

A Nonlinear Dynamic Model of a Vinyl Acetate Process

by

Rong Chen

Kedar Dave

Thomas Mc Avoy*

Department of Chemical Engineering/Institute for Systems Research

University of Maryland, College Park MD 20742

Michael Luyben

E.I. Dupont & Co.

Wilmington, DE

ABSTRACT: This paper discusses a public domain model of a vinyl acetate (VAC) process. The VAC process is large enough to be representative in its essential features of many chemical processes. The process model contains 246 states, 26 manipulated variables, and 43 measurements. Parts of the model, e.g. the azeotropic distillation tower, are highly non-linear. An earlier model of the same process was published, but it was implemented in proprietary software. Both the source code used to generate the results in this paper and a compiled version are available to the public and the paper discusses how they can be obtained. The modeling approach used and differences with the earlier VAC model are discussed in detail. Simulations are presented which show the agreement with earlier results. The goal of the paper is to provide a readily available simulation that can be used as a testbed for advanced control approaches, such as plantwide control design and optimization, model predictive control and process monitoring.

INTRODUCTION

For several years, researchers in plantwide design, optimization and control areas have expressed interest in getting realistic test problems for assessing new technologies. The most frequently studied test problem so far is the Tennessee Eastman Challenge (TEC) process¹ which has been utilized by many different researchers in studies ranging from plantwide control to optimization to fault detection. In 1998, an additional model of a large, industrially relevant system, a vinyl acetate monomer (VAC) manufacturing process, was published by Luyben and Tyreus². The VAC process contains several standard unit operations that are typical of many chemical plants. Both gas and liquid recycle streams are present as well as process-to-process heat integration. Luyben and Tyreus presented a plantwide control test problem based on the VAC process. The VAC process

*Corresponding Author: Phone 301 405-1939, FAX 301 314-9920, email: mcavoy@eng.umd.edu

was modeled in TMODES, which is a proprietary DuPont in-house simulation environment, and thus, it is not available for public use.

This paper presents a first-principle nonlinear dynamic model for the VAC process as well as associated software, based on the design details provided in references^{2,3}. The model of the VAC process is developed in MATLAB, and both the steady state and dynamic behavior of the MATLAB model are designed to be close to the TMODES model. Since the MATLAB model does not depend on commercial simulation software and the source code is open to public, the model can be modified for use in a wide variety of process control research areas. To obtain a reasonable simulation speed, compiled model written in C is also available to researchers. Details on how the various programs can be obtained are given at the end of this paper.

In this paper, design details of the MATLAB model are presented. For each unit, design assumptions, physical data, and modeling formulations are discussed. There are some differences between the TMODES model and the MATLAB model, and these differences together with the reasons for them are pointed out. Steady state values of the manipulated variables and major measurements in the base operation are given. Production objectives, process constraints, and process variability are summarized based on the earlier publication^{2,3}. All of the physical property, kinetic data, and process flowsheet information in the MATLAB model come from sources in the open literature.

PROCESS DESCRIPTION AND MODELING ASSUMPTIONS

In the VAC process, there are 10 basic unit operations, which include a vaporizer, a catalytic plug flow reactor, a feed-effluent heat exchanger (FEHE), a separator, a gas compressor, an absorber, a carbon dioxide (CO₂) removal system, a gas removal system, a tank for the liquid recycle stream, and an azeotropic distillation column with a decanter. Figure 1 shows the process flowsheet with locations of the manipulated variables. The numbers on the streams are the same as those given by Luyben and Tyreus². There are seven chemical components in the VAC process. Ethylene (C₂H₄), pure oxygen (O₂), and acetic acid (HAc) are converted into the vinyl acetate (VAc) product, and water (H₂O) and carbon dioxide (CO₂) are by-products. An inert, ethane (C₂H₆), enters with the fresh C₂H₄ feed stream. The following reactions take place:



Readers are referred to section 2 in reference² for a detailed process description, including the reaction rate expressions and the major aspects of each unit operation. Totally, the MATLAB model includes 246 states, 26 manipulated variables, and 43 measurements. The base operation discussed here, in which the peak temperature in the reactor is below 162 °C, is the same as that discussed in Chapter 11 in reference³.

The following assumptions are the same in both the TMODES model and the MATLAB model:

- (1) There are no light components (O_2 , CO_2 , C_2H_4 , and C_2H_6) in the distillation column. The reason for this assumption is that in reality the light components are very small in concentration and they remain essentially in the vapor phase within the tower. They leave the tower via a venting system in the decanter. Including the light components in the tower model would make the model much stiffer, without altering its essential control characteristics. To realize this assumption, a gas removal system is implemented to remove all the light components in the column feed stream before they enter the column. The gas removal system does not exist in the real plant, but is mathematical in nature. As described in section 4.1 in reference² an ideal component separator is implemented to completely separate the seven components into a gas stream and a liquid stream. The liquid stream containing VAc, H_2O and HAc only feeds the column. The gas stream, containing only the four light components, is sent back and combined with the vapor produced from the separator to form the vapor feed to the absorber. The light components then exit in the two purge streams, rather than in the actual tower vent stream. Because of this assumption, a decanter vent valve is not included in the MATLAB model.
- (2) To simplify the modeling of the gas loop dynamics, it is assumed that the entire gas loop pressure drop only occurs in the reactor. There are two vapor effluent valves in the process, one in the vaporizer and the other in the separator. Based on the pressure drop assumption, the pressure drops across the two valves are assumed zero. The absorber pressure is assumed to be equal to the compressor exit pressure. Therefore, the absorber vapor effluent valve is not included in the MATLAB model.
- (3) To simplify the modeling of the gas loop dynamics, a total gas loop holdup, which is used to calculate dynamic change in the gas recycle stream pressure, is assigned only to the separator. In the MATLAB model, the separator pressure is a state variable. There is a pressure increase through the gas compressor, and the compressor exit pressure determines the gas recycle loop pressure.
- (4) Pump and compressor dynamics are ignored since they are fast. In addition, there is no deadtime associated with the flows between units. As a result, no pipe dynamics are included in the model.

To speed up the simulation, several additional assumptions, which are different from those given by Luyben and Tyreus², are made in the MATLAB model. These assumptions do not significantly affect the overall plant dynamics and the nature of the plantwide control problems encountered. These additional assumptions are:

- (5) In the TMODES model, water is the reactor coolant and it is allowed to boil and thereby provide a constant shell temperature for the reactor. A steam drum is included in the TMODES model to introduce some additional dynamics on the shell side. Two valves are used in the TMODES model: the reactor steam drum liquid makeup valve and the reactor steam drum vapor exit valve. Since a change in steam drum pressure causes a fast change in the shell temperature, the shell side dynamics are ignored in the MATLAB model and the shell temperature is assumed to be perfectly controlled. As a result in the MATLAB model, the shell temperature is a manipulated variable, and therefore the steam drum is not included.

- (6) In the TMODES model, the separator has a cooling jacket that removes the heat from the separator during the condensing process. The cooling jacket temperature is assumed uniform, and the coolant flowrate is a manipulated variable. Since a change in the coolant flowrate causes a fast change in the jacket temperature, the jacket side dynamics are ignored in the MATLAB model by assuming that the cooling jacket temperature is perfectly controlled. Therefore, the cooling jacket temperature is a manipulated variable in the MATLAB model.
- (7) In the TMODES model, the heaters and coolers are modeled by shell-and-tube heat exchangers, in which the steam or coolant streams are used as manipulated variables. Since the dynamics of these units are fast, to simplify the MATLAB model, a steady state energy balance is applied to the heaters and coolers to calculate the temperature changes of streams. To introduce some dynamics, a two-minute lag is added to the exit temperature of each heater and cooler. There are totally 5 such types of heat exchangers in the MATLAB model, and each of them has a state variable and the heat duty is used as a manipulated variable.
- (8) In the MATLAB model, molar flowrate (kmol/min), heat duty (kcal/min) and temperature ($^{\circ}\text{C}$) are directly used as manipulated variables and no valve dynamics are included. It can be noted that liquid flow meters normally read in mass/time, but in the simulation mole units were used since they had been used in the earlier work^{2,3}. Using moles does not change the essential nature of the plantwide control problem involved with the plant. Both minimum and maximum values for each manipulated variable are used and a pseudo linear valve operating between the two extreme values is defined.

MODELING THE VINYL ACETATE PROCESS

This section discusses design assumptions, equipment data, and modeling formulations for each unit operation. The design details differ slightly from those in Luyben and Tyreus² and why these differences occur is explained. In this section, the simulation model used for each major unit is discussed in detail after a brief discussion of the thermodynamics and physical property data. For each unit, the state and manipulated variables are identified.

a) Thermodynamics and Physical Property Data

In the MATLAB model, the vapor-liquid equilibrium (VLE) calculations are performed assuming an ideal vapor phase and a standard Wilson liquid activity coefficient model. The Wilson parameters and molar volumes are listed in Table 1, and they are obtained directly from the TMODES model. The molar volumes are different from what is given in Luyben and Tyreus², who only gave some of these values. The pure component physical property data are the same as those given in Table 2 in reference², except that the molecular weights are calculated to three decimal places rather than 2 decimal places. The reason of this change is that if the molecular weights given in reference² are used (2 decimal places) then a slight generation of moles results from the round off of the molecular weights and the overall material balance is not satisfied. The component vapor pressures are calculated using the Antoine equation, and the Antoine coefficients are the same as those given in Table 3 in reference². The equipment data are given in Table 2 and most of them are directly obtained from Tables 8 through 10 in reference². The properties of the three feed streams are given in Tables 6 and 7 in reference².

b) The Vaporizer

The vaporizer is implemented as a well-mixed system with seven components. It has a gas input stream (F_1), which is a mixture of the C_2H_4 feed stream and the absorber vapor effluent stream. It also has a liquid input stream (F_2), which comes from the HAc tank. There are 8 state variables in the vaporizer, including the liquid level, the mole fractions of O_2 , CO_2 , C_2H_4 , VAc, H_2O , and HAc components in the liquid, and the liquid temperature. The liquid level is defined by the ratio of the liquid holdup volume over the total working volume. Since the dynamics of the vapor phase are ignored, total mass, component and an energy balance are used to calculate the dynamics in the liquid as:

$$\rho_L^{VAP} \dot{V}_L^{VAP} = F_1^{VAP} MW_1^{VAP} + F_2^{VAP} MW_2^{VAP} - F_V^{VAP} MW_V^{VAP} \quad (3)$$

$$M_L^{VAP} \dot{x}_{L,i}^{VAP} = F_1^{VAP} (x_{1,i}^{VAP} - x_{L,i}^{VAP}) + F_2^{VAP} (x_{2,i}^{VAP} - x_{L,i}^{VAP}) - F_V^{VAP} (y_{V,i}^{VAP} - x_{L,i}^{VAP}) \quad (4)$$

$$Cp_L^{VAP} M_L^{VAP} \dot{T}_L^{VAP} = F_1^{VAP} (h_1^{VAP} - h_L^{VAP}) + F_2^{VAP} (h_2^{VAP} - h_L^{VAP}) - F_V^{VAP} (H_V^{VAP} - h_L^{VAP}) + Q^{VAP} \quad (5)$$

Vapor liquid equilibrium (VLE) is assumed in the vaporizer, and as a result, the vaporizer pressure and the vapor compositions are determined by a bubble point calculation. Two manipulated variables (Q^{VAP} and F_V^{VAP}) are available in the vaporizer. In the base operation, the liquid holdup, V_L^{VAP} , is 2.8 m^3 , which is 70% of the working level volume. The vaporizer is followed by a heater, and the heater duty is a manipulated variable. In the base operation, the heater exit temperature is specified to be $150 \text{ }^\circ\text{C}$.

c) Catalytic Plug Flow Reactor

The reactor is implemented as a distributed system with ten sections in the axial direction. Two irreversible exothermic reactions, given by Eqns. 1 and 2, take place. In the MATLAB model, the following assumptions are made for the purpose of model simplification:

- (1) Plug flow is assumed so that there are no radial gradients in velocity, concentration, or temperature. Diffusion occurring in the axial direction is considered negligible compared to the bulk flow. Potential and kinetic energy and work are considered negligible in the energy balance calculation.
- (2) It is assumed that the mass and heat transfer between the fluid and catalyst are very fast and therefore the concentrations and temperatures in the two phases are always equal.
- (3) Pressure drop is assumed linear along the length of a tube, and it is time-independent. Instead of using the momentum balance given by Equation 20 in reference², Eqn. 6 is used to calculate the pressure drop in each section:

$$\Delta P / \Delta Z = f * \rho_1^{RCT} * (v_1^{RCT})^2 \quad (6)$$

where: $\Delta P / \Delta Z$ is the pressure drop per unit length (psia/m), f is a constant friction factor, ρ_1^{RCT} is the mass density of the feed stream (kg/m^3), v_1^{RCT} is the volumetric flowrate of the feed stream (m^3/min). The value of f is taken directly from the TMODES model, instead of calculating it using the Ergun's method⁴.

- (4) As stated earlier, the shell temperature is assumed uniform, and it is used as a manipulated variable in the MATLAB model. Thus, the steam drum dynamics are not modeled.

Material and energy balances on the reactor, which are based on a tubular reactor dynamic model developed by Reyes and Luyben⁵, are given by Eqn. 7 and 8:

$$\varepsilon \frac{\partial C_{i,j}}{\partial t} = -\frac{\partial(C_{i,j}v_i)}{\partial z} + \phi_i \rho_b (\theta_{1,j}r_{1,i} + \theta_{2,j}r_{2,i}) \quad (7)$$

$$\left(\varepsilon \sum_{k=1}^7 C_{i,k} Cp_{i,k} + \rho_b Cp_b \right) \frac{\partial T_i}{\partial t} = -\frac{\partial(v_i \sum_{k=1}^7 (C_{i,k} Cp_{i,k}) T_i)}{\partial z} - \phi_i \rho_b (r_{1,i} E_1 + r_{2,i} E_2) - Q_i^{RCT} \quad (8)$$

where index i represents the section number and index j represents component j , ϕ_i is the catalyst activity in section i , given by Equation 8 in reference². $\theta_{1,j}, \theta_{2,j}$ are the stoichiometric coefficients for component j in the two reactions, $r_{1,i}, r_{2,i}$ are the reaction rates in section i , given by Equations 3 and 4 reference², and E_1, E_2 are the heats of reactions. Q_i^{RCT} is the external heat flux per unit volume in section i , and it is calculated by $Q_i^{RCT} = UA(T_i - T_s)$, where T_s is the shell temperature.

In the MATLAB model, the molar concentrations of components $\text{O}_2, \text{CO}_2, \text{C}_2\text{H}_4, \text{VAc}, \text{H}_2\text{O}$ and HAc and the tube temperature in each section of the reactor are state variables. Therefore totally 70 state variables are present in the reactor. The molar concentration of component C_2H_6 can be calculated based on the ideal gas law. Only one manipulated variable, T_s is available in the reactor. In the base operation, the reactor exit temperature is equal to 159.17 °C.

d) Feed Effluent Heat Exchanger (FEHE)

As stated in Chapter 5 in reference³, for the purpose of plantwide control studies, it is not necessary to rigorously model the dynamics of a process-to-process heat exchanger if it doesn't dominate the process response. In the MATLAB model, the NTU-Effectiveness method^{6,7} is used to calculate the steady state exchanger exit temperatures and the exact FEHE dynamics are not modeled. A small time constant is added to the exit temperature sensors to capture temperature dynamics. The inverse of the total thermal resistance, UA , is calculated by Eqn. 9, which shows that the effective UA is a function of the mass flowrates of the two streams:

$$UA = UA_0 * [(F_1^{FEHE} / F_{C_REF})^{0.8} + (F_2^{FEHE} / F_{H_REF})^{0.8}] / 2 \quad (9)$$

where F_1^{FEHE} is the mass flowrate of the cold stream and F_2^{FEHE} is the mass flowrate of the hot stream. There is one manipulated variable, the bypass ratio, and no state variable in the FEHE. In the base operation, the FEHE hot effluent temperature is equal to 134 °C.

e) Separator

In the MATLAB model, the separator is modeled as a partial condenser. At each point in time, a steady state equilibrium-flash calculation is carried out to obtain the flow rates and properties of the vapor and liquid streams immediately after the pressure letdown valve on the separator feed stream. The pressure letdown valve is not shown in the process flowsheet in Figure 1. A standard algorithm is used to solve the isothermal flash problem, assuming that the flash temperature and pressure are known⁸. In reality, the flash temperature cannot be easily obtained. The amount of the stream that condenses is a function of the heat removed, but the heat removed is a function of the flash temperature, which, in turn, is determined by the amount of the stream that condenses. In the MATLAB model, the flash temperature is approximated by adding 5 °C to the cooling jacket temperature, and the jacket is assumed well mixed so that the jacket temperature is uniform. Then the vapor and liquid streams are split into the vapor and liquid phases respectively. It is assumed that there is no driving force for material and heat transfer between the two phases. In the vapor phase, it is assumed that the vapor volume, which represents the total gas loop volume, is a constant. A mass balance is used to model the vapor pressure dynamics. In the MATLAB model, the separator vapor exit stream flowrate is fixed. In the liquid phase, a total energy balance determines the temperature dynamics.

There are 16 state variables in the separator, including the liquid level, vapor phase pressure, mole fractions of components O₂, CO₂, C₂H₄, VAc, H₂O, and HAc, and temperatures in both phases. The ideal gas law is applied to the vapor phase. In the separator, three manipulated variables are available, the liquid exit stream flowrate, the vapor exit stream flowrate, and the cooling jacket temperature. In the base operation, the liquid holdup is 4 m³, which is 50% of the working level volume. The separator pressure is 84.25 psia, and the separator liquid phase temperature is 40 °C.

f) Compressor

In the MATLAB model, the pressure increase across the compressor is calculated by Eqns. 10 and 11:

$$P_{OUT}^{COM} = P_{IN}^{COM} + \Delta P \quad (10)$$

$$\Delta P = \gamma \rho^{COM} \quad (11)$$

where γ is the compressor coefficient, ρ^{COM} is the compressor inlet stream density. The exit temperature is calculated by assuming an isentropic compression. The compressor is followed by a

cooler, and the cooler duty is a manipulated variable. In the base operation, the cooler exit temperature is 80 °C.

g) Absorber

In the MATLAB model, the gas absorber is divided into two parts. The top part contains six theoretical stages. Its inlet liquid stream is from the HAc tank, and its inlet vapor stream is from the top of the bottom part of the absorber. The bottom part contains two theoretical stages. Its inlet liquid stream is a combination of the liquid stream from the top part and a circulation stream. Its inlet vapor stream is from the compressor. It is assumed that the absorber pressure, which is specified at 128 psia in the base operation, is uniform in the two parts of the absorber.

On each stage, the liquid and vapor phases are not in equilibrium, and a rate-based model is implemented to capture the liquid phase dynamics. The vapor phase dynamics are ignored. On each stage, the mass transferred from the vapor phase to the liquid phase is given by Eqn. 12:

$$N_i = \min\{N_{MT} * (y_i - y_{INT,i}), 0.5 * F_{V,i} * y_i\} \quad (12)$$

where N_i is the molar flowrate of component i (kmol/min), N_{MT} is a constant mass transfer coefficient, y_i is the mole fraction of component i in the vapor inlet stream, $y_{INT,i}$ is the mole fraction of component i at the gas-liquid interface, which is obtained from an equilibrium calculation using the liquid phase compositions and temperature. $F_{V,i}$ is the mole flowrate of component i in the inlet vapor stream. To avoid a large mass-transfer rate between the two phases, it is assumed that the largest amount of component i transferred between two phases is the half of the amount of component i in the inlet vapor stream.

The heat transferred from the vapor phase to the liquid phase is given by:

$$Q_j = Q_{MT,j} * (T_{V,j} - T_{L,j}) \quad (13)$$

where Q_j is the heat transferred between the two phases on stage j (kcal/min), $Q_{MT,j}$ is a constant heat transfer coefficient, $T_{V,j}$ is the temperature of the vapor inlet stream, $T_{L,j}$ is the temperature of the liquid phase. During stage-to-stage calculations, total mass, component and an energy balance around the vapor phase are used to calculate the vapor exit stream flowrate, composition, and temperature. A total mass, component and an energy balance around the liquid phase, which are similar to Eqn. 3 to 5, are used to model the absorber dynamics. In the energy balance, the enthalpy of the material transferred between the two phases is calculated as a vapor phase enthalpy at the stage liquid temperature.

There are totally 72 state variables in the absorber, which are the liquid holdup, mole fractions of components O₂, CO₂, C₂H₄, VAc, H₂O, and HAc in the liquid phase, and liquid temperature on each stage. There are three manipulated variables, the liquid exit stream flowrate, the scrub stream flowrate, and the circulation stream flowrate. In the base operation, the liquid holdup is .25 m³, which is 50% of the working level volume. There are two coolers, which are installed on the scrub

stream and the circulation stream respectively, and the cooler duties are manipulated variables. In the base operation, the exit stream temperatures of the two coolers are 25 °C.

h) CO₂ Removal System

The CO₂ removal system is modeled exactly as described in Section 4.1 in reference². There is one manipulated variable, which is the inlet stream to the CO₂ removal system. In the base operation, the CO₂ mole fraction in the gas recycle stream is 0.73%. The system efficiency is given by Eqn. 14:

$$Eff = 0.995 - 3.14 \times 10^{-6} * (F_{CO_2} - 6.4136) - 32.5 * (x_{CO_2} - 0.01342) \quad (14)$$

where F_{CO_2} is the inlet stream flowrate (kmol/min), and the x_{CO_2} is the mole fraction of CO₂ in the inlet stream.

i) Gas Removal System

The gas removal system is designed to remove all the light components in the column feed stream before they enter the column. The system has two liquid inlet streams that come from the bottoms of the separator and the absorber. As described in Section 4.1 in reference², an ideal component separator, which can completely separate the seven components into two streams, is implemented here. The gas stream (O₂, CO₂, C₂H₄, C₂H₆) is sent back and combined with the vapor produced from the separator to form the vapor feed to the compressor. The liquid stream (VAc, H₂O, HAc) is the feed to the column.

j) Azeotropic Distillation Tower

The distillation column contains 20 theoretical stages, whose liquid holdup can vary. It should be noted that the stage efficiency coefficients (50%) given in Tables 9 and 10 in reference² should not be included in the calculations since theoretical stages are used in the model. It is assumed that the column is homogeneous, and only one liquid phase is present. To reduce the system stiffness, the pressure profile in the column is assumed known. A bubble-point calculation is used to determine temperature and compositions on each stage, and then the energy balance is used to solve for the vapor flowrate from stage to stage. The decanter is modeled in the same way as discussed in Section 5.5 in reference². Since the Wilson model can't be used in the decanter due to the liquid-liquid equilibrium, the equilibrium partition coefficients, β , used in the decanter are assumed constant and independent of temperature. It is also assumed that the temperatures of the two liquid phases in the decanter are always same.

There are totally 69 state variables in the distillation column. There are six manipulated variables, reflux flowrate, reboiler duty, condenser duty, organic product flowrate, aqueous product flowrate and bottom flowrate. In the base operation, the bottom liquid holdup is 2.33 m³, which is 50% of the working level volume. The organic liquid holdup and the aqueous liquid holdup are 0.85 m³,

which are 50% of their working level volumes. In the base operation, the decanter temperature is 45.85 °C.

k) HAc Tank

The HAc tank is only used to mix the liquid recycle stream and the fresh HAc feed stream. There are totally 4 state variables in the tank, which are the liquid holdup, mole fractions of VAc, and HAc in the liquid, and the liquid temperature. The flowrates of all the streams connected to the tank are manipulated variables. In the earlier publication on the VAC process^{2,3} an HAC tank was also used, but it was not shown in the process flowsheet given in these publications.

STEADY STATE DATA AND DYNAMIC SIMULATION

The steady state data for the VAC process are obtained after a control structure similar to that developed by Luyben et al³ is implemented. The control system used is shown in Figure 4 and since its major loops are the same as those used by Luyben et al its response should be similar to that given in reference³. However, there are some small differences due to the differences in control structure and how loops were tuned (see discussion below), and the simplifications used in the dynamic model presented in this paper and discussed above. The initial values of all the state variables and manipulated variables come from the TMODES results, and the MATLAB model converges to a steady state (the base operation) that is very close to the TMODES results. The steady state values of manipulated variables are given in Appendix 1. The control structure and controller parameters are given in Appendix 2. Steady state values for the measurements are listed in Appendix 3. The reactor temperature profile and column temperature profile are given in Figures 2 and 3 respectively, where they are compared with the results given in reference³. The temperature profiles from the two models are almost identical.

Four setpoint disturbances are used to illustrate the dynamic behavior of the MATLAB model with the control structure implemented. The transients agree with those published in reference³. In reference³ only a partial set of tuning parameters is given, whereas Appendix 2 gives a complete set. Note that in Appendix 2 several manipulated variables are listed as being fixed. These specifications match those used to generate the results in reference³. In the code for the VAC plant all manipulated variables can be used for control. The controller tuning parameters used here were determined by trial and error and they differ somewhat from those used to generate the results in reference³. In general in the responses to the four setpoint changes, better control of the column temperature is achieved compared to the results in reference³, and the response of the fresh HAC feed differs somewhat from that given in reference³. Results for the individual setpoint changes are as follows.

- (1) A step setpoint change in reactor exit temperature, from 159.17 °C to 151 °C is shown in Figure 5. As can be seen the reactor temperature response is very rapid and the new steady state is quickly achieved. Tighter control of the mole % H₂O is achieved here compared to the results in reference³. The oscillations in the tray 5 temperature are caused by the fact that the reflux is manipulated by a signal from the analyzer that measures the % H₂O in the bottoms. The analyzer has a 10 minute sampling dead-time and every time that the reflux changes as a result of a new analyzer reading it causes an upset in the tray 5 temperature.

The response of the tray 5 temperature is much faster than the response of the analyzer loop.

- (2) A step setpoint change in reactor exit temperature, from 159.17 °C to 165 °C is shown in Figure 6. Again the response of the reactor temperature is very fast and the new steady state is achieved very quickly.
- (3) A step setpoint change in %H₂O in the column bottom, from 9.34% to 18% is shown in Figure 7. As can be seen this set point change produces a very slow transient, which takes over 5 hours to die out.
- (4) A step setpoint change in total HAc recycle flowrate, from 2.19 kmol/min to 2.62 kmol/min is shown in Figure 8. As can be seen the transients take about 100 minutes to die out in this case. The peak in the mole fraction of H₂O is higher than that shown in Chapter 11. The changes in the fresh O₂ and C₂H₄ feeds are smaller and faster compared to the results in reference³.

In general the results presented here agree very well with the earlier results both in the shape and the time scale of the responses. Based on this agreement, it is concluded that the VAC model discussed here essentially captures the dynamics as the earlier model.

CONTROL REQUIREMENTS

Generally, one should be able to set the production rate of VAc while minimizing yield losses to CO₂. The control system must operate effectively as the catalyst activity decreases. The column must produce an overhead product with essentially very little HAc, and a bottoms product with very little VAc. The absorber must recover essentially all of the VAc, H₂O and HAc from the gas recycle loop to prevent yield losses in the CO₂ removal system and purge. Based on the discussion in the Section 4 in reference² and in reference³, a list of control requirements, which includes process constraints, production requirements, and process variability, is provided in this paper to define the plantwide control problem. Note that economic data for the VAC process is given in reference².

a) Process Constraints

1. The O₂ composition must not exceed 8 mol% anywhere in the gas recycle loop.
2. The pressure in the gas recycle loop must not exceed 140 psia.
3. The peak reactor temperature along the length of the tube must not exceed 200 °C .
4. Liquid levels in the vaporizer, separator, absorber base, column base, and decanter must operate within the limits of 10-90%.
5. Reactor feed temperature must be greater than 130 °C.
6. FEHE hot effluent temperature must be greater than 130 °C.
7. The HAc composition must not exceed 0.06 mol% in the decanter organic phase.
8. The VAc composition must not exceed 0.01 mol% in the column bottom.
9. All the analyzers used in the process, except the O₂ analyzer, have a 10 minute sampling frequency and 10 minutes of deadtime. The O₂ analyzer operates continuously.

10. No frequency constraints are applied to the variability of the three fresh feed streams.
11. Any violation of constraint 1, 2 or 3 will shut down the process via interlocks. These interlocks are not coded in the model.

b) Production Requirements

1. The organic product flowrate should be maintained at 0.829 kmol/min.
2. The VAc composition should be around 95 mol% in the organic product.

c) Process Variability

1. The control system should function well as the catalyst activity decreases to 0.8 at the end of a year.
2. Setpoint Changes: (1) change the VAc production design rate by at least 20% up or down over the course of 6 hours, (2) reduce the VAc production design rate by one half but at maximum selectivity.
3. Disturbances: (1) a step change in the composition of C_2H_6 in the fresh C_2H_4 feed stream from 0.001 to 0.003 mole fraction, (2) loss of column feed for 5 minutes, (3) loss of fresh HAc feed stream for 5 minutes, (4) Loss of fresh O_2 feed stream, (5) an analyzer is off-line (except the O_2 analyzer).

Disturbances 1 and 2 have been included in the MATLAB model.

THE COMPUTER CODE

The model equations for the Vinyl Acetate monomer process have been coded in MATLAB and then translated into the C programming language. The C-coded files have been written in such a way that they can be compiled into "MEX functions". As a result, the C-coded model becomes available from within the MATLAB environment. The purpose of providing an interface between the C-coded model and MATLAB is to have a very high execution speed and, at the same time, take advantage of the excellent graphing, data analysis and advanced control functionalities available in the numerous MATLAB toolboxes. The C-coded model runs at approximately 80 times real time, and it has been tested under Microsoft Windows NT, ME and Unix operating systems. When compiled from within the MATLAB environment, the model becomes available as a single .dll file in the MS Windows operating system and the corresponding file extension within Unix is .mexsol.

In MATLAB, the C-coded model can be called as follows:

```
[dstatedt, states, MVs, measurements] = VAModel (states, MVs, time, is_initial, disturbanceID)
```

The inputs, on the right hand side, are:

states = (246 x 1) vector containing the current states.

MVs = (26 x 1) vector containing the current values of the manipulated variables.

time = current time in minutes.

is_initial = binary flag. 1 when extracting steady state data. 0 when carrying out dynamic simulation.

disturbanceID = integer flag. 1. a step change in the composition of C₂H₆ in the fresh C₂H₄ feed stream from 0.001 to 0.003 mole fraction; 2. loss of column feed.

The outputs, on the left hand side, are:

dstatedt = (246 x 1) vector containing the current derivatives of the states.

states = (246 x 1) vector containing the current states (intended to be used for model initialization).

MVs = (26 x 1) vector containing the current values of the manipulated variables (intended to be used for model initialization).

measurement = (43 x 1) vector containing all the measurements.

It is important to note that the analyzer/transmitter time lags and time delays are not implemented in the VAModel subroutine, discussed above. The user needs to include the time lags and time delays in the code that is used to control the process. The m-file test_VAcPlant(t, ID) gives details on how to control the VAC process with a multiloop SISO architecture. In this routine the transmitter lags are assumed to be 3 seconds, and the two analyzers on the gas recycle and column bottoms also have a 10 minute time delay. An Euler integration approach with a 1/3 sec. time step is used to calculate dynamic responses. A 1 sec. sampling time is used for the controllers and transmitters, except for the controllers that are involved with the analyzers that have a 10 minute time delay. These controllers have a 10 minute sampling time. The MATLAB source code, the C source code, the .dll and .mexsol files, and the MATLAB files for sample simulations can be accessed through the Internet. The address is:

www.isr.umd.edu/~mcavoy/VACMaterial

Users are requested to send questions, comments and to share their experience using the model with the corresponding author.

SUMMARY

This paper has discussed a public domain model and simulation of a vinyl acetate (VAC) process. The model is large and in parts highly non-linear. Results calculated using the model are in close agreement with those calculated from an earlier model of the same process, which was implemented in proprietary software. Both the source code and a compiled version of the VAC model are available to the public, and details on how they can be gotten have been given. The modeling approach used and differences with the earlier VAC model have been discussed in detail. The VAC model can be used as a testbed for many advanced control approaches, such as plantwide control design and optimization, model predictive control and process monitoring.

ACKNOWLEDGEMENT

The authors would like to acknowledge the valuable suggestions and input of Bob Cox in the development of the MATLAB code discussed in this paper.

REFERENCES

- (1) Downs, J.; Vogel, E. A Plant-Wide Industrial Process Control Problem. *Computers Chem. Engng.* **1993**, 17, 245.
- (2) Luyben, M.; Tyreus, B. An Industrial Design/Control Study for the Vinyl Acetate Monomer Process. *Computers Chem. Engng.* **1998**, 22, 867.
- (3) Luyben, W.; Tyreus, B.; Luyben, M. *Plantwide Process Control*; McGraw Hill: New York, Chap. 11, 1999.
- (4) Ergun, S. Fluid Flow Through Packed Columns. *Chem. Eng. Prog.*, **1952**, 48, 89.
- (5) Reyes, F.; Luyben, W. Extensions of the Simultaneous Design of Gas-Phase Adiabatic Tubular Reactor Systems with Gas Recycle. *Ind. Eng. Chem. Res.*, **2001**, 40, 635.
- (6) Gebhart, B. *Heat Transfer*, 2nd Ed.; McGraw-Hill: New York, 1971.
- (7) Jones, W.; Wilson, J. An Introduction to Process Flexibility. 1. Heat Exchange. *Chem. Eng. Education*, **1997**, 31, 172.
- (8) Biegler, L.; Grossmann, I.; Westerberg, A. *Systematic Methods of Chemical Process Design*; Prentice Hall: New York 1997.

Appendix 1 Steady State Values of Manipulated Variables

MV	Description	Steady State	Range	Unit
1	Fresh O ₂ Feed	0.52343	0 – 2.268	Kmol/min
2	Fresh C ₂ H ₄ Feed	0.83522	0 – 7.56	Kmol/min
3	Fresh HAc Feed	0.79003	0 – 4.536	Kmol/min
4	Vaporizer Steam Duty	21877	0 – 1433400	Kcal/min
5	Vaporizer Vapor Exit	18.728	0 – 50	Kmol/min
6	Vaporizer Heater Duty	9008.54	0 – 15000	Kcal/min
7	Reactor Shell Temp.	135.02	110 – 150	°C
8	Separator Liquid Exit	2.7544	0 – 4.536	Kmol/min
9	Separator Jacket Temp.	36.001	0 – 80	°C
10	Separator Vapor Exit	16.1026	0 – 30	Kmol/min
11	Compressor Heater Duty	27192	0 – 50000	Kcal/min
12	Absorber Liquid Exit	1.2137	0 – 4.536	Kmol/min
13	Absorber Circulation Flow	15.1198	0 – 50	Kmol/min
14	Circulation Cooler Duty	10730	0 – 30000	Kcal/min
15	Absorber Scrub Flow	0.756	0 – 7.560	Kmol/min
16	Scrub Cooler Duty	2018.43	0 – 5000	Kcal/min
17	CO ₂ Removal Inlet	6.5531	0 – 22.68	Kmol/min
18	Purge	0.003157	0 – 0.02268	Kmol/min
19	FEHE Bypass Ratio	0.31303	0 – 1	
20	Column Reflux	4.9849	0 – 7.56	Kmol/min
21	Column Reboiler Duty	67179	0 – 100000	Kcal/min
22	Column Condenser Duty	60367	0 – 150000	Kcal/min
23	Column Organic Exit	0.8290	0 – 2.4	Kmol/min
24	Column Aqueous Exit	0.8361	0 – 2.4	Kmol/min
25	Column Bottom Exit	2.1584	0 – 4.536	Kmol/min
26	Vaporizer Liquid Inlet	2.1924	0 – 4.536	Kmol/min

Appendix 2 Control Structure and Controller Parameters

LOOP	Controlled Variable	Manipulated Variable	C.V. Value	Type	K _C	T _R (min)
1	%O ₂ in the Reactor Inlet	O ₂ fresh feed sp	7.5% (0 – 20)	PI	10	10
2	Gas Recycle Stream Pressure	C ₂ H ₄ fresh feed valve	128 psia (0 – 200)	PI	0.3	20
3	HAc Tank Level	HAc fresh feed valve	50% (0 – 100)	P	2	
4	Vaporizer Level	Vaporizer Heater Valve	70% (0 – 100)	PI	0.1	30
5	Vaporizer Pressure	Vaporizer Vapor Exit Valve	128 psia (0 – 200)	PI	5	10
6	Heater Exit Temp.	Reactor Preheater Valve	150 °C (120 – 170)	PI	1	5
7	Reactor Exit Temp.	Steam Drum Pressure sp	159.17 °C (0 – 200)	PI	3	10
8	Separator Level	Separator Liquid Exit Valve	50% (0 – 100)	P	5	
9	Separator Temp.	Separator Coolant Valve	40 °C (0 – 80)	PI	5	20
10	Separator Vapor Flowrate	Separator Vapor Exit Valve		Fixed		
11	Compressor Exit Temp.	Compressor Heater Valve	80 °C (70 – 90)	PI	1	5
12	Absorber level	Absorber Liquid Exit Valve	50% (0 – 100)	P	5	
13	Absorber Scrub Flowrate	HAc Tank Exit Valve 2		Fixed		
14	Circulation Stream Temp.	Absorber Scrub Heater Valve	25 °C (10 – 40)	PI	1	5
15	Absorber Circulation Flowrate	Absorber Circulation Valve		Fixed		
16	Scrub Stream Temp.	Circulation Cooler Valve	25 °C (10 – 40)	PI	1	5
17	%CO ₂ in the Gas Recycle	CO ₂ Purge Flowrate sp	0.764% (0 – 50%)	P	1	
18	%C ₂ H ₆ in the Gas Recycle	Purge Flowrate sp	25% (0 – 100%)	P	1	
19	FEHE Hot Exit Temp.	Bypass Valve	134 °C (0 – 200)	PI	5	10
20	%H ₂ O in the Column Bottom	Column Reflux Flowrate sp	9.344% (0 - 20)	PI	0.5	60
21	5 th tray Temperature	Reboiler Steam Valve	110 °C (0 – 120)	PI	20	30
22	Decanter Temperature	Column Condenser Duty	45.845 °C (40 – 50)	PI	1	5
23	Decanter Organic Level	Organic Product Flowrate	50% (0 – 100)	P	1	
24	Decanter Aqueous Level	Aqueous Product Flowrate	50% (0 – 100)	P	1	
25	Column Bottom Level	Column Bottom Flowrate	50% (0 – 100)	P	1	
26	Liquid Recycle Flowrate	HAc Tank Exit Valve 1		Fixed		

Appendix 3 Measurements at Steady State

Measurement	Description	Value	Unit
1	Vaporizer Pressure	128	Psia
2	Vaporizer Level	0.7	
3	Vaporizer Temperature	119.145	°C
4	Heater Exit Temperature	150	°C
5	Reactor Exit Temperature	159.17	°C
6	Reactor Exit Flowrate	18.857	Kmol/min
7	FEHE Cold Exit Temperature	97.1	°C
8	FEHE Hot Exit Temperature	134	°C
9	Separator Level	0.5	
10	Separator Temperature	40	°C
11	Compressor Exit Temperature	80	°C
12	Absorber Pressure	128	Psia
13	Absorber Level	0.5	
14	Circulation Cooler Exit Temperature	25	°C
15	Scrub Cooler Exit Temperature	25	°C
16	Gas Recycle Flowrate	16.5359	Kmol/min
17	Organic Product Flowrate	0.829	Kmol/min
18	Decanter Level (Organic)	0.5	
19	Decanter Level (Aqueous)	0.5	
20	Decanter Temperature	45.845	°C
21	Column Bottom Level	0.5	
22	5 th Tray Temperature	110	°C
23	HAc Tank Level	0.5	
24	Organic Product Composition (VAc, H ₂ O, HAc)	0.949786	%mol
25		0.049862	%mol
26		0.000352	%mol
27	Column Bottom Composition (VAc, H ₂ O, HAc)	0.000010	%mol
28		0.093440	%mol
29		0.906550	%mol
30	Gas Recycle Composition (O ₂ , CO ₂ , C ₂ H ₄ , C ₂ H ₆ , VAc, H ₂ O, HAc)	0.055664	%mol
31		0.007304	%mol
32		0.681208	%mol
33		0.249191	%mol
34		0.001597	%mol
35		0.000894	%mol
36		0.004142	%mol
37	Reactor Feed Composition (O ₂ , CO ₂ , C ₂ H ₄ , C ₂ H ₆ , VAc, H ₂ O, HAc)	0.075000	%mol
38		0.006273	%mol
39		0.585110	%mol
40		0.214038	%mol
41		0.001373	%mol
42		0.008558	%mol
43		0.109648	%mol

Table 1 Wilson parameters and molar volumes (ml/mol)

		Data						
a(i,j)		O ₂	CO ₂	C ₂ H ₄	C ₂ H ₆	VAc	H ₂ O	HAc
	O ₂	0	0	0	0	0	0	0
	CO ₂	0	0	0	0	0	0	0
	C ₂ H ₄	0	0	0	0	0	0	0
	C ₂ H ₆	0	0	0	0	0	0	0
	VAc	0	0	0	0	0	1384.6	-136.1
	H ₂ O	0	0	0	0	2266.4	0	670.7
	HAc	0	0	0	0	726.7	230.6	0
Vi	O ₂	64.178						
	CO ₂	37.400						
	C ₂ H ₄	49.347						
	C ₂ H ₆	52.866						
	VAc	101.564						
	H ₂ O	18.01						
	HAc	61.445						

Table 2 Equipment Data

Equipment	Data	Variable	Value
Vaporizer	Total Volume		17 m ³
	Working Level Volume		4 m ³
Reactor	Catalyst Density	ρ_b	385 Kg/m ³
	Catalyst Heat Capacity	C_{pb}	0.23 kcal/(kg*°C)
	Catalyst Porosity	ε	0.8
	UA per section	UA_rct	269.84 kcal/(min*°C *m ³)
	Tube Number	N	622
	Tube Length	L_{Tube}	10 m
	Tube Diameter	d	0.0371 m
	Friction Factor	F	0.000795 psia*(min) ² /(kg*m ³)
	Heat of Reaction	E_1 E_2	-42100 kcal/kmol -316000 kcal/kmol
	FEHE	Reference UA	UA ₀
	Reference Mass Flowrate of Cold Stream	F_{C_REF}	498.95 kg/min
	Reference Mass Flowrate of Hot Stream	F_{H_REF}	589.67 kg/min
Separator	Vapor Volume		170 m ³
	Working Level Volume		8 m ³
	UA	UA_sep	9075.18 kcal/(min*°C)
Compressor	Compressor Coefficient	γ	15000
Absorber	Working Level Volume		8.5 m ³
	Bottom Section		2 theoretical stages
	Top Section		6 theoretical stages
	Tray Holdup		13.61 kmol
	Hydraulic time	τ_{abs}	0.1 min
	Material transfer coefficients for both sections	Nmt	27.22 kmol/min
	Heat transfer coefficients for the bottom section	Qmt_bot	100.8 kcal/(°C *min)

Table 2 Equipment Data (Continued)

Equipment	Data	Variable	Value
Absorber	Heat transfer coefficients for the top section	Qmt_top	50.4 kcal/(°C *min)
Column	Theoretical Stage Number		20
	Feed Stage		15 from bottom
	Tray Holdup		2.3 kmol
	Hydraulic time	τ_{col}	0.1 min
	Top Pressure		18 psia
	Bottom Pressure		29.4 psia
	Reboiler Pressure		30 psia
	Base Working Level Volume		5.66 m ³
Decanter	Equilibrium Partition Coefficient	β_{VA_C} β_{H_2O} β_{HA_C}	395 0.05 1
	Organic Working Level Volume		1.7 m ³
	Aqueous Working Level Volume		1.7 m ³
HAc Tank	Working Level Volume		2.83 m ³

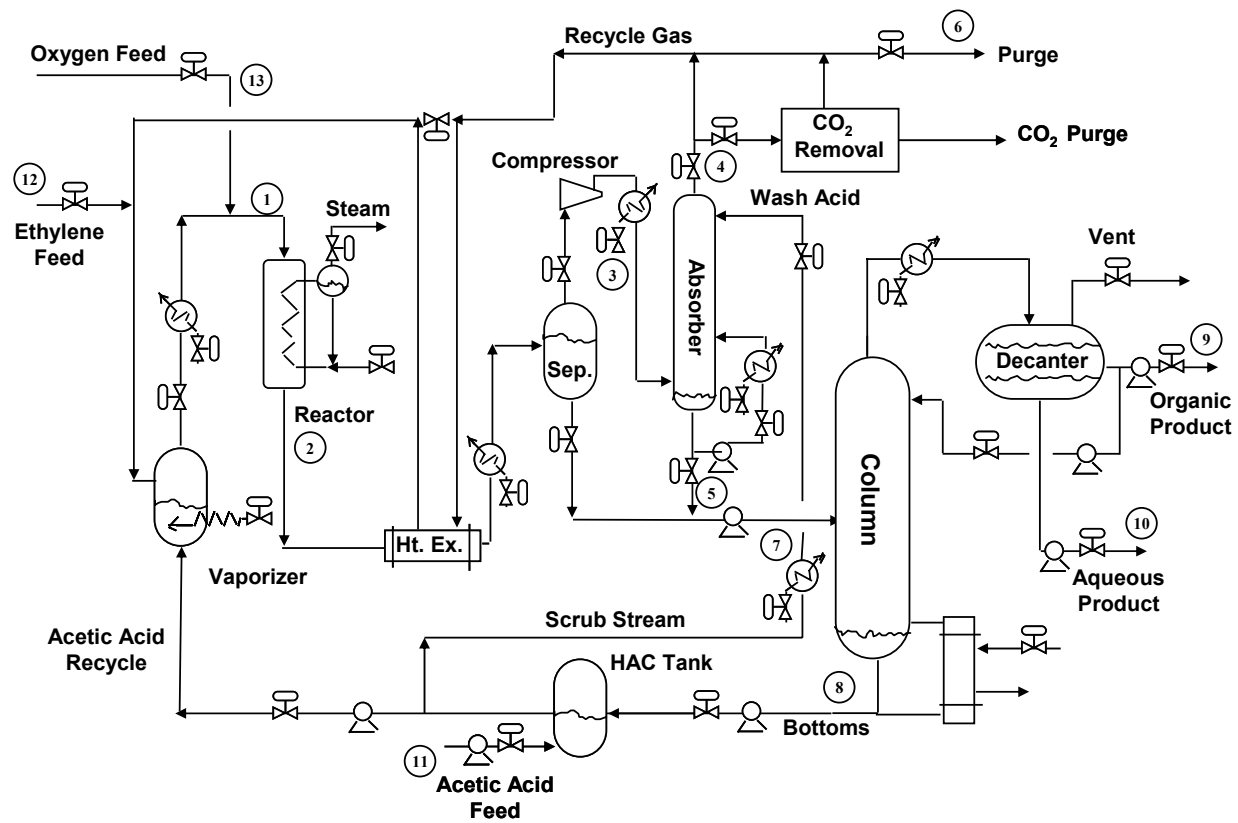


Figure 1. VAC Process Flow Sheet

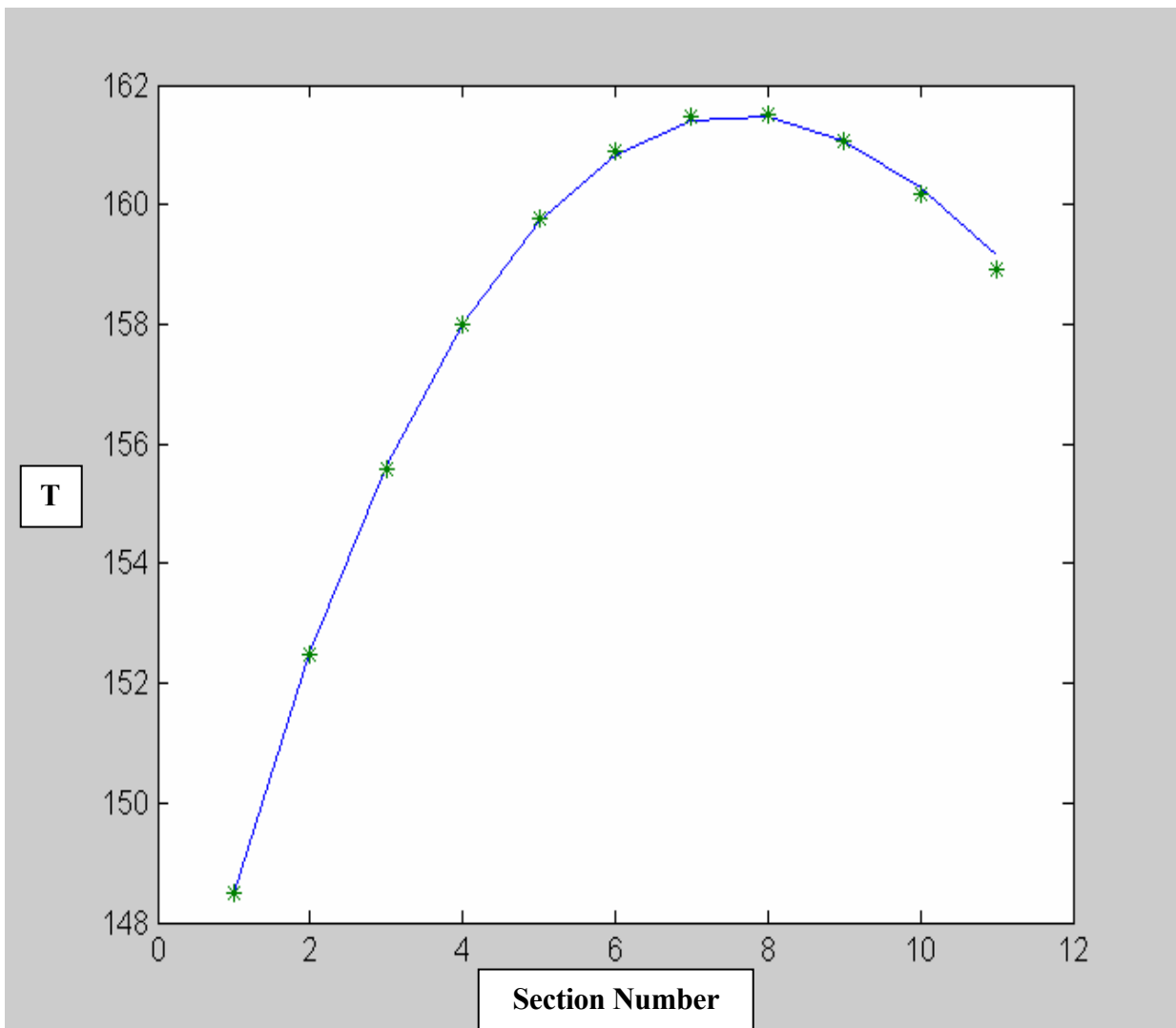


Figure 2. Reactor Temperature Profile. * Calculated in This Paper, — TMODES

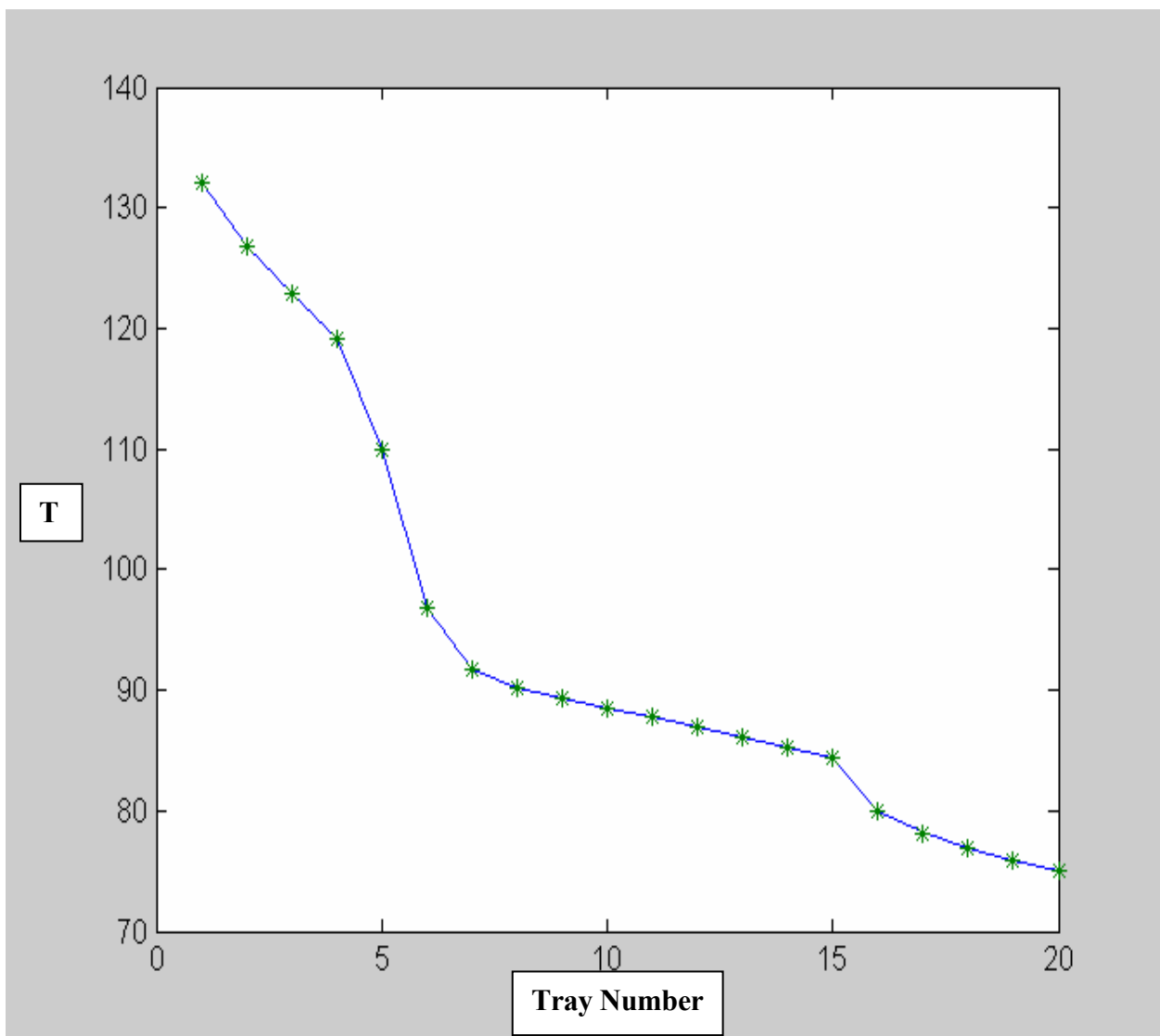


Figure 3. Column Temperature Profile. * Calculated in This Paper, — TMODS

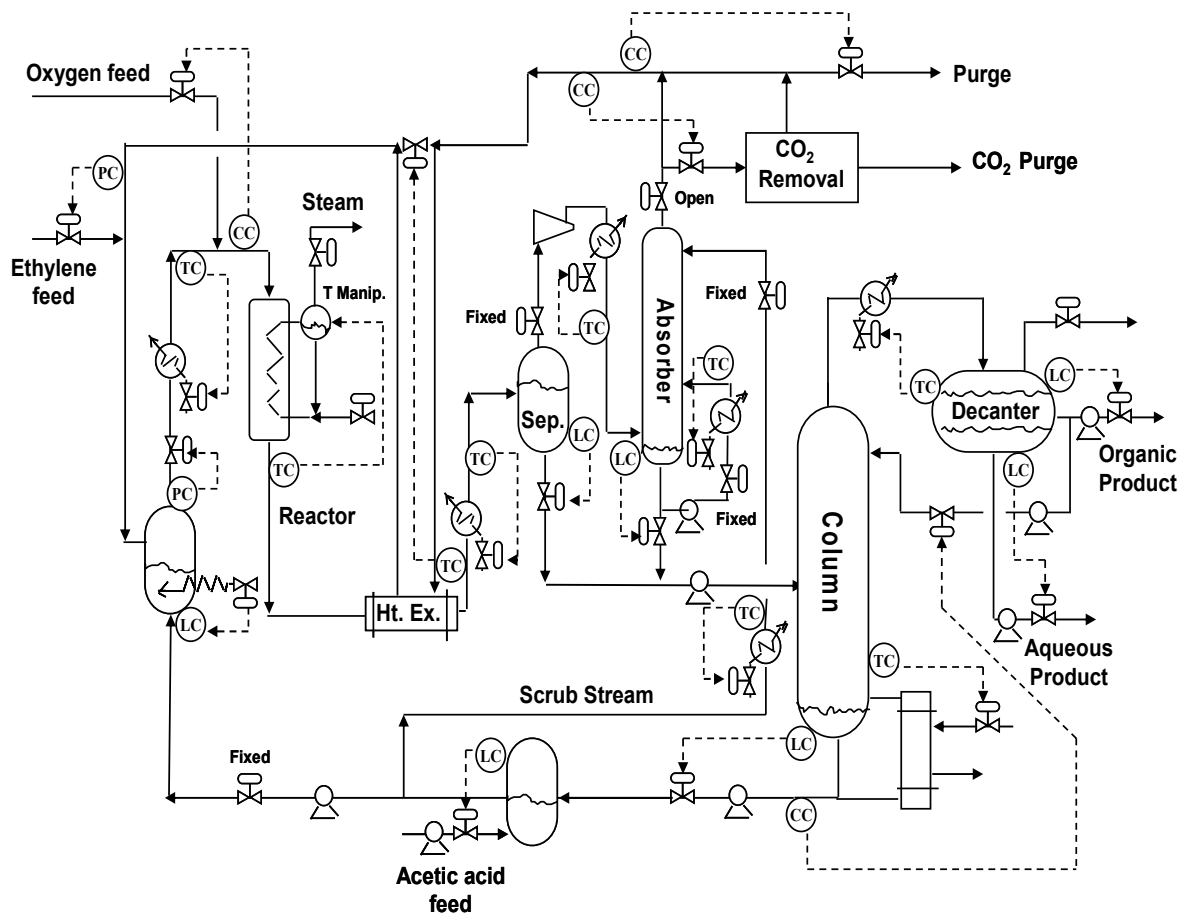


Figure 4. SISO Plantwide Control System

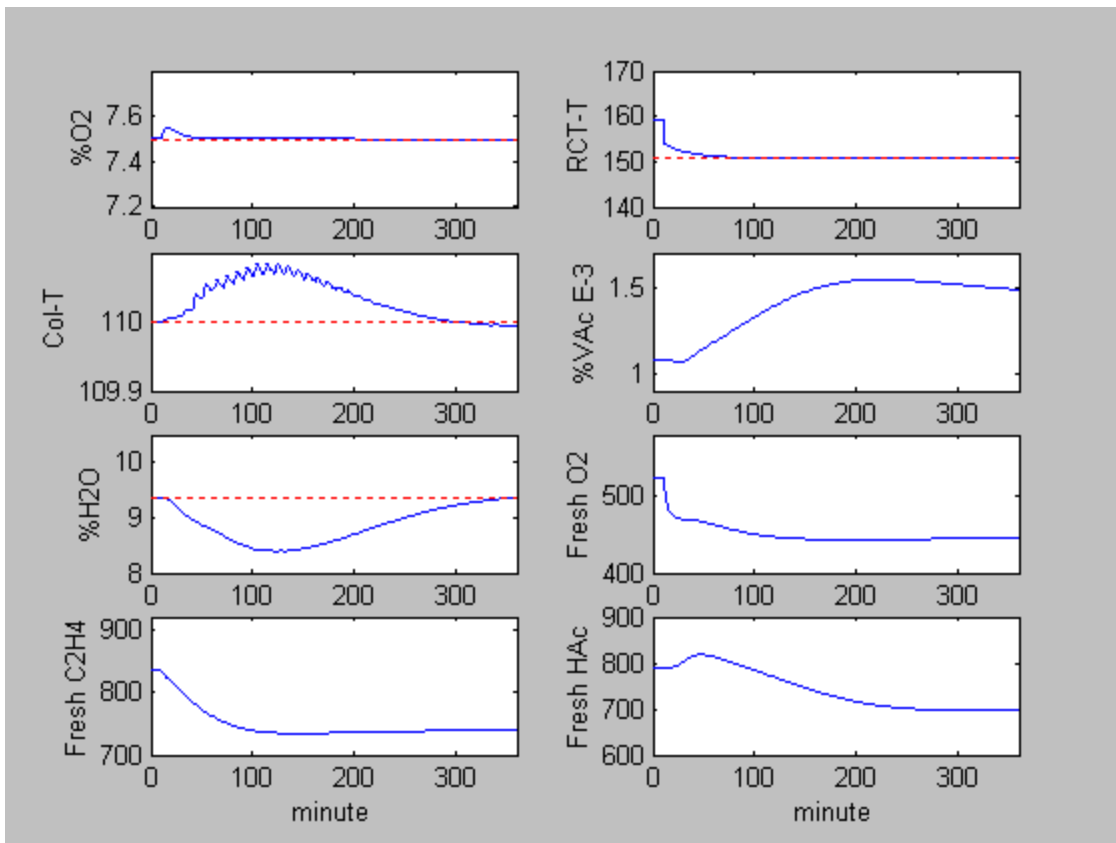


Figure 5. Reactor Outlet Temperature Decrease of 8 °C

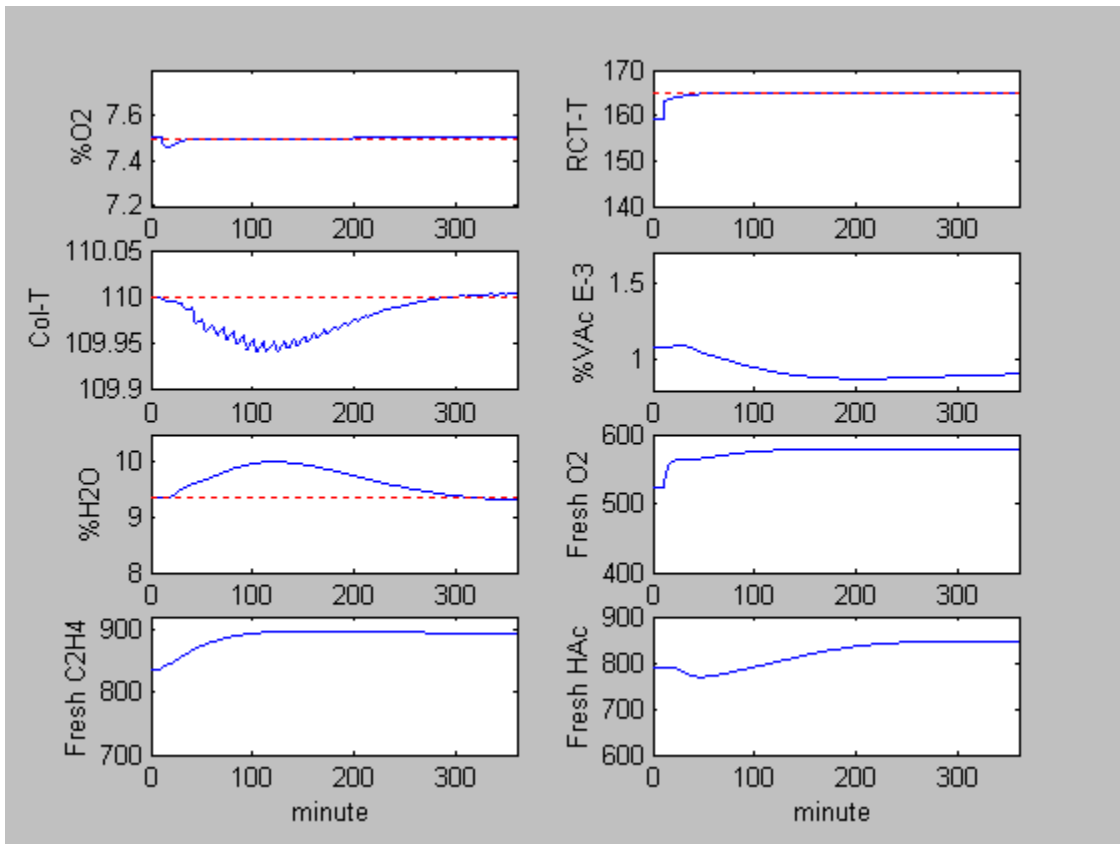


Figure 6. Reactor Outlet Temperature Increase of 6 °C

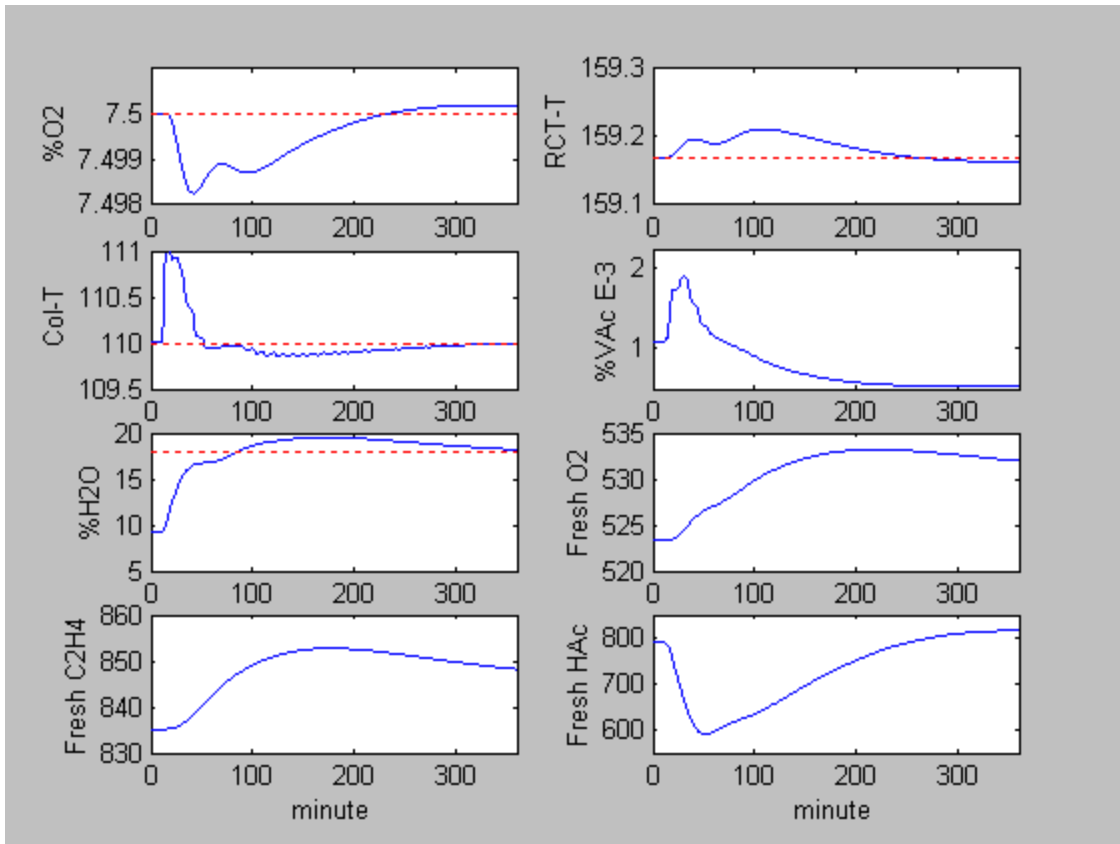


Figure 7. Water Concentration Increases 9% In Bottoms

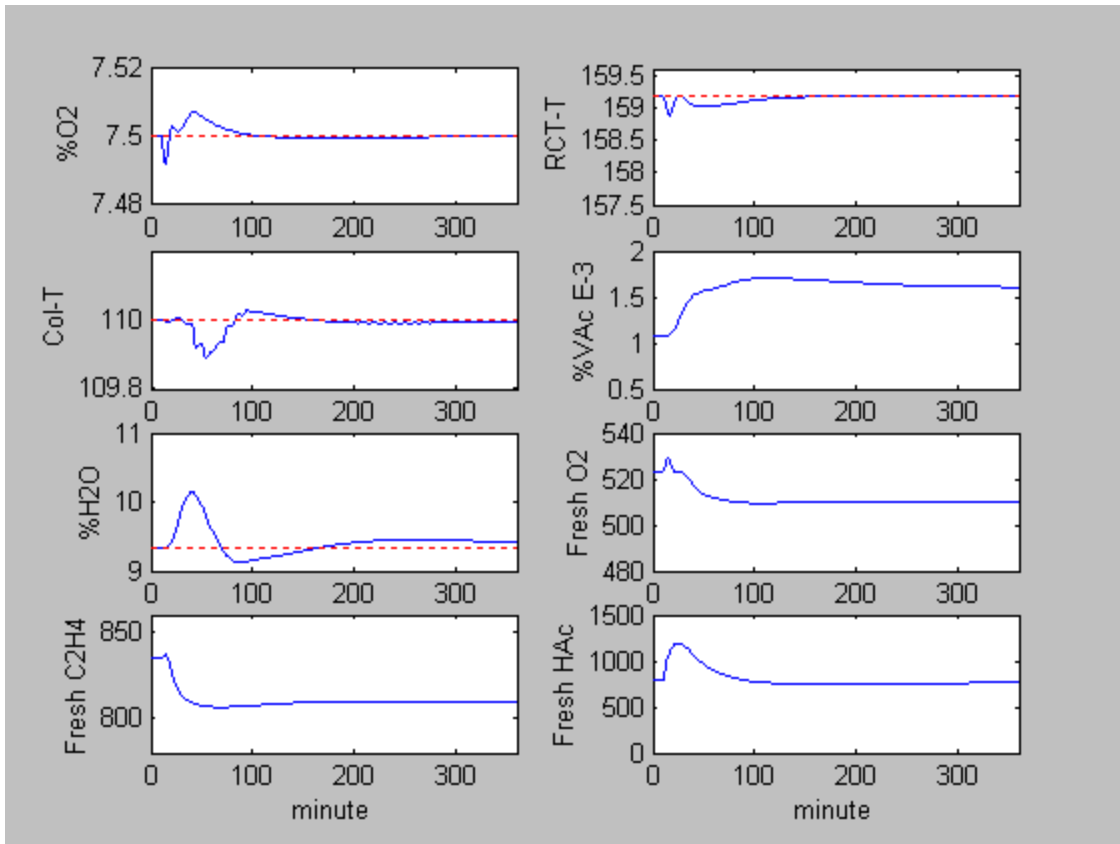


Figure 8. Liquid Recycle Flowrate Increases 20%