from solution and the recovered water is recycled back to the process via the bioreactor. Necessarily, a small slipstream of the solvent is bled from the system to prevent an accumulation of salts (as side products). Figure 2 is a schematic flow diagram of the THIOPAQ Process.

THIOPAQ O&G acts as a mixture of acid gas removal and sulfur recovery units when  $\text{CO}_2$  removal is not required. If  $\text{CO}_2$  removal is needed, treating the amine unit off gas as a single THIOPAQ unit may work [29]. THIOPAQ could be a good alternative for removing only up to 100 tons of sulfur/day [9, 29].  $\text{H}_2\text{S}$  removal efficiency reaches 99.99%. However, the products of this process include by-products such as unwanted biomass products and salts (sulfates and thiosulfates), which are sent for disposal. The



**Fig. 2** Schematic flow diagram of the THIOPAQ Process [11]

**Table 1** Feed condition & composition

Total Feed gas flow (for 2 trains)	5.1824	MMSCFD
Feed gas flow (for 1 train)	2.592	MMSCFD
Pressure	1.1	Barg
Temperature	40	°C
Methane	0.0199	Mole fractions
Ethane	0.0002	Mole fractions
i-Pentane	0.0013	Mole fractions
H <sub>2</sub> S	0.2462	Mole fractions
CO <sub>2</sub>	0.6137	Mole fractions
H <sub>2</sub> O	0.1187	Mole fractions

selectivity of sulfur production mainly depends on the bacterial species, whether it functions in both anaerobic and aerobic processes (main species: *Thioalkalivibrio sulfidiphilus*) [30–32]. Furthermore, the selectivity depends on the oxygen concentration of the air supplied to the bioreactor [33, 34]. The final sulfur purity is approximately 95–98% by weight [4, 24, 27, 28].

This paper covers the following studies: process flow diagrams, simulations, operating conditions, feed flow rates, simplicity, safety, and economics of Claus and THIOPAQ processes, and the importance and usage of bio-sulfur.

## Methods

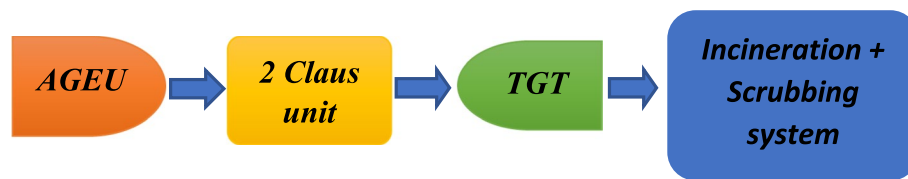
A current natural gas plant was selected as a case study to compare the Claus and THIOPAQ methods (see Table 1).

### Claus process

#### Design of one Claus train by Aspen HYSYS simulation and discussion

The origin of the feed gas is amine acid gas and its flow represents the flow of one sulfur recovery train, whereas there are two sulfur recovery trains in the plant with a total feed gas flow of 5.1824 MMSCFD. So, one train has been studied.

Claus sulfur recovery train consists of an acid gas enrichment unit (AGEU), two Claus (thermal & catalytic reactors) units, a tail gas treatment unit, and incineration section & scrubbing system (Fig. 3).



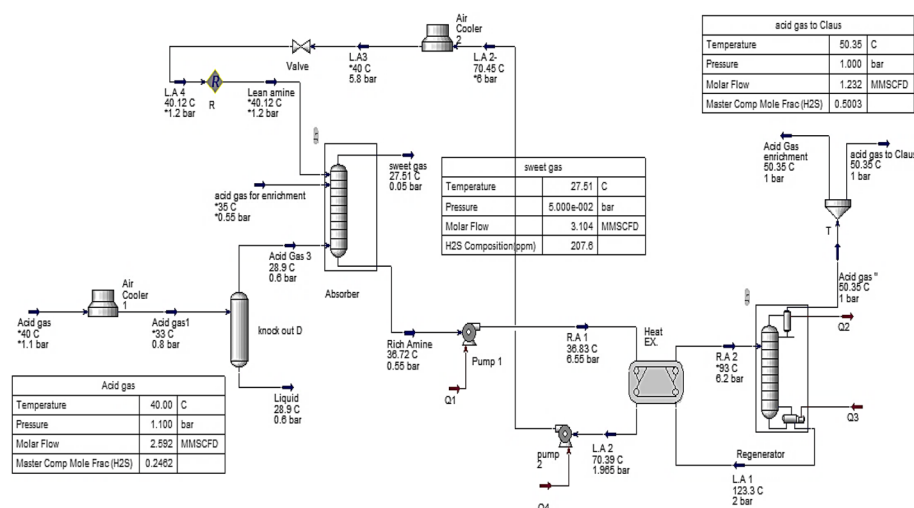
**Fig. 3** Block diagram of the main sections of Claus process

**Table 2** Products & some results (using Aspen HYSYS)

Parameters	One SRU train (half)	Two SRU trains (Total)	Unit
Total Acid gas from AGEU	1.232	2.464	MMSCFD
Acid gas to one Claus	0.616	-----	MMSCFD
H <sub>2</sub> S concentration to Claus	50.03	-----	Mole %
Temperature inside Thermal Reactor	1081	-----	°C
Sulfur produced	22.464	44.928	ton/day
Sulfur purity	99.98	-----	Mole %
Flue gas to incinerator	3.104	6.208	MMSCFD
H <sub>2</sub> S in flue gas	207.6	-----	ppm
Claus Section efficiency	95.08	-----	%
Amount of efficiency increased by TGTU	4.82	-----	%
Total H <sub>2</sub> S Removal Efficiency	99.90	-----	%

**Process description:** In this case study, the sour gas produced from the AGR unit enters the AGEU to absorb H<sub>2</sub>S and increase the H<sub>2</sub>S content in the acid gas for the Claus section. The acid gas is fed into the cooler to condense liquid droplets (sour water and hydrocarbon) as foaming can occur in the amine system. Three feeds enter the absorber: Amine acid gas, a slipstream from the reflux drum and a stream from the TGTU to increase the H<sub>2</sub>S concentration, and the lean amine that absorbs the H<sub>2</sub>S. Two products from the absorber: sweet gas (CO<sub>2</sub>, HC gases) sent to the incinerator, and rich amine (lean amine after H<sub>2</sub>S absorption). The rich amine is preheated in the heat exchanger with the hot lean amine from the regenerator. After preheating, the rich amine is fed into the regenerator to remove H<sub>2</sub>S and convert the rich amine to lean amine, which is sent back into the absorber. The acid gas leaving the regenerator is cooled to separate any liquid before being fed into the thermal reactor. The H<sub>2</sub>S concentration was increased to 50.03% mole percent, as shown in Table 2. Figure 4 illustrates the flowchart of the simulation process of AGEU.

After the H<sub>2</sub>S content has risen, the acid gas is distributed to two Claus units to produce sulfur and water vapor. The TGTU is needed to increase the overall H<sub>2</sub>S removal efficiency to about 99.9% (using HYSYS), increase the sulfur product, and meet environmental regulations. In addition, the TGTU converts all sulfur compounds in the process gas from the Claus section that cannot be converted to sulfur, such as SO<sub>2</sub>, CO, COS, CS<sub>2</sub>, and S<sub>x</sub> to H<sub>2</sub>S and then passes them back to the AGEU. Two feed into the Claus train: The acid gas from the AGEU and the air stream enters an electric heater to increase the temperature for thermal stability reasons, as the H<sub>2</sub>S concentration is less than 60% [35]. After preheating, the acid gas and air enter the thermal reactor, where the H<sub>2</sub>S reacts with the oxygen in the supplied air to form sulfur and water vapor at a



**Fig. 4** Process flow diagram of AGEU

high temperature of 1081 °C. The flue gases are sent to a waste heat recovery unit. It is then fed into a waste heat boiler to lower the temperature and generate steam, which is then fed into a condenser to condense the sulfur and water vapor. The flue gases from the thermal part contain the remaining H<sub>2</sub>S and SO<sub>2</sub> that have not been reacted and the sulfur compounds, which are preheated by a slipstream from the WHB to raise the temperature for catalyst activation reasons and eliminate sulfur condensation, then enter the first catalytic reactor where the hydrolysis (main reaction) and Claus reactions take place, and then into a condenser to condense the resulting sulfur and water vapor. The flue gases from the condenser were only slightly preheated (higher than the sulfur dew point) to eliminate the sulfur condensation. They then enter the second catalytic reactor, where the Claus reaction (main reaction) and the hydrolysis reactions take place. The TGTU consists of an inline heater (sub-stoichiometric burner) to generate reduction gases (H<sub>2</sub>, H<sub>2</sub>O) and to preheat the feed from the Claus section, the hydrogenation reactor, the cooling stage, and the recycling facilities. The hydrogenation reactor contains a cobalt-molybdenum-based catalyst that performs both the hydrogenation and hydrolysis functions [36]:

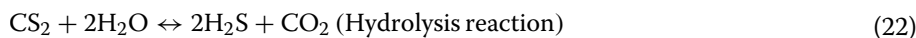
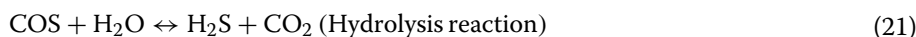
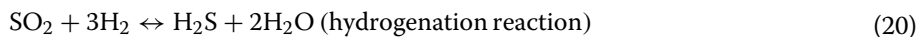
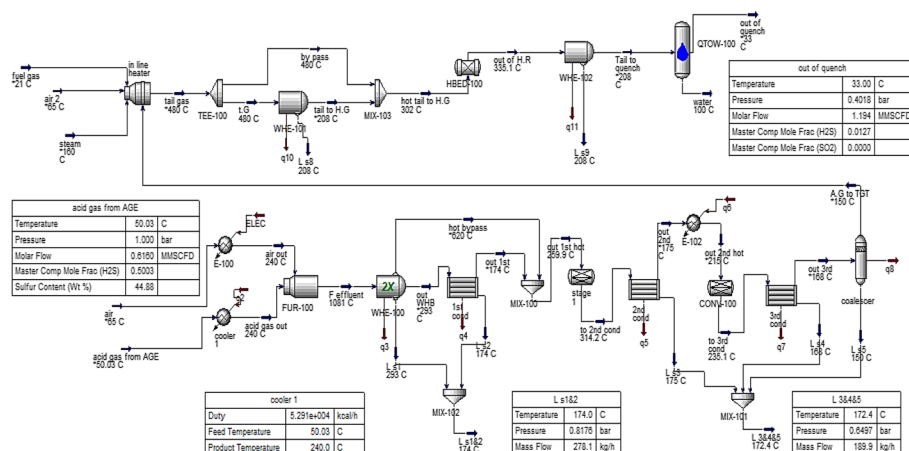


Figure 5 illustrates the sulfur production operation (one Claus unit) and the TGTU simulation using Aspen HYSYS. Table 2 show the results and the operating parameters of the Claus unit and the TGTU.

### Results and discussion

In the AGE section because of the Knowhow of the proprietary additives and composition of the commercial lean amine, more trial and error of the lean amine flow rate





**Fig. 5** Process flow diagram of one Claus unit & TGT unit

**Table 3** Actual field data

Average daily Acid gas flow rate to AGEU	Average daily sulfur (Outlet) production rate	Average H <sub>2</sub> S concentration	Inlet sulfur load in the feed acid gas (using Aspen HYSYS)	The total Conversion Efficiency (%)
2,344 MMSCFD	21.49 Ton/D.	24.4 mol%.	21.943 Ton/D.	97.93%.

(73,000 kg/hr.) and its compositions ( $\text{H}_2\text{O}$  – MDE Amine – ME Amine) conducted to reach the minimum  $\text{H}_2\text{S}$  content (207.6 ppm) in the sweet gas exit from the absorber to incinerator. Figure 4 shows the process flow diagram of AGEU using Aspen HYSYS, the increase of  $\text{H}_2\text{S}$  concentration to Claus, and the optimum operating conditions.

To achieve a maximum sulfur recovery efficiency of 68.3% (with HYSYS) in the thermal reactor, the air and acid gas inlet temperatures are 240 °C. Two catalytic reactors with titanium and alumina catalysts and inlet temperatures of 215–260 °C are used to achieve a maximum sulfur recovery efficiency of 26.78% of the total efficiency. The total sulfur recovery efficiency (thermal + catalyst) is 95.08%. The sulfur produced from one SRU train (2 Claus units) is 2\*468 kg/h. (22,464 tons/day). TGTU increases the overall H<sub>2</sub>S removal efficiency to 99.9%.

**Actual field data along 1 year of observation**

Table 3 shows the data obtained from the field to compare it with the results from Aspen HYSYS simulations.

The inlet sulfur load in the feed acid gas(using Aspen HYSYS) = (sulfur content Wt%/100)

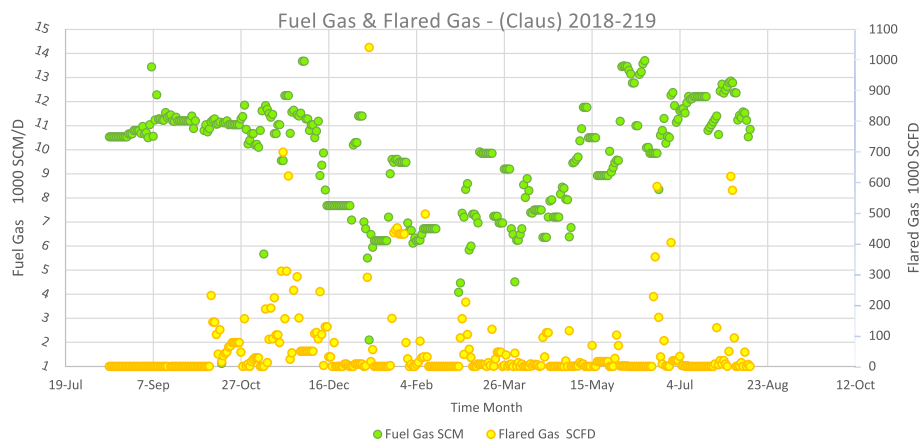
\*feed mass flow rate (kg/hr) =  $(21.14/100) * 4471 * 24 = 21.943$  Ton/D.

The total conversion efficiency (%) = (outlet sulfur produced/ inlet sulfur load) \* 100

$= (21.49/21.943) * 100 = 97.93\%$ . (it is a large difference with the theoretical efficiency from the simulation)

Figure 6 illustrates field fuel gas consumption flow rate and flared gas along 1 year and shows the average required fuel gas throughput = 9526.34674 SCM/D (increases operating costs), and the average throughput of flared gas = 50,119.8034 SCF/D (flared gas is the acid gas that is sent from the Claus plant to the flare when malfunctions occur,





**Fig. 6** Actual fuel gas daily consumption and flared gas to the incinerator

which means that many malfunctions occur). Before the flue gases from the incinerator are released into the atmosphere, they are normally sent to the  $\text{SO}_2$  scrubber using the exhaust fans to further reduce the level of sulfur oxides in the flue gases to the level required by environmental regulations.

Other equipment exists downstream two trains:

Sulfur degassing and storage section: The principle of the degassing process of the liquid sulfur is to release the dissolved  $\text{H}_2\text{S}$  gas and to accelerate the decomposition of the polysulfides to  $\text{H}_2\text{S}$  then send to the thermal reactor.

Utilities: (Steam / Condensate / Blowdown).

Chilling Unit: a dedicated demineralized water-cooling medium system (chilled water) required for quench water coolers and lean amine coolers.

Solidification Unit: to convert the liquid sulfur from the storage section to solid sulfur and form sulfur pills.

#### **Claus operation simplicity study**

- 1- It contains a large number of transmitters and many complex control loops, which are shown in Table 4. This makes the operation of Claus more difficult for the operator to follow and increases CAPEX and OPEX (maintenance).
- 2- The problem is that liquid sulfur turns into solid sulfur at low temperatures, causing blockages in process lines. Therefore, the steam jacket covers all liquid sulfur process lines. If a steam leak occurs or the steam stops for some reason, the system will shut down.
- 3- Since the panel station contains a large number of phase plates, it is difficult to focus on all these parameters and the changes that occur (requiring multiple panel operators).
- 4- There are many startup sequences in which three units would restart to enter the operating mode. Table 5 shows two startup modes, one when the system is hot after a sudden unit shutdown and one when the system is cold after a scheduled shutdown.

**Table 4** Actual field numbers of transmitters, control loops and shutdown valves in one Claus train

Equipment	No. of Transmitters	No. of control loops	No. of SDVs
AGE K.O Drum	11	3	1
Absorber	16	2	----
regenerator	15	4	----
Reflux Drum	17	7	----
Lean Amine Loop	13	3	----
Filtration	6	1	----
Analyzers	2	----	----
Acid Gas Separator	2 × 20	2 × 5	2 × 2
Thermal reactor	2 × 56	2 × 25	2 × 5
WHB	2 × 9	2 × 3	-----
Catalytic Reactor	2 × 16	2 × 3	-----
TGT interconnecting	12	8	4
Sub-stoichiometric in line Heater	11	3	1
TGT WHB	6	2	1
Hydrogenation Reactor	11	3	----
Quench tower	17	8	1
incinerator	23	14	8
Air Blowers	5	4	1
Analyzers	12	-----	-----
<b>Total</b>	<b>379</b>	<b>128</b>	<b>31</b>

**Table 5** Time required for the startup of one Claus train

Unit	Startup (Hot Mode) time required	Startup (Cold Mode) time required
AGE	1 h. to ramp up the temperature in the regenerator from 89 °C to 126 °C.	3.5 h. to ramp up the temperature in the regenerator from 27 °C to 126 °C.
Claus	The temperature of the thermal reactor is increased from 905 °C to 1150 °C in almost 2 h, then starts to enter acid gas.	The temperature of the thermal reactor is increased by fuel gas from 30 °C to 1150 °C in almost 12 h. before acid gas enters.
Incinerator	It takes almost 2 h. to ramp up the temperature from 400 °C to 650 °C.	It takes almost 7 h. to ramp up the temperature from 30 °C to 650 °C.
TGT	The temperature is increased from 340 °C to 420 °C and the H <sub>2</sub> concentration in the loop reached > 3% mole almost in 1 h.	The temperature is increased from 27 °C to 420 °C and the H <sub>2</sub> concentration in the loop reached > 3% mole almost in 6 h.

This means that a hot mode startup will take almost 2 h and a cold mode startup will take 12 h.

- 5- Approximately four operators are required on site for operation and startup, and experienced panel operators (at least two) are required to meet these parameters.

### Safety study

This section describes the most apparent risks in operation and maintenance present in the Claus process. Table 6 lists the most common risks in the process and the equipment that causes these risks. It is found that there is no high pressure in all units in the Claus process, but there are high temperatures and free H<sub>2</sub>S in most equipment in all units.

**Table 6** Most common hazards and risks in Claus process

Unit	Hazard/Risk	Equipment
AGE	High pressure	No high pressure.
	High temperature	1- Amine regenerator. 2- Heat exchanger.
	Free H <sub>2</sub> S	Free H <sub>2</sub> S gas is included in all unit equipment.
Claus	High pressure	No high pressure.
	High temperature	1- Pre-Heater. 2- Thermal reactor. 3- Waste heat boiler. 4- Catalytic reactor. 5- Condenser. 6- Seal legs. 7- Steam loop.
	Free H <sub>2</sub> S	Free H <sub>2</sub> S gas is included in all unit equipment.
TGT	High pressure	No high pressure.
	High temperature	1- Tail gas coalesce. 2- In-line heater. 3- TGT WHB. 4- Hydrogenation reactor.
	Free H <sub>2</sub> S	Free H <sub>2</sub> S gas is included in all unit equipment.
Incineration Section	High pressure	No high pressure.
	High Temperature	Incinerator.
	Free H <sub>2</sub> S	Incinerator.

**Table 7** Capital cost and operating cost calculated by Aspen HYSYS

Summary	
Total Capital Cost [USD]	2,188,000
Total Operating Cost [USD/Year]	1,003,860
Total Raw Materials Cost [USD/Year]	0
Total Product Sales [USD/Year]	0
Total Utilities Cost [USD/Year]	64,077.7
Desired Rate of Return [Percent/Year]	20
P.O.Period [Year]	0
Equipment Cost [USD]	120,600
Total Installed Cost [USD]	323,100

This means that most equipment will have free H<sub>2</sub>S and high temperature.

### **Economic study**

Using Aspen HYSYS to calculate the operating cost and capital cost (for one train):

- 1- AGE Unit capital cost and operating cost is showed in Table 7:
- 2- Claus unit, TGT unit, and incinerator:

Aspen HYSYS doesn't support the capital cost and operating cost of the Claus sulfur recovery unit.

**Table 8** The vendor capital and operating costs

H <sub>2</sub> S treated gas from AGE Absorber ppm		H <sub>2</sub> S & SO <sub>2</sub> in Incinerator stack flue gas in case of SO <sub>2</sub> scrubber package not in operation.		H <sub>2</sub> S & SO <sub>2</sub> in Incinerator stack flue gas in case of SO <sub>2</sub> scrubber package in operation	
< 70 ppmV		< 4 ppmV & < 130 ppmV		< 4 ppmV & < 55 ppmV	
Sulfur load total (ton/D)	No. of trains	Sulfur load per train (ton/D)	Capex M€ Including (AGEU, 1 <sup>st</sup> Claus, 2 <sup>nd</sup> Claus, TGT, Incinerator, installed cost).	Opex (365 DOS) M€	Manhour's cost (per year) K€
50	2	25	98 (49 M€ for one train)	4.2	150

**Table 9** The operating cost analysis

Type	Consumption rate	Annual consumption
Electric Power	1.8 MW/h	15,120 MW
Fuel Gas	510 kg/h	4,284,000 kg
Boiler Feed Water	13 m <sup>3</sup> /h	109,200 M <sup>3</sup>
MP Steam	6.1 t/h	51,240 ton
LP Steam	-1.3 t/h	-10,920 ton
20% w/w Caustic Solution	50 l/h	420 M <sup>3</sup>
Activated alumina Cat. (H <sub>2</sub> Reactor)	1.2 m <sup>3</sup>	Vendor to guarantee a lifetime of 4 years.
Activated Carbon	3.5 m <sup>3</sup>	Vendor to guarantee a lifetime of 2 years.
Activated Alumina catalyst	5.2 m <sup>3</sup>	Vendor to guarantee a lifetime of 4 years.
Titania catalyst	4.2 m <sup>3</sup>	Vendor to guarantee a lifetime of 4 years.
Hydrogenation Catalyst	3.5 m <sup>3</sup>	Vendor to guarantee a lifetime of 4 years.
Pure MDEA	41,600 kg	It decreases by 10% every 6 months.
Pure antifoam	276 kg	As order.

3- The capital cost and operating cost from the vendor (KT Kinetics Technology S.P.A.) data for two Claus SRU trains, Table 8 shows the vendor data of the H<sub>2</sub>S and SO<sub>2</sub> emissions and the cost required for the Claus process shows that the Capex and Opex for one Claus train 49 million euro and 2.1 million euro.

4- Economic analysis (operating cost parameters):

The common parameters that affect the operating cost calculations in one Claus train including (AGEU, 1<sup>st</sup> Claus, 2<sup>nd</sup> Claus, TGT, Incinerator), showed in Table 9.

### THIOPAQ process

#### *Design of THIOPAQ process by Aspen HYSYS simulation and discussion*

The acid gas composition and properties are the same as shown in Table 1, and the feed gas flow rate from the amine AGRU is 1.296 MMSCFD flowing to one THIOPAQ sulfur recovery train for early production facility. Moreover, this unit will be used as a case study, and to get a comparison, four trains of THIOPAQ (as an alternative cost-effective option) will be equivalent to two Claus trains.

THIOPAQ sulfur recovery train consists of absorption section, bio-reaction section and sulfur separation section showed in Fig. 7.

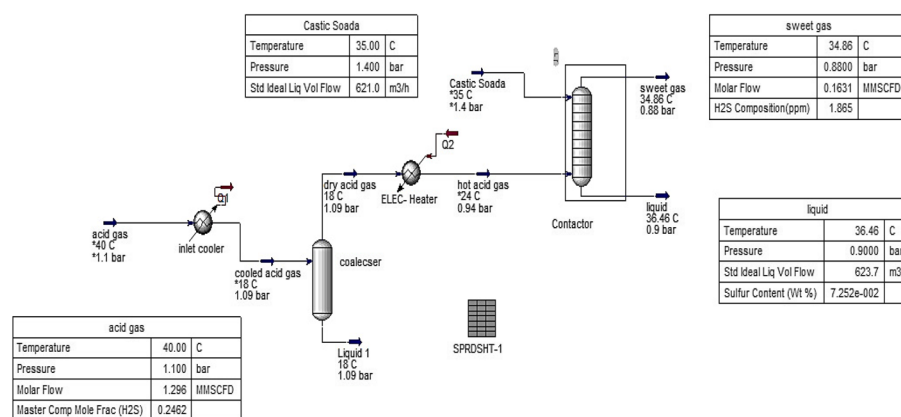


**Fig. 7** Block diagram of the main sections of THIOPAQ

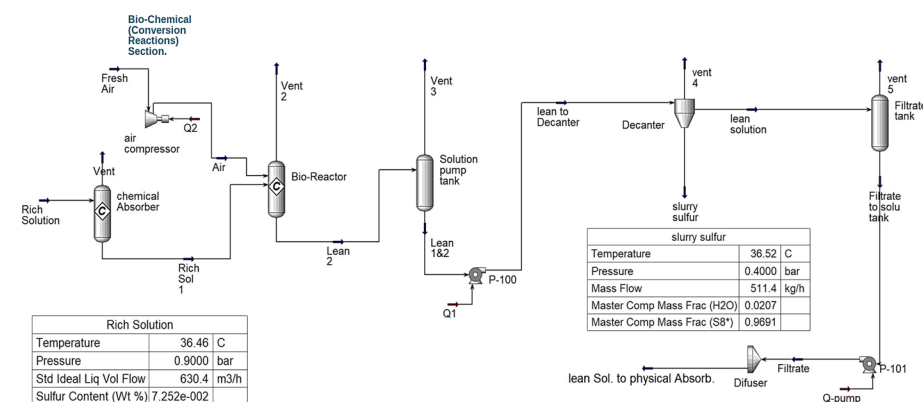
**Process Description:** After the acid gas is produced from AGRU directly enters THI-OPAQ, it first enters the cooler to condense any liquids in the feed gas. These liquids would be collected in a coalescer. The dry acid gas is then heated with an electric heater to remove condensation of hydrocarbon gas in the system for foaming purposes. The acid gas enters at the bottom of the absorber and flows countercurrently with the lean solution from the top of the absorber, absorbing  $H_2S$  and some  $CO_2$ . After  $H_2S$  absorption, a rich solution comes out from the bottom. Also, sweet gases ( $HC$ ,  $CO_2$ ) come out from the top. It is then sent to a thermal oxidizer (burner), where the sweet gas is combusted before being released into the atmosphere. The rich solution was sent to a bioreactor for biochemical reactions that converted the  $H_2S$  to elemental sulfur and sulfate (byproducts) and chemical reactions that produced the byproducts thiosulfate and sulfate. The rich solution regenerated into lean solution in the bioreactor, where sulfur-oxidizing bacteria convert ( $HS^-$ ) to biosulfur ( $S$ ), as shown in Figs. 8 and 9. Aspen HYSYS is used to simulate the THIOPAQ sulfur recovery process with a feed flow rate of 1.296 MMSCFD, and HYSYS has (Acid gas-caustic wash) fluid package that supports the physical reactions of acid gas with an alkaline solution (caustic wash).

So, the process theoretically will be divided into two parts: THIOPAQ physical absorption with the acid gas-caustic wash fluid package for physical reactions, and THIOPAQ chemical absorption with the Peng-Robinson fluid package for biochemical and chemical reactions, as shown in Figs. 8 and 9.

After the acid gas is physically absorbed in the alkaline solution with an  $H_2S$  removal efficiency of 99.999% (using Aspen HYSYS), the  $H_2S$  content in the sweet gas from the absorber is 1.865 ppm. Chemical reactions occur in the same contactor (the absorber), but we use a conversion reactor as a chemical absorber to simulate the chemical reactions inside



**Fig. 8** Absorption section (physical reactions) simulation



**Fig. 9** Chemical absorption section, bio-reactor and sulfur separation section simulation

the absorber. The rich solution flows to the bioreactor, where the bio-sulfur is formed, and the solution becomes a lean solution, as shown in Fig. 9. The lean solution and the bio-sulfur slurry are sent to the solution pump tank (settler) to allow the bio-sulfur grains to be collected with each other; the sulfur slurry is separated from the lean solution through a sulfur decanting centrifuge separator. Aspen HYSYS doesn't support some components and compounds, so we use hypothetical solid components and compounds (an additional option in HYSYS where there is a component list in which there are three options: pure components, hypothetical, and hypothetical solids). A hypothetical solid is selected, then a new solid is added and identified by its molecular weight and density. These compounds are NaHS\*, Na<sub>2</sub>CO<sub>3</sub>\*, NaHCO<sub>3</sub>\*, Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>\*, Na<sub>2</sub>SO<sub>4</sub>\*, and S<sub>8</sub>\*. Figure 9 illustrates the flow diagram of the chemical absorption in the absorber, sulfur conversion in the bioreactor, and sulfur separation section.

### Results and discussion

The purity of the produced sulfur (S<sub>8</sub>\*) is selected 96.91% [4, 24, 27, 28] and the side products are Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>\*, H<sub>2</sub>O, NaOH, Na<sub>2</sub>SO<sub>4</sub>\*.

Table 10 shows the operating parameters where the THIOPAQ process is operating under an atmospheric pressure and temperature of 36.66 °C, which supports low risks and low capital and operating costs. Also, the H<sub>2</sub>S emissions in the sweet gas of just 1.865 ppm

**Table 10** Products & some results (using Aspen HYSYS)

Parameters	One THIOPAQ train (half)	Two THIOPAQ trains (total)	Unit
Feed acid gas flow rate	2.592	5.182	MMSCFD
Feed gas flow (for half train simulation)	1.296	-----	MMSCFD
Temperature inside bio-Reactor	36.66	-----	°C
Salary sulfur produced	2*11.894 (23.788)	47.577	Ton/day
Sulfur purity	96.91	-----	%
Sweet gas to thermal oxidizer	2*0.1631 (.3262)	.6524	MMSCFD
H <sub>2</sub> S in the sweet gas	1.865	-----	ppm
H <sub>2</sub> S removal efficiency	99.999	-----	%

are a perfect result of H<sub>2</sub>S removal exceeding a great deal in the Claus process, which affects finally the sulfur recovery efficiency to reach 99.999%.

**Actual field data along 1 year of observation**

Table 11 shows the data obtained from the field to compare it with the results from Aspen HYSYS simulations

The Average H<sub>2</sub>S (ppmV) in the sweet gas = 30 ppmV.  
The inlet sulfur load in the feed acid gas (using Aspen HYSYS) = (sulfur content%/100) \* feed mass flow rate = (20.45/100) \* 2420 \* 24 = 11.877 Ton/D.

The field total sulfur conversion efficiency almost same the theoretical efficiency in the simulation 99.999%.

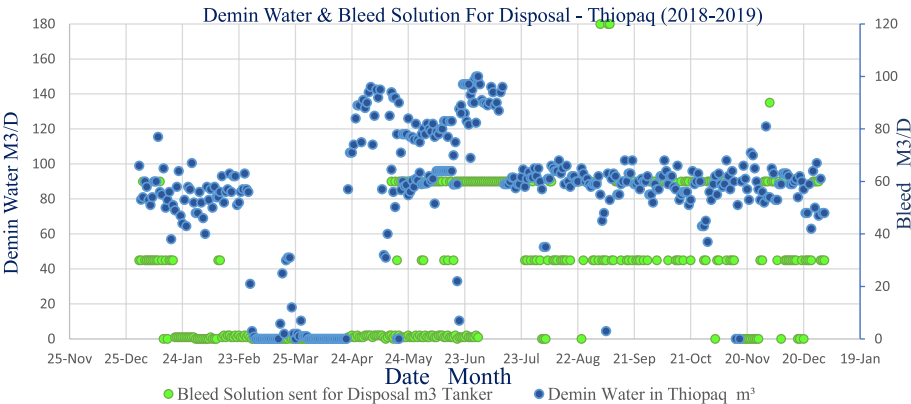
Table 12 shows the data obtained from the field shows the chemicals, demineralized water consumption and the bleed solution sent to disposal.

**Table 11** Actual field data

Average daily acid gas flow rate	Average daily sulfur (outlet) production rate	Average H <sub>2</sub> S concentration
1.269 MMSCFD	11.45 Ton/D	24.4 mol%.
Inlet sulfur load in the feed acid gas (using Aspen HYSYS)	Average bleed solution contains side products (Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> *, H <sub>2</sub> O, NaOH, Na <sub>2</sub> SO <sub>4</sub> *, H <sub>2</sub> O)	The total conversion efficiency (%)
11.877 Ton/D	55 M <sup>3</sup> /D	99.999%

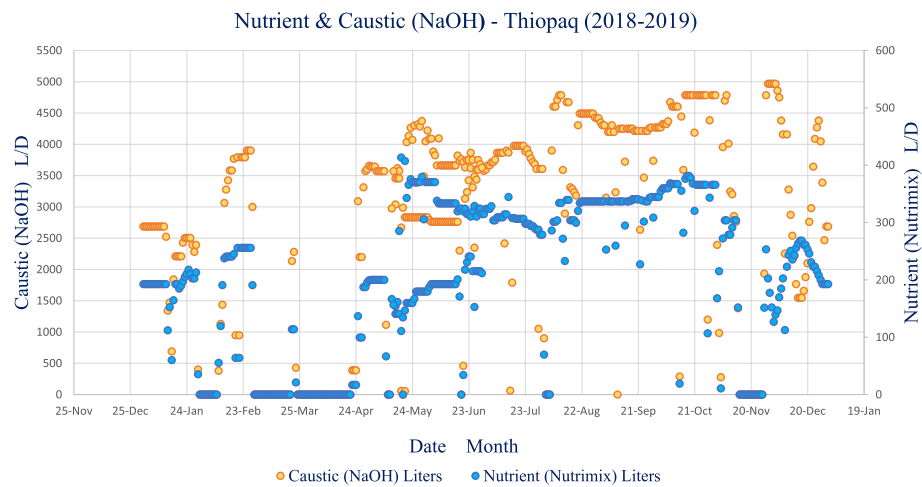
**Table 12** Actual field data over one year

Demineralized water consumed	The average daily flow rate of Demin. water	Total bleed solution
22,978.031 M <sup>3</sup>	76.6 M <sup>3</sup> /day	16,882 M <sup>3</sup> .
Nutrient consumed	Average nutrient flow rate	caustic soda (20%) consumed
89.21M <sup>3</sup>	297.37 L/day	1118.35 M <sup>3</sup>



**Fig. 10** Demineralized water consumed and bleed solution for disposal





**Fig. 11** Nutrient and caustic (NaOH) consumed

**Table 13** Actual field numbers of transmitters, control loops and shutdown valves in one THIOPAQ train

Equipment	No. of Transmitters	No. of control loops	No. of SDVs
Inlet gas cooler, Coalescer, Heater	16	5	5
Contactor, Outlet gas scrubber	10	3	2
Solution pump tank	13	5	1
bio-reactor	6	4	-----
Air compressor	4	3	-----
Decanting centrifuge	4	1	4
Caustic / Nutrient tank	5	2	-----
Thermal oxidizer	7	4	7
Chilled water tank	1	-----	-----
Filtrate tank	1	2	2
Analyzers	7	-----	-----
<b>Total</b>	<b>74</b>	<b>29</b>	<b>21</b>

The following Figs. 10 and 11 illustrate the chemicals, demineralized water consumption rate and the bleed solution sent to disposal, that used as indication for operating cost calculation.

Average Nutrient (Nutrimix) consumption rate = 244.4 L/day. Average Caustic (NaOH) consumption rate = 3064 L/day.

Average Demin Water in THIOPAQ consumption rate = 63 M<sup>3</sup>/day.

The actual data of demineralized water consumed and bleed Solution for disposal is illustrated in the following Fig. 10.

The actual data of nutrient and caustic (NaOH) consumed along 1 year of operation is illustrated in Fig. 11.

#### THIOPAQ operation simplicity study

- 1- It contains a smaller number of transmitters and simple control loops, as shown in Table 13. This makes the THIOPAQ operation simpler to follow up by the operator

**Table 14** Most common hazards and risks in the THIOPAQ process

Unit	Hazard/Risk	Equipment
THIOPAQ	High pressure	No high pressure.
	High temperature	No high temperature.
	Free H <sub>2</sub> S	Only in the absorption section (Inlet gas cooler, Coalescer, Heater, Absorber).

and decreases the CAPEX and OPEX (maintenance). Table 13 shows that the total transmitters are 74, and the control loops are 29 less than the Claus 379 transmitters and 128 control loops.

- 2- The bio-sulfur is found in the system as small solid grains suspended in the solution at ambient temperature that don't show hydrophobic behavior or need a high temperature, which is typical of the chemically produced sulfur from Claus, so it is a stable suspension without clogging or other nuisances.
- 3- The panel station includes a low number of phase plates (about 12 phase plates), so it is easy to focus on all parameters (one panel operator is enough).
- 4- Little startup sequences are required; less than 1 h is required to enter the acid gas into the unit (only three parameters need to be handled: PH, redox (mV), and conductivity (mS/cm)).
- 5- Approximately the number of field operators required for operation and startup is one or two, and only one panel operator is enough to follow all parameters.

### Safety study

In this section, we will also discuss the most important risks that were apparent in the operation and maintenance of the THIOPAQ unit. Table 14 shows the most common risks in the process and the equipment that contains these hazards. Since the THIOPAQ is operated under atmospheric pressure and ambient temperature, the risk of high pressure and temperature does not exist, except if there is a flash vessel, then the risk of high pressure exists only in this vessel. And for free H<sub>2</sub>S, it exists only in the first four vessels. When the H<sub>2</sub>S is still gas, it becomes in the liquid phase when soluble in the alkaline solution in the absorber, where the risk of free H<sub>2</sub>S leakage disappears.

**Table 15** Capital cost and operating cost of physical section calculated by Aspen HYSYS

Summary	
Total Capital Cost [USD]	2,773,020
Total Operating Cost [USD/Year]	1,005,540
Total Raw Materials Cost [USD/Year]	0
Total Product Sales [USD/Year]	0
Total Utilities Cost [USD/Year]	66,944.9
Desired Rate of Return [Percent/Year]	20
P.O.Period [Year]	0
Equipment Cost [USD]	182,100
Total Installed Cost [USD]	652,700

**Table 16** Capital cost and operating cost of chemical, bio-reactor, and sulfur separation section calculated by Aspen HYSYS

Total Capital Cost [USD]	6,759,000
Total Operating Cost [USD/Year]	2,350,990
Total Raw Materials Cost [USD/Year]	0
Total Product Sales [USD/Year]	0
Total Utilities Cost [USD/Year]	489,389
Desired Rate of Return [Percent/Year]	20
P.O.Period [Year]	0
Equipment Cost [USD]	1,909,600
Total Installed Cost [USD]	3,249,000

This means that no high pressure or temperature existed; free H<sub>2</sub>S only existed in four pieces of equipment in the absorption section.

### Economic study

Using Aspen HYSYS to calculate the capital cost and operating cost:

- 1- The first part (physical absorption section) showed in Table 15:
- 2- Second Part (chemical absorption section & bio-reactor & sulfur separation) showed in Table 16:

Tables 15 and 16 shows the capital cost and operating cost for each physical and chemical sections that show the following calculation:

The capital cost (half train) = 9,532,020 USD.

The capital cost (one train) = 2 \* 9,532,020 = 19,064,040 USD.

The total capital cost (two trains) = 38,128,080 USD.

The total operating cost does not match the actual OPEX. This may be because HYSYS does not support all information about this process, as it is new, which appeared when unsupported hypothetical compounds were added to HYSYS, so not all information about the process is available, especially the chemicals used in this process, the products made from it, and their prices. And also, in actuality, there is one section in which physical and chemical reactions occur. All this affects the calculation of the process costs.

- 3- The capital and operating costs from vendor (Pacell) data for the two THIOPAQ O&G trains are shown in Table 17. The capital cost of one THIOPAQ train is 22 million euros for 25 tons per day of sulfur and almost 1.78 million euros in operating costs

**Table 17** The vendor capital and operating costs

Sulfur Load Total (ton/D)	No. of trains	Sulfur Load per train (ton/D)	H <sub>2</sub> S treated gas ppm	Capex& installed cost M€	Opex (365 DOS) M€	Manhour's cost (per year) K€
50	2	25	< 20	44	3.56	48

(including nutrient (Nutrimix), caustic (NaOH), anti-foam agent (TOG), and demineralized water in THIOPAQ). So, it becomes less in capital and operating cost of the Claus train (49 M€ Capex and 2.1 M€ Opex) for the same sulfur production rate.

This means that the Capex was calculated by Aspen HYSYS close from the actual vendor data.

#### 4- Economic analysis (operating cost parameters:

The common parameters that affect the operating cost calculations of one THIOPAQ train, including those shown in Table 18.

#### **Bio-sulfur importance and uses**

Once the bio-sulfur removed from the system, there are several outlets for the sulfur. Conventional options, such as a melting process to obtain 'Claus spec' sulfur or land-fill are available. Solid biological sulfur is considered a nonhazardous refinery waste, but landfilling is an undesirable option, partly because acidification by oxidation has to be prevented. But bio-sulfur is also the basis for a range of new agricultural products designed to act as liquid fertilizers and fungicides. The very small particle size adds to the appeal since it guarantees an even distribution over crops and easy absorption by the plants and soils it is used on [27, 29, 37].

General Valid Advantages of the bio-sulfur:

- 1- Bio-sulfur is very easily converted by microorganisms in the soil and is therefore very quickly available for the plant. This is in contrast to the usual hydrophobic sulfur from other sources [38].
- 2- With a diameter of about 5  $\mu\text{m}$ , it has very low phytotoxicity [27].
- 3- Sulfur is one of the few components that are permitted in organic farming [27].
- 4- The product is an ideal fertilizer for plants that need a lot of sulfur [39].
- 5- Finally, it is worth mentioning that the bio-sulfur can be used to lower the pH in the soil [27].

Figure 12 shows the visible difference between hydrophilic and hydrophobic sulfur; Claus sulfur is hydrophobic, which means that it cannot be soluble in water, but THIO-PAQ sulfur (bio-sulfur) is hydrophilic and can be soluble in water, making it easy to use

**Table 18** The operating cost analysis

Type	Consumption rate	Annual consumption
Electric Power	0.8 MW	6720 MW
Fuel Gas	450 kg/h	3,780,000 kg
Nutrient (Nutrimix)	297.38 L/D	89.22 M <sup>3</sup>
Caustic (NaOH)	3727.83 L/D	1118.35 M <sup>3</sup>
Anti-Foam Agent (TOG)	-----	33.25 M <sup>3</sup>
Demin Water in Thiopaq	76.6 M <sup>3</sup> /D	22,978.1 M <sup>3</sup>
sulfide-oxidizing bacteria	-----	One time



**Fig. 12** Visible difference between Claus sulfur and THIOPAQ sulfur [38]

in agriculture, although the bio-sulfur can convert to Claus sulfur properties (but not vice versa) via a melting process.

## Conclusions

The Claus technique simulated with a property package Sulsim (sulfur recovery) that is integrated within Aspen HYSYS to create an accurate model, but Aspen HYSYS does not support the capital cost and operating cost of the Claus section, TGTU, and the incineration section. TGTU increases the  $H_2S$  removal efficiency from 95.08% to 99.90%, but it also increases the total cost of a Claus unit. The actual acid gas removal efficiency in the field is 97.93% less than the efficiency of 99.90% by using Aspen HYSYS. By using trial and error of the lean amine flow rate (73,000 kg/hr) and the composition of the lean amine ( $H_2O$ , MDE Amine, and ME Amine), they affect the  $H_2S$  content (207.6 ppmV) in the sweet gas. The vendor's total capital cost of the two Claus trains is 98 million euros, and the operating cost is 4.2 million euros for producing about 50 tons per day of sulfur product. The Claus process required much more transmitters and control loops and took a long time to startup. Free  $H_2S$  and high temperatures exist in most equipment of the Claus process. In a new addition to our study, Aspen HYSYS used to create a new model for the absorption section and the bioreaction section of the THIOPAQ technique, so new evidence will be applied by Aspen HYSYS V11. As a result, it is used for the development of the physical and chemical absorption of the THIOPAQ technique. Acid Gas-Caustic Wash Property Package is used for physical reactions (the absorption process), and Peng-Robinson is used for chemical reactions and conversion reactions. The Aspen HYSYS can calculate the total capital cost and operating cost for the THIOPAQ unit, but the operating cost did not match the actual (vendor) operating cost. THIOPAQ does not need any additional units to achieve the  $H_2S$  removal efficiency of 99.999%. Actual acid gas removal efficiency in field 99.999%, which is the same as the efficiency of 99.999% by using Aspen HYSYS. The vendor's total capital cost of the two THIOPAQ trains is 44 million euros, and the operating cost is 3.56 million euros for producing about 50 tons per day of sulfur product. The THIOPAQ is a small unit in volume with low numbers of

equipment, low transmitters and control loops, a short time to startup, no high pressure or temperature, and free H<sub>2</sub>S only existed in the absorption section. Finally, the THIO-PAQ technique has demonstrated lower capital and operating costs, easier operation, and a safer unit. Bio-sulfur is the basis of a range of new agricultural products designed to act as liquid fertilizers and liquid fungicides.

#### Abbreviations

AGR	Acid gas removal
SRU	Sulfur recovery unit
AGRU	Acid gas removal unit
AGE	Acid gas enrichment unit
TGT	Tail gas treatment
TGTU	Tail gas treatment unit
WHB	Waste heat boiler
O&G	Oil and gas
MDE Amine	Methyl di-ethanol amine
ME Amine	Methyl ethanol amine
K.O Drum	Knock out drum
SDVs	Shut down valves
NO.	Number
hr.	Hours
CAPEX	Capital cost
OPEX	Operating cost

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#### Authors' contributions

MF: Conceptualization, Methodology, Writing – Original. TM: Investigation, Formal analysis. MA: Investigation, Formal analysis. All authors have read and approved the manuscript.

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#### Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

#### Declarations

##### Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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