

## REDUCING SULFUR EMISSION IN A COMMERCIAL SCALE SULFUR RECOVERY UNIT (SRU) USING ARTIFICIAL NEURAL NETWORK: R APPLICATION

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### Abstract

The main objects of this research are developing an artificial neural network (ANN) model in R environment for simulating an industrial scale modified Claus unit, and optimizing temperatures of Claus catalytic reactors to achieve the minimum sulfur emission. The input layer of the ANN model consists of acid gas flow rate, reactors inlet temperatures, and H<sub>2</sub>S and SO<sub>2</sub> contents of acid gas, while the output neuron is the sulfur content of the tail gas (TG). The results confirmed that the AAD% (average absolute deviation) of the ANN model for training and validating data are 1.164% and 3.429%, respectively. After validating the model, optimal temperatures of input streams to the catalytic bed are calculated considering the sulfurous compounds of the TG as the objective function. Simulation results prove that the sulfur content of the TG decreases from the average value of 0.6 mol% to about 0.3 mol% that considerably reduces the sulfur emission of the target Claus process.

**Keywords:** Claus process; artificial neural network; optimization; sulfur emission; R software.

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## 1. Introduction

H<sub>2</sub>S (hydrogen sulfide) is a byproduct of processing natural gas and refining high sulfur crude oils [1]. This species is a toxic gas, and consequently, its present in any exhaust gas is under stiff environmental regulations. To meet these requirements, H<sub>2</sub>S must be converted to elemental sulfur which is mainly accomplished in a sulfur recovery unit (SRU). To do such a task, Claus process is widely used nowadays in a modern configuration succeeds the one pioneered in 1883 based on the reaction of H<sub>2</sub>S over a catalyst with air (oxygen) to produce sulfur and water [2] that typically recovers 95 to 97 percent of the H<sub>2</sub>S feed stream [3]. Approximately 90 to 95 percent of recovered sulfur is produced by this technology.

Few articles have been reported on the fundamental modeling and simulation of the Claus process [4-7]; however, there are scarce studies on using black-box modeling approach for this process. In recent years, there has been a significant increase in the use of artificial intelligence based techniques in the field of industrial control. Different intelligent techniques (black-box modeling approaches) like fuzzy logic, artificial neural network (ANN) and other hybrids were successfully implemented for various control problems. Prakash et al. studied optimal control of incinerator in a sulfur recovery unit by an ANN model [8]. This model had four inputs including O<sub>2</sub> and SO<sub>2</sub> concentration, the temperature of the incinerator and total flow rate of the tail gas. The outputs of the network controlled parameters of the process i.e., fuel gas and air flow rates and oxygen concentration in the input air. It was found that the model was able to successfully approximate the training data and generate the inputs for the plant for its optimal operation. Abdali et al. studied estimation of the temperature of Claus furnace using ANN and control of the Claus reactions. In this research, the amount of H<sub>2</sub>S content was

adjusted aware of the air flow rate [9]. Moreover, output temperatures of Claus reactors was estimated via adaptive linear neuron networks.

In this research, based on data collected from a modified SRU in an Iranian gas refinery, the sulfur content of the tail gas (TG) is modeled applying the ANN modeling methodology. This model which is implemented in R has a feed-forward structure, and it is composed of 6, 3 and 1 neurons in the input, hidden and output layers, respectively. By using the proposed model, the sulfur emission from the target plant is minimized. Due to the importance of the SRU from the environmental viewpoint, this research and the proposed methodology is significant.

## 2. Process description

A block flow diagram of the understudy Claus process unit is presented in Figure 1. The acid gas from the upstream (i.e., sweetening unit) is directed to the acid gas KO drum D-01. The gas comes from this KO drum is warmed up to 220°C in the E-01 by using HP steam, and then goes through the reaction furnace burner (H-01). By using the compressor (K-01 A/B), the required air is introduced to the pre-heater (E-02), and heated by high pressure steam before entering the furnace burner H-01.

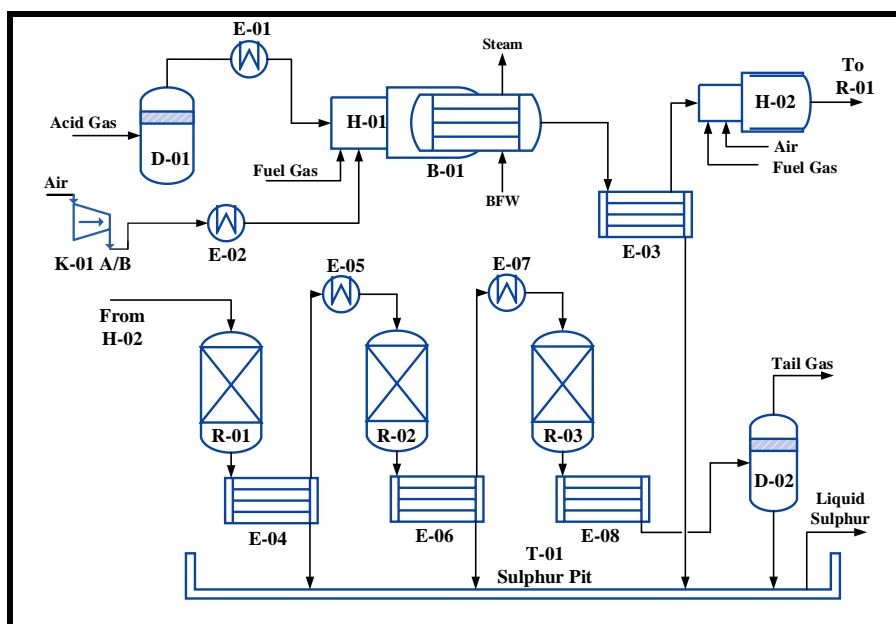


Figure1. Block flow diagram of the target Claus unit

The process gas from furnace burner (H-01) is cooled in passing through the reaction furnace boiler B-01, and then routed to the 1<sup>st</sup> condenser (E-03) where the gases are cooled, and sulfur is formed. The condensed sulfur is removed through a seal pot to the sulfur degassing pit (T-01). The process gases pass through the auxiliary burner (H-02), where they are reheated up to 235°C before introducing the first catalytic reactor (R-01). In this reactor, additional sulfur is produced and scraped in the vapor phase by the process of hot gases. Due to the exothermic nature of Claus reactions, the temperature increases across the catalytic bed. However, the inlet temperature of the reactor should be adjusted such that the outlet temperature of the reactor reaches to at least 300°C for promoting the hydrolysis of COS and CS<sub>2</sub>.

The hot gases leaving the 1<sup>st</sup> reactor are cooled in the 2<sup>nd</sup> condenser (E-04) by generating LP steam. The condensed sulfur is also sent to the sulfur degassing pit (T-01). The gases leaving the condenser (E-04) first are reheated up to 215°C by the heater (E-05) and then routed to the 2<sup>nd</sup> catalytic reactor converter (R-02). The hot gases leaving the second converter (R-02) are cooled in the 3<sup>rd</sup> condenser (E-06) by generating LP steam. The condensed sulfur flows through a seal pot to the sulfur degassing pit (T-01). Process gases leaving the

3<sup>rd</sup> condenser is reheated up to 200°C by HP steam in the heater (E-07), prior to entering the 3<sup>rd</sup> catalytic reactor (R-03). The process gases pass downwards through the catalyst bed of third reactor (R-03). The gases are then cooled to 130°C in the last condenser (E-04) in order to condense the sulfur and to decrease its losses by vapor pressure in the tail gas. The last condenser ensures cooling of gases by generating very low LP steam, which is condensed in an air cooler. Prior to being incinerated, the effluent gas from E-03 introduces to the last equipment (D-02), and any mists and drops of liquid sulfur are collected.

### 3. Process modeling and optimization

The modeling strategy used in this research is the same as the methodology presented in the previous works [10–11]. In the current study, an ANN model is implemented in R software (Version 3.4.1, Copyright 2017) for simulating the sulfur content of TG (i.e. sum up molar percentages of H<sub>2</sub>S and SO<sub>2</sub>). R is a language and environment for statistical computing and graphics which was developed at Bell Laboratories (formerly AT&T, now Lucent Technologies) by John Chambers and colleagues. The significant benefit of using R is its opportunity as free software under the terms of the Free Software Foundation's GNU General Public License in source code form. To develop and construct the corresponding ANN model, the package 'neuralnet' (Version 1.33) obtained from the CRAN repository is applied. This package permits resilient settings through custom-choice of error and activation function, and also the calculation of generalized weights is implemented [12]. The globally convergent algorithm of 'neuralnet' is based on the resilient back-propagation without weight back-tracking, and additionally modifies one learning rate, either the learning rate associated with the smallest absolute gradient [13].

The most widely employed networks have one hidden layer only. Each node within a given layer is connected to all of the nodes of the previous layer. The node sums up the weighted inputs and bias, and it passes the result through a linear function as follows [14]:

$$a_j = \sum_{i=0}^m w_{ji} y_i + b_j \quad (1)$$

In Eq. (1),  $w_{ji}$  is the weight that goes from the input ( $i$ ) to the hidden neuron ( $j$ );  $b$  is the bias to the node, and  $y_i$  is the input unit of the neuron. By utilizing an activation function( $f$ ), the output of the neuron can be written as follows:

$$Z_j = f(a_j) \quad (2)$$

By using 'neuralnet' package, a feed-forward ANN with one hidden layer is developed to simulate the sulfur content of the TG. The input layer of Claus ANN model is contained in acid gas flow rate ( $F_{in}$ ), temperatures of input streams to the catalytic reactors ( $T_{inR1}$ ,  $T_{inR2}$ , and  $T_{inR3}$ ), and H<sub>2</sub>S and SO<sub>2</sub> contents of acid gas. The output layer has one node which is the summation of SO<sub>2</sub> and H<sub>2</sub>S contents of the TG ( $InH_2S$  and  $InSO_2$ ). To inhibit from memorizing instead of learning, only 3 neurons are selected for the hidden layer such that the number of coefficients should be less than the number of real data points obtained from the target SRU plant.

To evaluate the accuracy of the model, the absolute average deviation (AAD%) and mean squared error (MSE) between the actual and predicted data are calculated as follows:

$$AAD\% = \frac{\sum_{n=1}^{N_t} \sqrt{\frac{(y_n^{actual} - y_n^{model})^2}{y_n^{actual^2}}}}{N_t} \times 100 \quad (3)$$

$$MSE = \frac{1}{N_t} \sum_{n=1}^{N_t} (y_n^{actual} - y_n^{model})^2 \quad (4)$$

where  $y$  and  $N_t$  are output variables (i.e. sulfur content of TG) and number of data points, respectively; superscripts *actual* and *model* indicate the actual (or real) and the simulated values, respectively.

Finally, to optimize the target modified Claus process, the sum of H<sub>2</sub>S and SO<sub>2</sub> content of the TG is minimized by manipulating the temperatures of the input temperatures to the catalytic reactors (1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> Claus reactors). To be engaged such a problem, 'optimx' package available in R software is applied. This package provides a replacement and extension of the

'optim' function of R toolbox to unify and streamline optimization capabilities for smooth [15]. In this study to carry out the optimization procedure, Large-scale Bound-constrained (L-BFGS-B) method is utilized which is a limited-memory quasi-Newton code for bound-constrained optimization.

## 4. Results and discussions

### 4.1. Modeling the target SRU

During 90 days, 90 data points including acid gas flow rate, temperatures of input streams to the catalytic reactors, H<sub>2</sub>S and SO<sub>2</sub> contents of acid gas, and sulfur content of TG were obtained from the understudy Claus plant. These data points were divided into two categories: training and validating groups which were included 63, and 27 points (70% and 30% of total data), respectively. The topology of ANN constructed by R for modeling sulfur content of TG is demonstrated in Figure 2. As seen, each network has 21 weights and 4 biases, totally 25 coefficients; therefore, the number of model variables is less than the half of the training data, and the degree of freedom (DOF) for training the network is equal to 38. It seems that this positive DOF restrains from over learning instead of training.

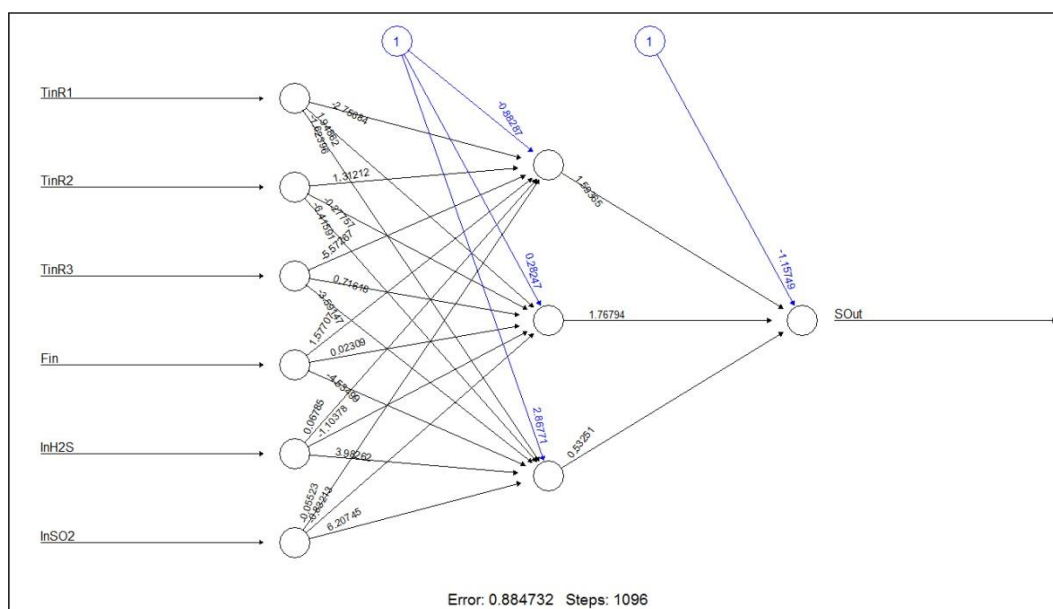


Figure 2. The topology of ANN constructed by R for predicting total sulfur of TG in the target SRU

The results obtained for simulating the sulfur content of TG is presented in Table 1. From this table, it is concluded that the AAD% and MSE of training data (1.164% and 0.057, respectively) are less than those of belong to testing or unforeseen data points (3.429% and 0.095, respectively), and they are relatively close together. To further explore the validity of the model, in Figure 3, the actual data on the sulfur content of TG are sketched against trained and validated values obtained from the developed ANN model. As seen, the ANN model has acceptable accuracy, and therefore it can be applied for the purpose of optimizing the modified Claus process. We believe that the observed deviation is mainly due to the error of measuring H<sub>2</sub>S and SO<sub>2</sub> concentrations of the TG.

Table 1. AAD% and MSE of the ANN model in comparison to the actual data

Product	Training data (%)	Validating data (%)
AAD%	1.164	3.429
MSE	0.057	0.095

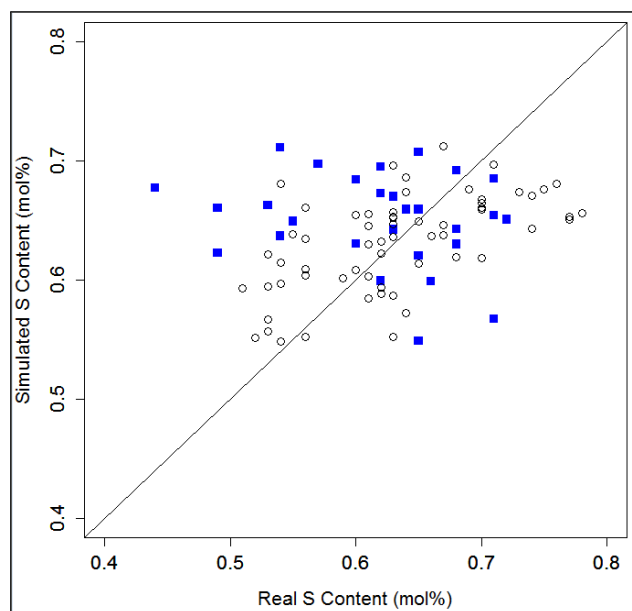


Figure 3. Comparison of real and simulated (trained (○) and validated (■)) values of the total sulfur of TG

#### 4.2. Reducing the sulfur emission

After developing and validating the ANN model for the target modified Claus process, this model is ready for being applied to reduce the sulfur emission of the SRU. Therefore, for all 90 test runs, the optimized feed temperatures of Claus convertors are calculated by using the proposed model. Then, for each case, temperatures are adjusted to minimize the sulfur content of the TG. After applying the optimal decision variables, the comparison between the optimal values of TG sulfur content versus the current (actual) ones is presented in Figure 4. As seen, the obtained optimum inlet temperatures can decrease the sulfur content of TG by approximately 0.3 mol% which is equal to 50% decrease in sulfur emission. It is obvious that lowering the concentration of sulfurous compounds in the TG is a momentous achievement both from the environmental and economic viewpoints.

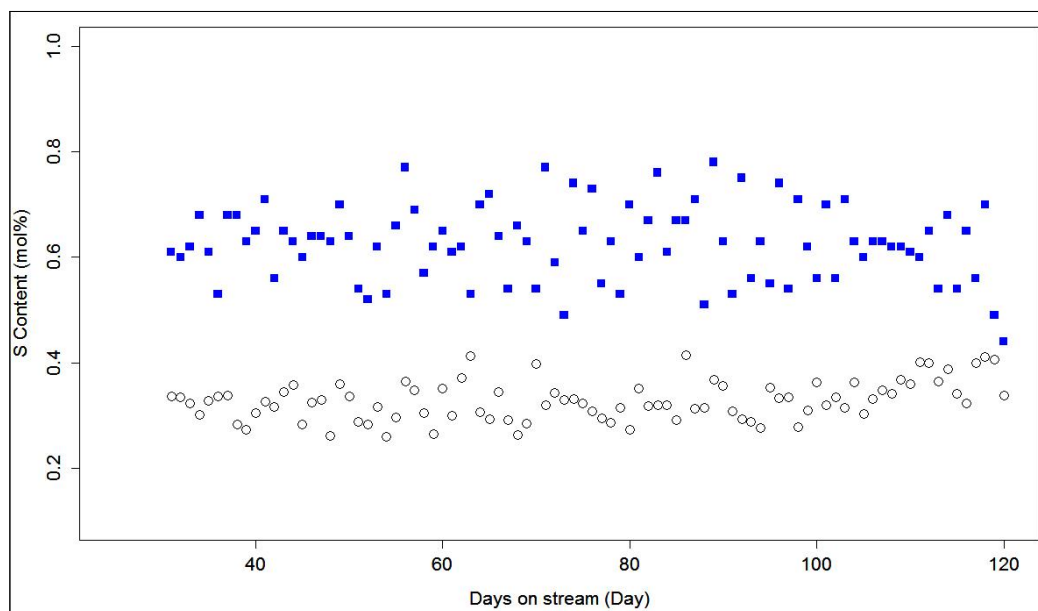


Figure 4. Comparison of the actual (■) and optimized (○) values of the total sulfur of TG

The results related to the optimum values of the input stream temperatures of catalytic beds against the actual values are presented in Figures 5 to 7. As seen in Figure 5, the optimum temperature of the first convertor should be adjusted to the values lower than the actual temperature. This phenomenon is expectable due to the exothermic nature of the Claus reactions; therefore, a lower temperature is desired to supply a suitable thermodynamic condition for higher  $\text{SO}_2$  and  $\text{H}_2\text{S}$  conversion [16]. On the other hand, in the 2<sup>nd</sup> and 3<sup>rd</sup> catalytic reactors, the degree of temperature reduction is limited by the sulfur dew point such that below this constraint, the sulfur conversion decreases because of the blockage of the catalyst surface, and the plausible deactivation [17]. Therefore, in opposite to the thermodynamic equilibrium, the temperature of the feed introduced to the 2<sup>nd</sup> and 3<sup>rd</sup> convertors should be meticulously elevated (see Figures 6 and 7).

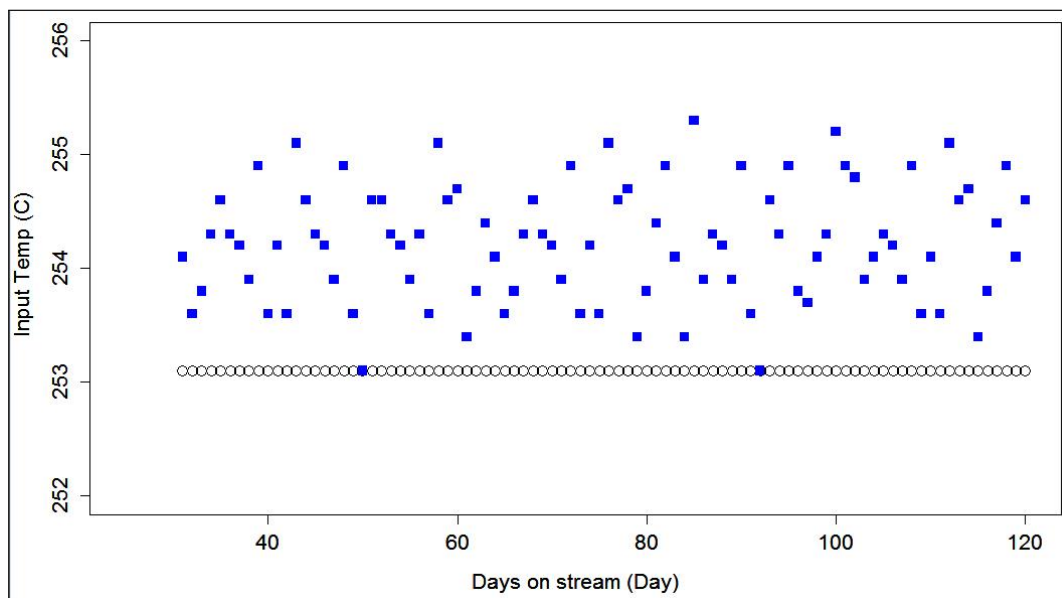


Figure 5. Comparison of the actual (■) and optimized (○) values of the inlet temperature to the 1<sup>st</sup> Claus reactor

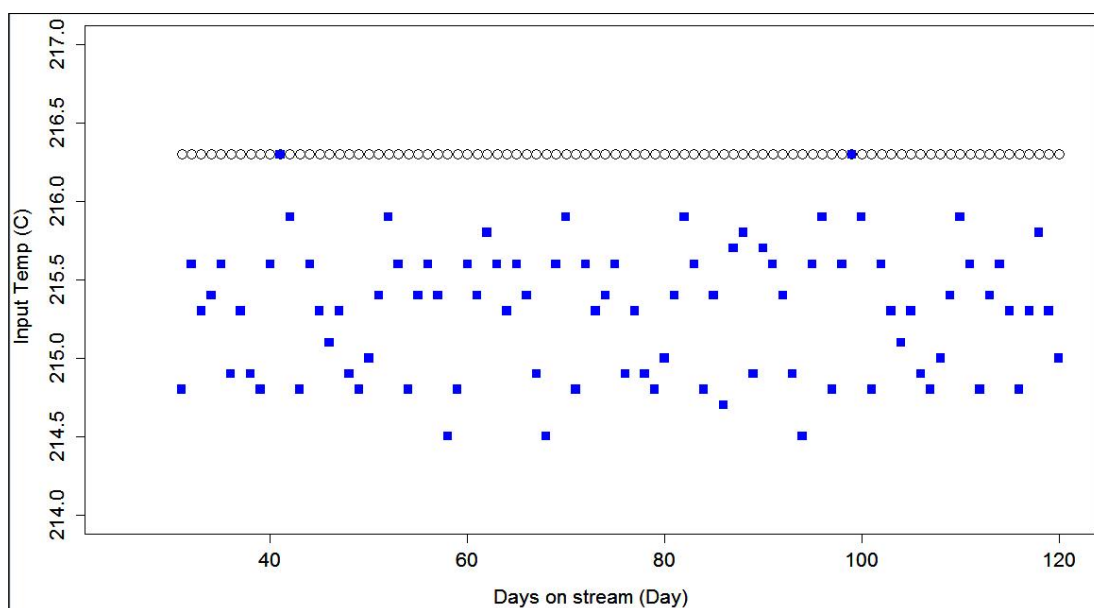


Figure 6. Comparison of the actual (■) and optimized (○) values of the inlet temperature to the 2<sup>nd</sup> Claus reactor



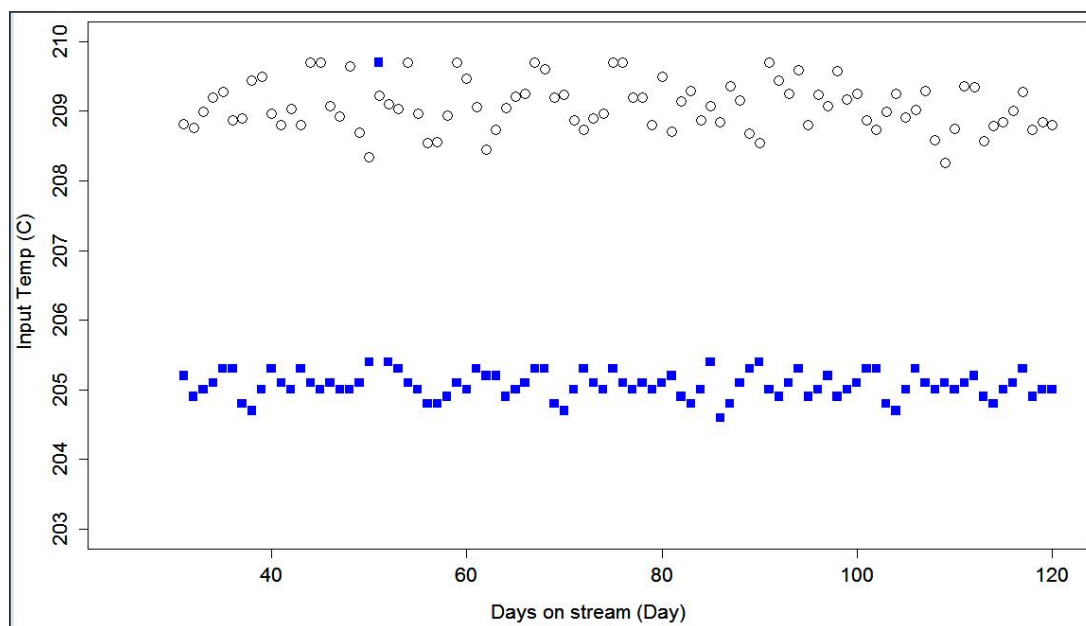


Figure 7. Comparison of the actual (■) and optimized (○) values of the inlet temperature to the 3<sup>rd</sup> Claus reactor

## 5. Conclusions

A commercial-scale modified Claus unit was modeled based on the ANN methodology developed in R software. This model was a feed-forward neural network including six neurons in the input layer (i.e. acid gas flow rate, temperatures of input streams to the catalytic reactors, and H<sub>2</sub>S and SO<sub>2</sub> contents of acid gas), three neurons in the hidden layer, and one neuron in the output layer (i.e. sum of SO<sub>2</sub> and H<sub>2</sub>S contents of TG). The model was trained and validated against the actual data points which were gathered from the target plant during 90 days. The results showed that ANN could simulate the sulfur content of training and validating data (63 and 27 data points, respectively) with an AAD% of 1.164% and 3.429%, respectively. The MSEs of the model for those data were 0.057 and 0.095, respectively. These results confirmed the reliability of the constructed ANN model.

To suggest an application for the developed ANN model, the sulfur content of the TG was minimized by manipulating the temperatures of feed streams entered to the catalytic converters of Claus process. The results proved that within the admissible range for the adjusted variables, the average sulfur content of the TG could be decreased from 0.6 mol% to 0.3 mol% (about 100% reduction in sulfur emission).

It is supposed that applying optimal decision variables in the target modified Claus process can decrease sulfur contaminant emission which has a great impact on the environmental and economic aspects of the process.

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