Nonlinear Model Predictive Control for Modified Claus Process

Jialin Liu,a,\* Hao-Che Chien,b David Shan-Hill Wong,b Chun-Cheng Changc

aResearch Center for Smart Sustainable Circular Economy, Tunghai University, Taiwan

bDepartment of Chemical Engineering, National Tsing Hua University, Taiwan

cGreen Energy and System Integration Research and Development Department, China Steel Corporation, Taiwan

jialin@thu.edu.tw

Abstract

The Claus process is the most significant gas desulfurizing process in industry, recovering elemental sulfur from gaseous hydrogen sulfide. However, the reactor temperature suffers a nonlinear process gain to the sulfur recovery because the Claus reaction is exothermic and reversible. In field operations, a traditional feedback controller for the outlet of SO2 is left in open loop, because the disturbances cannot be compensated by the linear controller. In this study, a nonlinear controller is designed by the differential evolution algorithm with a surrogate model that is constructed by the sequence-to-sequence network. The results show that the process nonlinearity can be conquered and the emitted SO2 concentration can be controlled by the reactor temperature.

**Keywords**: Nonlinear process control, modified Claus process, surrogate model.

* 1. Introduction

In the iron and steel industry, raw materials for steel production are coal and iron ore. The iron ore is sintered and taken to the blast furnace where it is mixed with coke, lime and blown air to produce pig iron. On the other hand, the coal is carbonized to produce coke at temperatures higher than 1000 °C in coke oven units. Coke oven gas (COG) is a by-product of the coke-making process, where volatile coal matter is generated as COG, leaving carbon intensive coke behind. The raw COG is carried out to washing stages where the regenerated water from the H2S stripper is introduced at the top of the absorber to remove hydrogen sulfide from the COG. The captured H2S is cracked and converted into H2O and elemental sulfur through the Claus process. The cleaned COG, which is mainly composed of hydrogen and methane, is presently used for heating purposes in coke batteries or furnaces. The original Claus process produces elemental sulfur by oxidation of hydrogen sulfide with air at elevated temperatures using a metal oxide catalyst: H2S + 1/2 O2 → 1/8 S8 + H2O. However, the associated temperature rise could not be controlled when significant amounts of H2S are present. The drawback is overcome by dividing the reaction into thermal and catalytic steps, which is usually referred to as the modified Claus process. In the process, almost one third of H2S is oxidized into SO2 in the thermal reactor (furnace); thereafter, the serial catalytic reactors convert the left part of H2S with SO2 into elemental sulfur and H2O (Clark, 2000).

* 1. Process description

Claus process consists of a thermal reaction furnace, a waste heat boiler (WHB), and a series of catalytic reactors and sulfur condensers, as shown in Figure 1. Due to the upstream load variations, a flow ratio controller is installed to maintain the primary air (F2) to the sour gas (F1) ratio. In the furnace, temperatures are usually in the order of 1000–1400 °C. A controller maintains the top temperature of burner (T8) through altering the set point of the fuel flow rate (F5) where an air-to-fuel ratio is regulated by adjusting the set point of the combustion air flow rate (F4). The oxidizing reaction, H2S + 3/2 O2 → SO2 + H2O, is exothermic and without any thermodynamic restriction. At the high temperature, some of the sulfur dioxide produced here reacts with hydrogen sulfide inside the furnace according to reaction: 2 H2S + SO2 ↔ 3/2 S2 + 2 H2O to produce elemental sulfur. The hot outlet gas passes to the WHB at the end of combustion chamber and cools down to 300 °C by converting high pressure steam to the medium pressure.



Figure 1. Flow diagram of modified Claus process.

The remaining H2S, from the furnace, is reacted with the SO2 at lower temperatures (about 200–300 °C) over an Al2O3 catalyst to make more sulfur: 2 H2S + SO2 ↔ 1/2 S6 + 2 H2O. In the catalytic stage mostly S6 is produced, which is an exothermic reaction whereas in the thermal stage S2 is the major product and the reaction is endothermic (Clark, 2000). The catalytic stages are operated at successively lower temperatures to achieve increased conversion to elemental sulfur. In the Claus reaction, the furnace produces SO2 used to react with H2S in a ratio of 2 (H2S/SO2). Therefore, a proper amount of oxygen, typically in the form of air, must be introduced to maintain the stoichiometric number. A tail gas analyzer is installed after the final catalytic stage, as shown in Figure 1. The set point of the secondary air flow rate (F3) is determined by the output of the molar ratio controller (H2S/SO2) from the tail gas analyzer. A condenser is installed downstream each converter to condensate the elemental sulfur. Thereafter, a reheater heats the effluent gases before entering the next converter. As shown in Figure 1, the inlet temperature of the second converter (T2) is controlled by bypassing the outlet flow from the previous converter, whereas the inlet temperature of the first converter (T1) is not regulated. The SO2 concentration in the tail gas determines the set point of T2 controller. However, the inlet temperatures of the gases have a positive process gain to the sulfur recovery when the reaction temperatures are far below that of the equilibrium states, i.e., the rising of inlet temperatures may result in the reduction of the SO2 concentration in the tail gas. On the other hand, the process gain of inlet temperatures to the sulfur recovery becomes negative once the reaction temperatures are higher than that of the equilibrium states due to the exothermic reaction. Therefore, the practical operation of a Claus converter is an interplay between equilibrium limitations and reaction kinetics.

* 1. Process model

In the present work, Aspen Plus V14 was used to simulate the Claus process in Figure 1 where the ideal gas model was applied. The reactions and their kinetic parameters for the furnace and converters were retrieved from the work of Nabikandi and Fatemi (2015). The feed conditions of the sour gas, combustion air, and fuel were also adapted from the same paper. The furnace length and diameter were 6.5 and 3.4 (m), in which the burner and combustion chamber are included. The adiabatic plug flow reactor (PFR) was applied to simulate the furnace and a heat exchanger was used for the WHB. For each catalytic converter, the length and diameter respectively were 6.0 and 1.4 (m), in which the catalyst loading is 36,000 (kg) with the particle density of 1.3 (g/cm3). The temperatures of the sulfur condensers were maintained at 120 °C and the inlet gas temperatures for the converters were set at 230 and 210 °C, respectively. Table 1 lists the reported industrial data and the simulation results where the relative errors for the outlet temperatures and total molar flows are below 10%. In addition, the simulation results of H2S and SO2 (mol/s) from the reactors are comparable with the reported industrial data.

Table 1. Simulation results and reported industrial data (Nabikandi and Fatemi, 2015)

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| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | Furnace | | | | Converter 1 | | | | Converter 2 | | | |
|  | Temp. (°C) | Total (mol/s) | H2S (mol/s) | SO2 (mol/s) | Temp. (°C) | Total (mol/s) | H2S (mol/s) | SO2 (mol/s) | Temp. (°C) | Total (mol/s) | H2S (mol/s) | SO2 (mol/s) |
| Industrial case data | 1000 | 355.82 | 12.28 | 9.73 | 307 | 335.29 | 5.41 | 2.70 | 222 | 331.93 | 1.45 | 0.73 |
| Simulation result | 1048 | 354.65 | 13.45 | 7.51 | 298 | 309.28 | 6.44 | 1.09 | 214 | 301.80 | 5.51 | 0.62 |

|  |  |
| --- | --- |
| (a) | (b) |

Figure 2. Results of sensitivity analysis on varying the air flow rate and T2, (a) the H2S conversions, (b) correlation between the H2S conversion and the H2S/SO2 in the tail gas.

As mentioned earlier, a proper amount of oxygen is needed in the furnace to produce SO2 that maximizes the H2S conversion to elemental sulfur in the Claus process. Figure 2 shows the results of sensitivity analysis on varying the molar flow ratio of the air to the sour gas and T2, whereas T1 is fixed at 230 °C. As shown in Figure 2(a), there exists an optimal temperature that maximizes the H2S conversion for each molar flow ratio. Once the inlet temperature below that of the maximum conversion, the inlet temperature has a positive gain to the H2S conversion; on the other hand, the process gain is negative. As shown in Figure 2(b), the H2S conversions of maintaining T2 at 180 °C are higher than those of the counterparts operating at 160 and 200 °C. Figure 2(b) also shows that the maximum conversions can be found, once the H2S/SO2 in the tail gas are kept around 2. Therefore, maintaining the H2S/SO2 in the tail gas at 2 by adjusting the air flow rate, Figure 3 shows the SO2 concentrations in the tail gas under varying T1 and T2. The concave surface shows that the SO2 concentration (controlled variable, CV) suffers the nonlinear difficulty with the manipulated variable (MV) and disturbance variable (DV) where are T2 and T1, respectively.

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| Figure 3. SO2 concentrations in the tail gas on varying T2 and T1. | Figure 4. Sour gas composition variations for dynamic simulation. |

* 1. Industrial example
     1. Dynamic model

A pressure-driven dynamic model was developed by Aspen Plus Dynamics V14 based on the steady-state model described in the previous section. The configurations of control loops for the dynamic model are shown in Figure 1. In addition, the pressure controllers for the furnace, WHB, and both converters were implemented by regulating the corresponding outlet flows. The set point of T2 was determined by the tail gas analyzer controller (AIC) for maintaining the SO2 concentration in the tail gas. However, the operators complained that the AIC is not functional; consequently, the AIC and T2 controllers were kept in open loop in the daily operations. The inlet compositions of the sour gas were not analyzed in the industrial process, only the data of pressure, temperature, and volume flow rate are collected. In the present work, the molar flows of H2S and CO2 were estimated from the previous scrubbing process and the balanced H2O was estimated by the measured data of F1. Figure 4 shows the variations of the sour gas compositions for the dynamic simulation where a Gaussian distribution is applied with the standard deviation set as 2.5% of the average value for each component.

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| --- | --- |
| (a) | (b) |

Figure 5. Dynamic simulation results, (a) the temperature variations of T1 and (b) SO2 concentrations in the tail gas

In Figure 5, the ideal case simulated the inlet concentrations of sour gas by the means without variations in Figure 4. The base case simulated the field operations where T2 and the SO2 concentration in the tail gas were not regulated, and the variations of the sour gas were introduced after one hour. In the cascade case, the set point of T2 was determined by the controller of the tail gas analyzer for maintaining the SO2 concentration. Figure 5 shows the dynamic simulation results for the three cases where T1 were nearly unchanged for the ideal case in Figure 5(a). On the other hand, the temperature variations were between 224 and 248 °C for the other cases once the variations of inlet components were introduced. Figure 5(b) shows that the cascade controller for the SO2 concentration in the tail gas was not practicable, because a linear controller cannot tackle the nonlinearity between CV (SO2) and MV (T2), as shown in Figure 3.

* + 1. Surrogate model

The SO2 concentration in the tail gas, which is one of the CVs in the Claus process, exhibits a nonlinear characteristic for the MV (T2), because the Claus reaction is an exothermic and reversible reaction. In this study, a simplified nonlinear controller was designed based on a surrogate model that is constructed by the sequence-to-sequence (Seq2seq) network. The model contains the CV, MV, and DV, which are the SO2 concentration, T2, and T1, respectively. The process dynamics could be extracted by the encoder of Seq2seq, where the gated recurrent unit (GRU) was applied and trained by the data of CV, MV, and DV from the cases of Aspen dynamic simulations. The input layer of the decoder connected to the hidden state output from the encoder and the data of MV, whereas the output layer is linked to the data of CV. The details of training the surrogate model by Seq2seq can be found in the previous work (Liu et al., 2023). The modeling window length for the encoder was determined by the open-loop tests of MV and DV regarding the CV. The window length for training the encoder was selected as six where ten-minute data were averaged as a training sample. The decoder was trained using the data immediately after training the encoder. The next three step data of MV and CV were connected to the input and output layers of the decoder, respectively. Meanwhile, the hidden state output from the encoder was also linked to the input layer of the decoder. Thereafter, the network parameters of encoder and decoder were simultaneously adjusted by minimizing the root mean squared error (RMSE) of the predicted CV through backpropagation. The R2 of the training and test data were 0.92 and 0.88, respectively; meanwhile the mean absolute percentage error (MAPE) for the test data is around 5%. Therefore, the Seq2seq network is accurate to apply for the surrogate model of designing a model predictive controller (MPC).

The future MVs, which refer to the set points of T2, were evaluated by the differential evolution algorithm (DEA). The SO2 concentrations were predicted by the Seq2seq network where the input layer of the decoder received the designed set points of T2. There were three options for each step of the MV, the current temperature ±0.5 °C and keeping unchanged. Since the next three steps of SO2 were predicted in an evaluation, there were 27 possible combinations for the future set points of T2. The sums of the predicted SO2 concentrations were minimized by selecting a proper combination for the future set points. Thereafter, only the first value of the best combination was implemented into Aspen Dynamics as the set point of T2 and integrated to the next time step. Figure 6 compares the simulation results of the base case, cascade case, and DEA where the input variations were introduced after one hour. As mentioned earlier, because T2 is not regulated in the daily operations (base case), the SO2 concentrations in the tail gas suffer variations under process disturbances, as shown in Figure 6(a). In addition, the figure also shows that the cascade controller fails to regulate the SO2 concentrations after the ninth hour, because the linear controller cannot tackle the nonlinearity between the CV and MV. As shown in Figure 6(b), the MV reaches its minimum setting after the ninth hour. On the other hand, the SO2 concentrations of DEA results tend to a lower value in Figure 6(a). Figure 6(b) shows that T2 was guiding toward a lower value by the DEA, gradually.

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| (a) | (b) |

Figure 6. Results of base case and DEA, (a) SO2 concentration in tail gas, (b) the inlet temperature of the second converter.

* 1. Conclusions

The Claus reaction exhibits the exothermic and reversible characteristics that leads a nonlinear process gain for the reactor temperature to the reaction conversion. The CV suffers the process disturbances that cannot be compensated by a traditional proportional–integral–derivative (PID) controller; therefore, the field operators chose to keep the SO2 concentration controller in open-loop. In this study, the nonlinear difficulty between CV and MV was tackled by a simplified nonlinear surrogate model with the DEA. The plantwide controller for the industrial process is undergoing where the overall surrogate model is prepared to interact with the deep reinforcement learning based controller to maximize the sulfur recovery and minimize the SO2 emission, simultaneously.

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