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Dynamic Simulation and Control of an Isopropanol-Acetone-Hydrogen Chemical Heat Pump

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ABSTRACT: In this paper, the study was focused on the dynamic behavior of isopropanol-acetone-hydrogen chemical heat pump system (IAH-CHP). The model for the IAH-CHP was developed and investigated under the environment of Hysys.Plant. The most important process variables, that affect the stability of the system, are the pressure in distillation column and at the outlet of the compressor. With plant-wide control concept, eight control loops were required for the system. These were 2 pressure control loops for distillation column and compressor, 2 level control loops for condenser and reboiler, 2 temperature control loops for reactors, concentration control loop for isopropanol in the distillate, and heat flow control loop for exothermic reactor. All control loops were tuned and tested for stability.

KEYWORDS: Isopropanol-Acetone-Hydrogen, chemical heat pumps, energy recovery, dynamic simulation, control system.

INTRODUCTION

The increase in energy consumption in every society leads to serious environmental problems, especially global warming and resource depletion. Thus, in order to reduce such problems, energy recovery and energy conservation are inevitable. At present, there are large quantities of low-temperature energy released to the environment such as industrial waste heat and solar energy. These can be utilized by transforming them to high-temperature energy. This goal can be achieved by using heat pumps. There are various types of heat pump.[1] The most widely used heat pump in industry is mechanical heat pumps. However, their performance is quite limited. Even the most advanced systems can deliver thermal energy at maximum temperature of 110°C,[2] which cannot be used as a practical heating source. Moreover, they are often with high operating cost and low efficiency. Another type is absorption heat pumps. They deliver high temperature energy and can be designed at large-scale for industrial application. Their limitation is the requirement of large pressure shifts among system components, which result in high operating costs and maintenance problems. To overcome the drawbacks of these two types of heat pump, chemical heat pumps (CHPs) have been proposed[1,3] and more than 250 chemical processes have been investigated.

The chemical heat pumps can provide high temperature energy required for industrial sectors by taking advantage of the heat of reversible catalytic reactions.[4] Among many CHP systems proposed to date, one promising reaction is the isopropanol/acetone/hydrogen system (IAH-CHP). The system was first proposed by Prevost and Bugarel.[5] Both reactions were carried out in the gas phase, with significant amount of energy consumed in vapor compression work. Later on, Saito and coworkers[6] proposed a modified system with a distillation unit allowing the separation of acetone and hydrogen from isopropanol. The dehydrogenation reaction took place in the liquid phase and the hydrogenation reaction took place in the gaseous phase. Gandia and Montes[7] developed a mathematical model for IAH-CHP to estimate the optimal ranges for system control variables. The reaction rate of dehydrogenation reaction was investigated by Kim *et al.*[8] Gastauer and Prevost[9] proposed to conduct the dehydrogenation reaction in the gas phase at low temperature with nickel catalyst and obtained a constant initial reaction rate with higher acetone concentration. Chung and coworkers[2] investigated for optimal design of IAH-CHP by simulation. The optimal values for design variables such as reaction temperature, reflux ratio, and feed position were obtained. Since acetone is a strong inhibitor in the dehydrogenation reaction, a reactive distillation column was used to separate acetone from the reaction field, by vaporizing to the top product. This approach can achieve complete conversion of the dehydrogenation.[10] Recently, the dehydrogenation reaction was investigated again with

10% wt Ru-Pt/activated C to study the influence of reaction temperature, catalyst concentration, nitrogen flow, and acetone concentration in liquid reactant.[11]

The literature reviewed above showed the development of the IAH-CHP, in the area of the reaction improvement and system design for steady state condition. The objective of this paper is to study dynamic behavior and to design a control system for the IAH-CHP. The process flowsheet was developed under Hysys environment, a chemical process simulator by Hyprotech.[12] The control loops were also developed, tuned, and simulated to study its responses under load changes.

The Isopropanol-Acetone-Hydrogen Chemical Heat Pump

The IAH-CHP, shown in Fig 1, is composed of two reactors, one for endothermic and the other for exothermic reactions. The endothermic reaction is dehydrogenation of isopropanol taking place at 80–90 °C and the exothermic reaction is hydrogenation of acetone taking place at 170– 210°C. The reaction equations are shown in Eq. (1).[12]

$$(CH_3)_2CHOH\,(l) \rightarrow (CH_3)_2CO\,(g) + H_2\,(g)$$
$$\Delta H = 100.4\ \mathrm{kJ/mol} \qquad (1.A)$$

$$(CH_3)_2CO\,(g) + H_2\,(g) \rightarrow (CH_3)_2CHOH\,(g)$$
$$\Delta H = -55.0\ \mathrm{kJ/mol} \qquad (1.B)$$

Since the Gibbs free energy of dehydrogenation reaction ($\Delta G°$=13.9 kJ/mol) is positive, the reaction can be proceeded catalytically. When it takes place, the low-temperature heat will be absorbed in the endothermic reactor. Acetone and hydrogen are produced. The absorbed heat will be carried by these chemical species and is transported to the exothermic part of the system by separating these two chemical species from isopropanol in a partial-condensation distillation column. Isopropanol as a bottom product of the distillation is fed into the endothermic reactor. Acetone and hydrogen are then sent, compressed, and reacted in the exothermic reactor, where the hydrogenation taking place, to produce isopropanol. The compressor is to compensate for the pressure drop in the system. The high-temperature heat will be released from the system to a process. The product from the reactor exchanges heat with reactor feed through a heat exchanger and is fed into distillation column.

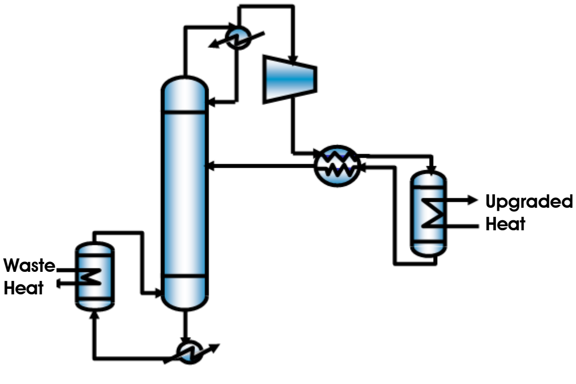


Fig 1. Isopropanol-Acetone-Hydrogen Chemical Heat Pump.

SIMULATION STUDY

Steady-State Simulation

A model of IAH-CHP for studying its behavior was developed under the Hysys.Plant environment. Fig 2 shows the IAH-CHP process flowsheet. Uniquac-Virial equation was selected for physical property calculation. Both the exothermic and endothermic reactors were modeled by using the equilibrium reactor model. Eq. (1) was specified for the reaction equation in the reactor. The conditions of the reactors were also specified so that the corresponding reactions were taking place. In the simulation, the reactions were carried out in vapor phase. The conversions were predicted according to the chemical equilibrium conditions. A column model with full reflux mode was used for the distillation column. Besides the specification of inlet streams, pressure profiles, number of stages and feed stage, two more specifications need to be specified for column with reboiler and condenser. These could be the duties, reflux rate, composition fraction, etc. For our design, the fractions of isopropanol in both top and bottom products were specified. The rest of the equipment, compressor and heat exchangers, in the process was quite straightforward to be modeled.

Dynamic Simulation

After solving the process for steady-state condition and in order to continue simulating the process dynamically, more information for the simulator is required. The information, on the initial conditions of the process or time constants of unit operations, needs to be known such that a set of differential equations with respect to time can be solved. Typically, the initial conditions could be obtained from steady state solutions of the system. The solutions provided temperature, pressure, composition, and flow rates for each stream and each unit operation.

To obtain a time constant for a unit operation, one has to know its size. In general, for complex unit operations, such as the distillation column, Hysys. Plant provides sizing tools for these unit operations. For example, the tray-sizing utility is used for calculating tray diameters, tray spacing, weir length and height for

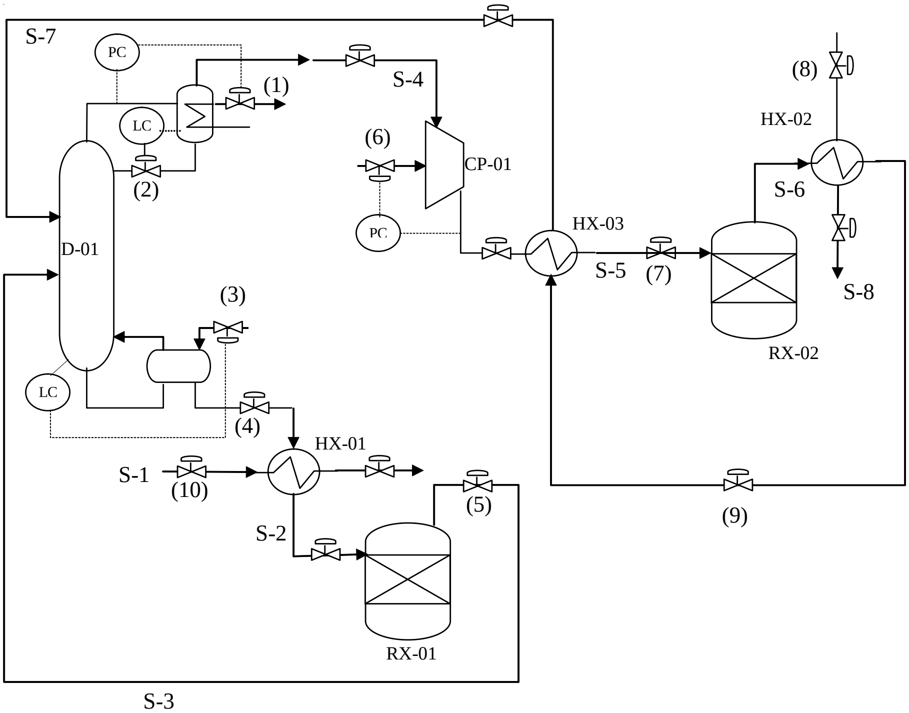


Fig 2. IAH-CHP with testing control loops.

a column. For a simple equipment, such as a tank, Hysys. Plant provides sizing guideline, such as time for vessel holdup. The equipment with chemical reaction such as a reactor, its size is dependent on chemical reaction kinetics. Thus, the size can be determined from the kinetic data. However, the correct kinetic data is not easily obtained and, in many situations, the values reported in literatures use different units, different bases and different composition dependency.[9] The highly nonlinear relationships used in kinetic expression make the results extremely sensitive to any mistakes in converting units of reaction rates. Thus, to cope with the problem and still support dynamic response of the reactors, the reactor sizing was also calculated by specifying time for vessel holdup according to Hysys.Plant guideline. The size of each unit operation was shown in Table 1.

Since the flows in and out from the equipment affected mass accumulation of unit operations, they also affected the process responses. Therefore, the pressure drop along each stream became important for dynamic simulation. Hysys provides advanced method for calculating pressure and flow profiles. Volume balances, defining material balance at pressure holdups, and resistance equations, defining flow between pressure holdup, are set-up and users provide the pressure-flow specifications. In general, a flowsheet will require one pressure-flow specification per flowsheet boundary stream. A flowsheet boundary stream is the one that crosses the model boundary and is attached to only one unit operation. Before switching from the steady state mode to the dynamic mode, the flowsheet should be set up so that pressure drops across the process were specified. The pressure-flow specification should be properly selected for the P-F solver to converge. The equations are solved simultaneously to find unknown pressure or flow rates. Pumps or valves, if necessary, should be added to the flowsheet. In this simulation, linear valve was used and sized with 50% opening at nominal steady state flow rates. All vessels were set to be 50% liquid level.

RESULTS AND DISCUSSION

First, the steady-state simulation was carried out and the solution was shown in Table 2. The reaction conversions in endothermic and exothermic reactions were 7.9% and 17.9%, respectively. The amount of low-temperature heat absorbed was 758 kW, while that of high-temperature heat released was 64.8 kW. The simulation shows the heat ratio of 0.06 or 6%. The heat ratio, representing the thermal effectiveness, is defined as the ratio of heat upgraded by the cycle to waste heat supplied to the cycle. After solving the process for the steady-state solutions, the dynamic simulation would be started and the responses would be observed under different control loops and different loading conditions.

Investigation on the Effects of Control Loops

Setting up all specifications necessary for running

Table 1. The size of unit operations in the IAH-CHP.

Equipment	Parameter	Size
HX-01	Area = 0.7 m^2	L = 11.7 m
HX-02	Area = 1.92 m^2	L = 30.6 m
HX-03	Area = 3.08 m^2	L = 49.0 m
Distillation D-01	Diameter	1.067 m
	Weir Height	0.051 m
	Weir Length	0.626 m
	Tray spacing	0.607 m
Condenser	Volume	5.769 m^3
Reboiler	Volume	2.442 m^3
Endothermic Reactor (RX-01)	Volume	31.69 m^3
Exothermic Reactor (RX-02)	Volume	22.88 m^3

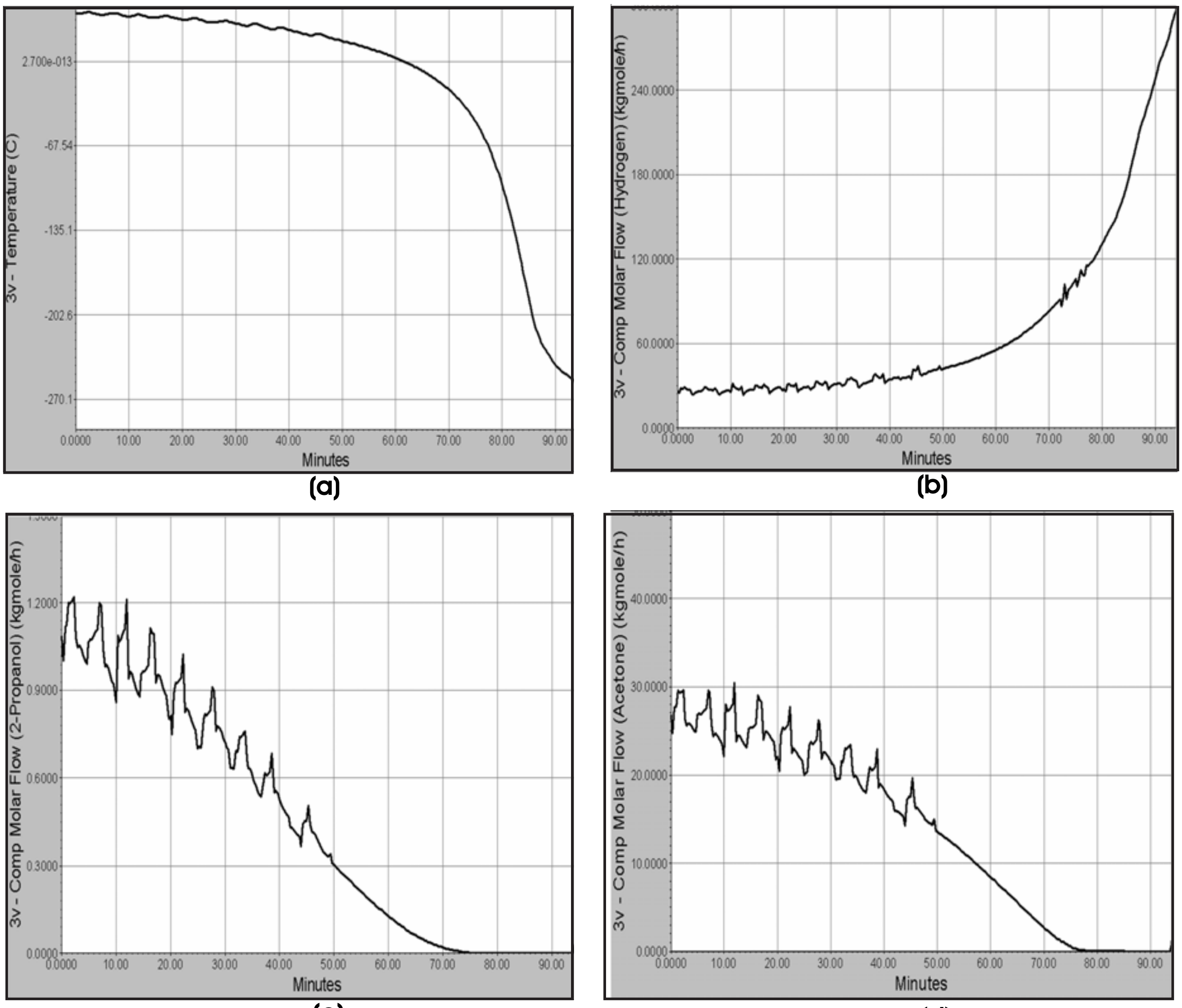
Table 2. Steady state operation condition of IAH-CHP.

Endothermic Reactor			Exothermic Reactor		
Waste heat temperature	C	130	Total flow of feed stream	mol/s	15.1
Conversion of isopropanol (equilibrium)		7.9%	Acetone to hydrogen ratio in feed stream		1.0
Heat absorbed	kW	758	Conversion of acetone (equilibrium)		17.9%
			Temperature	C	221
			Heat released	kW	64.8
Compressor			**Distillation column**		
Total pressure drop	Atm	0.46	Number of stages		15
Work	kW	20.2	Feed stage		8
			Condenser temperature	C	38.5
Heat exchanger			Condenser duty	kW	1074
Duty	kW	74.7	Molar reflux ratio		2.18
UA	kJ/C.h	26170	Isopropanol mole fraction in distillate		0.02
COP		3.22	Reboiler temperature	C	83.0
Heat Ratio		0.06	Reboiler duty	kW	301.6
			Distillate to feed ratio		0.46
			Acetone mole fraction in bottom		0.02

Table 3. Controller parameters for the IAH-CHP system.

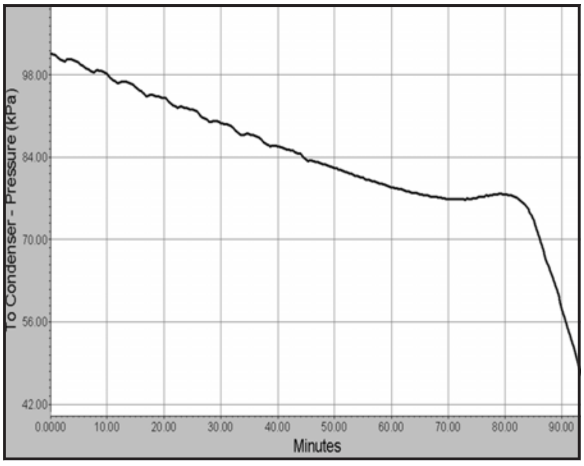
Control loop	Controller Type	Controller Action	Controller Gain, Kc	Integral time, I (min)
HT Steam temperature	PI	reverse	3.102	0.984
HT Steam Flow	PI	reverse	0.1	0.2
Column Pressure	PI	direct	2.0	2.0
Conc. of distillate	PI	direct	2.638	16.938
Level of condenser	P	direct	1.0	-
Level of reboiler	P	direct	1.0	-
Process pressure	PI	reverse	4.774	0.139
Temperature inlet stream to endothermic reactor	PI	reverse	2.523	0.576

in the dynamic mode, IAH-CHP process was simulated dynamically as an open loop system. It was found that the process was unstable as shown in Fig 3. Temperature, pressure, and component flows in the column were decreased steadily until their values were negligible. It implies that the process cannot operate without a control system. Thus, control loops were introduced into the process flowsheet and they were tested to observe their influence on the response of the system. Since the pressure and flow are strongly



(a) (b)

(c) (d)



(e)

Fig 3. The responses of IAH-CHP system when operated without control system (a) Temperature of top product (b) Pressure in the column (c) Concentrations of isopropanol (d) Concentrations of acetone (e) Concentrations of hydrogen.

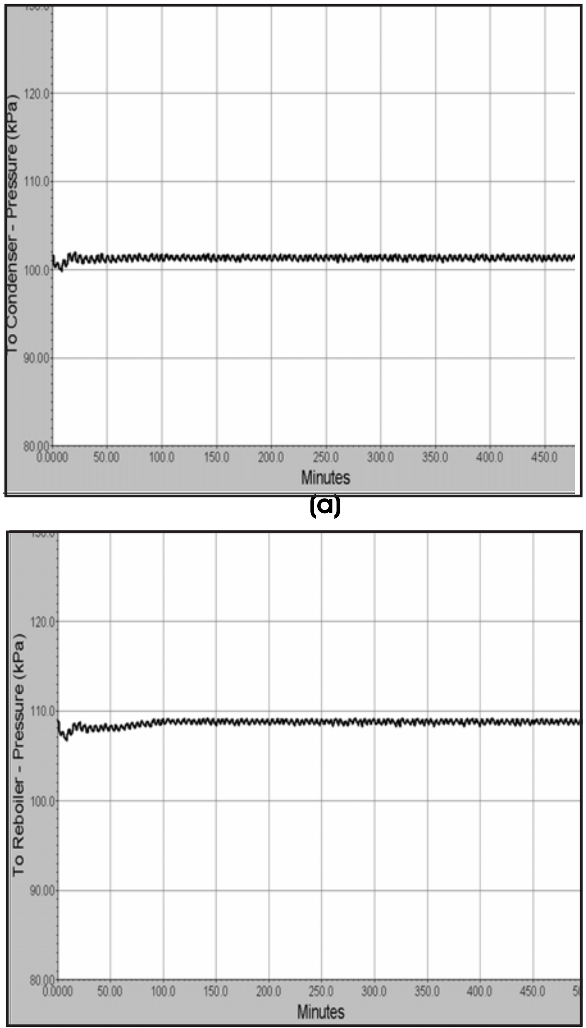


Fig 4. Pressure responses at (a) the top and (b) the bottom of the column.

coupling, one needs to keep these variables within an extent. Therefore, the pressure was selected to be the controlled variable, since, by nature, the process pressure should be kept unchanged. At least, its change should not occur so often. On the other hand, the flow would normally be used for adjusting in order to maintain the level or the energy in a system. Five sets of control loops were developed and tested. These are (1) column pressure control loop, (2) process pressure control loop, (3) both of column and process pressure control loops, (4) set (1) plus condenser and reboiler level control loops, and (5) combination of (2) and (4). Fig 2 describes locations and their physical connections of these control loops.

For control set 1, Fig 4 (a and b) show the pressure responses at the top and the bottom of the column. The responses were stable, but oscillated with amplitude and frequency of 0.22 rpm. Consequently, the molar flow of the top product of the column was also stable, but oscillated with a very high frequency at 0.38 rpm, as shown in Fig 5(a-c). This oscillation carried out through the process. The observation shows that small oscillation of the pressure in the column could generate

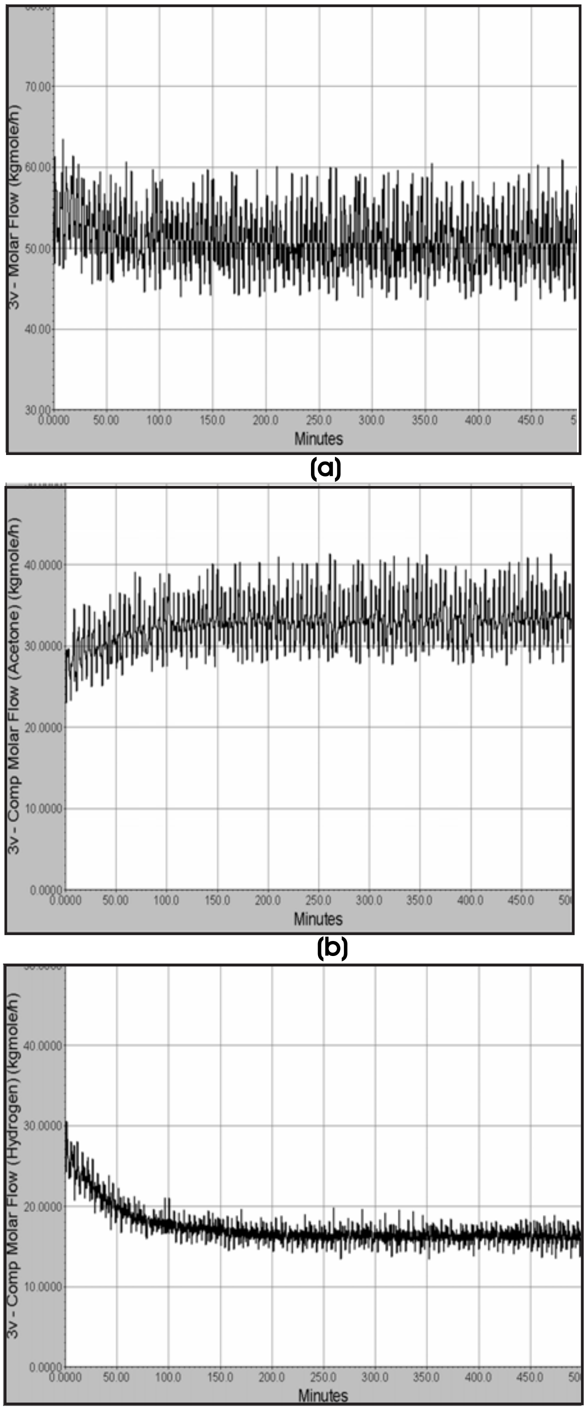
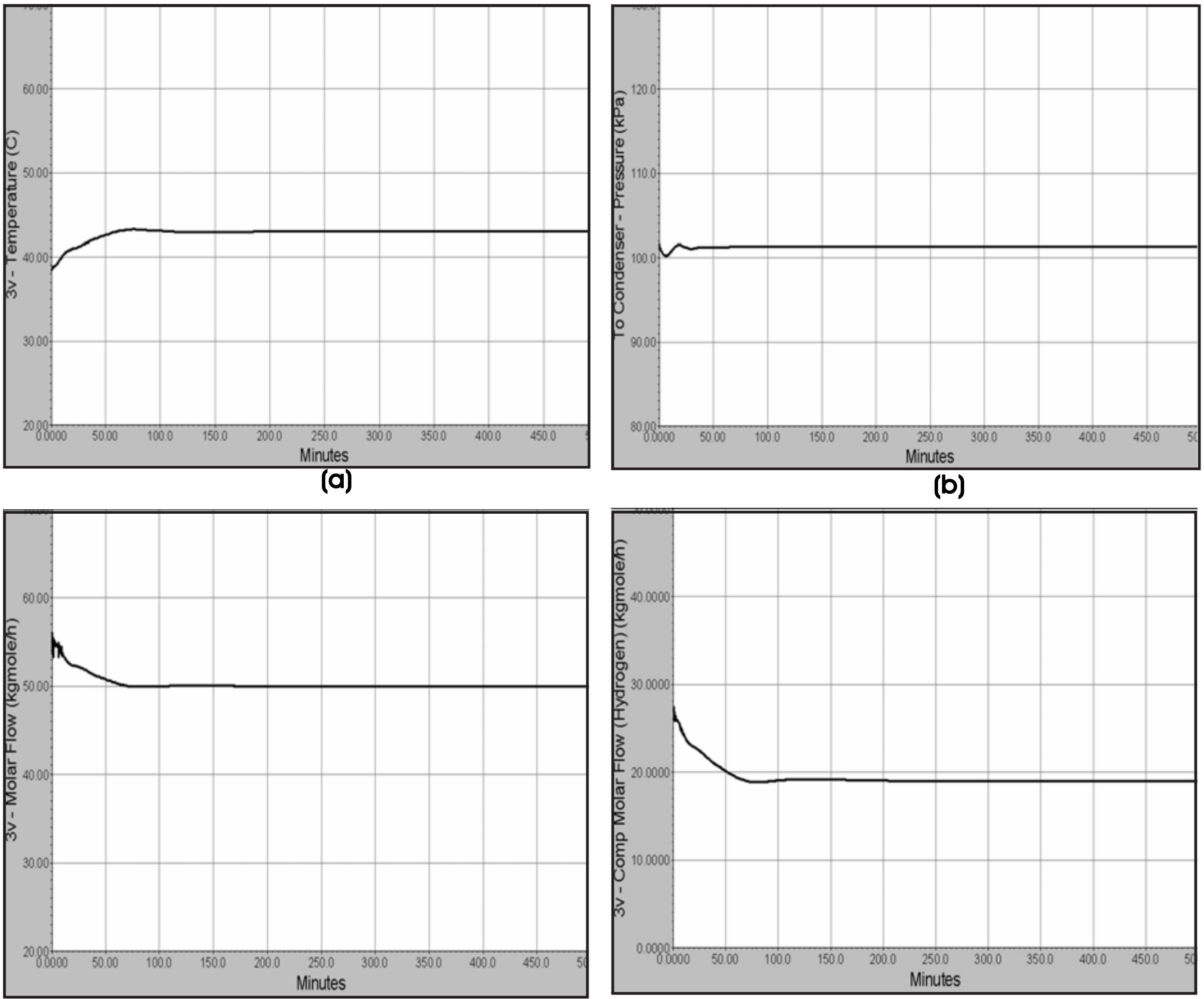
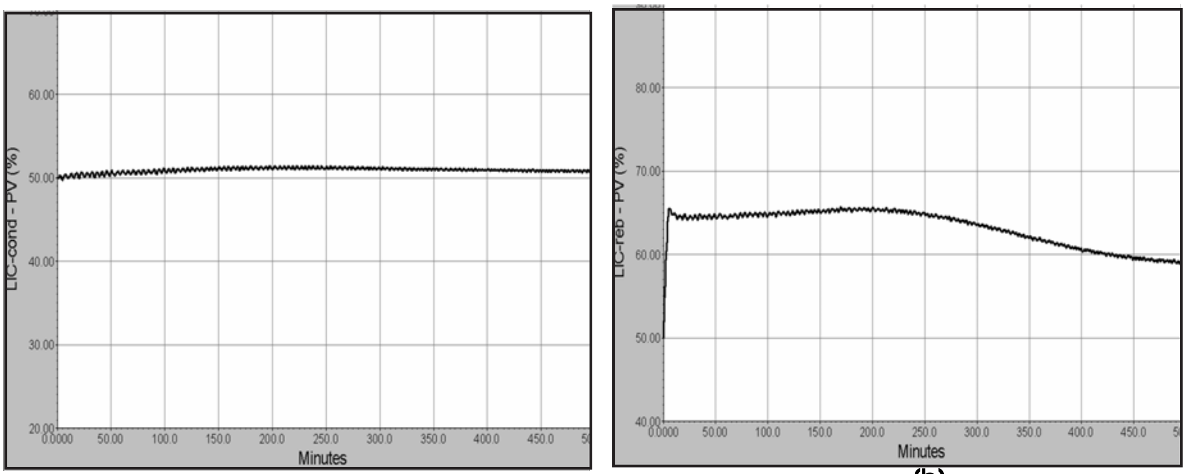


Fig 5. Molar flow responses of (a) isopropanol (b) acetone (c) hydrogen.



(a) (b) (c) (d)

Fig 6.(a) Top temperature (b) Column pressure (c) Molar flow of top product (d) Molar flow response of hydrogen.



(a) (b)

Fig 7.(a) Level of product in condenser (b) Level of product in reboiler.

larger oscillation in the process. On the other hand, control set 2 has demonstrated a contradicting result with control set 1. The system was unstable. The temperature and pressure in the column were steadily decreased. While acetone and isopropanol compositions of the top product were decreased until reaching zero values, the hydrogen composition was increased. It is implied that only the pressure control loop of the compressor could not keep the process stable. Control set 3 was proposed by combining both the column pressure and process pressure control loops. The responses were displayed in Fig 6 (a-d). It was found that the responses were stable and were not highly oscillating as shown in control set 1. They were slightly overshoot and then leveled off to a new steady state. The pressure of the column was returned to a set point within 50 minutes. Component flows and temperature in the top of the column reached new steady state values approximately within 90 and 120 minutes, respectively.

For control set (4), besides the column pressure control loop, the level control loops for condenser and reboiler were also included. The reason is that normally all inventories have to be controlled so that the controlled quantity would not run dry or flooding. It was found that the responses were similar to those in control set 1. Therefore, the response will not show here. Fig 7 (a and b) show the condenser and reboiler levels during the transient operation. The condenser level had slightly changed. On the contrary, the reboiler level changed rapidly at the beginning of flow setpoint change. Then, it was slowly returned to the level set point. Since these inventory control loops did not change the system responses, they were not the key control loops. As expected, by combining control sets 2 and 4, the observed responses were the same as that reported for control set 3. By this investigation, it was concluded that in order to run IAH-CHP smoothly and stably, two control loops has to be installed. These are the column pressure and the process pressure control loops.

Complete Control Structure

To operate the IAH-CHP practically, one might like to change the temperature or the amount of heat demand. In order to achieve these functions, a number of control loops were required, in addition to the two key control loops as mentioned in the previous section. The concept of plant-wide process control procedure[14] was adopted and applied to design the IAH-CHP control system. Its procedure satisfies two fundamental chemical engineering principles of the overall conservation of mass and energy. Moreover, the procedure accounts for nonconserved entities within a plant such as chemical components and entropy. There are 9 design steps involved. Five of the nine steps (Steps 3 – 7) deal with plantwide control issues that would not be addressed by simply combining the control systems from all of the individual unit operations. Steps 1 and 2 establish the objectives of the control system and the available degrees of freedom. Step 3 ensures that heat production in the process is properly dissipated. Steps 4 and 5 achieve the business objectives concerning production rate, product quality, and safety. Step 6 involves total mass balance control. Step 7 accounts for nonconserved chemical components. Step 8 is to complete the control systems for individual unit operations. Finally, Step 9 uses the remaining degrees of freedom for optimization and improved dynamic controllability.

Step 1. For IAH-CHP, one must be able to set the temperature or the amount of heat demand on the high temperature heat exchanger side, HX-02. When the heat demand is changed in term of either quantity or quality, the process has to adjust external heat supplies accordingly to achieve the target.

Step 2. There are 10 control degrees of freedom in this process. These include two feed valves for distillation column, condenser and reboiler valves, reflux, distillate and bottom products valves, waste heat and upgraded heat valves, and power for compressor.

Step 3. Energy management is critically important because the process is involved with heat absorption and heat liberation from reactions. In general, heat generation on the hydrogenation side will be controlled since it involves product quantity. It is removed by manipulating low quality steam flow rate, valve (8) in Fig 2. The other variable that reflects the amount of accumulation energy in the system is the column pressure. To maintain the energy in the system, the column pressure is controlled by adjusting a cooling water valve of the condenser, valve (1).

Steps 4 and 5. The objective of the IAH-CHP system is to produce heat at a specified temperature with the required amount. Two control loops play the major role for these requirements; one for quantity and the other for quality. The former is to control the input flow rate of the HX02 heat exchanger by using valve (8). The latter is to control the exit temperature of the HX02 heat exchanger. There are two options for the controller. One is by adjusting valve (6) and the other is by using valve (7).

Step 5. In addition to control loops in the previous steps, the control loop pertaining to safety, operation, and environmental constraints are also important. In this process, since most of the equipment in the process was operated under vapor phase condition, the pressure could reflect energy accumulation. To maintain the balance of energy, pressure control loop is necessary,

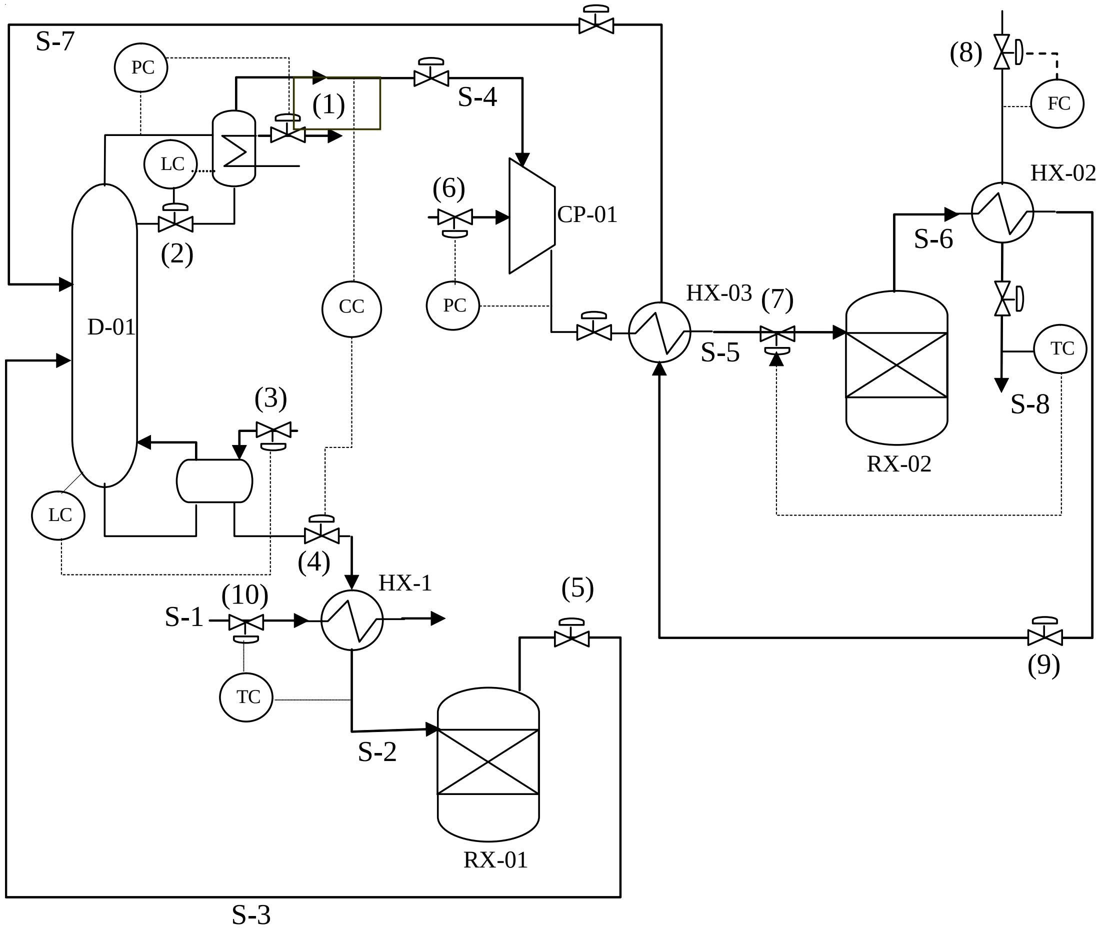


Fig 8. IAH-CHP with complete control loops.

valve (1). Moreover, isopropanol concentration of the top product has to be controlled, otherwise, the production quantity and quality will not be achieved. The bottom rate, valve (4), was used for manipulating the isopropanol concentration.

Steps 6 and 7. The IAH-CHP is a closed system in the view of material balance, since it is a cycle device: no mass in and out of the system. Once, isopropanol, acetone and hydrogen were charged into the system, ideally, the system could be operated indefinitely. Thus, there is no accumulation of the related chemicals in the system.

Step 8. Besides the control loops established along steps 1-7, there are some control loops necessary to operate each of the individual unit operations. In this case, level control loops for both condenser and reboiler were specified for the distillation column, by adjusting valves (2) and (3), respectively.

Step 9. Up to this point, all basic regulatory strategies have been set. Eight out of ten control valves were used to control eight controlled variables. There are two degree of freedom left. According to the design above, two valves that were left were the valves for adjusting the column feeds. The flows in these streams were determined by the pressure differences between those in the reactors and in the distillation column. The pressure in the column was controlled. By observation, the pressure in endothermic reactor was almost unchanged and that in the exothermic reactor was varied according to the operating condition in the direction that was benefit to the operation. Thus, it was decided not to install any control loop for these streams.

Test for System Responses to Load Changes

After applying the plant-wide control procedure above, the control system for the IAH-CHP was obtained as shown in Fig 8. Then, a controller in each of control loop was tuned by using Cohen-Coon[15]. Table 3 shows the values of control parameters for each control loop. The process was tested to check whether the control objectives can be achieved. The first objective is to supply heat for a certain demand change. The test was

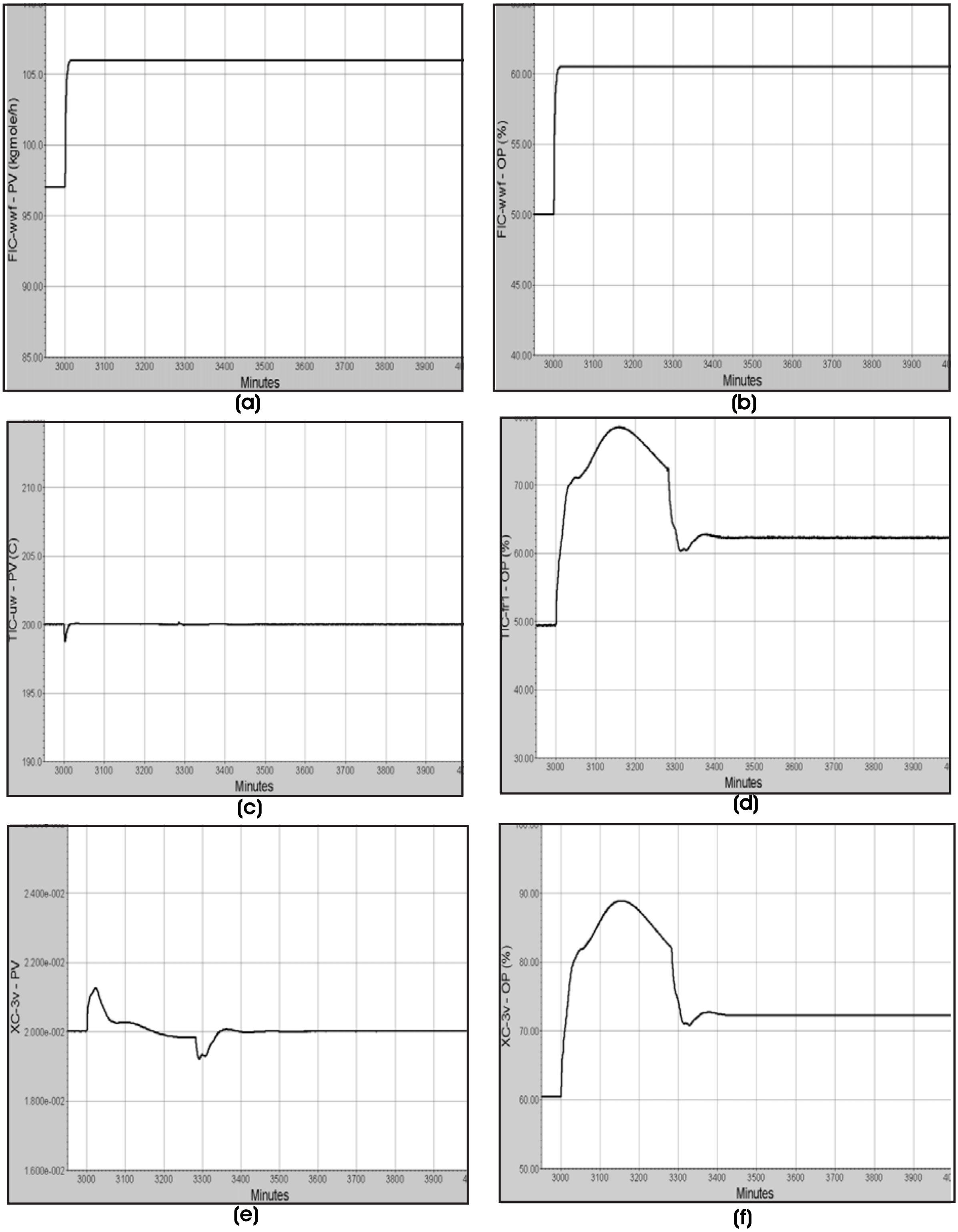
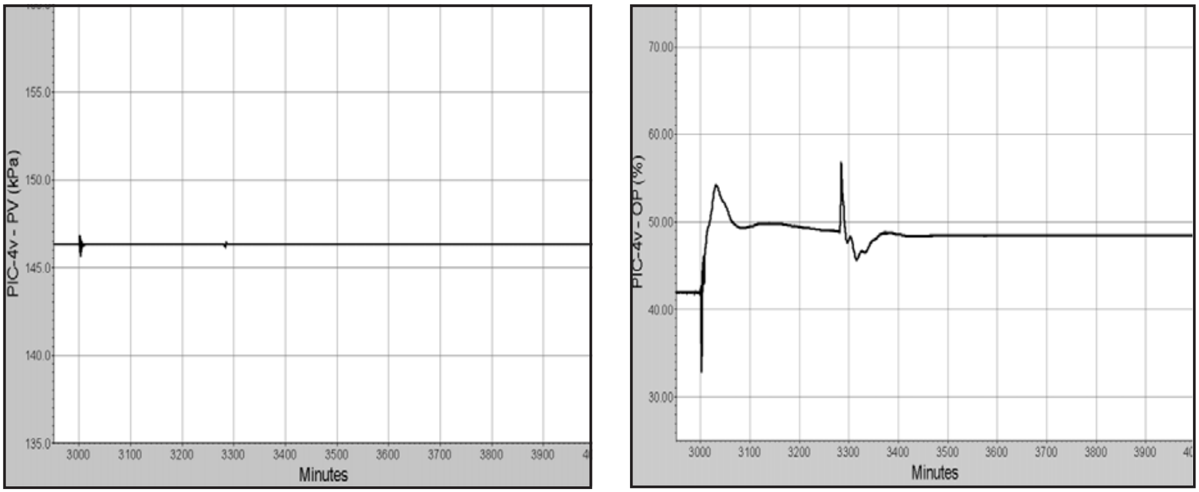
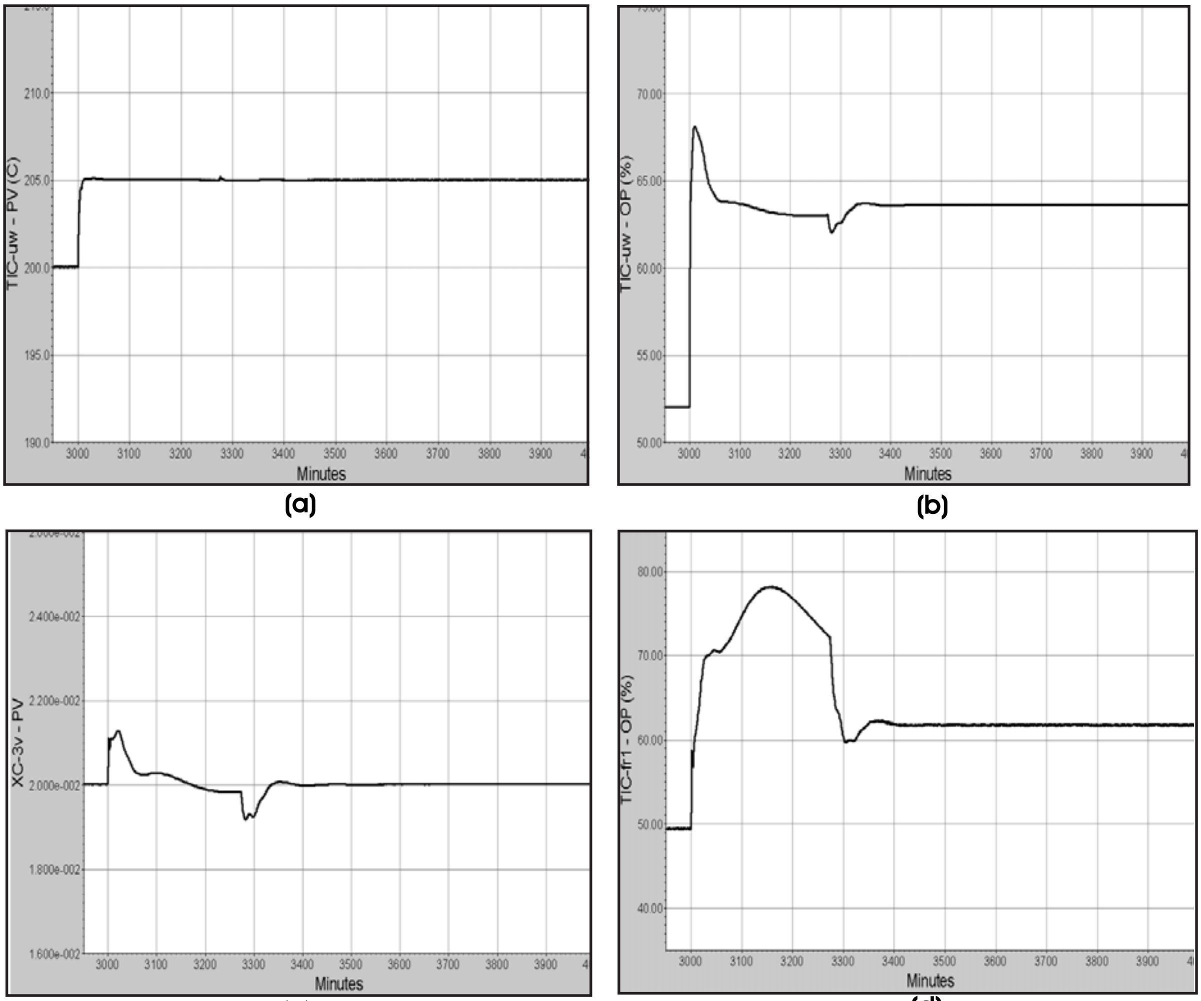


Fig 9. The effect of an increase in heat demand by increasing the flow of high temperature steam on exothermic reactor by 9 kmol/h where (a) HT steam flow, (b) Valve position of HT steam, (c) HT steam Temperature, (d) Valve position of LT steam, (e) Isopropanol concentration, (f) Valve position of bottom produc, (g) Pressure column and (h) Valve position of compressor.



(g) (h)

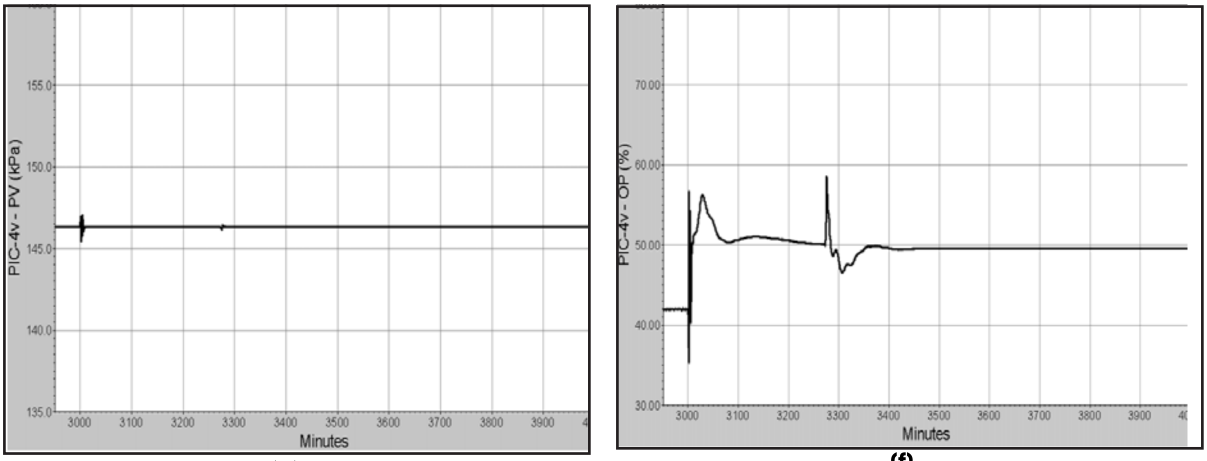
Fig 9 (continued).



(a) (b)

(c) (d)

Fig 10. The effect of an increase in heat quality by increasing the HT steam temperature on exothermic reactor by 5 deg C where (a) HT steam temperature, (b) Valve position of HT steam, (c) Isopropanol concentration, (d) Valve position of LT steam (e) Pressure column, (f) Valve position of compresso.



(e) (f)

Fig 10 (continued).

conducted by increasing the heat demand by 9 kmol/h of high-temperature steam. Fig 9 (a-g) show the responses of control valves and key parameters in the system to the change. It was found that the system moves to the new set point within 15 minutes as shown in Fig 9a. The response was not oscillated. The valve position of the product flow, valve (8), was changed from 50 to 61% as shown in Fig 9b. The temperature of the product, in Fig 9c, dropped slightly in the beginning and returned to set point. The valve for controlling temperature inlet of the endothermic reactor, in Fig 9d, was opened wider by changing from 49% to 62%. The isopropanol concentration, Fig 9e, was changed rapidly in the onset of the flow set point change and was returned to the setpoint in 150 minutes. The valve for the bottom flow, Fig 9f, was opened wider from 60% to 72%. The flow of the waste heat steam on the endothermic reactor side was also increased corresponding to the increase of heat demand as shown in Fig 9f. Fig 9g showed the increase in power supply to compressor by 7%. The pressures of the column and the process were slightly changed for a short period of time and returned to their set points.

The second objective of the IAH-CHP system is to move the system to the specified temperature, when the quality of heat was changed. The test was conducted by increasing the temperature of the high-temperature steam by 5 degree Celsius. Fig 10(a-f) also show the responses of control valves and the key parameters in the system. Fig 10a showed that the response had reached the new set point (205°C) within 15 minutes which was the same duration of time when the flow was changed. The exothermic reactor feed was also increased with an overshoot. Its valve position was changed by 12%, as shown in Fig 10b. The concentration of isopropanol was increased rapidly at the onset of the change in temperature set point and was returned to the same value as the steady-state value. The flow of the waste heat steam on the endothermic reactor side was also increased corresponding to the higher temperature requested as shown in Fig 10d. The column pressure was disrupted suddenly at the onset of setpoint change and was returned to its setpoint rapidly, as shown in Fig 10e. The power supply for the compressor was increased by 8% as shown in Fig 10 f. The results above showed that, in both cases, the control system can move the process to the specified conditions.

CONCLUSION

In conclusion, dynamic model of IAH-CHP was developed under the Hysys.Plant environment. Major equipment was sized to obtain proper dynamic response of the system. The necessary control loops for keeping system stability were pressure control loops of the column and the process. To achieve the functional objectives, load or quality changes, the concentration control loop was required to be installed so that stable responses with required objectives could be achieved. There were 8 control loops installed in the system. Each controller was tuned and the tests were conducted to verify its performance. It was found that the performance was satisfactory.

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