



# Process Revamping of H<sub>2</sub>SO<sub>4</sub> Plant to Double Contact Double Absorption (DCDA) Using ASPEN HYSYS to Reduce SO<sub>2</sub> Emission: Case of Awash Melkassa Sulfuric Acid Factory

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**Abstract.** Sulfur dioxide (SO<sub>2</sub>) is identified as among one of the major air pollutant gases in the globe. SO<sub>2</sub> cause severe adverse effects on the respiratory system of all living things and causes several difficulties in the environment such as acid rain and plant growth retardation. This work presents a virtual experimental investigation of the simulation of H<sub>2</sub>SO<sub>4</sub> using Double contact double absorption (DCDA) to reduce the SO<sub>2</sub> emission. Awash Melkassa Aluminum Sulfate and Sulfuric Acid Share Company (AMASSASC) produce Sulfuric acid by using single contact single absorption (SCSA) which releases up to 3% SO<sub>2</sub> to the atmosphere. In this work, the simulation uses ASPEN HYSYS with Antoine based fluid package to predict several values. The major unit operation involved in the design includes four consecutive converters and absorption columns including the proposed intermediate absorber and secondary absorber. Different recycle setup configuration was considered to achieve the best possible SO<sub>2</sub> reduction. The effect of inlet temperature, split ratio of mass to the modeled absorber in response to the final SO<sub>2</sub> amount was studied. From the optimal design it was able to reduce SO<sub>2</sub> to 0.12 mol%. The trade-off between utility power consumption and SO<sub>2</sub> emission for different design options was also studied to obtain optimal design setup.

**Keywords:** SO<sub>2</sub> emission · H<sub>2</sub>SO<sub>4</sub> · Aspen HYSYS · Simulation · SCSA · DCDA

## 1 Introduction

Sulfuric acid is one of the most widely produced chemical in the world and sulfuric acid production is a good indicator of a nation's industrial strength [1, 2]. It can be produced by different methods and it is very crucial chemical in process industries [3]. It has a vast application in different areas like fertilizer manufacture, mineral processing, oil refining, wastewater processing, chemical synthesis, domestic acidic drain cleaners, as an electrolyte in lead-acid batteries, and in various cleaning agents [4, 5]. Although the universal use of sulfuric acid has made it indispensable, its production process releases gaseous pollutants such as sulfur oxides (SO<sub>x</sub>), nitrous oxides (NO<sub>x</sub>) and acid mist (H<sub>2</sub>SO<sub>4</sub>) to the environment, which ecologically threaten the standard of living [6].

The federal Clean Air Act (CAA) requires the United States Environmental Protection Agency (U.S. EPA) to set National Ambient Air Quality Standards (NAAQS) for six criteria pollutants that are considered harmful to public health and the environment in 1970. These pollutants are: particulate matter, carbon monoxide, ground-level ozone, nitrogen dioxide, SO<sub>2</sub>, and lead. The NAAQS set limits for the criteria pollutants in the ambient air. Limits established to protect human health are referred to as primary standards while limits established to prevent the environmental damage are referred to as secondary standards. The primary NAAQS for SO<sub>2</sub> measured over a 1-h period is set at 75 parts per billion parts of air. A secondary NAAQS for SO<sub>2</sub> measured over a 3-h period is set at 0.5 parts per million parts of air. This standard cannot be exceeded more than once per year [7]. High level of sulfur dioxide emission is associated with various ecological damages and health problems. The major health issues due to the availability of sulfur dioxide in the atmosphere causes a series adverse effect on the respiratory system of all living beings [8, 9]. Short-term exposure to SO<sub>2</sub> may cause wheezing, chest tightness and shortness of breath. Longer-term exposure to sulfur dioxide causes series respiratory problems. The environmental effects of SO<sub>2</sub> are detrimental. SO<sub>2</sub> is dissolved in water and can be easily oxidized in to sulfurous acid and sulfuric acid in the water vapor in the air which will falls as an acid rain [10]. Therefore, SO<sub>2</sub> can be a major factor for the acid rain that is harmful for vegetation, agriculture and, corrosion of buildings and constructions [11].

Hence, strategies for mitigating these emissions become a major concern for sustainable development. Over the years, the design and modification of the sulfuric acid production process have received considerable attention to minimize air pollution. The lead chamber process was the earlier design employed for large scale production and this production process gradually outdated and replaced by single contact single absorption (SCSA) process [12, 13]. 97 to 98% of the SO<sub>2</sub> is oxidized to SO<sub>3</sub> in the SCSA process while the remaining 2 to 3% is vented to the environment [14]. To enhance the oxidation efficiency of SO<sub>2</sub> to SO<sub>3</sub> and to obtain a higher grade of sulfuric acid, the SCSA process was modified to the double contact double absorption (DCDA) process by Bayer in 1963 [14]. Currently, the DCDA process is the most preferable and widely used sulfuric acid production process. Basically, the operating principle of DCDA process is same as that of the lead chamber and SCSA processes [15]. The distinguished characteristics of the DCDA process over other types of production processes are the presence of double reactive-absorption towers and four or more converter beds in the catalytic reactor. These enhance the oxidation of SO<sub>2</sub> to about 99.5% or higher in the catalytic converters, and an acid concentration in the range of 98 to 99.9% in reactive-absorption towers [13]. The unreacted SO<sub>2</sub> and SO<sub>3</sub> from the process are emitted through the absorber exit gases to the environment and its emission should be below the permissible limit.

Several studies have been conducted by numerous researchers to reduce the emission of SO<sub>2</sub> to environment. Kiss et al. proposed a model to simulate a DCDA process with five converter-beds; air flow split to catalyst beds 3 and 4, and the burner. Their results showed that SO<sub>x</sub> emissions are significantly reduced approximately by 40% by optimizing air flow rate to the burner or split fractions [16, 17]. Gomez-Garcia et al. also developed a rigorous heterogeneous model for the analysis of an industrial adiabatic multi-bed catalytic reactor for SO<sub>2</sub> oxidation based on the Maxwell-Stefan

diffusion model. The implementation of the model in steady state simulation allows realistic prediction of each catalytic bed in terms of SO<sub>2</sub> oxidation, pressure and temperature profile, and the effectiveness factor variation along the reactor [18]. Many other studies were conducted and reported by different authors to reduce the emission of SO<sub>2</sub> to the atmosphere [19–23].

Awash Melkassa Aluminum Sulfate and Sulfuric Acid Share Company (AMAS-SASC) is the only sulfuric acid and aluminum sulfate factory in Ethiopia and it was established 1982 E.C. The annual production rate of the factory is 13,600 and 17,000 tons for aluminum sulfate with concentration of 17% and sulfuric acid with a concentration of 98.5%, respectively [24]. This factory produces sulfuric acid by using single contact single absorption (SCSA) process. Due to SCSA process, 2 to 3% SO<sub>2</sub> toxic gas is emitted into the environment and the release of this SO<sub>2</sub> emission into the environment has a tremendous impact on the health of all living things nearby. This study aimed at minimizing the emission of SO<sub>2</sub> gas from AMASSASC factory by improving the oxidation of sulfur dioxide to sulfur trioxide using double contact double absorption process through ASPEN HYSYS simulator.

This study mainly focuses on reducing the emission of SO<sub>2</sub> into the environment by increasing the oxidation of SO<sub>2</sub> to SO<sub>3</sub> and thereby improving the production of Sulphuric acid in AMASSASC. The specific objectives are to investigate the effect of recycle stream configuration to different converters on the final SO<sub>2</sub> amount, split ratio supplied to intermediate absorber on to the amount of SO<sub>2</sub> released to the environment and to study the tradeoff between percent emission reduction and utility consumption.

## 2 Methodology

### 2.1 Materials

Aspen HYSYS V10 was used to simulate the modified H<sub>2</sub>SO<sub>4</sub> plant. Simulation data were obtained from the Awash Melkassa chemical factory.

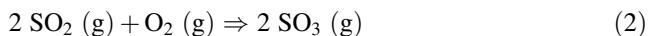
**Problem Definition:** The process of sulfuric acid plant uses elemental sulfur (99.99%). The solid sulfur changes to liquid form in melting section. In this section the solid sulfur is fed into the feed hopper from the storage area and enters into the melting pit. As the coil is heated the solid sulfur become melted and change to black color liquid sulfur at a temperature between 135 °C–145 °C and at a pressure of 0.6 Mpa.

Molten sulfur is pumped by a screw pump to the burner through a jacketed pipe which used to prevent the solidification of molten sulfur inside the pipe. The liquefied Sulfur is oxidized in to SO<sub>2</sub> in burning section with temperature range between 700 °C to 1040 °C in the presence of dried air with temperature of 235 °C from directed from air drying tower. Then the molten sulfur is converted to SO<sub>2</sub> gas with 11% SO<sub>2</sub> gas according to reaction in Eq. 1.



The converter section has four fixed catalytic beds with its own heat exchangers. The gases with 11% SO<sub>2</sub> from the waste heat boiler is charged with a temperature of

425 °C at the top of the converter and the dried air from the drying tower also directed to the converter. Then the Sulfur dioxide gas is oxidized to sulfur trioxide in the presence of vanadium pent oxide ( $V_2O_5$ ) catalyst according to the Eq. 2 shown below.



All the reactions in the converter are exothermic and reversible in nature. The converters are operating in the temperature intervals of 400–600 °C and 1–2 atm.

**First Bed:** The gas with a temperature of 425 °C is directed to the 1st bed which is filled with the vanadium penta oxide catalyst and the first converter has a conversion of 63%. 1st converter bed with a temperature of 597 °C is sent to the gas-gas heat exchanger, where it's cooled down to a temperature of 440 °C.

**Second Bed:** The gas with the temperature of 440 °C is supplied to the 2nd bed where the unconverted gas further converted to  $\text{SO}_3$ . The conversion of 2<sup>nd</sup> bed is around 87% and gas lift from 2<sup>nd</sup> bed with the temperature of 505 °C sent to gas-water heat exchanger where it was cooled down to a temperature of 430 °C.  $\text{SO}_2$  gas with the temperature of 430 °C was sent to 3<sup>rd</sup> bed converter.

**Third Bed:** The unconverted gas from 2<sup>nd</sup> bed converter is expected to be further converted to  $\text{SO}_3$ . The grade of conversion in the 3rd bed converter reaches to 96%. Gas leaving from 3<sup>rd</sup> bed with the temperature of 453 °C to 420 °C and sent to 4<sup>th</sup> bed converter.

**Fourth Bed:** The unconverted gas from 3<sup>rd</sup> bed further converted by mixing of the dried air with gas. Finally, the grade of conversion after 4th bed was 98%. Now the  $\text{SO}_3$  gas with the temperature of 424 °C was sent to economizer (E501-3) or gas water heat exchanger where  $\text{SO}_3$  gas cooled to a temperature of (180–200) °C. Finally, this  $\text{SO}_3$  with temperature of 180–200 °C was sent to absorption tower.

**Absorption Column:** In an acid plant absorption tower,  $\text{SO}_3$  gas was charged to the bottom of the tower from the converter and 10% concentrated sulfuric acid with 98.5% concentration from the circulation tank which is filled with 70% of concentrated sulfuric acid showers at the top of absorption tower by a circulating pump by reducing its temperature from 80 °C to 60 °C by acid cooler. The rate of absorption can be improved by rushing ring which maximizes surface contact area between gas and liquid while minimizing pressure drop. Sulfuric acid has the ability to absorb  $\text{SO}_3$  gas. These acids contain 1.5% water which reacts with the  $\text{SO}_3$  gas. Once  $\text{SO}_3$  comes in contact with the acid, it is immediately absorbed by circulating acid and produces 99.1% concentrated sulfuric acid with a temperature of 89.3 °C. Here the top exit gas contains  $\text{SO}_2$  is emitted to the environment at the concentration of 2–3%.

**Dilution Tank:** Finally, oleum enter into dilution tank to reduce the concentration from 99.1% to 98.5% by adding water or by an acid that return from drying tower with

concentration of 98.3% according to Eq. 3. After mixing in the dilution tank the acid has concentration of 98.5%. Then enter into circulation tank with temperature of 80 °C. Figure 1 shows the overall Block flow diagram for the production of H<sub>2</sub>SO<sub>4</sub> and revamped process synthesis.

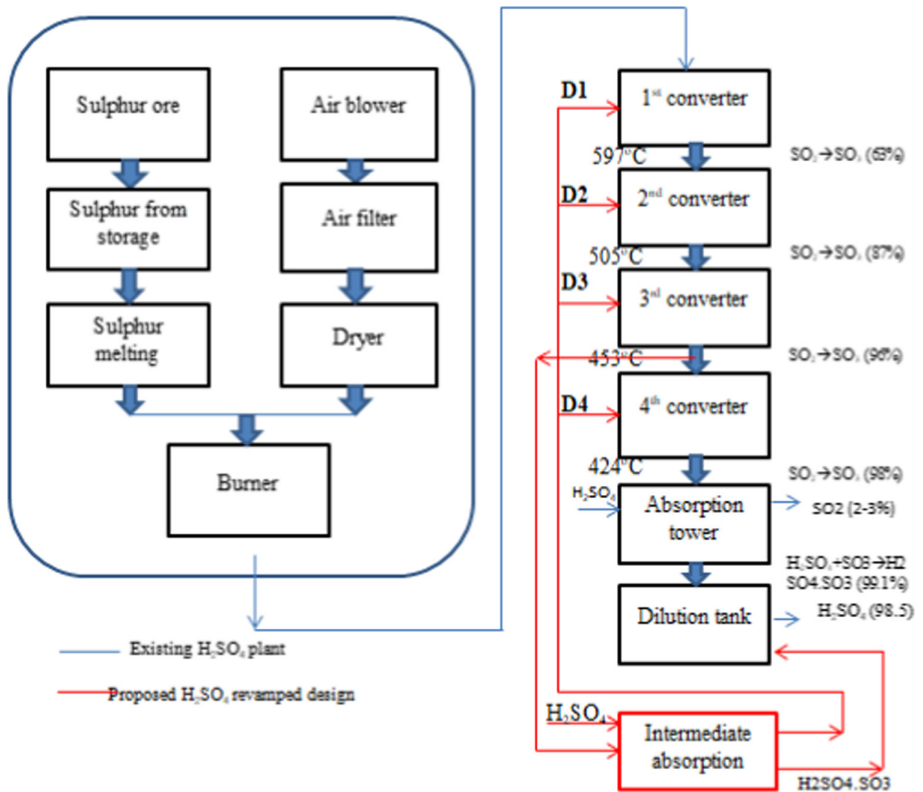
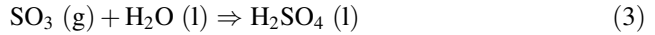


Fig. 1. Existing plant SCSA and the revamped proposed of H<sub>2</sub>SO<sub>4</sub> using DCDA

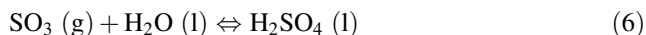
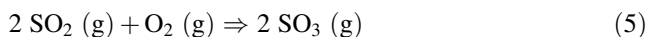
## 2.2 The Proposed Intermediate Absorption

In this study some portion of the exit gas of the third converter will be taken to the proposed absorption column where H<sub>2</sub>SO<sub>4</sub> is fed at the top of the column to react with SO<sub>3</sub> and leaving the column at the bottom as oleum (H<sub>2</sub>SO<sub>4</sub>.SO<sub>3</sub>) increasing the sulphuric acid concentration then enters in to the dilution tank along with the oleum

exiting the secondary absorption column (existing). The top stream of the absorption column containing  $\text{SO}_2$  will be recycled to the subsequent converter to study the effect of the configuration. Figure 1 shows the existing and revamped process synthesis. There are three proposed configuration of the entrance of the exit unconverted  $\text{SO}_2$  from intermediate absorption column. From figure these are designated as a, b and c that corresponds to that the exit gas  $\text{SO}_2$  from the intermediate absorption column entering into 1<sup>st</sup> converter, 2<sup>nd</sup> converter and 4<sup>th</sup> converter respectively.

### 2.3 Reactions Package

Sulphuric acid is manufactured from elemental sulphur in a three-stage process. These stoichiometric were entered to the basic environment of the simulation. For the simulation modeling Conversion reactor model was selected for Eq. 4 and 5 that takes place in burner and converter 1 respectively. The reaction between  $\text{SO}_3$  and  $\text{H}_2\text{O}$  was modeled using equilibrium reactor model using dilution tank according to the relation presented by Eq. 6.



### 2.4 Fluid Package

For the simulation of the modified design options, Antoine model was used. This model is applicable for low pressure systems that behave ideally. The equations are traditionally applied for heavier hydrocarbon fractionation systems and consequently provide a good means of comparison against rigorous models. They should not be considered for VLE predictions for systems operating at high pressures or systems with significant quantities of light hydrocarbons.

### 2.5 Simulation of Feed Conditions

For the implementation of the revamped  $\text{H}_2\text{SO}_4$  plant simulation in ASPEN HYSYS, two feed streams were considered including sulfur and air having the process conditions and parameter specifications. The data were provided from Awash Melkasa sulfuric acid and aluminum sulfuric factory, Adama, Ethiopia. Table 1 presents process conditions and component composition considered for the simulation.

**Table 1.** Feed stream conditions

Stream name	Sulphur	Air
Vapour/phase fraction	0.00	1.00
Temperature [C]	25.00	25.00
Pressure [kPa]	1500.00	1500.00
Molar flow [kgmole/h]	27.12	51.99
Mass flow [kg/h]	869.5	1500.00
Std ideal liq vol flow [m <sup>3</sup> /h]	0.48	1.73
Molar enthalpy [kJ/kgmole]	-77327.98	-94.07
Molar entropy [kJ/kgmole-C]	448.15	129.07
Heat flow [kJ/h]	-2096952.45	-4891.41
Major components (mole fraction)		
S	1	0
O <sub>2</sub>	0	0.21
N <sub>2</sub>	0	0.79
H <sub>2</sub> O	0	0
H <sub>2</sub> SO <sub>4</sub>	0	0
SO <sub>2</sub>	0	0
SO <sub>3</sub>	0	0

### 3 Result and Discussion

#### 3.1 Column Design Analysis

A SO<sub>3</sub> absorption column is a unit where gas flows up and liquid (H<sub>2</sub>SO<sub>4</sub>) flows down. SO<sub>3</sub> is transferred from the gas phase to the liquid phase where it reacts with the H<sub>2</sub>SO<sub>4</sub> to form oleum (H<sub>2</sub>SO<sub>4</sub>.SO<sub>3</sub>). The gas and liquid phases are made to get in contact by the help of column plates or random or structured packing. In this work plate type absorption column is selected for making ease of maintenance. The numbers of stages are 10 with 1.5 m internal diameter and tray spacing of 0.5 m. Figure 2(a) shows the temperature and pressure profiles of the intermediate absorber. Across the column from top to down arrangement, the pressure is 1 atmosphere however there is a slight rise in temperature in the first two stages from 156.9 to 159.9 °C. One of the most decisive considerations in designing gas absorption towers is to study whether or not temperatures in the tray along the height of the tower is varied. The solute solubility usually depends strongly on temperature. From stage 1 to stage 7 the temperature of the trays were isothermal at 159.4 °C. The equilibrium shows that the remaining stages including 7, 8, and 9 to 10 are at a high tray operating temperature with the corresponding values of 159.3, 159.4, 160 and 170 °C respectively. Figure 2 (b) and (c) presents the net liquid and vapor at every tray positions and the pictorial appearance of the intermediate absorption column.

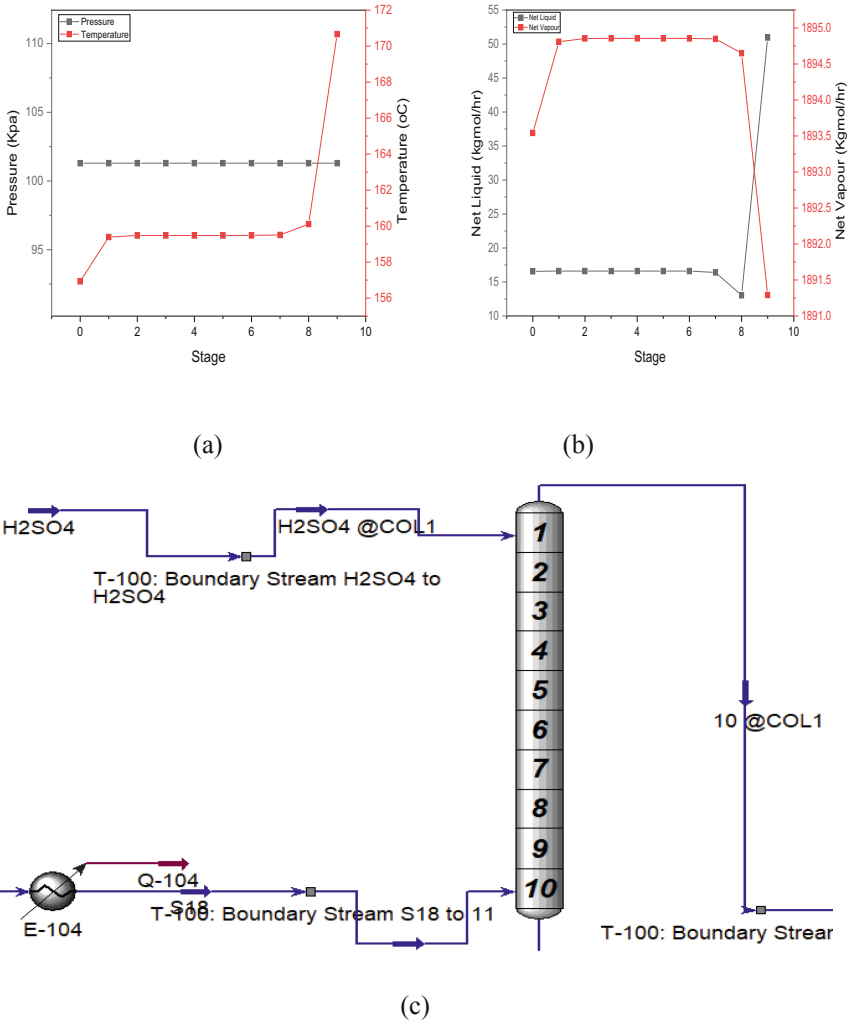
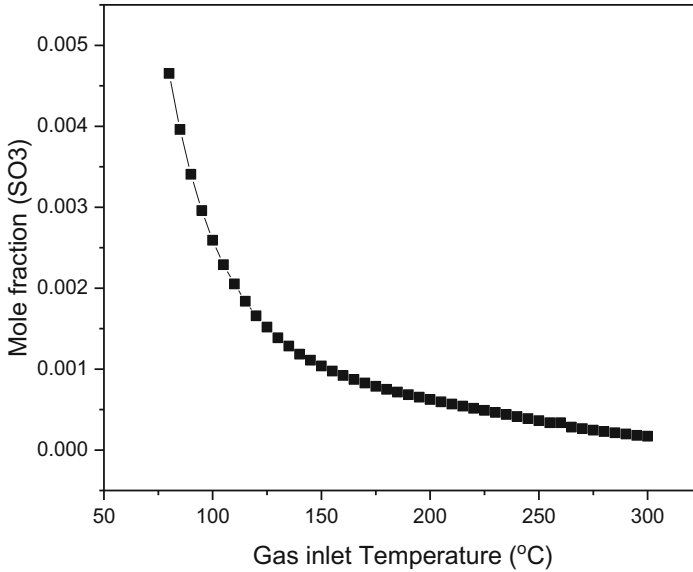


Fig. 2. Parameters profile for the intermediate against tray position (a) Temperature and pressure (b) net liquid and vapor (c) simulated unit block of absorption column

### 3.2 Effect of Inlet Temperature on SO<sub>3</sub> Absorption

An increase in gas and liquid inlet temperature leads to improve the absorption of SO<sub>3</sub> at equilibrium. Simulation results based on constant stage efficiency are shown in Fig. 3. As the temperature of the inlet gas stream is increasing from 80 °C to 300 °C,

the column exhibits a significant absorption of SO<sub>3</sub> in the range of 0.0046 to 0.00016. In practice, a higher temperature gives a higher absorption and reaction rate [25]. The result obtained by the simulation sensitivity analysis on the inlet temperature against SO<sub>3</sub> removal proved similar trend.



**Fig. 3.** Effect of inlet temperature on SO<sub>3</sub> removal in the absorption column

### 3.3 The Effect of Different Types of Recycles Setup

The top product from absorption column containing O<sub>2</sub>, N<sub>2</sub> and SO<sub>2</sub> was recycled to in different design options. The first design option (Fig. 4 (a)) is building the recycle stream to the first converter. This configuration is by far results a higher removal of SO<sub>2</sub> which is 0.12%. The second design option (Fig. 4 (b)) fed the recycle stream directly to the second converter. These options dropped the removal capacity to 0.84%. Equal amount of removal of SO<sub>2</sub> was predicted by the third converter (Fig. 5 (c)). However the setting up the recycle to the last converter (Fig. 4 (d)), the removal of SO<sub>2</sub> was lower than the other subsequent converters that resulted a 1.08% SO<sub>2</sub> removal. High removal of SO<sub>2</sub> at the secondary absorber was noted because the unconverted SO<sub>2</sub> was subjected to a sequence of beds that increase the reaction time of SO<sub>2</sub> with excess O<sub>2</sub> at each bed. The recycled stream containing a large amount of SO<sub>2</sub> and other

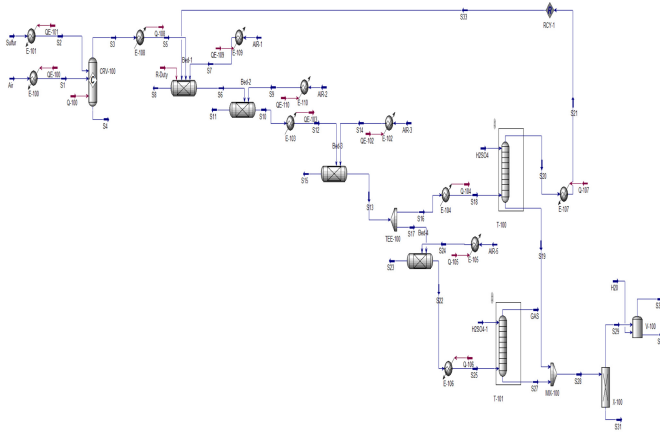
gases gas requires additional dosage of O<sub>2</sub>. In every column, there is an excess O<sub>2</sub> supplied from air dryer to every converter to facilitate a complete conversion of SO<sub>2</sub> to SO<sub>3</sub> because according to Le Chatelier's Principle, Increasing the concentration of oxygen in the mixture causes the shifting of the equilibrium position towards the product side. Therefore the need of shifting the equilibrium position to the right side in order to produce the maximum possible amount of sulfur trioxide in the equilibrium mixture is the other design consideration (Table 2).

**Table 2.** Mole fractions results obtained at different recycle setup

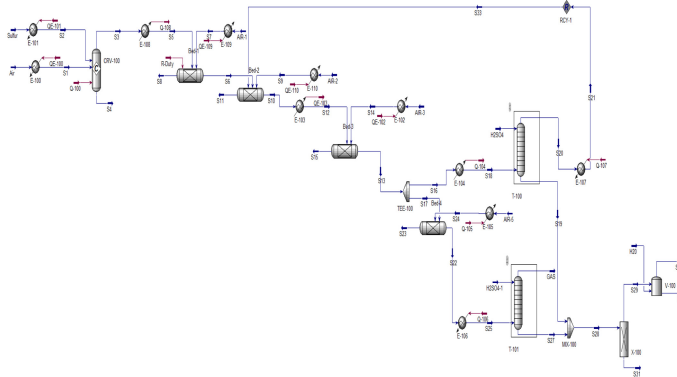
Components	Design option 1	Design option 2	Design option 3	Design option 4
H <sub>2</sub> O	0.0000	0.0000	0.0000	0.0000
SO <sub>2</sub>	0.0012	0.0084	0.0084	0.0108
SO <sub>3</sub>	0.0245	0.0142	0.0142	0.0183
H <sub>2</sub> SO <sub>4</sub>	0.0021	0.0012	0.0011	0.0036
Nitrogen	0.7942	0.7949	0.7949	0.7926
Oxygen	0.1779	0.1814	0.1814	0.1723
S_Liquid	0.0000	0.0000	0.0000	0.0025
Energy consumption				
Cooling duty (KJ/hr)	8.327E + 07	8.167E + 07	8.11E + 07	6.435E + 07
Heating duty (KJ/hr)	8.122E + 07	8.062E + 07	8.015E + 07	6.345E + 07
Total	1.6449E + 07	1.6229E + 07	1.6125E + 07	1.278E + 07

### 3.4 Effect of Split Ratio on the Amount of SO<sub>2</sub>

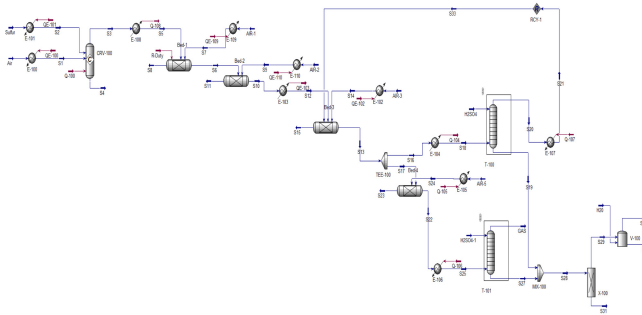
Varying the split ratio of the 3<sup>rd</sup> converter product stream into intermediate absorption column and 4<sup>th</sup> column brought a significant change in the final concentration of the product. Figure 4 shows the variation of split ratio from 0.1 to 0.9 with respect to its corresponding calculated SO<sub>2</sub> mole fraction. Increasing the split ratio from 0.1 to 0.2% lifted the SO<sub>2</sub> amount from 1% to 1.6%. Further increased split ratio from 0.2 to 0.9 reduces the amount of the SO<sub>2</sub> amount to 0.002 (0.2%) which at the end give the optimal point of value set for split ratio. In overall perspective, the amount of SO<sub>2</sub> decreased significantly within the entire range of the split ratio. The Le Chatelier's principle states that equilibrium favor to forward side when removing the product SO<sub>3</sub> from catalytic converter.



(a)



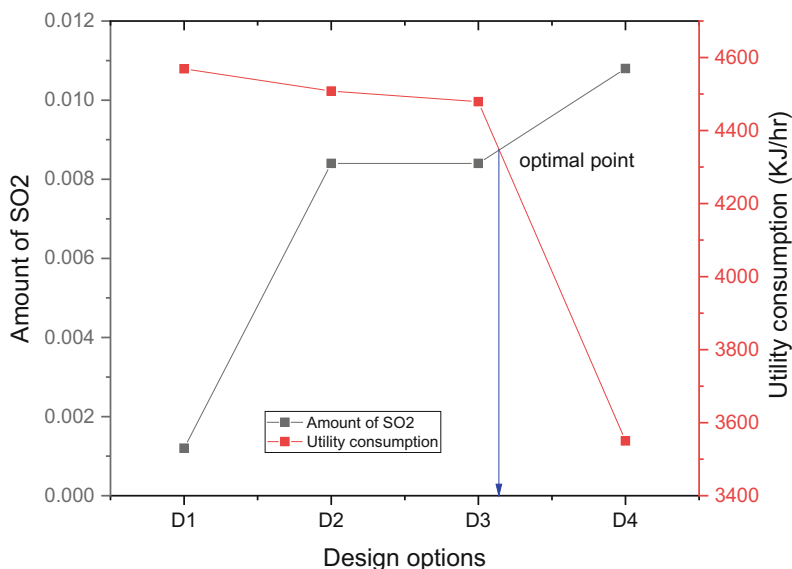
(b)



(c)

**Fig. 4.** Design options for H<sub>2</sub>SO<sub>4</sub> production where unconverted SO<sub>2</sub> is recycled to (a) to 1<sup>st</sup> converter (b), to 2<sup>nd</sup> converter (c), to 3<sup>rd</sup> converter and (d) to 4<sup>th</sup> converter





**Fig. 6.** Design options and their Trade-off between mole percentage of SO<sub>2</sub> amount and utility consumption

## 4 Conclusion

This research mainly emphasis on the reduction of SO<sub>2</sub> emission that contributes to the acid rain and substantially plant growth. The Antoine property package was used due to its compatibility with the actual process condition to handle the chemical reacting system and the separation process as well. It was proved that retrofitting the existing H<sub>2</sub>SO<sub>4</sub> plant from SCSA to DCDA potentially has shown a promising result. Additionally, four different case studies were considered for setting up the recycle stream that contains SO<sub>2</sub> gas after concentrating SO<sub>3</sub> with H<sub>2</sub>SO<sub>4</sub>. Hence design option 1 (configuring gas effluent from the absorber to the 1st converter) that was able to drop the SO<sub>2</sub> amount to only 0.12% which is much lower that what is being released now a days using SCSA. The trade-off between power consumption and SO<sub>2</sub> emission reveled that setting up the recycle stream to directly the 3<sup>rd</sup> converter give an optimal power consumption and SO<sub>2</sub> emission. However, controversy between environment and cost should not be compromised for sustainability and a cleaner production.

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