



## Using TSWEET Process Simulator to Optimization of Sulfur Recovery Unit

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### Abstract

In this sulfur recovery unit (SRU), hydrogen sulfide (H<sub>2</sub>S) is converted to elemental sulfur using modified sulfur recovery unit. In the present study, application of different alternatives for increasing the reaction furnace temperature and sulfur recovery of Claus sulfur recovery units (SRUs) are investigated by TSWEET process simulator. This simulator based on Gibbs free minimization. The usefulness of different techniques such as acid gas enrichment, Acid gas flow of AG splitter, oxygen enrichment and air dry flow for increasing the furnace temperature and sulfur recovery are determined by proposed simulator. It is shown that furnace temperature increases up to maximum temperature, increasing fraction of AG splitter flow to main burner and then decreases by a sharp slope. In the case of concentration of H<sub>2</sub>S and oxygen concentration, temperature of main burner increases monotonically. Also in this paper it is found that sulfur recovery increases up to a maximum value and then decreases as H<sub>2</sub>S concentration (in all three concentrations of oxygen) increases While in the case of oxygen concentration is not so.

**Keywords:** Sulphur recovery, H<sub>2</sub>S Concentration, H<sub>2</sub>S/CO<sub>2</sub> ratio, oxygen concentration, Claus unit.

### Introduction

Sulphur recovery is an important refinery processing unit (SRU). It removes H<sub>2</sub>S from acid gas streams before they can be released into the atmosphere [1]. Furthermore, in industrial world H<sub>2</sub>S exists mainly as an undesirable by-product of gas processing [2]. Different processes are used for recovering sulphur from H<sub>2</sub>S. The most widely used process is Claus process [1,3]. Claus method was developed by Carl Friedrich Claus in 1883 [4] in which an overall sulfur recovery of 94-97 % was achieved. Several modifications were developed on the main process in order to increase the overall sulfur recoveries which are mentioned here. The sulfur recovery requirements range from 97.5% to 99.8+% for gas processing and refining facilities processing 10 LT/d and greater [5]. The higher recovery of 99.8+% is required for most facilities with 20 LT/d and higher [3,6]. Changing the number and type of beds in the rich acid gas feed, the sulphur recovery rate increases from 96.1% to 99.3% while, in the case of lean acid gas feed, using modifications such as acid gas bypass and acid gas bypass with oxygen enrichment, causes to enhancement of the sulphur recovery from 96.1% to 96.6% [7,8].

Recently, a number of studies have been performed on the reaction furnace and sulfur recovery which are mentioned below. Wen et.al [9] studied empirical prediction of CO, COS, CS<sub>2</sub> and H<sub>2</sub> through Gibbs free minimization method. In 1990, Dowling et al. [10] conducted a study on the

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conversion of H<sub>2</sub>S into hydrogen and sulfur. This reaction is one of reactions occurring in the main burner and the kinetics indicated that the reaction rate is very fast. Low temperature is favorable for the forward reaction and the reverse reaction is enhanced at high temperature. Both forward and reverse reactions are first order reactions.

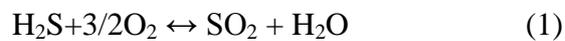
Reaction kinetic of carbonyl sulfide formation has been determined through combination of carbon monoxide and hydrogen sulfide [11]. Zarenezhad and Hosseinpour [12] compared real data and data obtained from Gibbs free minimization method and concluded that reaction furnace temperatures obtained from this model are in good agreement with experimental data.

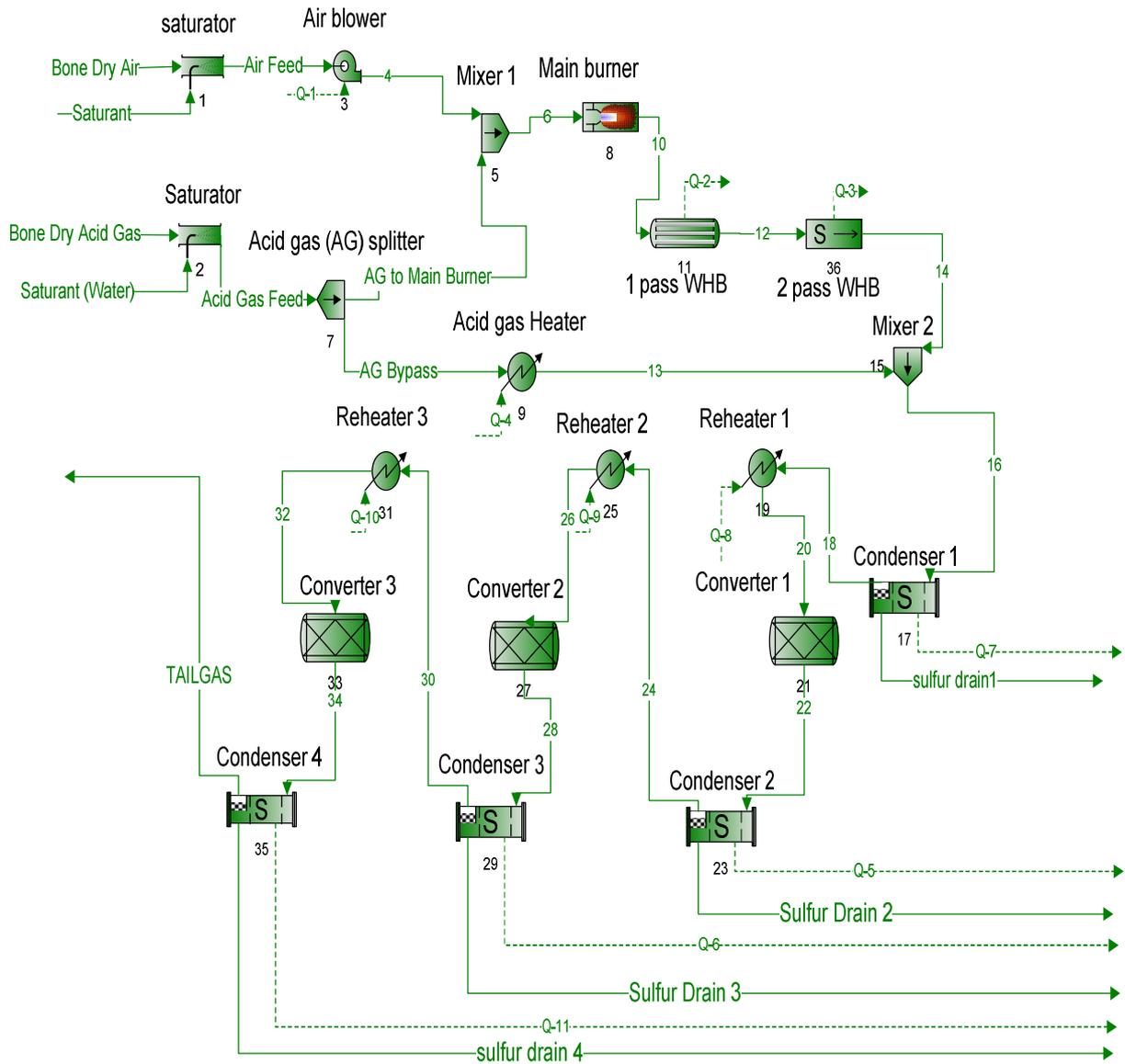
In this paper, we present a simulation for Claus process, then concentration effect of H<sub>2</sub>S, acid gas flow of AG splitter and oxygen concentration were studied. For the purposes of this article, intake air and acid gas feed are classified into three different categories in terms of composition. The first types of intake air and acid gas feed are containing 21 mole % O<sub>2</sub> and 30 mole % H<sub>2</sub>S. The second Types are containing 50 mole % O<sub>2</sub> and 50mole % H<sub>2</sub>S. The last types are containing 85 mole % O<sub>2</sub> and 90 mole % H<sub>2</sub>S.

### ***Methodology***

Research methodology consists of review and comparison of the effect of H<sub>2</sub>S concentration, fraction of acid gas flow to main burner and oxygen concentration (in the tail gas ratio of 2.0) on the reaction furnace temperature and sulfur recovery using a process simulator called TSWEET.

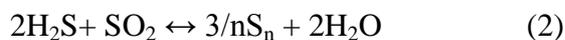
Fig. 1 shows the flow diagram of the Claus unit. In this process, the acid gas (acid gas + fuel gas) enters SRU and is divided into two streams in the AG (Acid Gas) splitter. A part of stream comes to main burner, and another part comes to acid gas heater (for hydrolyzing sulphur components to H<sub>2</sub>S). 1/3 H<sub>2</sub>S in the acid gas is oxidized to SO<sub>2</sub> at the main burner using outlet air of air blower:





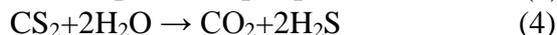
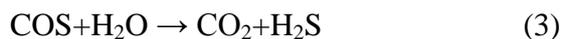
**Fig. 1:** Flow diagram of simulated Claus process.

This combustion generates a large amount of heat. Further, the combustion products undergo Claus reaction between  $\text{H}_2\text{S}$  and  $\text{SO}_2$ .



Where  $n$  is in the range of 6-8. Reaction (2) is exothermic and reversible, thus, processing under adiabatic conditions increases temperature. The effluent gas from the main burner (reaction furnace) passes through the first pass waste heat boiler (1<sup>st</sup> pass WHB) to recover heat and produce high pressure steam [1,13]. The second pass of the waste heat boiler is where the redistribution of  $\text{S}_2$  to  $\text{S}_8$  is the primary reaction. Furthermore, side reactions involving hydrocarbons and  $\text{CO}_2$  in the acid gas feed can result in the formation of carbonyl sulphide ( $\text{COS}$ ), carbon disulfide ( $\text{CS}_2$ ) in the output of the main burner [12]. The effluent streams from the 2<sup>nd</sup> pass WHB and acid gas heater is cooled in condenser 1 to condense and recover sulphur. The effluent gas of the condenser 1 is preheated in the reheater 1 and is sent to three catalytic converters where Claus reaction occurs at lower temperature. This leads to higher equilibrium conversion because Claus reaction is exothermic.

Typically, COS and CS<sub>2</sub> are also hydrolyzed in the first catalytic converter according to the following exothermic reactions [2]:



The first converter normally operates at temperature high enough to hydrolyze COS and CS<sub>2</sub>. Usually, the catalysts, based on - Al<sub>2</sub>O<sub>3</sub> or TiO<sub>2</sub>, are used in the process [14]. Sulphate is formed on the - Al<sub>2</sub>O<sub>3</sub> catalyst when a mixture of H<sub>2</sub>S/SO<sub>2</sub> or H<sub>2</sub>S and SO<sub>2</sub>, separately, are passed on the catalyst at industrial conditions [15]. Sulphur is recovered after each catalytic stage by cooling converter effluent gas in a sulphur condenser. Effluent gas from the process is incinerated to SO<sub>2</sub> and vented to atmosphere.

The simulation was performed using the TSWEET (copyright©2007, Bryan Research & Engineering, Inc.) version 2.0 (PROMAX). PROMAX is a powerful and jack of all trades stream based process simulation pack. It is used by engineers around the world to design and optimize gas processing, refining, and chemical processes.

TSWEET will assume that the Claus beds operate at 95 percent of equilibrium conversion and 4 (lb) sulfur/100 Moles of gas are entrained in the sulfur condensers [16]. This simulator uses three unit operations (main burner, 1<sup>st</sup> pass WHB, 2<sup>nd</sup> pass WHB) to model the burner/waste heat boiler [8,16]. The burner unit operation simulates the combustion of acid gas, with COS and CS<sub>2</sub> formation being calculated by NSER 1993 correlation [9]. This correlation predicts the concentration of COS, CS<sub>2</sub>, CO, H<sub>2</sub>, and/or S<sub>2</sub> out of the burner. The correlation appears to be most accurate when no sour gas (NH<sub>3</sub>) is present.

The reaction furnace (modeled as the first pass of the waste heat boiler) is where large amounts of free sulfur, hydrogen and water recombine into H<sub>2</sub>S and SO<sub>2</sub>. The typical output temperature is about 600°C since all significant reactions other than sulfur redistribution have ceased in 600°C. The 2<sup>nd</sup> pass WHB is where the redistribution of S<sub>2</sub> to S<sub>8</sub> is the primary reaction, which the typically outlet temperature is about 315°C.

### ***Results and discussion***

The first role of the Claus unit reaction furnace is to partially oxidize H<sub>2</sub>S in the acid gas feed to SO<sub>2</sub> and it will also produce a significant segment of the total sulphur product. A second role of the reaction furnace is to ensure destruction of many contaminants in the acid gas feed stream. This will ensure that these compounds do not break through the downstream process unit, where they can have a significant detrimental effect on the Claus catalyst activity. Therefore, it is critical to understand the effect of furnace operating parameters on the reaction furnace temperature and overall sulphur recovery.

There are different techniques to change the furnace temperature and sulphur recovery, including, concentration of H<sub>2</sub>S in the acid gas feed, fraction of AG splitter flow to main burner and oxygen concentration. The usefulness of all of these methods in changing the furnace temperature and sulphur recovery are investigated by a process simulator. All results of this research were obtained in constant conditions of acid gas flow rate (108 lb mole/ hr).

### ***Acid gas enrichment***

In this section, using TSWEET, concentration effect of H<sub>2</sub>S in acid gas feed in different concentrations of oxygen (in input air) on temperature of main burner and sulfur recovery was studied.

With the advent of selective amine systems, it is possible to enhance the quality of an acid gas stream in an amine treating unit [17]. Providing a high quality acid gas for processing in a Claus SRU as well as reducing the acid gas flow rate and the volume of SRU are two important

advantages of acid gas enrichment. Enrichment of acid gas should be utilized with care, considering the Claus process features. If a lack of air occurred, less than 1/3 of H<sub>2</sub>S would react and the H<sub>2</sub>S would then be in excess.

There are different forms of carbon and stainless steel corrosion namely, sulfide stress cracking, hydrogen stress cracking, localized corrosion [18-20] and cracking in the welded region. H<sub>2</sub>S increases the speed of the hydrogen absorption in steel [21]. Hydrogen sulfide reacts with iron in the presence of water and the produced hydrogen moves to the high concentration regions in the metal. When there is hydrogen in steel, it facilitates the motion of the linear effects in the crystalline structure of metal. So, the ratio of tail gas (H<sub>2</sub>S/SO<sub>2</sub>) was fixed at 2.0. The ratio is controlled by manipulating the flow rate of the inlet air from the blower to the main burner [7].

The effect of increasing the H<sub>2</sub>S content of the acid gas on the main burner is predicted by the process simulator in Fig. 2a and b. It is obvious that increasing the H<sub>2</sub>S content of the acid gas will significantly increase the main burner temperature. As seen in the Fig. 2a and b, when concentration of H<sub>2</sub>S in acid gas feed is 5 mole%, temperature of main burner in all three concentrations of oxygen in input air is the same and equal to 538°C. As H<sub>2</sub>S concentration increases the difference between curves are increased.

For a given H<sub>2</sub>S concentration, as the concentration of oxygen in the intake air increases, the curves become closer together which means the temperature variations in the main burner are smaller.

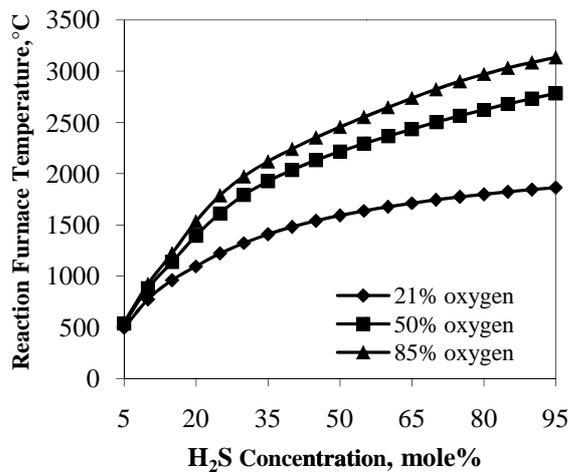
For example when concentration of H<sub>2</sub>S in the acid gas feed is 50% mole, temperature of main burner is 1593°C (21% mole oxygen), 2216°C (50% mole oxygen) and 2454°C (85% mole oxygen) respectively.

When concentration of oxygen in input air into simulated Claus unit is 21% mole, by changing concentration of H<sub>2</sub>S in the range 5-95%, temperature of main burner increases from 538°C to 1864°C, indicating an increase of 1326°C. However; in the cases in which oxygen concentrations in input air are 50% and 85%, temperature of the main burner (by rising concentration of H<sub>2</sub>S) has increased from 538°C to 2783°C and 538°C to 3149°C respectively which represents increases of 2245 and 2611°C respectively. So, as oxygen concentration increases, temperature of the main burner (by changing H<sub>2</sub>S) also increases. But there are limitations in using Fig. 2a and b since the temperature of reaction furnaces used in the Claus sulphur recovery units are at least ~1700 °F (for flame stability) and maximum ~2700-3000°F [12]. So according to the Figures, concentration variations of H<sub>2</sub>S should be almost in the range 15-55 mole% (21 mole% oxygen), 11-25 mole% (50 mole% oxygen) and 10-22 mole% (85 mole% oxygen).

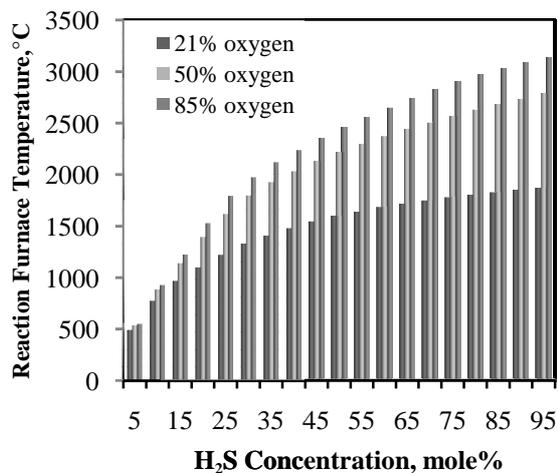
As studied in Fig. 2a and b, the temperature of main burner inconsistently increases as H<sub>2</sub>S concentration existing in acid gas feed increases while sulfur recovery increases up to a maximum value then decreases (as seen in Fig. 2c and d). Column graph (Fig. 2d) is used to shed light on variability of sulfur recovery relative to H<sub>2</sub>S concentration. As seen in the graph, maximum recovery percent of sulfur is 98.58% (21% mole oxygen), 98.64% (50% mole oxygen) and 98.66% (85% mole oxygen) the maximum of sulfur recovery has been obtained in the range 25-30% mole H<sub>2</sub>S (in all three concentrations of oxygen). Also as seen in Fig. 2d, in concentrations lower than 35% mole, the sulfur recovery in the case of 85% mole oxygen has been doubled with respect to the other two. However, in concentrations higher than 35% mole H<sub>2</sub>S in acid gas feed, the sulfur recovery in 21% mole oxygen is more than the other two. Other results can be derived from this figures which are obvious.

The output value of H<sub>2</sub>S and CO<sub>2</sub> from stripper tower of gas sweetening unit is different depending on amine type used in this unit. So the effect of the ratio H<sub>2</sub>S/CO<sub>2</sub> on the sulfur recovery is examined to optimize H<sub>2</sub>S/CO<sub>2</sub> ratio and recovery rate of sulfur according to this ratio (Fig. 3a and b). As seen in this figures, the sulfur recovery first increases up to a maximum value and then decreases by different slopes (in proportion with different concentrations of oxygen). Maximum value of sulfur recovery in each concentration of oxygen is 98.58% (21% mole oxygen), 98.64% (50% oxygen) and 98.66% (85% mole oxygen), respectively. These

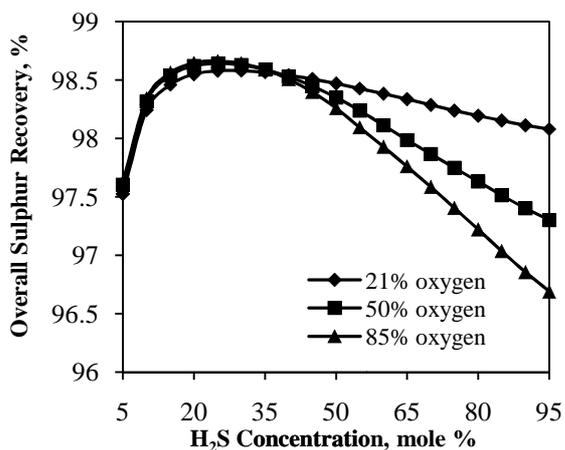
values have been obtained in ratios of 0.45, 0.33 and 0.33 respectively which are the same in two concentrations of 50% and 85% mole oxygen.



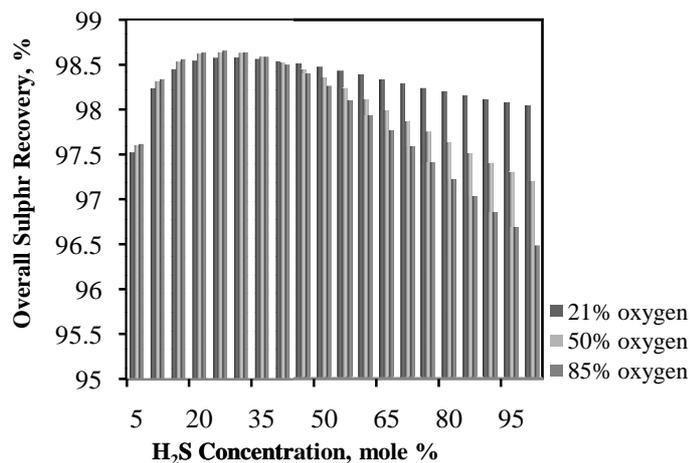
(a)



(b)

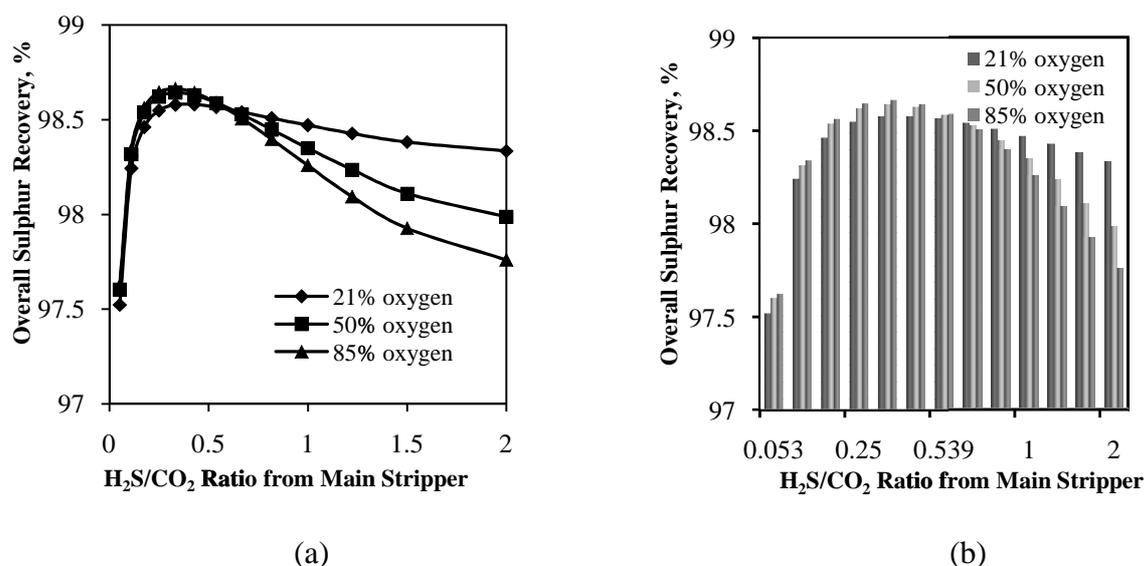


(c)



(d)

**Fig. 2:** Effect of H<sub>2</sub>S concentration on the reaction furnace temperature (a,b), and overall sulphur recovery (c,d).



**Fig. 3:** Effect of H<sub>2</sub>S/CO<sub>2</sub> ratio obtained from main stripper of amine sweetening unit on the sulfur recovery (a,b).

### *Acid gas flow of AG splitter*

Next the effect of the fraction of the AG splitter flow that goes to the main burner on the main burner temperature (for 31 mol% H<sub>2</sub>S in the acid gas feed) was studied and is shown in Fig. 4a and b. For all three concentrations of oxygen, an increase in the fraction of acid gas flow to the main burner initially caused an increase in the main burner temperature but a further increase resulted in a sharp decrease.

When all the acid gas flow enters into the bypass flow below the splitter, the main burner temperature in all cases is 104°C. Since oxidization of H<sub>2</sub>S does not occur in the main burner, the main burner temperature should be equal to the output temperature of the air blower (104°C).

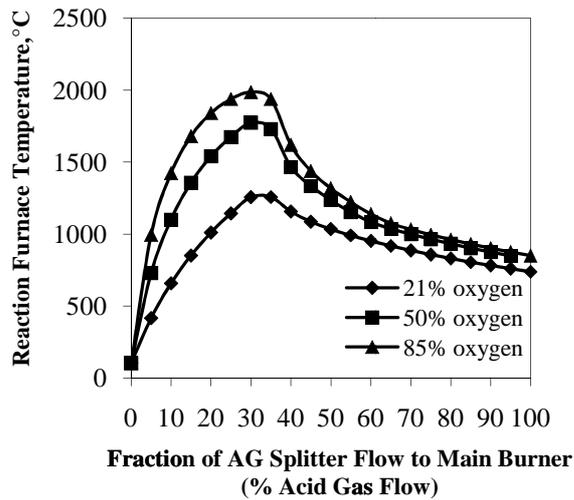
As seen in Fig. 4a and b, when the oxygen concentration in the input air is 21 mol%, the main burner temperature increases rapidly to 1258°C (maximum) which is obtained for a 35% acid gas flow from the splitter to the main burner. In concentrations above 35%, the main the burner temperature decreased sharply changing from 1258 to 740°C. If the oxygen concentration in the input air is 50mol%, the maximum temperature (1773°C) of the main burner in obtained for 30% acid gas flow to the main burner. For concentrations above 30%, the main burner temperature decreases from 1773 to 850°C. Also, for an oxygen concentration of 85 mol%, the main burner temperature increased to 1986°C (at 30% acid gas flow) and then decreased to 852°C. It can be concluded that when the splitter flow is below the maximum value, increasing the acid gas flow to the main burner results in the curves diverging from each other. Whereas for fractions in which the maximum temperature value has been reached, the curves converge.

So far the effect of fraction of AG splitter flow to main burner (Fig.4a and b) on the reaction furnace temperature (in the range of 0-100%) was investigated and the results were analyzed. But there are limitations in using Fig. 4a and b which was mentioned in the previous section. So the fraction of AG splitter flow to main burner must be in range 17-100% (21mole% oxygen), 7-22% and 37-80% (50 mole% oxygen), 5-15% and 40-85% (85 mole% oxygen).

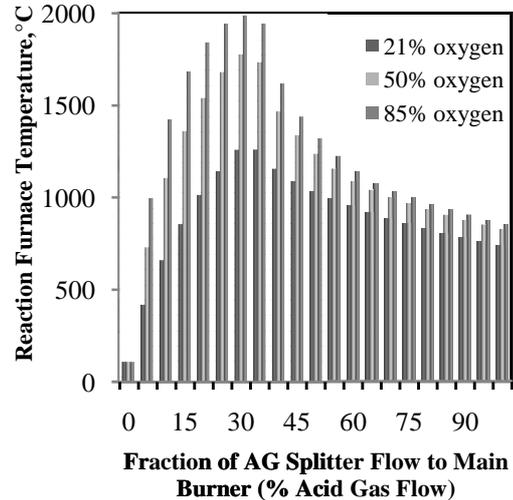
As said previously, increases in fraction of AG splitter flow to main burner might lead to increase in temperature of main burner up to maximum temperature then it decreases by different slopes (depending on oxygen concentration). However, sulfur recovery has decreased to a minimum value then increased by a deep slope which is different for each concentration of oxygen (as seen in Fig.4c and d). However, this increase continues by a deep slope to a fraction

of AG splitter flow to main burner then increases by a very slow slope (slight increase in recovery rate of sulfur). Sulfur recovery in fraction of AG splitter flow lower than 30 mole % (70% splitter flow enter bypass) in concentration of 21% mole oxygen is higher than the other two. However, in concentrations 30mole % and higher of AG splitter flow fraction, sulfur recovery in concentration 85% mole oxygen is more than the two other.

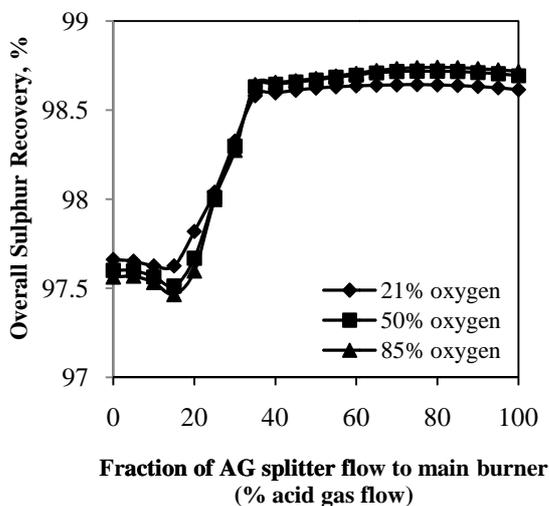
Overall, sulfur recovery for each concentration of oxygen has increased from 98.66% to 98.62% (21% mole oxygen), from 97.60% to 98.70 % (50% mole oxygen) and from 97.56% to 98.72% (85% oxygen), respectively. Also it is seen in the Fig. 4c and d, if total flow (without bypass) enter main burner, the sulfur recovery is less than the case in which flow is 70-85% (in the low concentrations of H<sub>2</sub>S).



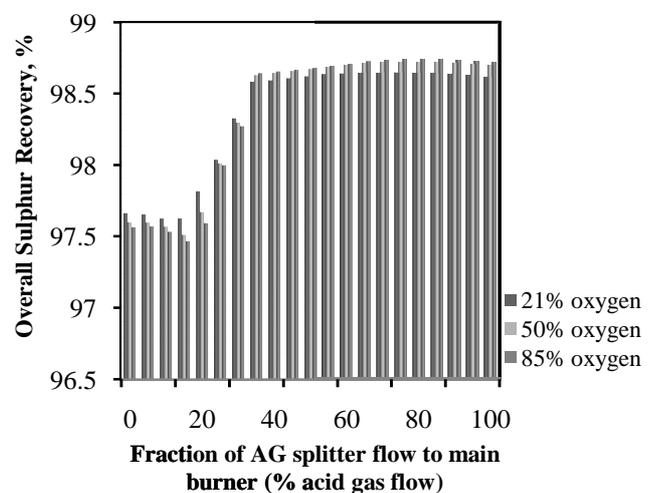
(a)



(b)



(c)



(d)

**Fig. 4:** Effect of fraction of AG splitter flow to main burner (% acid gas flow) on the reaction furnace temperature (a,b), and sulfur recovery (c,d).

### Oxygen enrichment

Many units are using oxygen enriched of air dry flow in their SRUs for many reasons [15,22–24]. In most cases, oxygen enrichment is used to raise the throughput capacity of the plant. However, in some particular cases, oxygen enrichment has been used to better the reaction

furnace temperature. In some cases this has allowed for the processing of highly lean acid gas feed and in others it has utilized for better overall furnace and burner operation. The oxygen enrichment should be used with care. The accuracy control of the mixing of combustion gases is an essential factor in avoiding oxygen breakthrough into the catalytic converters. The poor concord in the combusting gases led to unreacted oxygen being accessible in the gas to further oxidize  $\text{SO}_2$  to  $\text{SO}_3$  either before the gas stream reached the catalyst bed or on the catalyst bed itself. This  $\text{SO}_3$  then reacts with the alumina catalyst and depraves it by forming aluminum sulphate. The  $\text{SO}_3$  may also reacts with  $\text{H}_2\text{O}$  to produce sulphuric acid, which is a corrosive operative capable of damaging steel equipments depending on acid concentration and temperature [25].

As seen in the Fig. a and b, by increasing oxygen concentration in tail gas ratio of 2 (optimal ratio), reaction furnace temperature increases. Increased  $\text{H}_2\text{S}$  concentration in acid gas feed led to increase the reaction furnace temperature. In 30% mole  $\text{H}_2\text{S}$ , temperature of main burner increased from  $598^\circ\text{C}$  to  $2016^\circ\text{C}$ . In concentrations of 50 and 90%  $\text{H}_2\text{S}$ , reaction furnace temperature was changed from  $598^\circ\text{C}$  to  $2498^\circ\text{C}$  and  $624^\circ\text{C}$  to  $3143^\circ\text{C}$ , respectively. Comparing these data with real ones indicates that when ratio of tail gas is 2, main burner temperature increases more. However, it is a general diagram because main burner temperature should be in the range  $700^\circ\text{C}$  to  $2000^\circ\text{C}$ . The advantage of this study compared to the other is that first general changes of main burner temperature with respect to oxygen concentration then allowable range of oxygen concentration (in proportion to allowable range of reaction furnace temperature) are specified.

In this section the effect of oxygen concentration on sulphur recovery rate while changing the temperature of reaction furnace mentioned in the pervious section has been studied. The main purpose of Claus unit is optimal sulphur recovery and optimal temperature of reaction furnace is a temperature in which more optimal sulphur recovery is obtained than other temperatures. So, in this study change rate of sulphur recovery was studied with respect to oxygen concentration in the intake air into Claus unit. As seen in Fig. 5 c and d, in 30% mole  $\text{H}_2\text{S}$  concentration, sulphur recovery was increased relatively however in 50% and 90% concentrations first it was increased to maximum then decreased with a sharp slope. It is concluded that in 30% mole  $\text{H}_2\text{S}$  concentration, the more reaction furnace temperature is, the more optimum the temperature will be. However in 50% and 90% mole, optimum temperature was obtained in a specific concentration.

Also seen in Fig. c and d, as oxygen concentration changes in the range 95-5%, sulphur recovery in 30% mole  $\text{H}_2\text{S}$  concentration was increased from 98.2 to 98.57%. However in 50% and 90% mole  $\text{H}_2\text{S}$ , sulphur recovery had maximums in concentrations 10 and 15 % mole respectively. Optimal temperatures of reaction furnace corresponding to the values of sulphur recovery were  $1271^\circ\text{C}$  and  $1050^\circ\text{C}$  respectively. It is also concluded that increased concentration of  $\text{H}_2\text{S}$  in acid gas feed leads to decreased reaction furnace temperature.

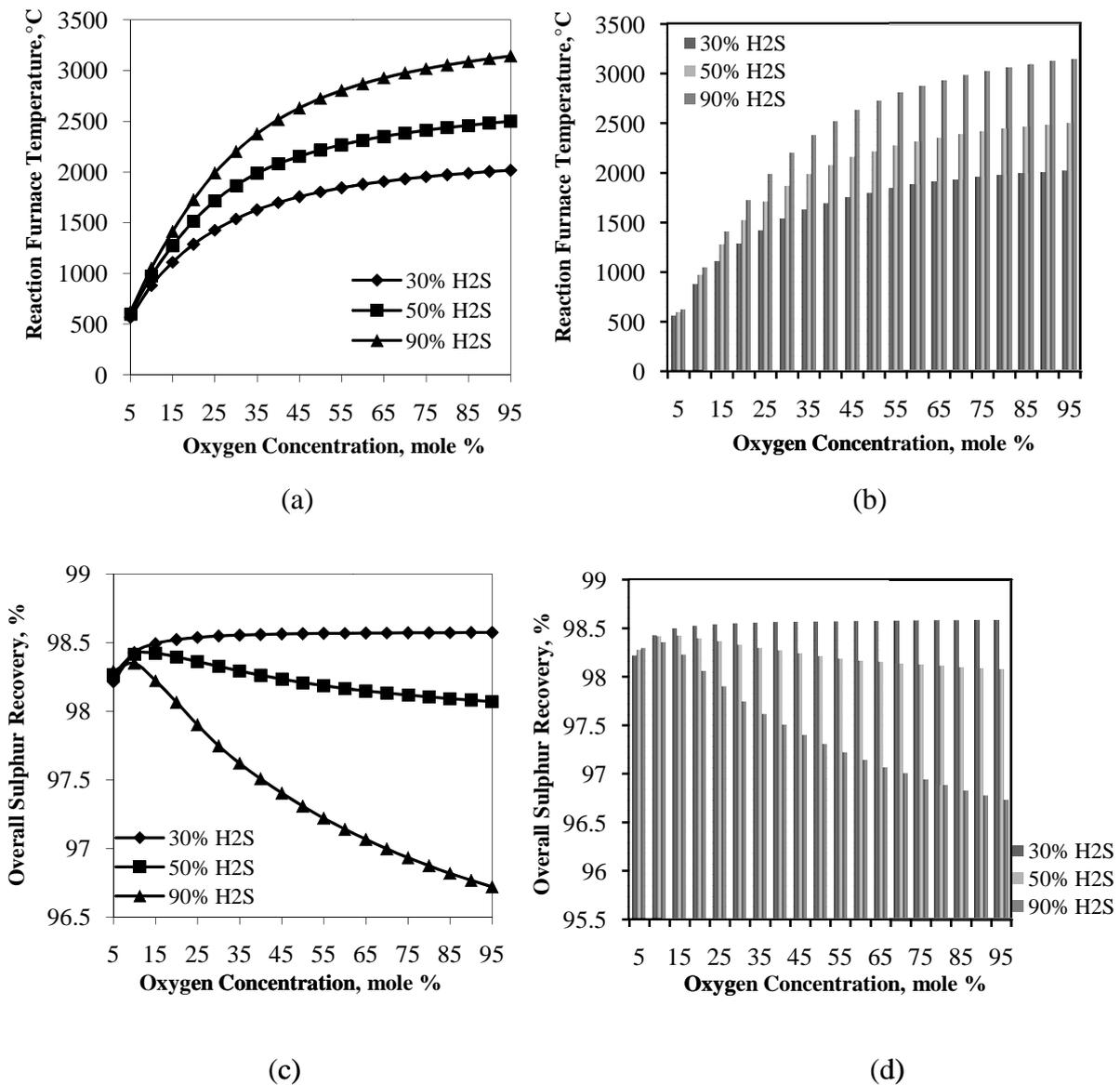


Fig. 5: Effect of oxygen concentration on the reaction furnace temperature (a,b), and sulfur recovery (c,d).

### Conclusion

Many Claus sulphur recovery units suffer from poor flow stability and hydrocarbon destruction in the reaction furnace due to a low flame temperature in reaction furnace. In order to reduce this problem, various techniques were investigated by a process simulator, TSWEET, to optimize the furnace temperature, including, changing the concentration of H<sub>2</sub>S, fraction of AG splitter flow to main burner and oxygen concentration. Also this process simulator is used for prediction of overall sulphur recovery and the predicted results are compared. In this study it is concluded that the main burner temperature increases up to a maximum value and then decreases by a sharp slope (Increasing fraction of AG splitter flow to main burner). However, increasing concentration of H<sub>2</sub>S and oxygen, temperature of main burner has increased monotonically (significant direct effect). As oxygen concentration in the input air has increased, permissible range of H<sub>2</sub>S concentration has decreased. In addition it is concluded that sulfur recovery has increased to a maximum value and then has decreased, increasing H<sub>2</sub>S concentration and H<sub>2</sub>S/CO<sub>2</sub> ratio. Also if 70-85% acid gas feed entered main burner, more optimal sulfur recovery than other flows would be achieved (tail gas ratio of 2.0). Whereas in the case of oxygen, it concluded that as oxygen concentration changes in the range 5-95%, sulphur recovery in 30% mole H<sub>2</sub>S was increased from 98.2 to 98.57%. Also The optimal reaction furnace temperatures (

in 50% and 90% mole H<sub>2</sub>S) corresponding to the values of sulphur recovery were obtained 1271°C and 1050°C respectively.

### ***Acknowledgement***

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