Process Stability Identification Through Dynamic Study of Single-bed Ammonia Reactor with Feed-Effluent Heat Exchanger (FEHE)

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Abstract. In ammonia reactor system, a feed-effluent heat exchanger (FEHE) is typically installed to utilize reaction-generated heat to heat the reactor's feed. Utilizing energy from exothermic reaction to the incoming feed stream is often called "autothermic operation". Despite the advantage of FEHE, there is a risk of utilizing FEHE in a reactor system such as instability of process temperature or known as hysteresis. Hysteresis phenomena in chemical process could cause operational problems, for example it could damage the integrity of the equipment's material. This paper aims to evaluate the dynamic behavior of a single-bed ammonia reactor with FEHE, particularly to propose a way to prevent instability within the system. The dynamic simulation of the single-bed ammonia reactor with FEHE was performed with Aspen HYSYS v8.8. The result of the simulation result shows that hysteresis phenomenon in the ammonia reactor system occurs when the feed's temperature is below a certain value. If the feed temperature reaches that value, the temperature of the reactor's outlet oscillates. One of the solution to keep the feed temperature above that critical value is by installing a trim heater within the system. Based on the simulation, trim heater installation within the system is able to prevent hysteresis in the system evaluated.

1 Introduction

Ammonia production process is the one of the most important industrial-scale chemical process. Mostly, ammonia is used commercially in fertilizer and its remainder could be used as raw material for variety product, such as plastics, synthetic fibers and resins, pharmaceuticals, explosives, papers and refrigeration [1]. Despite of its importance, one part of the ammonia synthetic process, catalytic reforming reaction of nitrogen and hydrogen is the most energy consuming process that requires massive amount of heat to ensure the reaction is presented [2]. Therefore, the catalytic reforming reaction is usually supported by feed-effluent exchanger (FEHE) to make energy usage more efficient [2].

Feed-effluent heat exchanger (FEHE) is a kind of heat exchanger that widely used to utilize reaction-generated heat carried by reactor outlet as reactor inlet heating media. The hot reactor effluent is recycled back to feed preheater to provide all or a portion of the energy required to preheat the reactor feed [3]. Using FEHE is the one of the practical ways to optimize energy consumption in chemical plant. This system could be implemented in the process that involve exothermic reaction such as ammonia and methanol synthesis. Releasing energy from exothermic reaction to the incoming reactant stream is often called "autothermic operation" [4].

In steady-state study, designing heat integration in reaction system using FEHE is likely seen as interesting way to reduce energy consumption. However, some dynamic study shown that heat integration that involved FEHE could potentially lead to process instability in certain condition [5,6]. The example of process instability is the oscillation of ammonia reactor outlet temperature that lead to operation failure. The oscillation of ammonia reactor outlet temperature is shown in figure 1. Because of sudden pressure decreasing caused by decrease of fresh feed stream, oscillation of operation temperature in ammonia reactor occurred with the period of 6 minutes and range within 200°C. This oscillation condition could be damaging the catalyst in the reactor and harmful to the process. Therefore, we could investigate the dynamic behavior of ammonia reactor with the aid of simulation application such as Aspen HYSYS.

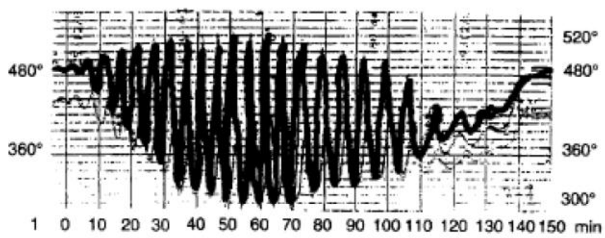

Fig. 1. Example record of industrial-scale ammonia synthesis reactor outlet temperature oscillation [1].

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2 Methodology

Ammonia reactor system is modelled using Aspen HYSYS v8.8 for steady state simulation and then converted to dynamic simulation. Peng-Robinson as fluid package is used for whole simulation. Process flow diagram scheme of the process is shown in figure 2 while simulation of the reactor system scheme is shown in figure 3.

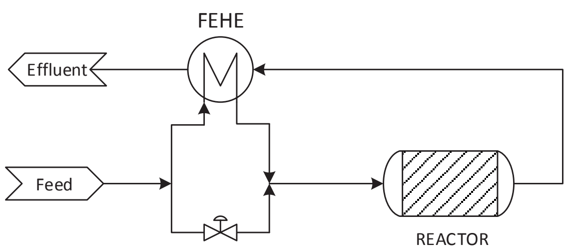


Fig. 2. Process flow diagram of single-bed ammonia reactor system with feed-effluent heat exchanger (FEHE)

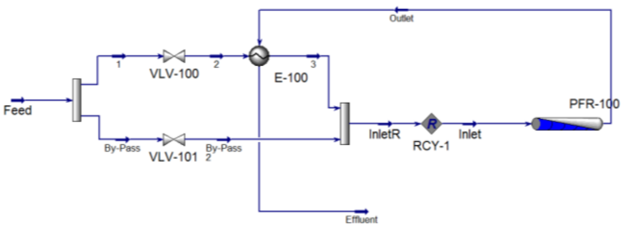


Fig. 3. Simulation scheme of autothermic single-bed ammonia reactor system with Aspen HYSYS v8.8.

Reactor feed is consisted of nitrogen, hydrogen and slightly amount of ammonia which specification is shown in table 1. Feed stream split into two part, one part which consist of 75% from total reactor feed amount is transferred to FEHE to be heated by reactor effluent and the other goes directly to the reactor after mixed with heated stream from FEHE. Specification of heat exchanger used as FEHE is shown in table 2.

Table 1. Ammonia reactor system feed stream specification.

Temperature	250°C
Pressure	163.5 bar
Mass Flow	200 ton/h
Composition	
N_2	75.6 %
H_2	16.3 %
NH_3	8.1 %

Heated feed then entered the ammonia reactor. The reaction undergoes in this reactor is assumed perfectly adiabatic. Reactor effluent that carried heat from exothermic reaction then flows through FEHE. Specification of single-bed ammonia reactor used in this study is based on [6] and presented in table 3.

Table 2. FEHE specification.

TEMA Type	BEM
Tube No.	379
Tube OD	19.05 mm
Tube Thickness	4.19 mm
Tube Pitch	23.08 mm
Tube Pattern	30-Triangular
Baffle Type	Double segmental
Baffle Cut Orientation	Vertical
Shell ID	600 mm
Shell OD	700 mm
Tube length	3500 mm
Baffle spacing	600 mm
Baffle No.	4
Shell in series	1
Shell in parallel	4

Table 3. Ammonia Reactor Specification.

Total Volume	30 m^3
Length	6.11 m
Diameter	2.50 m
Wall Thickness	5.00×10^{-3} m
Void Fraction	0.40

Reaction equilibrium and kinetics parameter, which also referred from [6], are shown in equation (1) and table 4 respectively.

$$N_2 + 3H_2 \rightleftharpoons 2NH_3 \quad (1)$$

Table 4. Reaction Kinetics Parameters.

Parameters	Forward	Reverse
N_2 Order	1.0	0
H_2 Order	1.5	-1.5
NH_3 Order	-1.0	1.0
A	23.62	3.40×10^{13}
E	87090 kJ/kmol	198464 kJ/kmol

A = Arrhenius Constant
E = Activation Energy

3 Result and Discussion

3.1 Instability phenomenon of single-bed ammonia reactor

The stability behavior of the ammonia reactor could be represented by outlet-feed temperature profile. Feed temperature is varied to obtain outlet temperature by using study case feature in Aspen HYSYS in forward step and backward step. Varying reactor inlet temperature is also performed to identify the feed-outlet temperature profile. Feed-outlet temperature profile is shown in figure 4. This relation was generated by varying reactor inlet temperature in such way to get the value of feed temperature and reactor conversion.

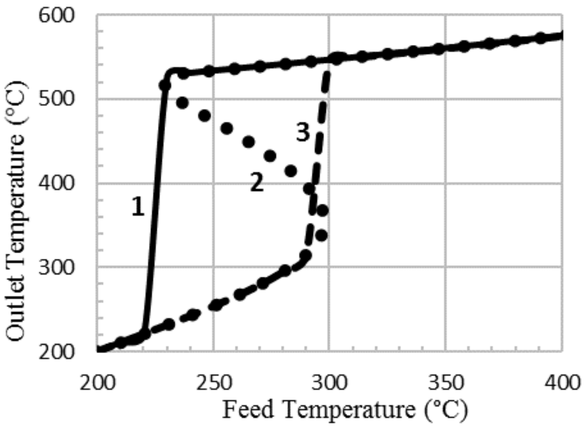


Fig. 4. Ammonia reactor feed-outlet temperature profile.

The 'S' curve, as seen in figure 4, implies that exothermic reactor could operate at several steady states condition under same operating condition depending on initial or feed conditions. There are three main information that could be obtained. First, from feed-outlet temperature profile generated by forward step (line 3), the reaction was activated when the feed temperature is above 290°C. Therefore, when doing the start-up, feed temperature must be set above 290°C to initiate the reaction. This temperature is often referred as "ignition temperature". Second, from feed-outlet temperature profile generated by backward step (line 1), it is shown that the reaction extinguished when feed temperature is below 230°C. The temperature value when the reactor extinguished often referred as "extinction temperature". Third, from feed-outlet temperature profile generated by varying reactor inlet temperature (line 2), it is shown that there is a transition zone between ignition temperature and extinction temperature. When reactor operated in the "upper outlet temperature" of the transition zone, once feed temperature goes below the extinction temperature, the outlet temperature would dramatically fall to "lower outlet temperature". After that moment, increasing the feed temperature back to its original value could not restore outlet temperature directly to "upper outlet temperature" path. However, the outlet temperature would follow "lower outlet temperature" path until feed temperature reach the ignition temperature. This transition zone that has non-reversible behavior could result instability of the process, known as hysteresis.

The stability of autothermic reactor as temperature margin have been introduced as the difference between the temperature of feed and a temperature near the extinction temperature of autothermic reactor [7]. Thus, to make sure that the reactor is safe from extinction, feed temperature should set away from extinction temperature. However, the desired operating temperature should be set to the as low value as possible that inside the region of multiplicity. Reactor operation is held close to the extinction temperature point because of the region is importance as economic optimum in commercial ammonia synthesis reactors [8]. Relation between reactor conversion and feed temperature shown in figure 5. As seen in figure 5, low operation temperature result higher conversion of reaction yet slight change of temperature at this region that may occur during operation could trigger the hysteresis phenomenon in the reactor. Assessment of reactor stability under operation in this region is necessary because slight process disturbances and changes in operating conditions could cause the reactor slide to extinction or to suffer growing oscillations.

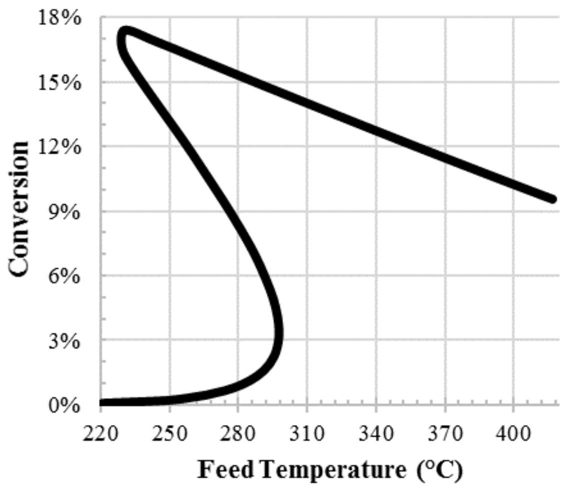


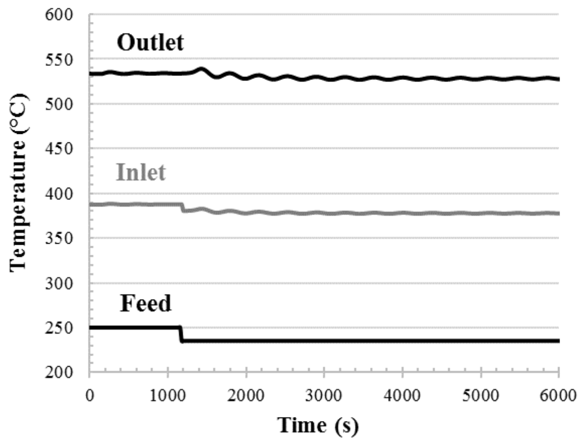
Fig. 5. Ammonia reactor feed-outlet temperature profile.

Furthermore, hysteresis phenomenon could be identified from dynamic simulation. Feed temperature was set at 250°C in steady operation condition and then suddenly dropped down to several temperatures around the extinction temperature which value is 230°C, referred as $\boldsymbol{T_E}$. The temperature drops performed with a step function after the reactor operated in 1,200 seconds. In this simulation, the final feed temperature would be $\boldsymbol{T_E}$ + 5°C, $\boldsymbol{T_E}$ and $\boldsymbol{T_E}$ - 5°C. Result of dynamic simulations to identify the instability behavior are presented in figure 6.

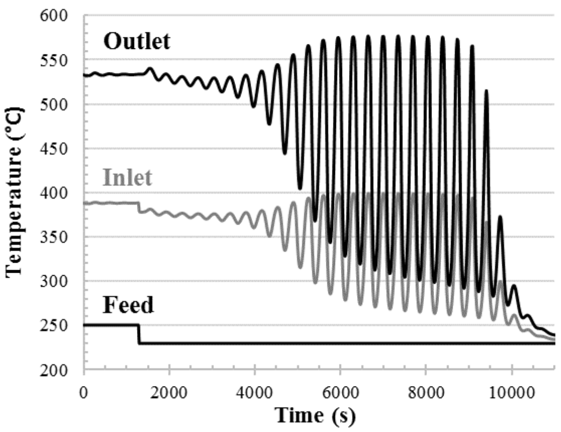
Reactor still in stable condition when feed temperature dropped from 250°C to $\boldsymbol{T_E}$ + 5°C (235°C) as shown in figure 6(a), although the outlet temperature was slightly oscillating in the beginning of temperature drop. Instability phenomenon could be observed when feed temperature dropped to $\boldsymbol{T_E}$ and $\boldsymbol{T_E}$ - 5°C, as shown in figure 6(b) and 6(c) respectively. After temperature have been dropped, outlet temperature was roughly oscillating in range of 300-570°C before finally extinguished the reaction. As shown in the figure 6(b) and 6(c), the obvious difference of these two instability phenomena is the duration of outlet temperature oscillating behavior. Reactor with final temperature of $\boldsymbol{T_E}$ had experienced oscillating behavior for roughly 9,000 seconds, while the other one was only 5,000 seconds.

Huge difference of oscillating temperature could damage equipment material because of temperature sudden change that could decrease equipment material integrity. Increased temperature within the reactor could change physical property of catalyst's support that affects the performance of the catalyst itself [9,10].

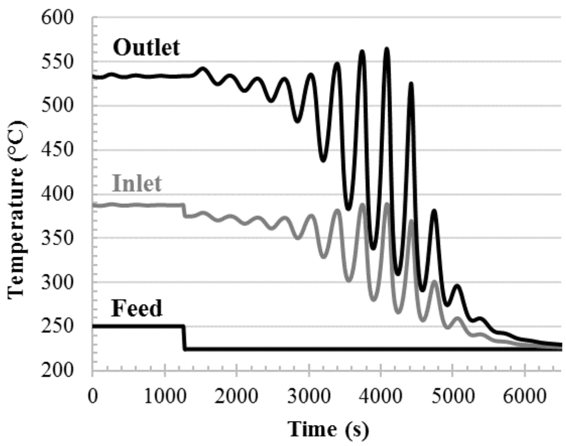
Unstable operation condition also could also cause problem to the next process; the uncertainty of the outlet stream specification from the reactor system may cause problems to the further process.



(a)



(b)

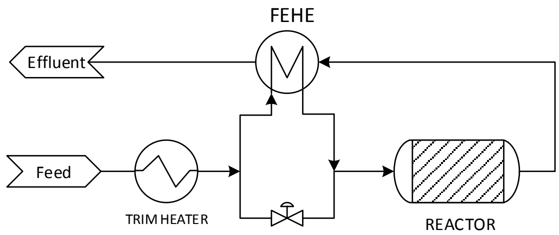


(c)

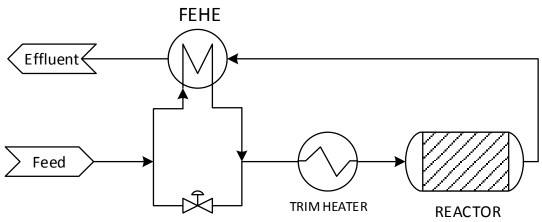
Fig. 6. Reactor temperature dynamic behavior of ammonia reactor system when feed temperature suddenly dropped from 250°C to (a) T_E + 5°C, (b) T_E and (c) T_E - 5°C

3.2 Prevention of hysteresis phenomenon in single-bed ammonia reactor by installing trim heater

Hysteresis phenomenon indicated by the oscillation of outlet temperature value is a dangerous event that could even causing loss or equipment failure to ammonia plant. One of the solution that proposed to prevent this phenomenon is installing trim heater in the reactor system. Trim heater used in ammonia reactor system is a kind of heat exchanger that keep reactor feed temperature above hysteresis temperature during the operation. Trim heater could be installed whether at the feed stream line or reactor inlet stream line. Each of this trim heater scenario illustrated in figure 7.



(a)



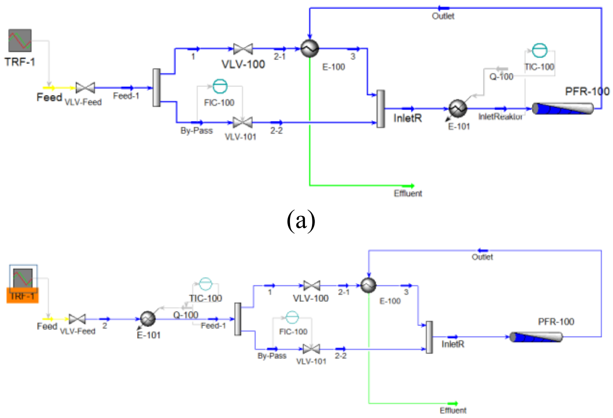
(b)

Fig. 7. Trim heater installation scenario (a) at feed stream line, (b) at reactor inlet stream line

The advantage of installing trim heater right before stream entered the reactor is the temperature of reactor inlet directly controlled and hysteresis zone could definitely be prevented, regardless whatever happens with stream in FEHE or its feed temperature. Conversely, installing trim heater before the FEHE only ensure the feed temperature to meet the feed temperature requirement. The inlet temperature still depends on FEHE performance. However, the advantage of installing trim heater before FEHE over before the reactor is that less heat requirement because of lower temperature target. Based on figure 4, to avoid the occurrence of instability behavior, minimum feed temperature required is 230°C, along with minimum inlet temperature requirement which is 400°C.

Usage of trim heater as unit that prevented hysteresis phenomenon is shown in dynamic simulation using Aspen HYSYS v8.8. Two simulation schemes, trim heater before the reactor and trim heater before FEHE, are presented in figure 8. Based on hysteresis zone data on figure 4, temperature control that govern the trim heater are set in 400°C and 240°C, respectively.

Simulation was performed with initial feed temperature 250°C and gradually decreased to 200°C in 250 minutes with ramp function. Dynamic simulation without the trim heater also performed as a comparison. Result of dynamic simulation of design case, installing trim heater before the reactor and installing trim heater before FEHE are presented in figure 9, 10 and 11, respectively.



(a)

(b)

Fig. 8. Trim heater installation simulation schemes, (a) trim heater before the reactor and (b) trim heater before FEHE.

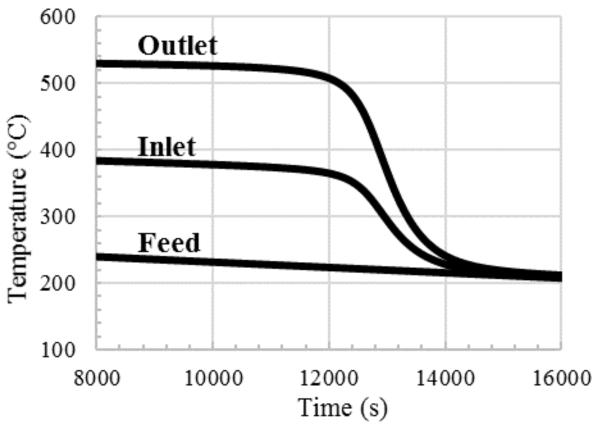


Fig. 9. Design case ammonia reactor temperature profile result.

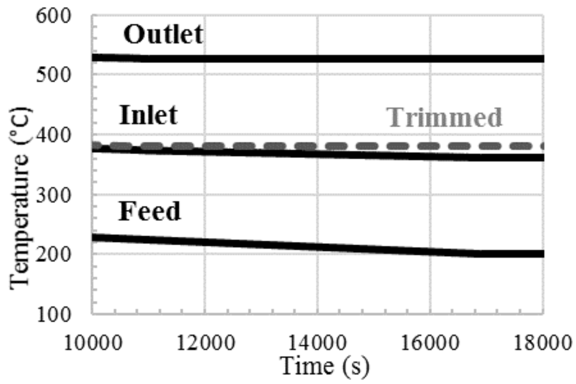


Fig. 10. Trim before the reactor case ammonia reactor temperature profile result.

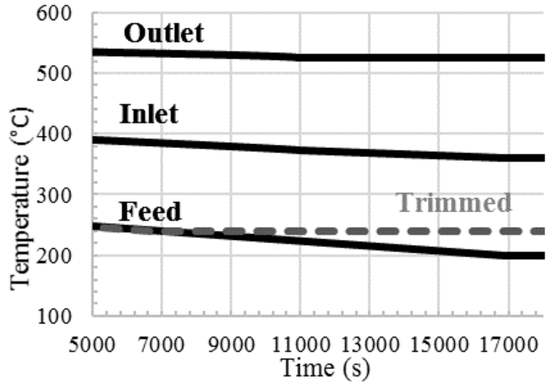


Fig. 11. Trim before FEHE case ammonia reactor temperature profile result.

Based on figure 10 and 11, trim heater is proven to prevent hysteresis phenomenon also avoid the reactor system to become extinguished as if design case in figure 9. Through the regulation of the feed or inlet temperature which value above the hysteresis temperature, the ammonia reactor should remain to work in stable condition without significant temperature fluctuations.

Energy usage of the trim heater from each scheme are analyzed with steady-state condition with feed temperature range of 150-220°C. The result of the energy analysis is presented in figure 12. From that figure, it is shown that case of installing trim heater before the reactor required less energy supply than installing trim heater before FEHE. However, trim heater that installed before the reactor should require more complex utility and tougher material since the stream must be heated until the temperature reach 400°C.

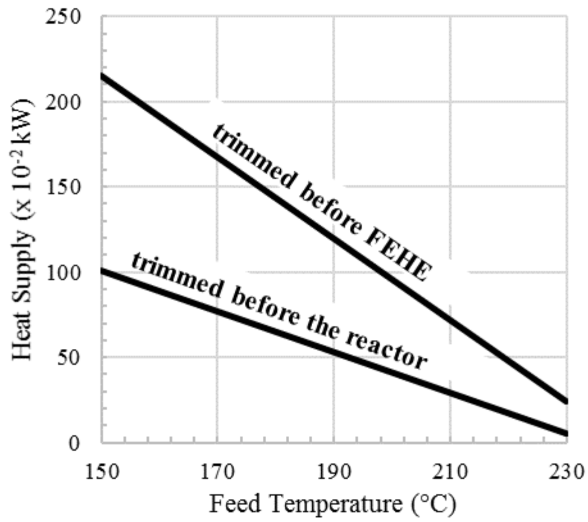


Fig. 12. Energy analysis of trim heater installation in ammonia reactor system.

4 CONCLUSION

Reactor that implemented autothermic configuration, like ammonia reactor, is at risk to become unstable when the reactor is operating. The instability often appears as hysteresis, the oscillation of operating parameter that could lead reactor to extinguish. Hysteresis could occur when reactor inlet temperature reaches certain value that made conversion of the reactor fall down. Hysteresis in single-bed ammonia reactor could be prevented by installing trim heater before stream entered the reactor or in the reactor system feed stream, before entering FEHE.

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