



Regular Article

Plantwide Control Structure of the Diethyl Oxalate Process Concerning: Safety and Process Improvement

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ARTICLE INFO

Article history:

Received: 2022-08-13

Accepted: 2023-04-15

Available online: 2023-04-15

Keywords:

Regeneration-Coupling Circulation,
Diethyl Oxalate Process,
Plant Wide Control,
Multi-Reactor Systems

ABSTRACT

During the regeneration-coupling process, a novel, plantwide control framework for the diethyl oxalate production unit is provided in this article. The advantage of this study is that it improves the process without the intricacy and expenses of the two structures described by Zhu and Luyben. The development of a plantwide control structure for this process was completed in two stages. The efficiency of the process was initially evaluated using a straightforward structure, with the primary goal being to prevent the usage of concentration controllers and complex cascading mechanisms to the greatest feasible extent. Due to the presence of persistent variations in the effluents of the process in the original structure, it was determined that there were numerous disruptions present that influenced the response during both recycle streams in the process and created variations. During the second phase, using trial and error to implement a functional adjustment in the process, the minimum amount of recycle stream, during which the variations were fully removed, was separated from the process, and a novel feed stream was inserted. Following the implementation of these modifications, it was discovered that variations in the effluent of the process are fully removed with just two concentration controllers, and this structure demonstrates instantaneous plantwide control over the received disturbances.

DOI: 10.22034/ijche.2023.355995.1455 URL: https://www.ijche.com/article_169891.html

1. Introduction

By advancing the development of industry and technology, the imbalance between supply and demand for energy resources leads the society to develop alternatives to fossil fuels [1]. The

C1 chemical technologies are considered as a potential method to solve this problem. The C1 technology is the science of using single carbon molecules. Although many compounds and ions contain only one carbon, four

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compounds of methane, carbon monoxide, carbon dioxide and methanol, which are of great industrial importance, are the focus of this technology's research. The process based on CO coupling to produce diethyl oxalate (DEO) is one of the most promising methods in C1 chemical technology [2, 3]. Diethyl oxalate is a chemical intermediate that has many uses in the industry, and its main use is in the pharmaceutical and paint industries. Diethyl oxalate is used in the pharmaceutical industry to produce API (the part of any drug that produces its effects), which can be used to produce high-value drugs [4]. This material is also used in the production of various colors and can be used as a solvent for a number of artificial and natural colors. Diethyl oxalate is also used as a cost-effective additive in dye-sensitized solar cells. Among its other uses, we can mention the production of pesticides [5]. Diethyl oxalate can be hydrogenated to produce ethylene glycol, an important chemical product used in the production of polyester fibers, antifreeze, unsaturated polyester resins, and plasticizers [5]. This chemical was formerly synthesized by an esterification technique leading to significant amounts of contamination and inadequate conversion levels [6]. Zhu et al [7] introduced a new flowsheet for the manufacturing of diethyl oxalate utilizing the carbon monoxide coupling process. The new method is more eco-friendly and more efficient than the conventional esterification. As a result of the existence of two strong recycle streams in this operation, every disturbance in the system multiplies the interaction effects into the process operations and increases the wastage of energy and material. In order to handle these challenges, a realistic simulation and efficient plantwide control structure are required. Zhu et al. [8] suggested the initial framework of

control for this process. This control system produced nearly satisfactory outcomes; however, the addition of four pricey combination controllers enhanced the structure's reaction duration and intricacy. Afterward, Luyben [9] attempted to change the process in order to decrease the number of concentration controllers and lower the cost of the process overall. Luyben accomplished this by employing cascading structures to mitigate the impacts of disturbances. As a consequence of successfully decreasing the three concentration control loops from the Luyben's flowsheet, the effluent findings were not as acceptable and precise as the design described by Zhu et al. [8], while the study leads to more deviation from the design parameters. A large, rapid, simple, and effective control structure is required for the process of the synthesis of diethyl oxalate owing to the availability of the significant volumes of hazardous gas and the significant ignitability of the compounds present in the reactors of this process as well as the existence of two large recycle streams in this process. Since any alteration to or disruption in the process could have significant impacts on the quality of the end product, leading to the wastage of energy and components or even triggering irreversible ecological and safety issues. The current research aims at developing an exhaustive, rapid, and efficient control structure that is both less expensive and simpler than earlier systems. Provided that the disturbances are decreased, this is achieved through modifying processes. For this operation, to implement the required structural improvements, we need a sustained and dynamic simulation of the process and the monitoring and analysis of the efficiency and response to disturbances of the process.

2. Diethyl oxalate process

The traditional technology for the synthesis of diethyl oxalate is the esterification process between oxalic acid and alcohol, using strong acids as catalysts, which causes serious environmental pollution and severe equipment corrosion. Also, due to the fact that this esterification reaction is reversible, and to reach equilibrium, it also proceeds in the opposite direction, so a large amount of toxic methylbenzene or benzene is required to achieve sufficient conversion. As a result, in addition to the existence of serious environmental risks, the amount of production is also low in this method [5]. The procedure suggested by Zhu et al. [7] for producing diethyl oxalate involves two reactive steps: coupling and regeneration. The coupling reaction occurs when the reactants carbon monoxide (CO) and ethyl nitrite (EN) are mixed to generate the major effluent of the process, diethyl oxalate (DEO), and the diethyl carbonate (DEC) as a by-product and the nitric oxide (NO) as a hazardous chemical. The other phase of the procedure involves a regeneration reaction. Recovering NO from the coupling

section causes it to react with ethanol (EtOH) and oxygen (O_2), resulting in the formation of EN and water (H_2O). The construction of a closed stream loop throughout this process, during which the created NO is recovered from the coupling reaction to the regeneration component to replicate EN, is the distinguished characteristic of the process. The EN created by this process is subsequently introduced into the partnering reactor to produce DEO and NO. The necessity to supply NO as a new feed to the system is a crucial aspect of the procedure. Considering an optimal process, when there are not any waste materials in the two intermediary reactants (nitric oxide and ethyl nitrite), the two chemicals disperse in the system cycle and manufacture and utilize one another. Therefore, it is unnecessary to provide these substances as new feeds to the process. Likewise, if there is no wastage of nitrogen (N_2), it merely travels in the recycle streams, as it is not engaged in any of the processes. Figure 1 depicts, with a few modifications, the flowchart for the process.

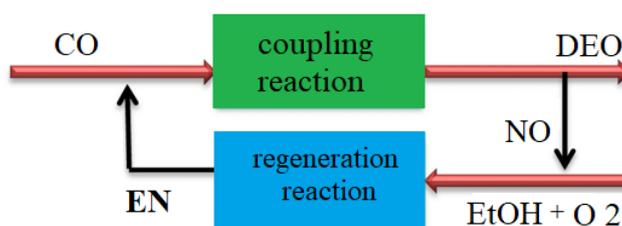


Figure 1. Schematic process of DEO [7].

In many cases, the procedure of the diethyl oxalate synthesis is conducted in low-pressure circumstances (maximum 5 atm), and because it involves the application of non-ideal solutions (water and ethanol), it is the optimal model to use for the NRTL procedure. Among many thermodynamic models for non-ideal

liquids and containing pressures below 10 atm, the NRTL activity coefficient model has the greatest efficiency. However, this model is insensitive to gaseous combinations. In this process, the dual thermodynamic model NRTL-RK has been examined due to the considerable gas mixture, which has been

employed in prior studies, being involved [10]. This model includes the subsequent stages.

1. Model of the NRTL activity coefficient regarding liquid phase.
2. Redlich-Kwong state equation regarding the vapor phase.
3. Henry's laws describe the tendency of non-condensable gases in liquids.

Hu et al. [11] developed the binary parameters that are not present in the Aspen database from earlier investigations. The

software database has been updated to include this simulation to further develop the thermodynamic model. The flowsheet of this process with a few changes can be seen in Figure 2.

2.1. Regeneration section

This part works toward achieving common purposes such as the synthesis of EN, the usage and avoidance of the wastage of NO during recycle streams, and the separation of surplus water and other pollutants from the process.

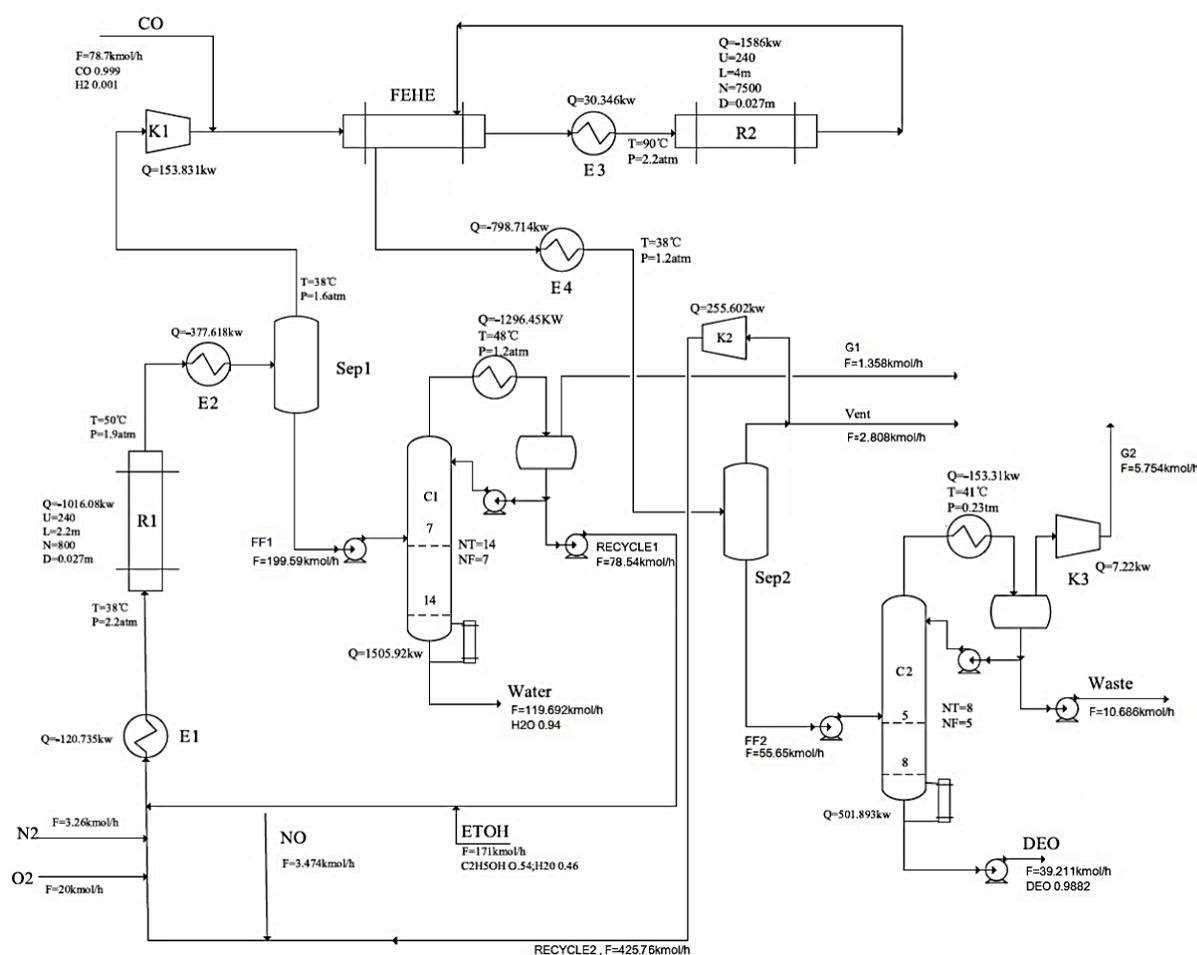
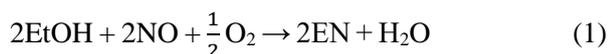


Figure 2. Flowsheet of the process of the diethyl oxalate synthesis [7].

This portion eliminates the need for adding a substantial quantity of additional EN to the process and recycling the majority of the

excessive effluent for reutilization. In this part, new feeds of oxygen (20 kmol/h), nitric oxide (648.1 kmol/h), nitrogen (23/3 kmol/h), as well

as a combination of ethanol and water (171 kmol/h) are mixed with recycle streams derived from Column C1 and the Sep2 reflux drum stream container reactor R1. The constituents of ethanol, nitric oxide, and oxygen react with one another in this reactor, producing ethyl nitrite and water in the reaction as follows (1).



This gas-phase reaction is exothermic and irreversible. The reaction is carried out at a low-pressure state employing inert nitrogen since there is a smaller amount of product than reactants. The regeneration reactor, or R1, is a tube-type reactor comprised of 800 tubes with 2.117 and 0.027 meters in length and diameter respectively. At 38 °C, the effluent in reactor R1 exits the heat exchanger E2 and is returned to the reflux drum Sep1 following the reaction and the generation of ethyl nitrite and water. The effluent from the Sep1 reflux drum is sent to the C1 distillation column, which has a capacity of 14 stages. The water generated during the reaction and exited from the bottom of the column has approximately 94 % concentration. This column functions using the cooling water in a condenser that is set to 49 °C and 1.2 bar. As for the feeding, the liquid containing a purity of 68 % moles of ethanol exiting the top of the column is recycled to the R1 reactor. In addition, to eliminate non-condensing light weight components infiltrating the column via the reflux drum, there is a small steam purification effluent (1.3 kmol/h) at the top of the column. From tray number 7, feed reaches the column. The reboiler duty, column diameter, reflux stream ratio, and distillation rate are 1411 kW, 0.717 m, 1, and 79.9 kmol/h respectively. The effluent gas in the Sep1 reflux drum achieves

49.2 bar pressure in the K1 compressor before entering the coupling section.

2.2. Coupling section

Following the compression by the K1 compressor, the Sep1 drum's exhaust gas stream is coupled with the fresh CO feed at a stream rate of 42.78 kmol/h. Before reaching the coupling reactor, it is heated to 90 °C using a heat exchanger 124 kW (E3). Diethyl oxalate, and nitric oxide are produced by the interaction between ethyl nitrite and carbon monoxide during the coupling reaction.



The reaction (2) in the gas phase is exothermic and irreversible. Attributable to a side reaction (3) in R2, extremely small quantities of diethyl carbonate are generated as well.



This reactor, which is similar to a tubular reactor has made up of 6500 pipes with the size of 4 meters in length and 0.027 meters in diameter. With a total heat transfer coefficient of 0.28 kWm⁻²K⁻¹, the reactor cooling load of the reactor is 1561 kW. The separation process begins when the effluent stream passes through the Sep2 reflux drum into the C2 distillation column. The column includes eight trays that operate under linear circumstances of 0.223 bar. With a stream rate of 392 kmol/h and a concentration of 98 % mol, the final product, diethyl oxalate, is released from the bottom of the column. At 41 °C, the reflux drum of the column generates two streams of steam (6 kmol/h) and distilled liquid (43.10 kmol/h) that are transported to a waste disposal facility. The boiling heat is 511 kW, and the column diameter is 0.8 meters as the feed

arrives from stage 5. The anticipated reflux stream ratio is 1.3. A ventilation stream (Vent) and an input recycle stream to the R1 reactor are both supplied by a portion of the exhaust gas coming from the Sep2 reflux drum. Before reaching reactor R1, the recycle stream should be compressed to 2.7 bar using K2 compressor. The kinetics of the coupling reaction have been reviewed in several studies [12-14]. During recent studies, there have been two types of kinetic expressions for the process including the Langmuir-Hinshelwood-Hougen-Watson method and power-law method. In this article, due to the investigation of the complete kinetic parameters and description of the side reaction, the power law method is used. The kinetics of the reduction reaction have also been studied by Chen et al. [15] and Fang et al. [3], and in the conclusion of their research, the power law method was chosen as the best method. The kinetic expressions of coupling, regeneration and side reactions are shown in the following equations:

Coupling reaction:

$$R_1 = K_1 p_{CO}^{1.03} p_{EN}^{0.76} p_{NO}^{-0.81} \quad (\text{Eq. 1})$$

Side reaction:

$$R_2 = K_2 p_{CO}^{0.88} p_{EN}^{0.93} p_{NO}^{-0.35} \quad (\text{Eq. 2})$$

Regeneration reaction:

$$R_3 = K_3 p_{NO}^1 \quad (\text{Eq. 3})$$

Coefficient K in the equations:

$$K_i = A \exp\left(-\frac{E_a}{RT}\right) \quad (\text{Eq. 4})$$

where, p is pressure (pa), R_1 and R_2 are the reaction rates in kmol/(kgcat.s) while R_3 is the reaction rate in mol/(m³.s). The parameters of

the reactions in this process are given in the Table. 1 [15, 16].

Table 1

Kinetic parameters of the process of diethyl oxalate reactions.

	A Factor	Ea (kJ/kmol)
R ₁	8.5686×10 ⁻⁸	18700
R ₂	2.2591×10 ⁻¹⁰	17900
R ₃	3.89×10 ⁻³	19400

2.3. Plantwide control

The PI integral proportional controller is used to inversely control the streams of CO, O₂, N₂, NO, and ETOH at the feedings of the process's five inputs. Because of their fast speed and the removal of the proportional controller's control drop, integral proportional controllers are ideal for regulating the flow rate of the process streams. It should be observed, nevertheless, that when an essential load is added, the response becomes more fluctuated, which is mitigated by reduced interest levels. Additionally, the response is accelerated by lowering the integral term duration. The gas and liquid separation activities that take place in the two reflux drums during the process are extremely important to the product separation and generation processes. The liquid that is discharged from either of the drums is fed directly into either of the distillation columns. In addition, the fluctuating levels of the liquid in the drums impede the separation between distillation columns C1 and C2. The liquid level in the two Sep1 and Sep2 tanks, with the recycle stream, is managed by regulating the exhaust fluid coming from the tank by employing a controlling valve on the way of the effluent stream and a proportionate controller. Using an integral proportional controller to regulate these tanks changes the system since the fluid level in these tanks fluctuates fast. In order to manage the liquid

level in the reflux drums, proportional controllers with linear performance and relatively low velocity are the best options. Using integral proportional controllers, the four heat exchangers in the process are regulated in reverse. By adjusting the converter's heat, the temperature of the effluent in each converter can be manipulated to offset disturbances. Controlling the temperature of the reactors, much like the heat exchangers, is accomplished via integral proportional controllers. By controlling the stream of the cooling water through the reactors, the temperature of the product can be manipulated. Owing to its simplicity and low maintenance cost, the single-point control is commonly employed in an industrial distillation processes. As suggested by Luyben [17], one of the better approaches is to choose trays in the column in which there are significant temperature differences from one tray to another. This approach involves drawing the temperature profile diagram under simulation settings and analyzing the slope of the diagram. The tray with the biggest fluctuation in temperature is indicated on the

graph by the spot that has the steepest slope. If there is a significant difference in temperatures from one tray to the next, this may be an indication that the concentration of critical components is shifting more quickly in that region. The distribution of the components of the composition in the distillation column are preserved by maintaining the tray's temperature at this location. This stops the light weight constituents leaving the bottom of the column and heavy components from the top. Based on the temperature profile for distillation columns which can be seen in Figure 3 and Figure 4, trays 13 and 6 are the optimal choices for managing the tray temperature in columns C1 and C2 respectively, since their temperature variations have the steepest slope in these diagrams. After selecting the appropriate tray within the columns, the temperature of the tray is inputted as a programmable parameter into a PI controller. This allows the PI controller to adjust the input heat concerning the boiler to maintain the favored temperature at the setting point for each column. In these loops, control is performed backward.

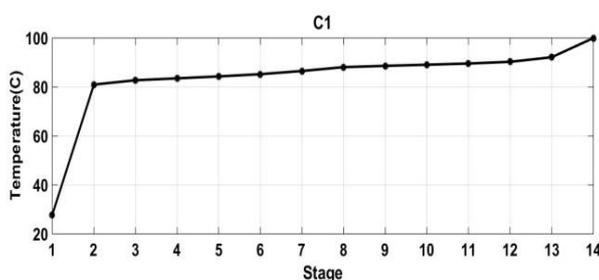


Figure 3. The temperature profile of column C1.

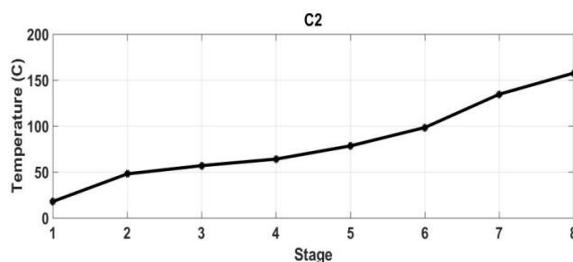


Figure 4. The temperature profile of column C2.

The following are additional control loops utilized in distillation columns C1 and C2: By employing a proportional controller, the liquid level in backward stream tanks in the cooling portion of columns C1 and C2 is controlled directly. The effluent controller in column C1 is linked to the pump. The effluent controller in column C2 is linked to the valve on the way of the effluent, allowing the favored effluent to be adjusted by regulating the pump shaft velocity and opening and shutting the valve. By adjusting the control valve located on the way of the lower effluent of the columns and using proportional controllers, the fluid level present at the base of columns C1 and C2 can be kept at the desired level. Utilizing an integral proportional controller and managing the cooling heat in reflux, the cooling pressure in both columns is regulated.

2.4. Concentration controllers

The concentration control enhances the precision of maintaining a steady ratio of the required streams at various points in the operation. However, as previously stated, these controllers have a slow response period and a considerable expense, which cause developers of control systems to eschew employing them whenever possible. In developing these controllers in the sequence mode and linking them to another controller as initial and secondary controllers, it is attempted to correct the issue of slow regulation of these controllers as much as feasible with the aid of the secondary controller, even if the direct concentration regulation is needed. The flammability and explosive strength of the reactants in the production of diethyl oxalate is one of the essential safety considerations mentioned. Because of its great degradability, the EN compound poses an explosion danger. The

breakdown temperature and critical degradation pressure of EN are low, according to the investigations conducted by Ochiumi et al. [18] and Zhang [19]. This endangers the entire factory and its personnel. It is important to keep in mind that the explosion in the process results in damage to the plant and its personnel and induces a substantial amount of nitrogen oxide to be released into the atmosphere, which pollutes the environment and creates a number of other ecological problems. Hence, we add nitrogen gas to the entire system, which is neither created nor absorbed by the reactions. This reduces the pressure of the reactants, so diluting them and therefor decreasing their concentration. This prevents the reactants from igniting and causing security concerns. By blocking the EN breakdown, even at small levels, nitrogen also suppresses the creation of negative reactions in the process. In addition, the pressure in the system is minimal, which lowers the partial pressure of the EN; yet, nitrogen's influence on the process to avoid a disaster should not be ignored. Therefore, it appears vital to regulate the nitrogen concentration in the process. Nitrogen makes up the majority of the exhaust gas released from the Sep1 reflux drum, and its concentration reaches 37.4 % mol in this stream before being introduced into the R2 reactor. This location is ideal for regulating the concentration of this compound. The concentration of nitrogen can be controlled by utilizing a PI controller. For the higher precision, the controller is employed both as part of a sequential control loop and as a setting point for the feed stream controller responsible for the supply of additional nitrogen. This will minimize the issue by adjusting the stream rate of the input feed in the occasion that the concentration of this component and the control structure is

perturbed. Another crucial element, carbon monoxide, of which the concentration must be tightly controlled at all times. Because of the significance of this compound in reactions and our decision to use oxygen stream as the “throughput manipulator” (TPM), we are required to pay closer attention to the quantity of carbon monoxide that is produced during the process to ensure that the process in the second reactor and the creation of diethyl oxalate is not disrupted. The recycle stream's carbon monoxide gets mixed with the new input stream's CO and reaches reactor R2. The product stream comprising 5.7 mol % carbon monoxide is discharged from the reactor following the coupling reaction. The molar concentration of CO in the effluent at the reactor shows how much of this molecule is utilized throughout the reaction, and provided there is any disruption (even a major temperature disturbance impacting the process), the concentration at this point will fluctuate significantly. We can regulate disruptions that directly and indirectly influence the concentration and stream of carbon monoxide and, eventually, the R2 reactor by altering the concentration at this point and sending the effluent to the CO feed freshwater stream controller as a cascading control. Similarly to the concentration of nitrogen, the concentration of carbon monoxide control loop employs a concentration-stream cascading control and an inverted integral proportional controller.

2.5. Reforming

By producing a 10 % disruption in the TPM, it is evident that the structure can effectively govern the system. In this instance, nevertheless, the effluent variations of the controllers are not entirely removed. Although the magnitude of these variations is small, they

may eventually disturb the process as larger disturbances develop. Therefore, it suffices to minimize the fluctuations. This becomes complicated by the presence of two large recycles that reflux these fluctuations to the system and eventually amplify them. Measurements of recycles reveal a considerable propensity for the recycle rate to fluctuate continuously. This results in significant fluctuations during the process. Even with slight variations in the feed, these systems experience wide fluctuations that frequently intensify with time [20]. Hence, the existence of two significant recycles in this process leads the system to fluctuate in an undesirable manner. This implies that the present variations are somewhat bigger than the structure's control potential and small adjustments are required in the process. This modification should be modest and should not affect the primary procedure or the anticipated effluents. In order to isolate the effluent at the top of column C, we need a thorough refurbishment and the installation of additional equipment for the process. The secondary recycle, which is the outcome of the Sep2 separation, is divided into two streams prior to being returned to the system using SPLIT. As a result, the stream from the RECYCLE2 stream is sent back into the system, while the stream from the Vent stream is transferred to the effluent. It is only possible to reduce the quantity of recycle by increasing the amount of spacing between these two points. Nevertheless, the substances introduced to the vent stream must be delivered into the system as a new feed so as not to affect the process's total efficiency and effluent. This must be accomplished as follows:

1. Do not eliminate costly chemicals, including EN and ETOH, from the system.

2. Failure to emit substantial amounts of NO.
3. Apply as fewest adjustments to the system as feasible.
4. This is accomplished through trial and error, emphasizing the separation of chemicals O₂, NO, and N₂, which constitute the majority of this stream.

Inappropriate variations are resolved by separating 3.2 kmol/h of RECYCLE2 and reintroducing it to the discharge, as determined by many testings. The separated stream is brought into the process as what is known as a "NEW" feed, which is a fresh input. This stream consists of 1.9 kmol/h of nitric oxide, 0.99 kmol/h of nitrogen, and 0.3 kmol/l of oxygen. A stream control loop has been incorporated into the incoming feed stream to improve the efficiency of the structure. The arrangement of other control circuits and the

types of controllers remain unchanged. Nevertheless, following each assessment, the settings of the temperature and concentration controllers are reset to new values. This fresh value, moreover, is not significantly distinct from the values that were found in earlier tests. The flowsheet of the final control structure in Aspen Plus Dynamics is elucidated in Figure 5, while the schematic diagram of the proposed control structure for better comparison with Figure 2 (diethyl oxalate process) has been depicted in Figure 5. Table 2 displays the ultimate adjustment constants for the temperature and composition controls. Based on the the final structure the fluctuations have been entirely removed, and as a result, we have developed a structure that is both transparent and durable. In the following, a comparison of the findings with the two preceding structures will be demonstrated.

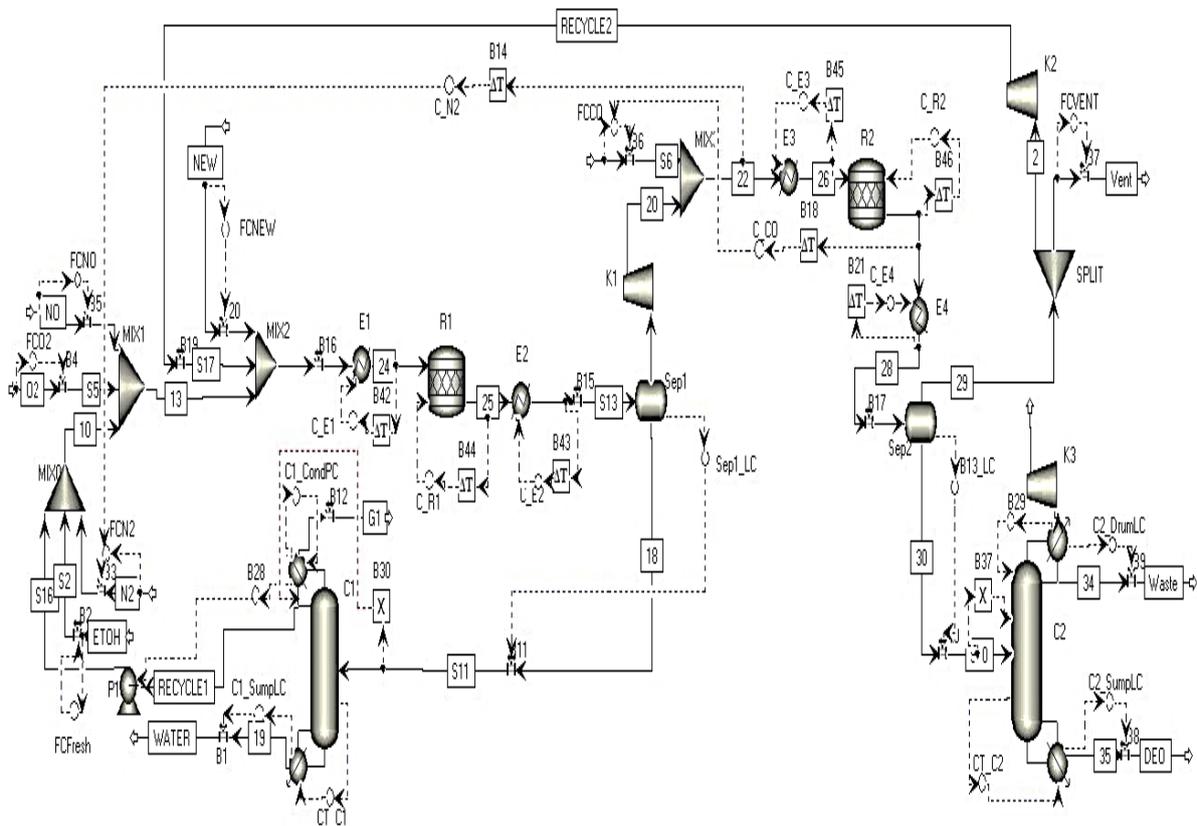


Figure 5. Flowsheet of the final control structure in Aspen Plus Dynamics.

Table 2

Final Control parameters of important control loops.

Controller	Action	K _C (%/ %)	τ (min)
C_E ₁	Reverse	0.516	5.28
C_E ₂	Reverse	1.224	5.28
C_E ₃	Reverse	0.665	3.96
C_E ₄	Reverse	1.226	9.6
C_R ₁	Reverse	1.128	3.96
C_R ₂	Reverse	0.517	5.26
CT_C ₁	Reverse	5.18	5.28
CT_C ₂	Reverse	2.46	9.24
C_CO	Reverse	0.76	19.8
C_N ₂	Reverse	0.8	21.3

2.6. Earlier constructions

The following is a comparison and discussion for the effectiveness of each control arrangement which was described in this paper. To facilitate comparisons, the structure described in this study is designated CS3, whereas the Zhu et al. [8] and Luyben [9] structures are designated CS1 and CS2 respectively. It is recommended that we begin by talking about the general control structure that was recommended by Zhu et al. [8]. As illustrated in Figure 6, the most significant aspect of this structure is the employment of four online mixture controllers to regulate the four mixtures of N₂, O₂, H₂, and CO in the process. By adjusting the four current streams of N₂, NO, Vent, and CO, these variables can be regulated. In this structure, the operational control stream is also regarded as the oxygen TPM. High conversion rates can be achieved with oxygen in the regeneration reactor (87 %). As a consequence of this, the composition of the gas stream that exists from the separator reflux drum following the reactor contains only 0.72 % mol of oxygen. This value is

manipulated in this system by adjusting the nitric oxide feed stream as a manipulating parameter and controlling it through the application of a composition controller. The controllers are regulated according to the Tyreus-Luyben standards, taking into account three minutes of the inactive time for mixture controllers and one minute for temperature controllers. By controlling the supply of carbon monoxide, the concentration of carbon monoxide exiting the R2 reactor can be adjusted (using the proportion of carbon monoxide to the O₂ feed). The removal of the fresh nitrogen feed after exiting the Sep1 reflux drum is another method utilized for controlling the concentration of nitrogen. By adjusting the Vent stream rate, the concentration of hydrogen in the exhaust gas stream coming from the reflux drum Sep2 (RECYCLE2) can be regulated. Zhu et al [8] has not justified this control loop. On the other hand, it is apparently not required in this process to maintain a steady concentration of H₂.

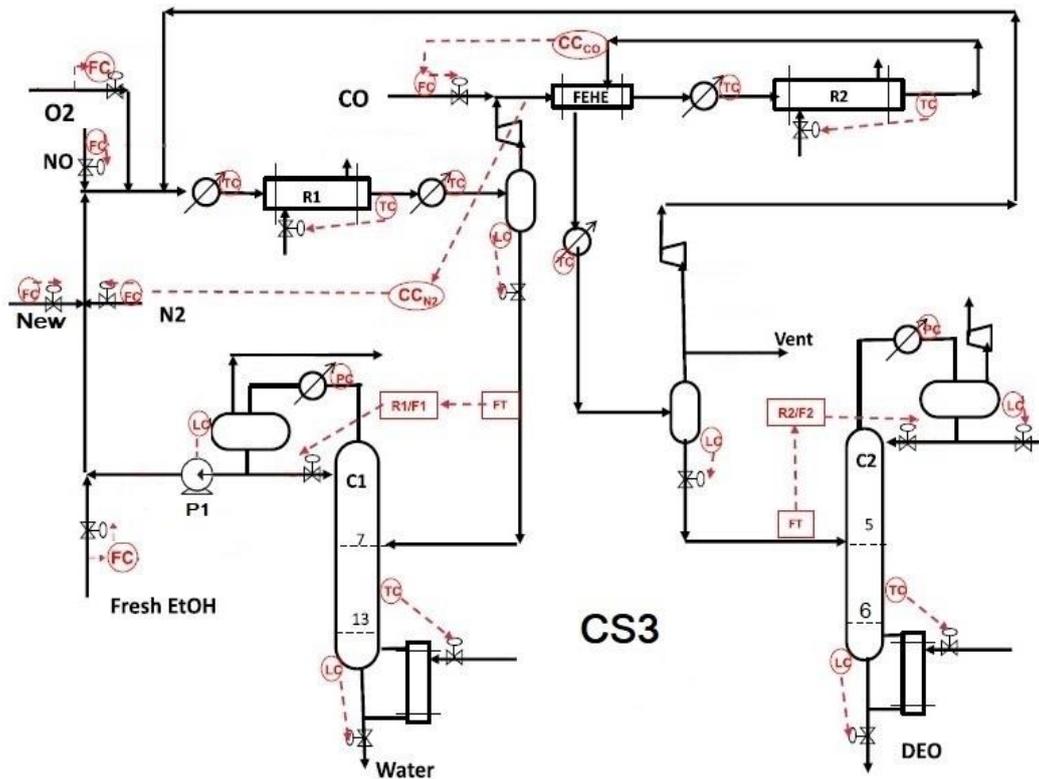


Figure 6. Plantwide control structure CS3.

Hydrogen has little influence on the primary reactions and effluents, and the loop is presumably designed to avoid the fluctuations of the RECYCLE2 recycle stream. Despite the fact that Zhu et al. have accomplished the same as we demonstrated in the preceding chapter, it does not appear to be required to create this concentration controlling loop. A similar controlling structure is utilized during the control of distillation columns in both columns, namely the constant ratio of the recycle stream rates to the feed stream and the temperature regulation of the bottom tray. The heat of the boiler is used to regulate the temperature of the tray that is indicated in both distillation columns. The pressure of the columns is also managed by adjusting the temperature of the heat of compression. Changing the effluent stream at the column's base controls the liquid level at the top of both columns. By managing the waste stream, the level of the reflux drum in column C2 is

regulated. Employing an uncommon cascading control loop, the reflux drum level in column C1 is controlled. During RECYCLE1, the liquid stream is managed by controlling the new ETOH feed. The point at which this controller is adjusted, on the other hand, is variable depending on the effluent signal received from the level regulator of reflux drum C1. In this configuration, the O₂ and CO input rates are set in order to minimize disturbance. In this regard, we will observe that this process results in dramatic fluctuations in the CO feeding when the O₂ feed stream changes. Whenever the disturbance enters, the existence of a cascading loop in the management of the concentration of oxygen generates a considerable divergence in the ultimate product, particularly DEO. Additional control loops, such as temperature, surface, and discharge loops, are typical and are built and calibrated in a manner to be analogous to that of the CS3 structure.

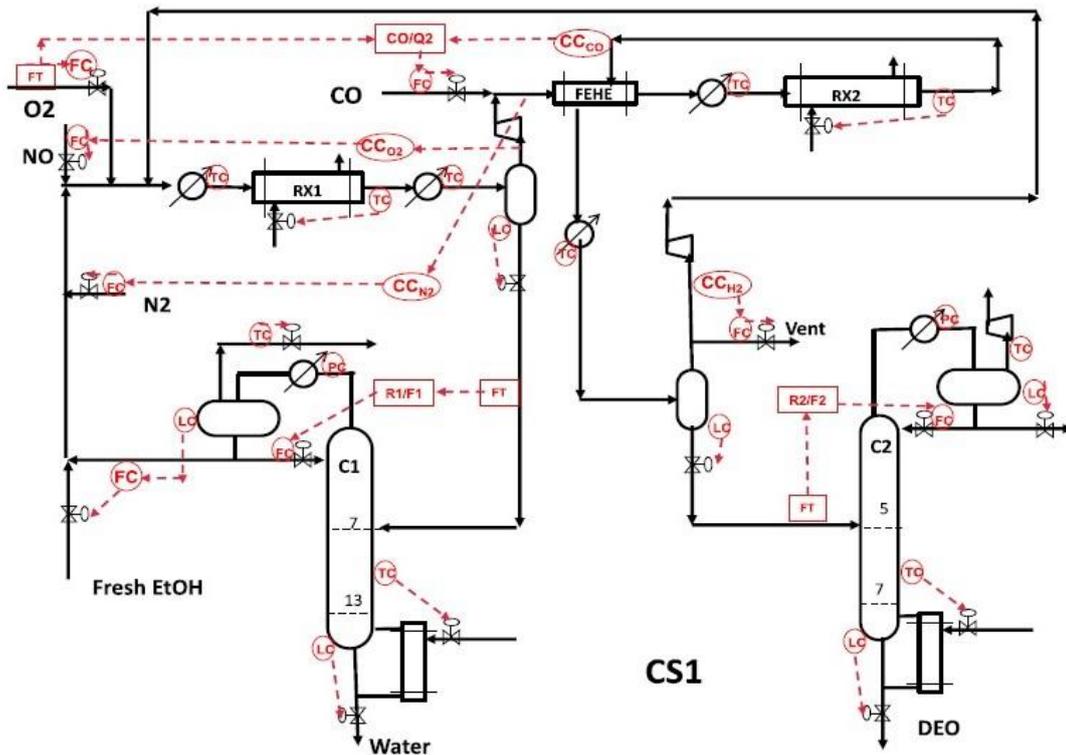


Figure 7. Plantwide control structure CS1.

2.7. Luyben's structure

Luyben's (2019) control system [9] for CS2 is depicted in Figure 8. The TPM in this structure is equal to the sum of the RECYCLE1 and ETOH streams. This stream has been given the name "Total" by Luyben by adding a control valve in order to manipulate the recycle and the feed streams. The effluent pump head of column C1 is enhanced to generate the sufficient pressure for this fresh valve to be utilized for Total regulation. Due to the fact that a stream controller in Total maintains a constant total flow rate, the decrease in the fresh feed causes an elevation in the amount of the fluid that is discharged from the reflux drum, which in turn allows the level of the fluid in the tank to decrease. The oxygen gas coming from the Sep1 reflux drum is the only compound that can be altered in the CS2 structure. This can be done by adjusting the NO feed stream (similar to the loop employed in CS1). Luyben's technique provided in this

structure, "maintaining a constant total stream while varying the feed", is a sophisticated cascading structure utilized in the management of numerous chemical processes involving recycle streams to successfully regulate the feed input. In this construction, four ratios are employed to modify the fresh feeds of oxygen, carbon monoxide, nitrogen, and the Vent effluent. Whenever the Total fluctuates, these feeds are modified to achieve the appropriate proportion. In other terms, the stream ratio of the mentioned streams to the TOTAL stream is regarded constant and reaches the stream regulators of these four streams as the input setpoint, allowing the process to be controlled in the event of a TOTAL structure alteration by adjusting these streams. Luyben explains the effectiveness of this strategy, which states that there are considerable ethanol, carbon monoxide, nitric oxide, ethyl nitrite, and nitrogen wastes in the four effluent streams of the process (G1, G2, Waste, and Vent) Due to

this characteristic, it is not required to maintain an accurate equilibrium between control variables to prevent accumulation and divergence from the set point. Despite the fact that this simplifies the structure, it is essential to assess the nitrogen content in this procedure owing to safety concerns. Luyben abstained from introducing it to the system and instead maintained a steady proportion of N₂ to Total. All other controlling loops are identical to the loops in the CS structure. The structural

performance of CS3 is compared to that of CS1 and CS2. Now because a comprehensive grasp of the two structures described in the previous section has been attained, it is necessary to evaluate the functionality of both structures using the broad control structure provided in this research. The disturbances are performed by lowering the purity of CO, lowering the purity of N₂, and enhancing the TPM flow rate. The consequences of these changes are documented as comparative diagrams.

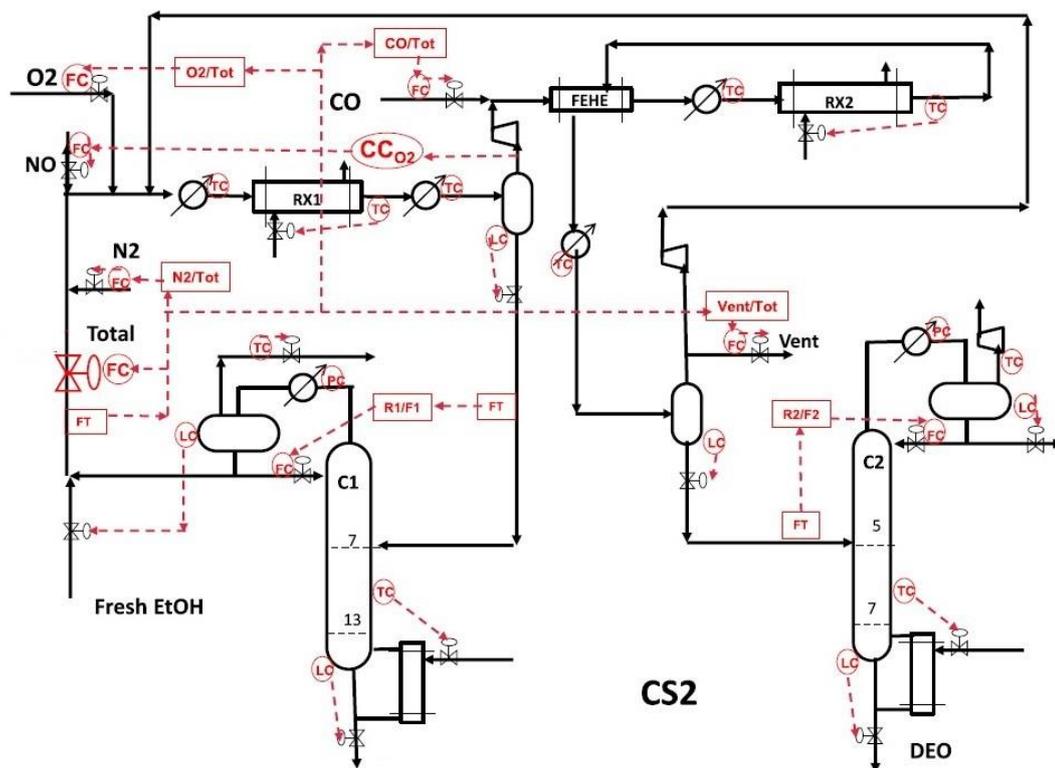


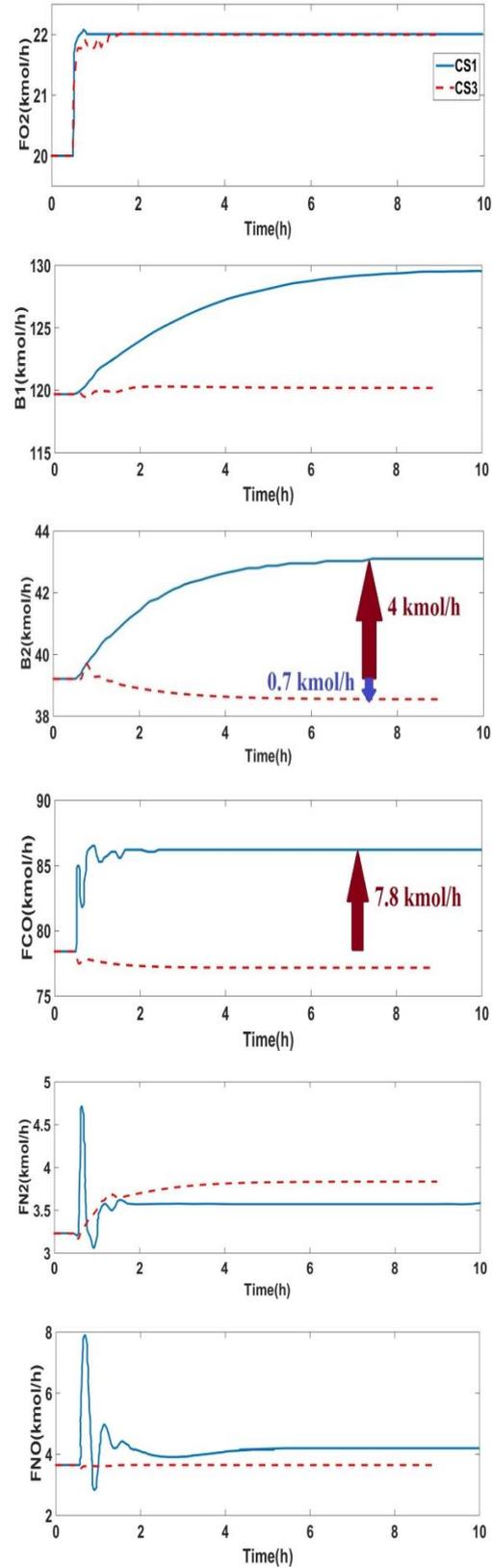
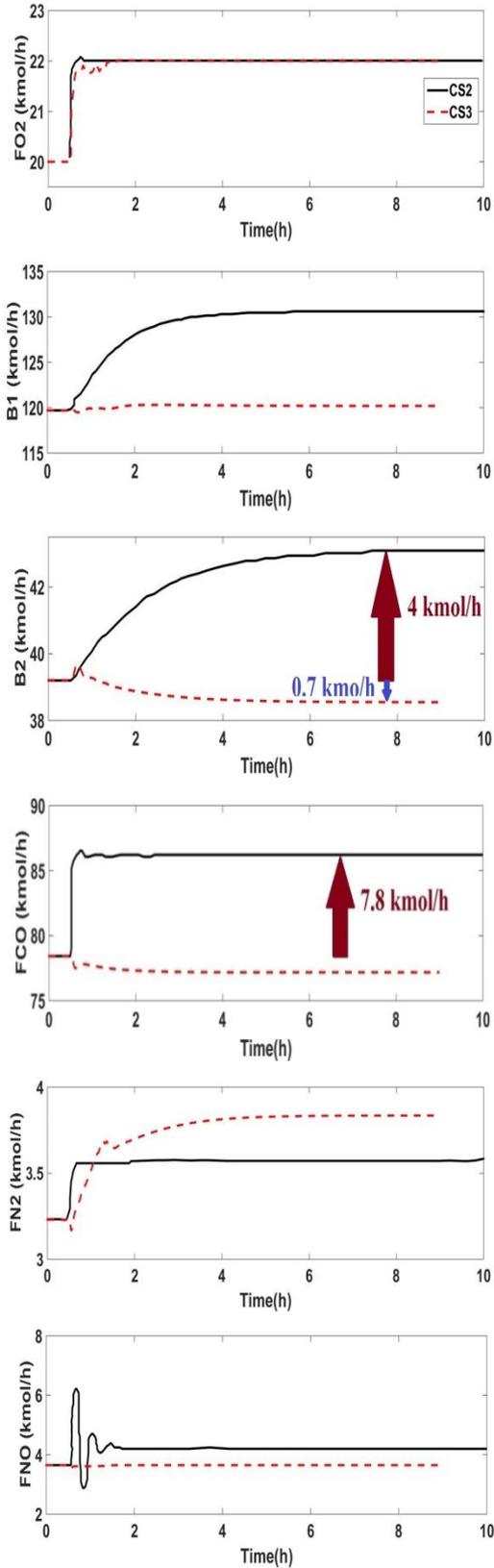
Figure 8. Plantwide control structure CS2 [9].

In the first phase, we follow the methodology of Luyben [9] and enhance TPM by ten percent. This allows us to evaluate how each structure reacts to the disturbance and how the controlling process is carried out. Regrettably, none of the suggested large-scale control structures could carry out the process of regenerating 10 % oxygen. Because there is less oxygen available, the concentration of this substance at the reactor's entrance has dropped, and as a result, the process has come to a halt.

This appears to be the cause of the inability of reactor R1 to continue operating normally. When it was thought that the reactions were irreversible, the Aspen Dynamics software integration proved incapable of controlling the concentration of reactants near zero. The outcomes of introducing this disturbance into the processes will be demonstrated in the following sections. On the left side of Figure 9 there is a comparison of the structure of CS3 with the structure that was represented by

Luyben (CS2), and on the right side there is a comparison of the structure of CS3 with the structure that was represented by Zhu et al. [8]. (CS1). The performance diagram for CS3 is

represented by a dashed line in all of the figures, whereas the performance of the other structures is represented by a solid line.



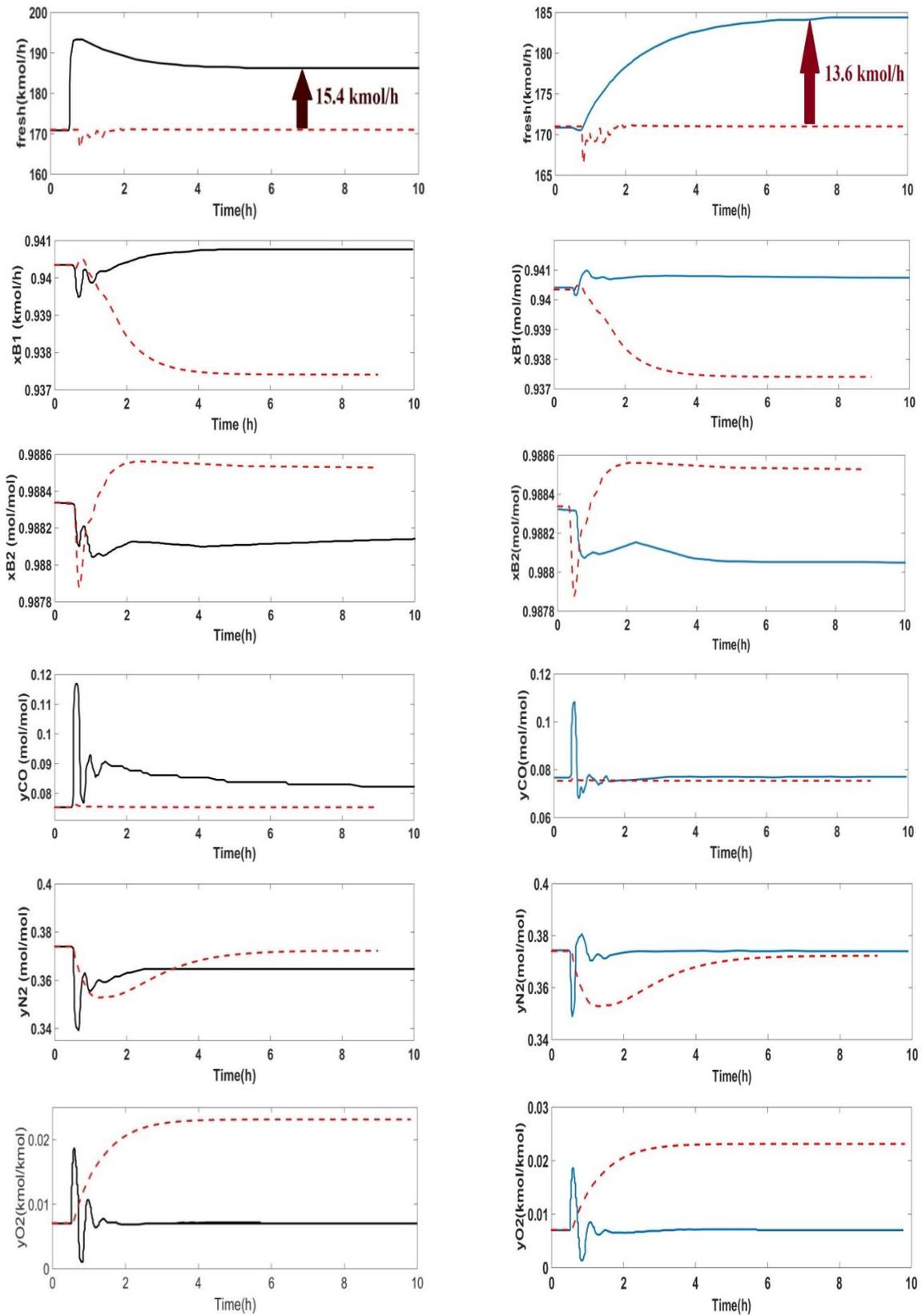


Figure 9. 10 % step increase in TPM.

Furthermore, to facilitate a comparison with the structure of Luyben, the feed of a mixture

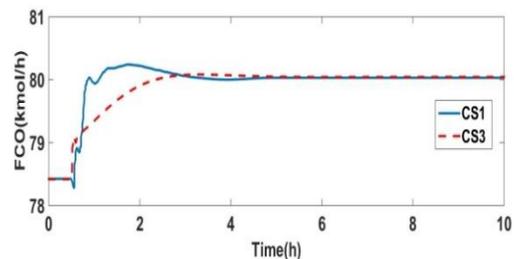
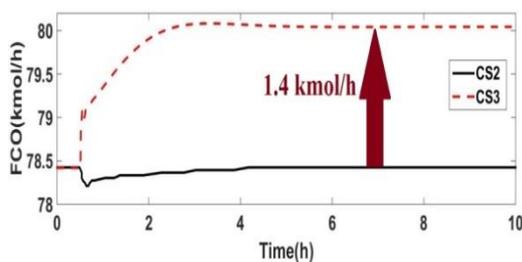
of ethanol and water is labeled fresh (fresh), and the lower effluent of column C1 (water) is

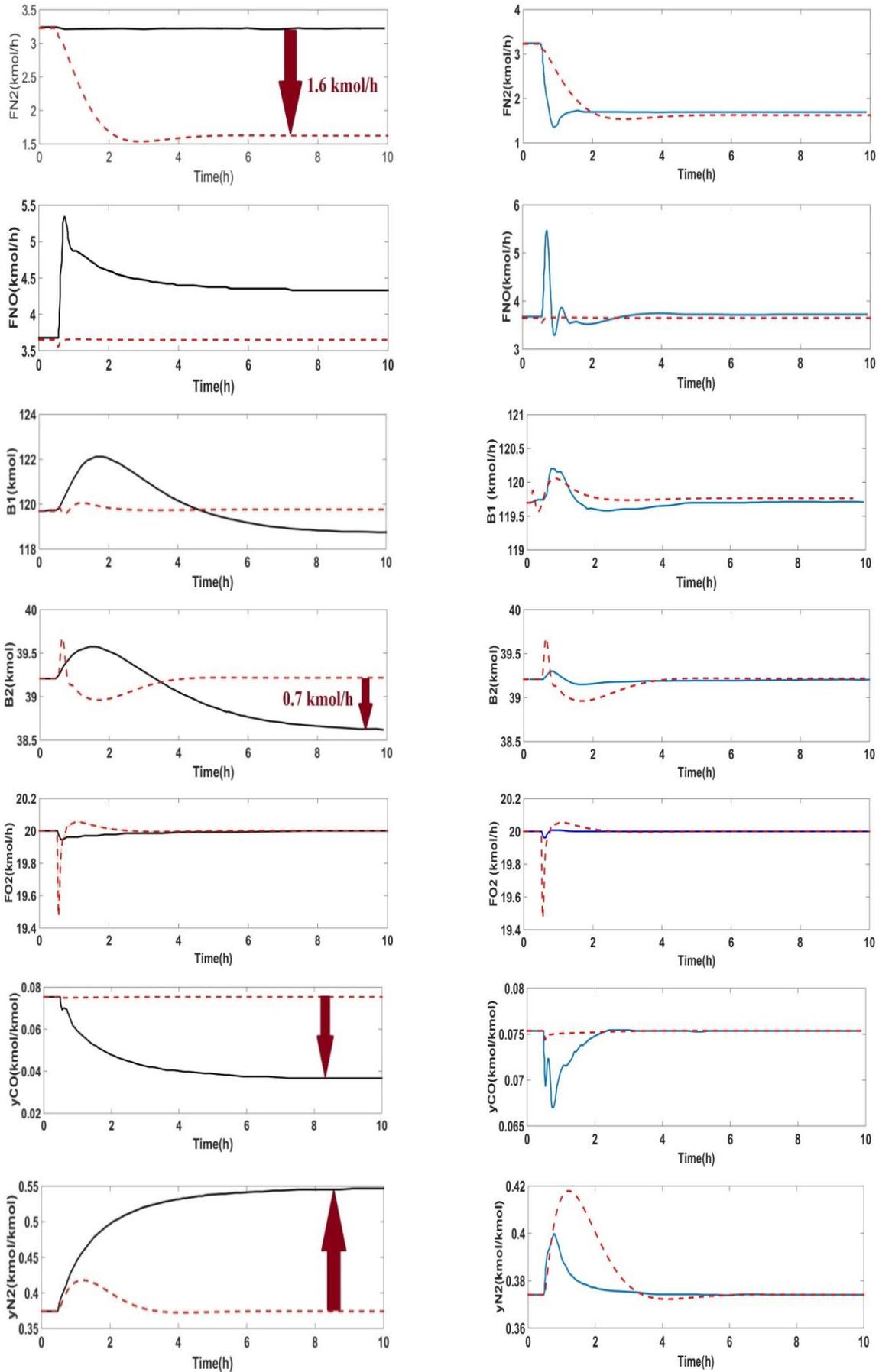
designated B2 in these diagrams. In addition, B2 displays the lower effluent of column C2, i.e., the ultimate product (diethyl oxalate). The concentrations of water and diethyl oxalate in streams B1 and B2 are denoted by x_{B1} and x_{B2} respectively. As depicted in Figure 9, to remove fluctuations triggered by disruptions, maintaining a constant ratio of the oxygen feed to the carbon monoxide feed in the CS1 structure, as well as a constant ratio of carbon monoxide and ETOH to the overall flow rate in the CS2 structure, led to a considerable increase in the carbon monoxide effluent in the fresh CO feed and the ethanol effluent in ETOH. This results in a considerable departure of the ultimate product stream, B2, from the set point. As a result, the end product stream (B2) increases by 4 kmol/h relative to the set points of the prior two structures. This is a result of the rise in the intake with the maximum feed in these structures, which includes an enhancement of the CO feed by 8.7 kmol/h in both preceding structures and an elevation of the ETOH feed discharged in the C1 and C2 structures by 13.6 kmol/h and 15.4 kmol/h respectively. Changes in the main feed discharge are minor in the CS3 structure owing to the instability of the feed ratio and proper management of the carbon monoxide content, and we do not witness numerous alterations in the ultimate product stream (B2). The product modification in the CS3 structure manifested as a drop of a B2 flow rate by 0.7 kmol/h compared to the set point. This is attributed to the decrease of the CO input by 0.6 kmol/h (to

manage the concentration of CO), while the utilization of the primary feed has not changed much. The generation of additional product (4 kmol/h) can be deduced from the fact that the stream of B2 has increased in the CS1 and CS2 structures. Owing to the deviation from the set point, which is modified according to the market requirements and is more than the market consumption, this additional product is undesirable. Due to the greatest increase of the amount of the feed (an increase of the CO feed by 7.8 kmol/h and the ETOH feed by 15.4 and 13.6 kmol/h) in these two structures, the expense of raw materials has enhanced, and notwithstanding the production of an additional product, it may not be marketed. During the commencement of perturbations, any deviation (reduction or increase) of the product from its set point is seen as waste and is undesired.

2.8. Reduced CO purity

The new carbon monoxide feed is designed to have 9.99 % mol/mol of carbon monoxide and 0.1 mol/mol of hydrogen. We generate a fresh disturbance in the process by introducing 2 % mol/mol of nitrogen impurities to this stream in order to assess the effectiveness of the three control structures. The reaction of the three control structures is compared in Figure 10. Based on Figure 10 the concentration adjustment in both CS1 and CS3 structures has resulted in a significant enhancement of the CO feed stream.





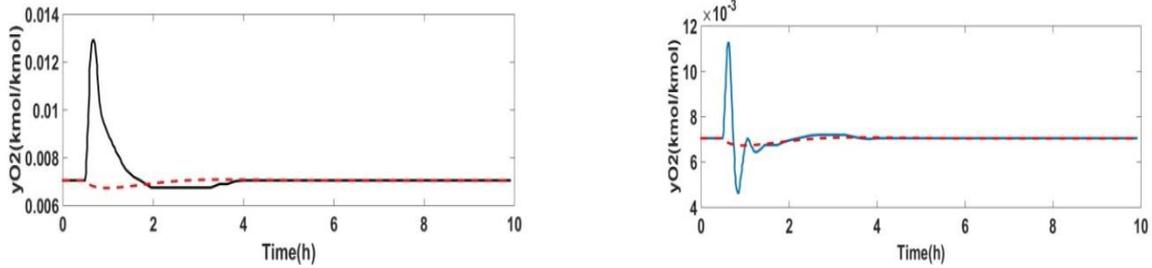


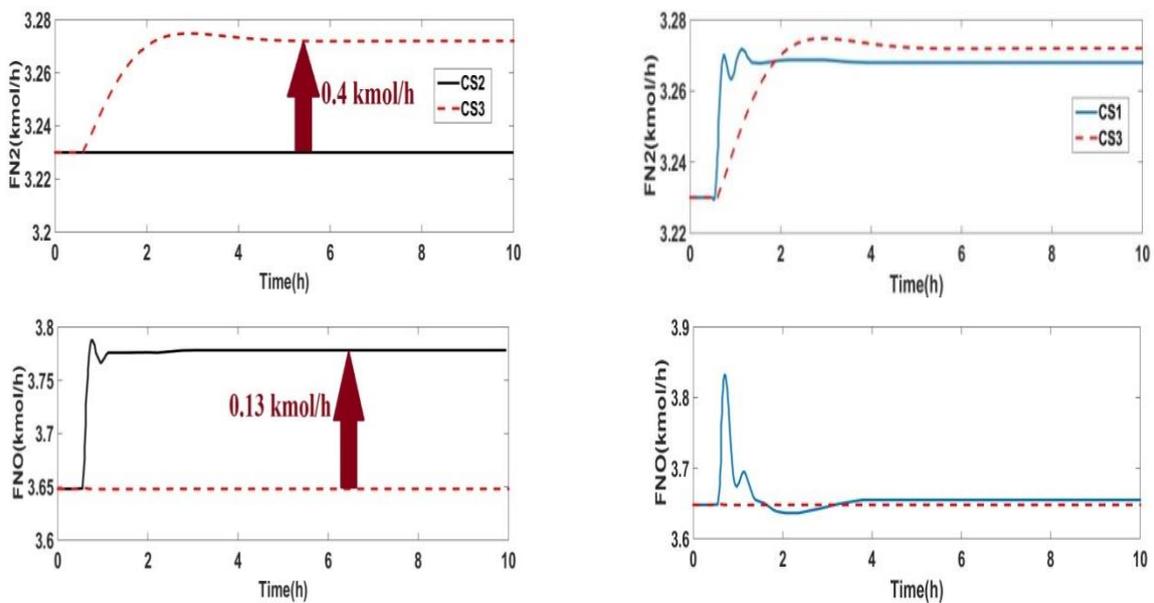
Figure 10. Reduced CO purity.

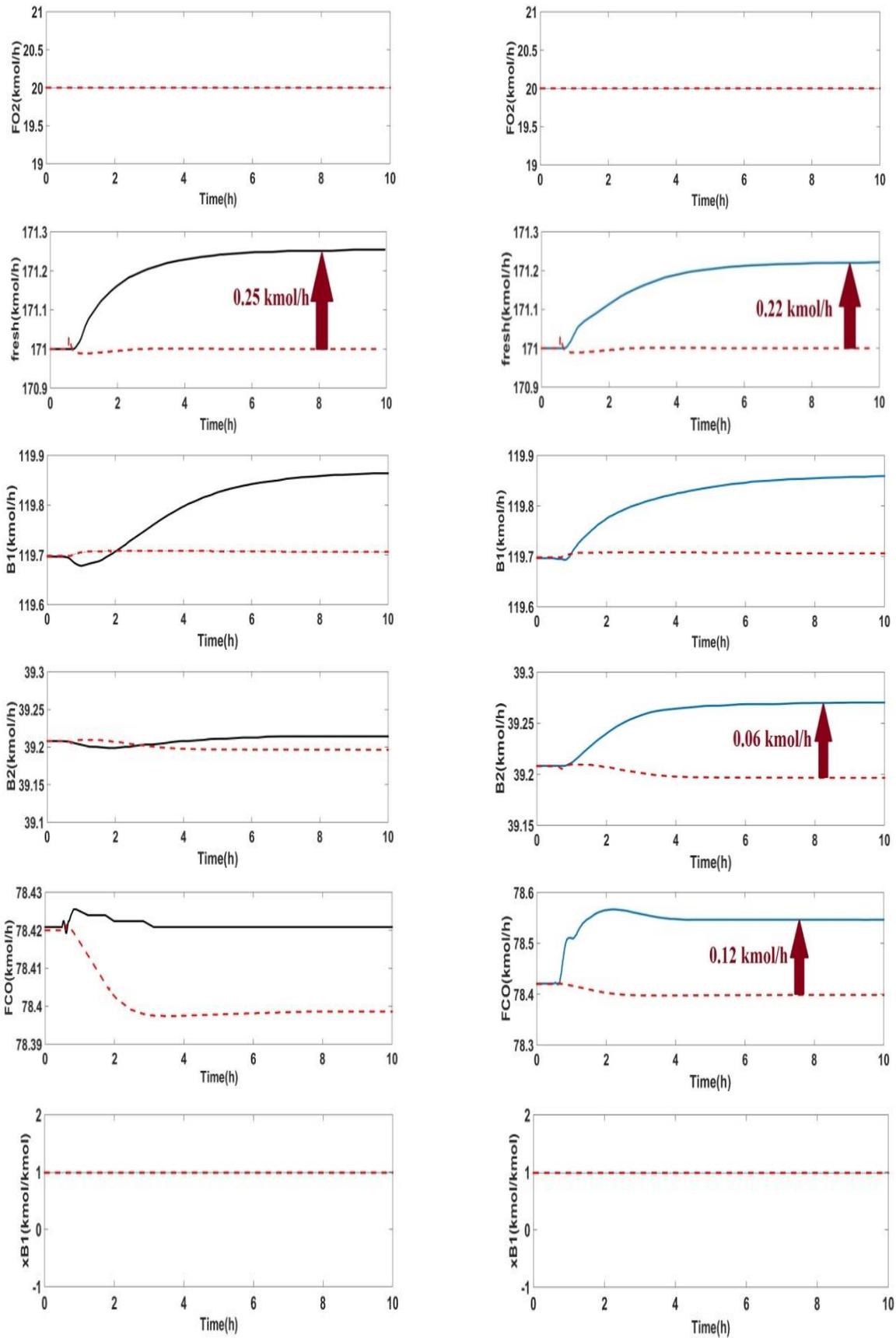
Since the addition of impurities lowers the concentration of carbon monoxide in the process, they raise the structure of the CO input stream by 1.4 kmol/h to avoid this impurity from affecting the reactions. Nevertheless, because the concentration of this compound cannot be controlled in the CS2 structure, the feed stream is not altered. On the other hand, the primary effluent stream (B2) is reduced to 0.7 kmol/h because the purity of CO in the feed decreases, and there is insufficient control in this structure. In this instance, the compound is nearly stable. Nevertheless, the B2 stream in both the CS1 and CS3 structures returns to the setpoint value following a minor adjustment and reaches a stable level. The nitrogen feed is lowered in both the CS1 and CS3 structures, which is necessary to cope

with the excessive nitrogen supplied from the carbon monoxide feed. The concentration of nitrogen in these two structures is controlled effectively. However, we notice an enhancement in the concentration of nitrogen without any oversight in the structure of CS2, which can be attributed to another structural flaw identified by Luyben [9]. In accordance with what is mentioned, if the concentration of nitrogen in this process is not monitored, it will result in difficulties with plant-level security.

2.9. Reduced N₂ purity

Reducing the purity of nitrogen in the feed and replacing the feed of the pure nitrogen with 99 % mol/mol of nitrogen and 1 % mol/mol of oxygen perturb the evaluated end compound.





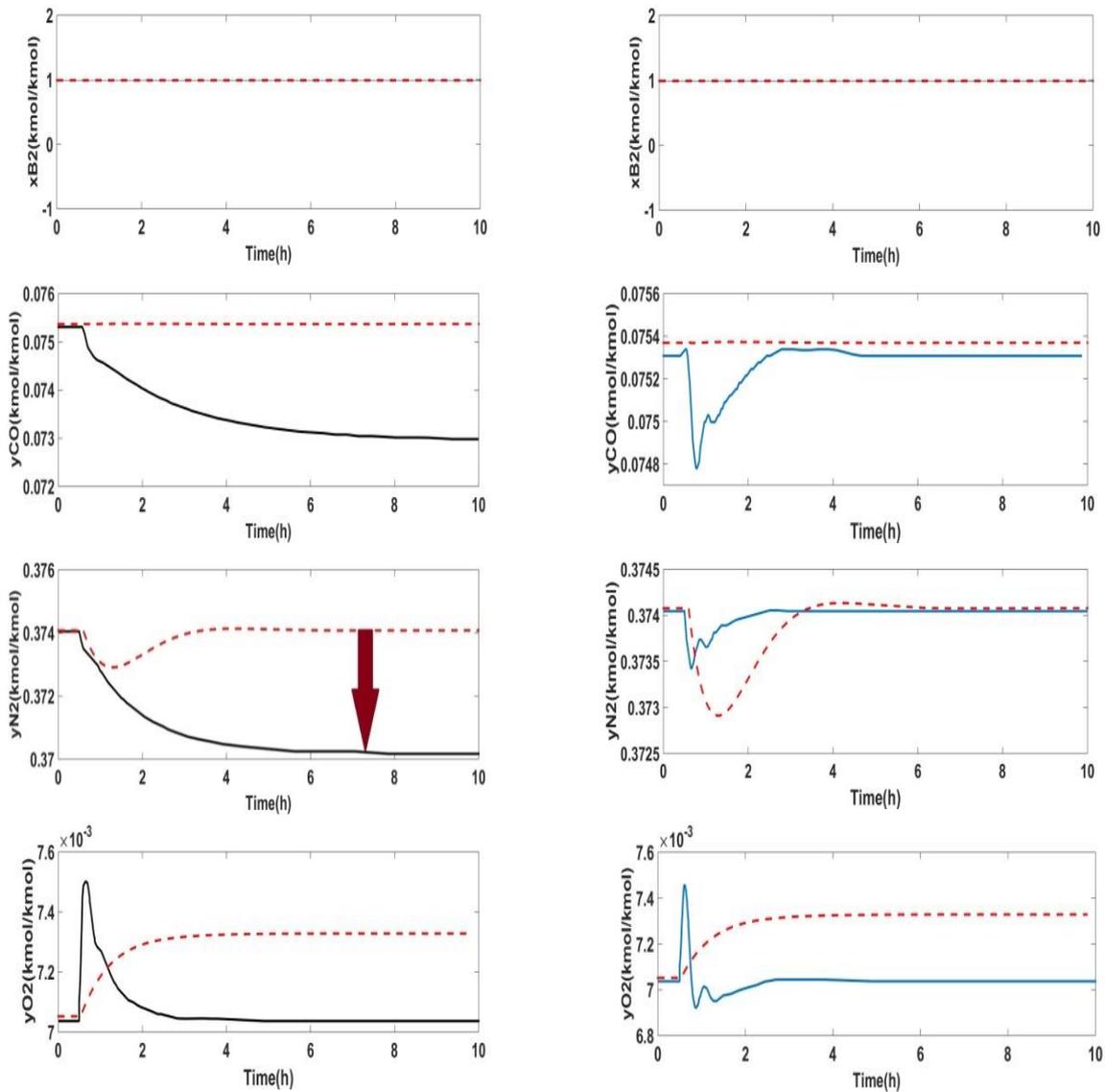


Figure 11. Reduced N₂ purity.

From Figure 11, it is evident that increasing the concentration of this compound in CS1 and CS3 structures causes the increase in the stream rate of the nitrogen feed. This is done to avoid any challenges. In contrast to these two structures, the CS2 structure is incapable of adequately controlling the nitrogen content and then endangers the process by introducing additional contaminants. While there was no variation in the product concentration in any of the three structures and a very little alteration in the stream of B2 in structures CS1 and CS3, the stream of the ultimate product (B2) enhanced in structure CS2. The concentration

of CO fluctuates as a result of the constant ratio of the feed streams to Total, and the operation of the R2 reactor is interrupted. Under these conditions, a comparable scenario occurred in the structure of CS1, leading to an increase in the product stream as a result of a rise of 0.25 kmol/h in the ETOHI stream after the cascading control structure in this structure (control loop in column C1). Even though this structure produces a negligible amount of the excess product, CS1 increases the quantity of the feed to a maximum during this operation, resulting in additional expenses. Nevertheless, in the CS3 structure, which is primarily related

to regulating the concentration of CO, there is no divergence in the effluent stream. There is only a little fluctuation in the concentration of oxygen, which does not have a significant impact on the process, is effectively regulated. Even though the oxygen content is well-controlled in the other two structures, it has influenced the NO feed rate and disrupted the input stream rate of the compound.

3. Conclusions

A new plantwide control structure for the coupling-regeneration process to synthesize diethyl oxalate is provided in this study. It was demonstrated that this particular structure does not require a high level of intricacy to get rid of the fluctuations brought on by the disruptions of the control loops. Furthermore, it was demonstrated that the efficient control over the introduction of potential disturbances could be obtained by merely modifying the process stream plate. Comparing this structure to those previously published by Zhou et al. [8] and Luyben [9], it was discovered that it does not appear required to employ four plantwide concentration controllers in the structure described by Zhu et al. [8], as the proportion of substances in this process is substantially sustained by an effluent-controlled structure. Nevertheless, this does not indicate that there is no requirement to regulate the concentration in the process; rather, it means that the variations produced by disturbances can be removed by altering the recycle stream and utilizing two concentration regulators. Despite the fact that managing the concentration of nitrogen does not have a considerable part in the chemical reactions of the process and that changing it does influence the end product, this step is important because of safety concerns. The possibility of a catastrophe should be prevented. In terms of safety, the structure

proposed by Luyben [9] is unreliable, and the concentration of nitrogen in this procedure is improperly regulated. Luyben was able to propose a less expensive construction than introduced by Zhou et al. [8] by using only one concentration controller. In this structure, nevertheless, the strategy of "keeping the overall stream constant and adjusting the feed" resulted in a substantial shift in the stream of the new feed in response to disruptions. This has a significant effect on the primary effluent, diethyl oxalate, in addition to enhancing the expense of basic materials. Using complicated cascade structures and maintaining a constant input stream rate to regulate variations and decrease disruptions in the two preceding structures are not required and increase the level of the variation of the basic material input. Although the process remains completely operational in all three structures, various configurations diverge from their design parameters in the two structures illustrated before in the situation of disruptions. Under these conditions, products and wastes undergo considerable modifications. Using the innovative structure provided in this work, however, the primary objective of improving process management through a simpler structure than earlier structures has been fulfilled. Considerable renovations were also made to change the input and effluent streams. Nevertheless, compared to those of the prior structures, the types of implemented controllers were not altered; specifically, their types were P and PI, and the process stream diagram underwent a minimal transformation.

Acknowledgement

Funding: This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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