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PERFORMANCE STUDY OF A SULFUR RECOVERY PLANT FROM FLARE GAS USING ASPEN HYSYS SULSIM SIMULATION

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Absract

Sulfur recovery is an essential process in the midstream and refining stages of the oil and gas industry. Changes in feed composition, temperature, and pressure; deterioration of equipment and catalysts; and suboptimal operation caused by inadequate control of temperature, pressure, and air flow rate are among the challenges that reduce the performance of sulfur recovery units. Poor performance can lead to safety and environmental problems. This study aims to improve the performance of a sulfur recovery unit at a gas plant. A modified three-stage Claus process model, using Aspen HYSYS V14 Sulsim, was used to simulate the behavior of an actual sulfur recovery unit and establish the base case. The base-case performance was 92.90%. Adding a selective oxidation converter increased performance to 98.46%. Adding a tail-gas section with a recycle stream further increased performance to 99.96%. The simulation procedure developed in this study can help process engineers operate sulfur recovery units effectively under challenging conditions.

***Keywords**: Alumina catalyst; Challenging feed; Sulfur recovery unit; Sulsim.*

الملخص- يعد استخلاص الكبريت عملية أساسية في مراحل التكرير والمعالجةالمتوسطة في صناعة النفط والغاز. ومن بين التحديات التي تُقلّل من كفاءة وحدات استخلاص الكبريت: التغيرات في تركيب المواد الخام ودرجة حرارتها وضغطها، وتدهور المعدات والمحفزات، والتشغيل غير الأمثل الناتج عن عدم كفاية التحكم في درجة الحرارة والضغط ومعدل تدفق الهواء. وقد يؤدي ضعف الأداء إلى مشاكل تتعلق بالسلامة والبيئة. تهدف هذه الدراسة إلى تحسين أداء وحدة استخلاص الكبريت في محطة غاز. تم استخدام نموذج مُعدَّل لعملية كلاوس ثلاثية المراحل، باستخدام برنامج Aspen HYSYS V14 Sulsim، لمحاكاة سلوك وحدة استخلاص كبريت فعلية وتحديد الحالة الأساسية. بلغ أداء الحالة الأساسية 92.90%. أدى إضافة مُحوِّل أكسدة انتقائي إلى زيادة الأداء إلى 98.46%. كما أدى إضافة قسم غاز العادم مع تيار إعادة التدوير إلى زيادة الأداء إلى 99.96%. يمكن أن تساعد إجراءات المحاكاة التي تم تطويرها في هذه الدراسة مهندسي العمليات على تشغيل وحدات استخلاص الكبريت بفعالية في ظل ظروف صعبة.

1-Introduction:

Natural gas may contain hydrogen sulfide (H_2S), carbon dioxide (CO_2), sulfur dioxide (SO_2), ammonia (NH_3), mercaptans, and other sulfur-bearing compounds. These impurities can lower gas quality, damage processing equipment and pipelines through corrosion, and contribute to environmental pollution, including acid rain [1]. Gas containing significant H_2S is commonly described as sour gas, while acid gas generally refers to streams containing acidic components such as H_2S and CO_2. Removing acid gases is necessary to meet safety and

environmental requirements, limit corrosion, protect equipment, improve the quality of liquid products, and achieve the required heating value for sales gas. A widely used treatment method is absorption with a regenerative aqueous amine solution. As the gas contacts the amine, H_2S and CO_2 are absorbed and separated from the natural gas, producing sweetened gas [2]. The amine solution is then heated to release the absorbed components, allowing the solvent to be reused.

In gas plants, after the acid gas cleaning, sulfur is removed. Based on H_2S content in acid gas, acid gas is classified as lean acid gas and rich acid gas. H_2S content in rich acid gas is greater than 50% and in lean acid gases, H_2S content is less than 50%. From the inception, Claus process is the most practiced process industrially for recovery of sulfur [3]. In Claus process, sulfur recovery from acid gases is a challenging task because of operational parameters and feed stock changes [4]. Process challenges can be handled by process modifications.

Process modification studies need the knowledge of thermodynamics and kinetics of chemical species involved in the process. Various studies on process modification and optimization are available to the sulfur recovery. Some of them are: A review for process modification technologies [5], Select process for lean acid gases processing [6], studies on effect of reaction parameters on the quality of captured sulfur [7], reaction with adsorption in a reaction furnace itself to overcome the Claus reaction equilibrium limitations [8], super Claus process using selective oxidation catalyst [9], designing new alumina catalysts with specific properties [10-11] and structures [12], lanthanum oxide based catalysts [13], ammonia pyrolysis and oxidation technique [14], benzene destruction technique [15], new tubular Claus catalytic reactor heat exchanger unit filled with a heat transfer enhancement medium [16], Claus recycle with double combustion process [17], reaction kinetic studies to reduce CS_2 formation [18], BTX oxidation by SO_2 in a BTX destruction unit placed between Claus furnace and catalytic units [19], toluene destruction using oxygen enriched air [20], maintaining suitable operating conditions in the reaction furnace for the presence of toluene and carbon dioxide [21], feasibility studies for lean acid gas feed containing mercaptans [22], coupled modification of GTU and SRU processes [23], thermal insulation to avoid thermal damage of Claus reaction chamber [24], reducing number of catalytic stages by following guidelines for the design of thermal section [25], introducing high temperature air combustion technology [26].

In the Claus process maximum possible sulfur is recovered from the thermal and catalytic sections. For a two-stage Claus process the maximum sulfur recovery is 95% and for a three-stage Claus process sulfur recovery is 98% [4]. These recovery efficiencies are not sufficient to meet the stringent environmental regulations. The new regulations demands, 100% recovery of sulfur from acid gases. To meet the new environmental standards, tail gas treatment units are attached to the sulfur recovery units. In tail gas treatment the remaining sulfur is recovered. Various technologies available for tail gas cleaning in literature are Beavon sulfur removal process [27], in which the tail gas is treated in two-steps. Sulfur contaminants are first catalytically hydrolyses and/or hydrogenated to hydrogen sulfide and the hydrogen sulfide is then converted to elemental sulfur and recovered, The Shell Claus Off-gas Treating (SCOT) Process [28], use of Cu- and Ag-exchanged Y zeolites as selective adsorbents for hydrogen sulfide [29], temperature swing adsorption systems for Claus tail gas clean up units [30], use of a re-generable solid sorbent [31].

Performance of the Claus units depends on thermodynamics and kinetic factors of the process [32]. Using thermodynamics and kinetic data models for the Claus process can be developped [25, 33-35]. These models are useful for the simulation of Claus process and its unit operations [36], Genetic algorithms [37], model-based optimization techniques [38], modeling and multi-optimization of thermal section [39] are some the techniques used for the improved performance of sulfur recovery units. The advantage of the simulations is one can predict the response of the process for challenging conditions and for operational changes [40-42].

The above studies motivated the present simulation work to improve the performance of the sulfur recovery plant with tail gas treating section. Results of this work are useful for process modification studies to recover the desired quantity of sulfur from acid gases without changing the process equipment.

2. Process description

Acid gas feed to the modified Claus three- stage process contains H_2S, CO_2, CH_4, C_2H_6, C_3H_8. Feed gas composition is given in Table 1. Acid gas is processed for sulfur recovery using the Aspen HYSYS Sulsim Claus three-stage process with tail gas treatment section and incinerator.

Table 1. Feed gas composition of sour gas

component	CO_2	H_2S	CH_4	C_2H_6	C_3H_8
Mole fractions	24.3	74.9	0.005	0.002	0.001

The Claus process generally consists of two main sections. The first is the thermal section, which includes the main burner, the reaction furnace, and the waste heat boiler. The second portion is the catalytic section, comprising two or three catalytic converters arranged in series. Each catalytic converter contains a catalytic reactor equipped with a layer of catalyst and a condenser. Between the catalytic reactors, re-heaters are installed to supply the heat required for the catalytic reactions. A schematic diagram illustrating the three-stage Claus process is presented in Figure 1. At the entry point of the thermal section, the main burner is located. Its primary function is to combust impurities, such as hydrocarbons, ammonia, and mercaptans, found in the acid gas stream coming from the acid gas cleaning unit. To facilitate this reaction, an external air stream is introduced. Immediately following the main burner is the reaction furnace, where one-third of the hydrogen sulfide (H_2S) present is converted into elemental sulfur. The relevant chemical reactions are shown as equations (1) and (2):

$$3H_2S+\frac{3}{2}O_2 \rightarrow SO_2+2H_2S+H_2O \quad (1)$$

$$2H_2S+SO_2 \leftrightarrow 3S+2H_2O+heat \quad (2)$$

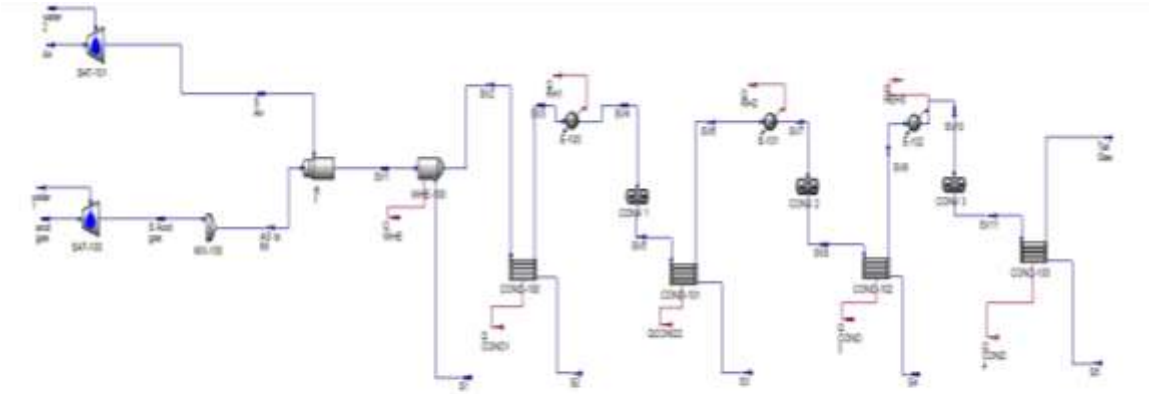


Figure 1. Process flow diagram for the base case three stage Claus process

The remaining two-thirds of the hydrogen sulfide (H_2S) is transformed into elemental sulfur utilizing catalytic converters, which is followed by the thermal section. The overall Claus reaction is represented by the equation:

$$2H_2S + SO_2 \rightarrow \frac{3}{n}Sn + 2H_2O \qquad (3)$$

During combustion in the reaction furnace, certain impurities such as carbonyl sulfide (COS) and carbon disulfide (CS_2) are produced. These impurities can reduce the sulfur recovery efficiency and must be addressed within catalytic converters through hydrolysis reactions. The specific reactions are:

$$COS + H_2O \leftrightarrow CO_2 + H_2S \qquad (4)$$

$$CS_2 + 2H_2O \leftrightarrow CO_2 + 2H_2S \qquad (5)$$

These hydrolysis reactions occur at elevated temperatures in the presence of a catalyst. In the thermal furnace, the initial reactions take place over approximately two seconds at temperatures ranging from 1000°C to 1400°C. The elemental sulfur formed is subsequently cooled in a waste heat boiler, allowing it to be separated through condensation. Heat from the reaction furnace gases is absorbed in the waste heat boiler to generate steam. Unconverted gases proceed to a re-heater, which connects the thermal and catalytic sections. The re-heated stream then enters the catalytic converter where sulfur formation continues, enhancing sulfur recovery. Catalytic reactors function at temperatures exceeding the sulfur dew point, ensuring sulfur can be effectively extracted from process gases at each catalytic converter system, elemental sulfur is condensed and collected within the condensers. Typically, alumina and titania catalysts are employed to facilitate this process. The gaseous effluent emerging from the final stage of catalytic condensation generally has a low concentration of sulfur-containing compounds. However, it may require further processing to meet the specifications for flare gas. This gas is routed to a tail gas treating unit, which may include various operations such as a hydrogenation bed, reducing gas generator, quench tower, amine absorber, incinerator, and flare system. The extent of sulfur recovery required

depends on the specific tail gas treatment system in place. Additionally, the recycled gas stream is fed back into the reactor furnace to maintain process efficiency. Any remaining process gas with low sulfur content is directed to the incinerator and flare for final disposal.

3. Process simulation

Acid gas stream from the amine regenerator unit in the acid gas cleaning unit is connected to the sulfur recovery unit as feed gas stream. The tail gas stream is connected to the outlet of the Sulsim sub-flow sheet. After the convergence of the Aspen HYSYS V14 Sulsim, sub-flow sheet cumulative sulfur recovery of the unit is checked. If the initial overall performance is not satisfactory a couple of modifications are performed to improve the performance of the sulfur recovery unit.

In the simulation, the main parameters considered are reaction furnace empirical model, H_2S to SO_2 ratio, the temperature of catalytic converters, incinerator checker wall kinetic parameters. Base case simulation uses the straight through acid gas empirical model. Selection of the empirical model depends on the percentage of the H_2S present in the feed gas. Here, straight through amine acid gas empirical model is selected and this option is suitable for feed containing the more than 50% of the H_2S. For further improvement in sulfur recovery, it can be tested with other empirical models available in Aspen HYSYS. Optimum H_2S to SO_2 ratio can be maintained by arranging air demand analyzer (ADA) and adjust operations. ADA is connected between the last condenser and the reaction furnace. Air demand analyzer (ADA) is useful in adjusting the air to fuel ratio and Adjust block is useful to adjust the converter temperature. To meet the tail gas specifications air demand analyzer controls air flow to the reaction furnace. To do this, in ADA internal options, tail gas is selected as the sample stream, target variable is selected as air demand percent and air demand target value is set as zero percent (it corresponds to the ratio of H_2S and SO_2 as 2). For convergence, it needs more iteration. After re-converging, performance will be improved.

Selective oxidation converter is linked to the vapor outlet stream from the final condenser, which is part of the catalytic converter in setup 3, as illustrated in Figure 2.

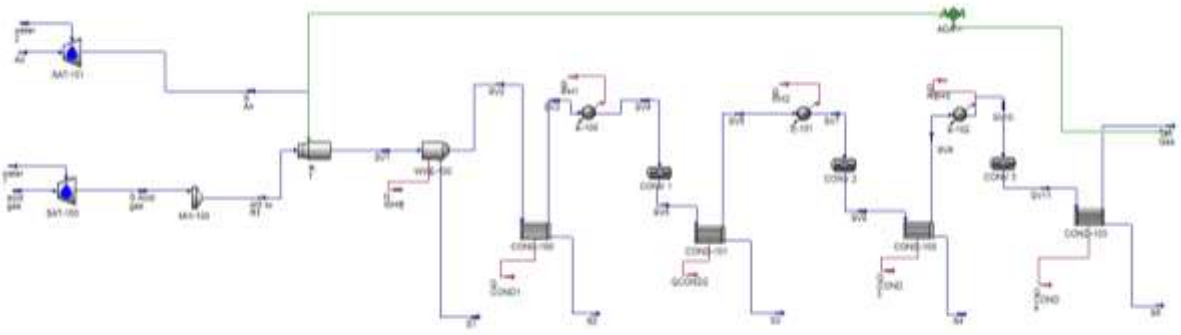


Figure 2. Sulfur recovery unit with selective oxidation converter

Feed composition fluctuations often occur in sulfur recovery units. In this study, simulations focus on low concentrations of H_2S in the feed stream to manage these challenging situations. The optimization process used here remains consistent with previous sections to ensure that the sulfur compound levels in the tail gas remain unchanged, without altering the process equipment. In Figure 3, the tail gas section is shown, which includes a reducing gas generator,

hydrogenation bed, quench tower, and a simple amine absorber. The reducing gas generator operates with a burn stoichiometry of 88% and a steam-to-fuel ratio of 93.72% by mass. The RGG Air, RGG Fuel Gas, and RGG Water streams are similarly defined. An adjustment block (ADJ-3) is incorporated to regulate the RGG Fuel Gas flow for achieving a target RGG Effluent temperature of 285°C. The specifics for the hydrogenation bed are provided next. The quench tower is set with a pressure drop of 3 kPa and an outlet temperature of 40°C. Data for the simple amine absorber are also included.

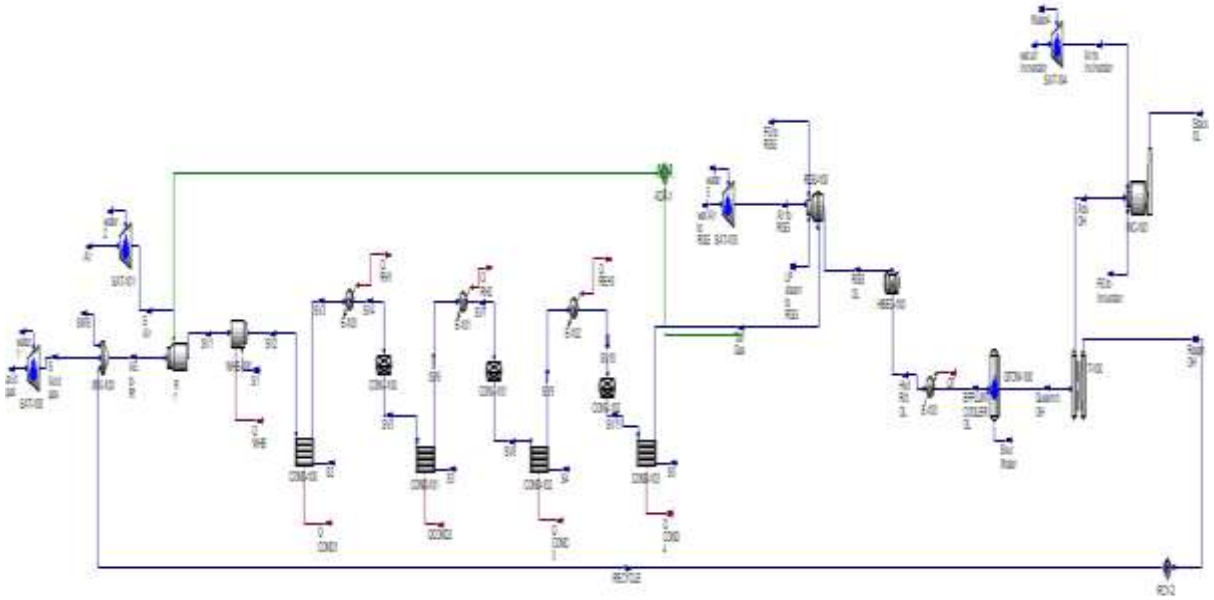


Figure 3. Sulfur recovery unit with tail gas treating section, incinerator and recycle stream

4. Result and Discussion

The composition (mole fractions) of the acid gas feed to the sulfur recovery unit is as follows: CO_2 0.242, H_2S 0.75, CH_4 0.005, C_2H_6 0.002, and C_3H_8 0.001. Simulations were conducted to optimize cumulative sulfur recovery in a three-stage Claus process. Key parameters focused on in the simulation included the H_2S/SO_2 ratio, the sulfur dew point temperature at the outlet, catalytic converter inlet temperatures, and the incinerator temperature. Given the potential to enhance sulfur recovery by optimizing hydrolysis reactions (specifically reactions (4) and (5)), and to achieve this in the first catalytic converter, the effect of elevated temperature on sulfur recovery was tested while maintaining the second and third converters at lower temperatures (as operating the final two stages at lower temperatures favors the Claus reaction). To further improve cumulative sulfur recovery or enhance hydrolysis reaction performance, an alumina catalyst was selected for the catalytic converter unit operation.

In Sulfur Recovery Units (SRUs) can be challenging due to fluctuations in feed composition; existing process configurations may fail to meet required specifications under these varying feed conditions. Consequently, process optimization and the calculation of optimal operating parameters are undertaken to meet the necessary operational requirements.

Table 2. Sulfur recovery efficiency summary for base case process

stage	Thermal reaction furnace	Catalyst convertor 1	Catalyst convertor 2	Catalyst convertor 3
Conversion %	66.97	56.52	51.43	1.42
Cumulative Conversion %	66.97	85.62	92.99	93.09
Sulfur recovery %	99.76	99.10	97.62	31.23
Cumulative Sulfur recovery %	66.81	85.45	92.81	92.90
Co hydrolysis %	N/A	94.90	26.04	9.57
CS2 hydrolysis %	N/A	87.47	6.56	1.91
Overall recovery efficiency %	---	---	---	92.90

To achieve Operational adjustments are essential to achieve a sulfur recovery rate of approximately 98.46%. To comply with flare specifications, the concentration of sulfur-containing compounds in the tail gas must also be kept below 1 kmol/h. These improvements in the sulfur recovery process can be implemented without requiring any modifications to the existing equipment. Through a process of trial and error, the converter temperature was optimized, and an air demand analyzer was introduced to effectively control the air-to-fuel ratio. This analyzer continuously monitors the process stream exiting the final condenser within the catalytic section. The operating temperature of the first catalytic converter is set at 359°C to ensure the effective decomposition of carbon disulfide (CS_2) and carbonyl sulfide (COS). For the second and third catalytic converters, their temperatures are maintained at least 10°C above the sulfur dew point. This precaution prevents liquid sulfur from depositing on the catalyst surfaces, which would otherwise impair the overall sulfur recovery efficiency. Figure 4.

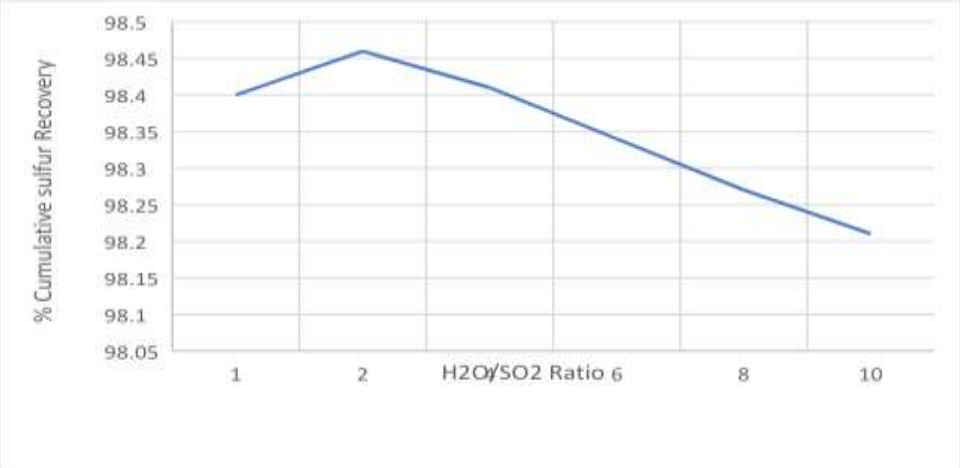


Figure 5. Variation of cumulative sulfur recovery percent with H2S/SO2

The relationship between H2S/SO2 ratios (expressed as air demand percentages) and their impact on sulfur recovery is illustrated in Figure 5. According to the Claus reaction, as represented by reaction (3), a closer alignment of the H_2S/SO_2 ratio to 2 enhances performance and increases sulfur recovery efficiency in the catalytic section. At zero percent air demand (where the ratio of H_2S to SO2 is 2:1), the cumulative sulfur recovery reaches its maximum value, achieving up to 99.96%, as noted in Table 3.

Table 3. Sulfur recovery unit performance summary for optimized process

stage	Thermal reaction furnace	Catalyst convertor 1	Catalyst convertor 2	Catalyst convertor 3
Conversion %	67.03	51.78	75.44	54.53
Cumulative Conversion %	67.03	84.09	96.08	98,21
Sulfur recovery %	99.52	97.96	96.81	82.23
Cumulative Sulfur recovery %	66.70	83.74	95.69	97.76
Co hydrolysis %	N/A	94.71	34.16	12.84
CS2 hydrolysis %	N/A	85.90	9.02	2.32
Overall recovery efficiency %	---	---	---	99.96

4.1 Effect of outlet sulfur dew point margin

To evaluate the effect of the outlet dew point temperature, an adjustment block is incorporated at the second catalytic converter. The outlet sulfur dew point temperature is modified by altering the target variable values, and the resulting variations in sulfur recovery are illustrated in Figure 6. It is noted that a lower sulfur dew point temperature margin leads to higher sulfur recovery rates. However, extremely low temperatures are detrimental to catalyst performance. The optimal dew point temperature margins are identified to be within the range of 18.65°C to 23.37°C.

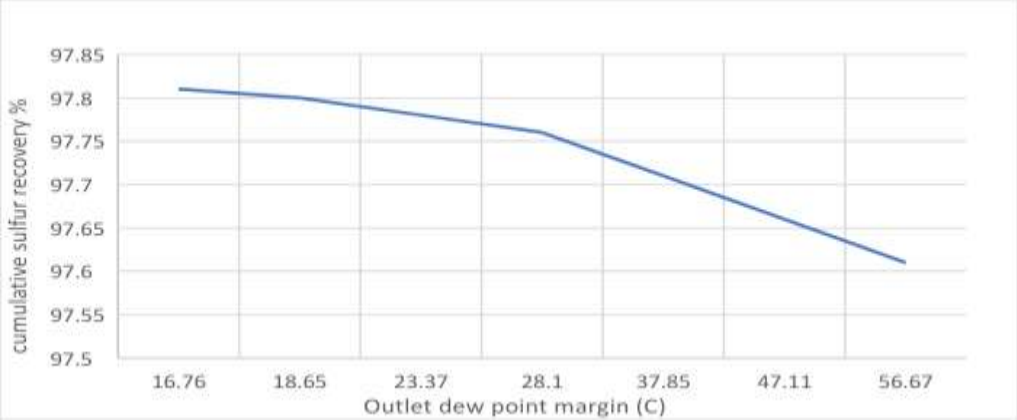


Figure 6. Variation of cumulative percent re-covery of sulfur with outlet dew point temper-ature margin at catalytic converter two

4.2 Effect of temperature

The products such as COS and CS_2 are formed in the reaction furnace due to side reactions. The presence of these compounds reduces sulfur recovery efficiency. To decompose these compounds, the temperature at the first catalytic converter must be raised. Figure 6 shows that increasing the temperature from 300°C to 350°C raises the cumulative sulfur recovery rate to 97.73% and 97.90%, respectively; this is attributed to the further decomposition of COS and CS_2 in the first catalytic converter. The recovery rate subsequently drops slightly at 400°C; however, at very high temperatures, the sulfur recovery rate remains very high, demonstrating the catalyst's activity even under high-temperature conditions.

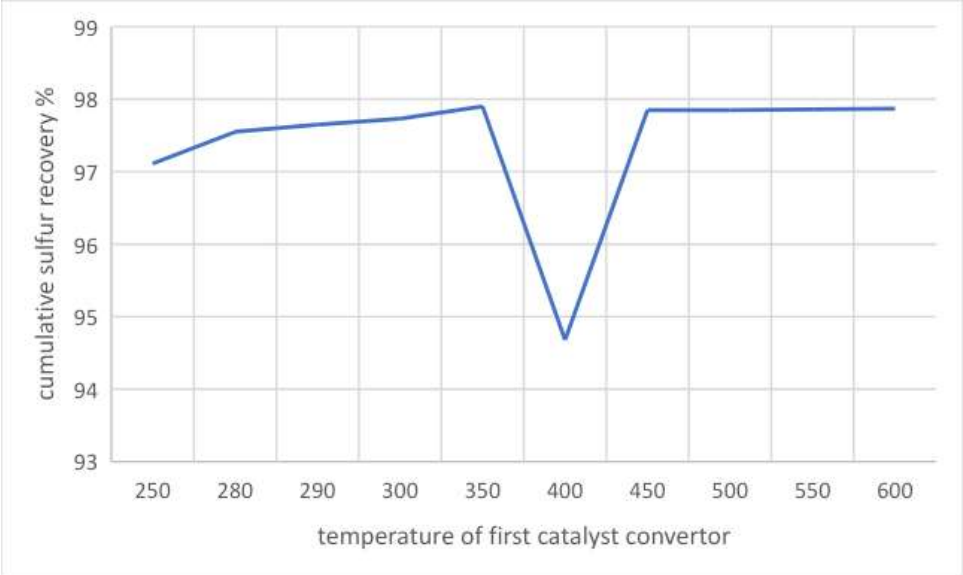


Figure 7. Variation of cumulative percent recovery of sulfur with varying temperature of first catalytic con-verter (for additional destruction of COS and CS2) .

5. Conclusion

- The cumulative sulfur recovery percentage reaches a maximum value of 99.96%.

- Increased sulfur recovery efficiency; simulations show that a significant portion of the H_2S in the flare gas is converted into elemental sulfur at a rate of 2,328 kg/hr, thereby enhancing recovery efficiency.

- Reduced hydrogen sulfide (H_2S) concentration in the flue gas or the unit's outlet gas, serving as a key indicator of the treatment process's success.

- Reduced emissions of sulfur dioxide (SO_2) and harmful gases, leading to improved environmental performance and decreased pollution resulting from flare gas combustion.

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