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Control Structure Design of Novel Microwave-Catalyzed Process for Simultaneous Production of Ammonia and Ethylene

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ABSTRACT

This work demonstrates the application of a pulsed microwave system for single-step co-production of ethylene and ammonia from methane. To mitigate inherent production fluctuations from pulsed microwave reactors, a staggered manifold configuration was utilized to stabilize effluent flow for industrial-scale compatibility. Dynamic validation of the ammonia and ethylene purification columns confirmed that a rigorously tuned control strategy effectively rejects ±10% feed disturbances while maintaining process stability and product purity. Ultimately, this systematic approach establishes a robust foundation for the sustainable, electrified production of foundational chemicals by bridging the gap between laboratory-scale pulsing phenomena and industrial-scale operational reliability.

Keywords: Process Control, Ethylene, Aspen Dynamics

INTRODUCTION

Ethylene (C_2H_4) is a simple, flammable, colorless, and highly reactive hydrocarbon containing a carbon–carbon double bond, which makes it the smallest and most important alkene. As one of the most widely produced organic chemicals, it is a foundational building block of the petrochemical industry and a crucial feedstock for manufacturing polyethylene plastics, ethanol, ethylene oxide, ethylene glycol, and vinyl chloride [1]. Ammonia (NH_3) is a colorless gas with a pungent odor, composed of one nitrogen atom bonded to three hydrogen atoms, with a boiling point of approximately −33°C at ambient pressure. It is highly soluble in water, forming ammonium hydroxide (NH_4OH), a weak base, and is a critically important industrial chemical with diverse applications across agriculture, energy, and industrial sectors [2]. Globally, more than 80% of ammonia production is used for nitrogen-based fertilizers such as urea, ammonium nitrate, and ammonium sulfate, which are essential for sustainable agricultural productivity, while additional applications include refrigeration systems, cleaning agents, and emerging roles as a hydrogen carrier for carbon-free energy storage, transportation, and fuel use [2]. Ethylene production currently tops 200 million metric tons annually, fueling a market exceeding $180 billion. This growth is sustained by a 4.1% rise in demand and a 6.2% increase in production capacity, largely propelled by the global requirement for plastics and consumer products [3]. In a similar vein, global ammonia output has surpassed 180 million metric tons per year, anchoring a fertilizer market valued at more than $100 billion while simultaneously representing nearly 2% of total global energy consumption [4]. The production of ethylene and ammonia represents the backbone of the modern chemical industry, though traditional methods remain significantly energy-intensive and carbon-heavy due to their reliance on high-temperature, endothermic reactions.

Microwave (MW) reactor technology represents a sophisticated process-intensification strategy that aligns with the global shift toward chemical process electrification as a replacement for traditional thermal heating [5]. By leveraging microwave catalysis, it is possible to simultaneously activate the highly stable CH_4 and N_2 molecules, enabling the direct co-production of ammonia and ethylene within a single reactor step [6]. This technology not only enhances energy efficiency and reduces CO_2 emissions by selectively heating the catalyst surface but also introduces operational flexibility through pulsed microwave energy. By deploying microwave reactor technology, it is possible to operate the reactor at two different temperature modes by pulsing the microwave power in periodic intervals. Under microwave power-on mode (heating), an endothermic reaction to produce ethylene can take place whereas under the microwave power-off mode (quenching), an exothermic reaction to produce ammonia can occur.

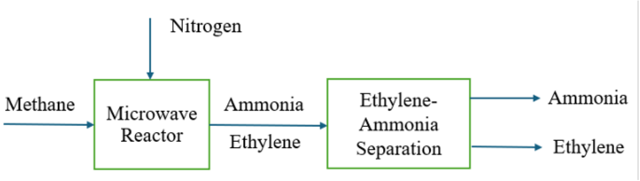


Figure 1. Block flow diagram of the simultaneous production of ethylene and ammonia.

The reactor outlet streams are combined and sent to the distillation column train for separation. During plant operation, due to switching between power on and power off modes in the two microwave reactors, the reactor outlets will not be steady. This fluctuation in the flowrate to the distillation column train leads to an inherently dynamic operation and hence it is necessary to design a control system for the separation section that keeps the quality of the products ethylene and ammonia at the desired levels. Based on the concept in Figure 1, a plant-wide steady-state model was constructed and subsequently converted into a dynamic model using Aspen Plus Dynamics. This model integrates a base control strategy consisting of tuned PID loops and parameters to provide a robust foundation for industrial-scale process simulations.

It was recently shown by our group that producing ethylene via novel microwave (MW) technology is economically competitive, with a levelized cost of USD 0.51/kg compared to USD 0.56/kg for conventional methods [7]. While a formal technoeconomic analysis (TEA) for the simultaneous production of ethylene and ammonia is not part of this study, this integrated process has the potential to achieve higher net present value (NPV) than standalone ethylene production since the hydrogen required for ammonia production is produced in the ethylene reaction cycle.

MICROWAVE REACTOR DESIGN ISSUES

Conventional processes to produce ethylene mostly operate at equilibrium conditions with a continuous heating process to supply the heat requirement for the reaction conditions. But a pulsed microwave heating process is a non-equilibrium process that has the potential to switch between exothermic and endothermic reactions together for simultaneous production of two commodity chemicals. In the pulsing process, the power-on mode (heating) favors endothermic reactions, and the power-off mode (quenching) favors exothermic reactions. The main reactions involved are as follows:

$$2CH_4 \rightarrow C_2H_4 + 2H_2 \text{ (endothermic, 500-700°C)}$$

$$3H_2 + N_2 \rightarrow 2NH_3 \text{ (exothermic, 250-400°C)}$$

As shown experimentally in Rahman et al. [7], small quantities of additional compounds such as acetylene, ethane, carbon dioxide, benzene, and toluene are also produced via side reactions, which need to separated from the main products via a train of distillation columns. The methane coupling reaction produces ethylene and hydrogen comparatively at higher temperature range (500-700°C) in power-on mode. The produced hydrogen reacts with nitrogen at lower temperature (250-400°C) to produce ammonia in the power-off mode. The transition from laboratory-scale pulsed microwave (MW) reactors to industrial-scale production introduces significant complexities in reactor design and process control, primarily due to the inherent fluctuations in product flow that alternately produce ethylene and ammonia. In a continuous industrial facility, equipment such as compressors, separation columns, and heat exchangers is designed for steady-state flows. Rapid fluctuations in mass flow can lead to severe pressure surges, thermal cycling fatigue, and significant challenges in maintaining product purity within distillation sumps. To rectify these fluctuations and stabilize the production rate, a process intensification strategy involving a series of MW reactors operating in a staggered, out-of-phase configuration must be implemented. By coordinating the pulsing intervals of multiple reactors, the peak production of one unit can compensate for the quenching phase of another, effectively dampening the oscillation and delivering a more consistent feed to the downstream processes. Despite this staggered operation, the outlet of the microwave reactor train can fluctuate up to ±10% and it is necessary to develop a control system for the separation train to work effectively despite this fluctuation.

METHODOLOGICAL FRAMEWORK

A systematic approach was adopted integrating steady-state modeling, dynamic simulation, and advanced process control. Initially, steady-state benchmarks for the intensified microwave-assisted routes were developed in Aspen Plus v14.0 using the NRTL property model [7]. This thermodynamic selection is critical to accurately account for the highly non-ideal liquid-phase activity coefficients of ammonia within the complex hydrocarbon and light-gas matrix. The outlet of the microwave reactor train has the composition of methane, ethylene, hydrogen, nitrogen, ammonia, acetylene, ethane, carbon dioxide, benzene, and toluene that were determined experimentally and fluctuates with microwave pulsing [5, 6, 7]. Then, a dynamic model in Aspen Plus Dynamics (APDs) v14, was constructed for the separation train based on the steady-state process model [8]. During this stage, all equipment in the process were appropriately sized. Building upon the steady-state architecture, a high-fidelity dynamic model was developed in Aspen Plus Dynamics (APD) v14.0, where all unit operations were sized based on established literature [7, 8]. This phase included the implementation of a foundational control layer and the rigorous fine-tuning of control parameters to achieve optimal response characteristics across diverse operational transients. To validate the model's reliability, it was subjected to extensive disturbance testing, specifically focusing on variations in feed flow rates and compositions [8]. The effectiveness of this control strategy was verified through robustness evaluations under dynamic scenarios, demonstrating a consistent ability to preserve optimal operating conditions.

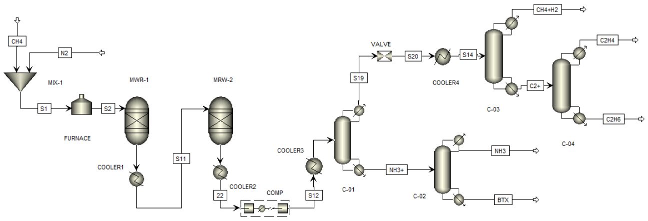


Figure 2. Process Flow Diagram of Microwave Assisted Process to Convert Methane to Ethylene and Ammonia.

Figure 2 illustrates the process flow diagram (PFD) that produces ethylene and ammonia simultaneously. In this industrial-scale simulation, a feed stream of methane and nitrogen is initially processed in a yield reactor at 600°C to facilitate co-production. To prevent ammonia decomposition, the effluent is immediately quenched to 280°C before entering a secondary hydrogenation reactor, which operates at the same temperature to convert byproduct acetylene into ethylene, thereby enhancing overall yield. The resulting stream is pressurized to 15 bar via a multistage compressor and fed into the first distillation column (C_01), operating at -15°C and 15 bar. The ammonia-rich bottom stream from C_01 is directed to a purification column (C_02) operating at 39°C and 15 bar to recover high-purity ammonia. On the other hand, the ethylene and methane-rich overhead stream is processed through two subsequent distillation columns (C_03 and C_04) for ethylene purification, following the same separation technique by Rahman et al [7]. A buffer tank to smooth out the reactor outlet fluctuations created by pulsing was not considered primarily because gas-phase streams are highly compressible and occupy large volumes, making physical storage for stabilization both economically and spatially impractical. To evaluate the direct conversion efficiency of the microwave-assisted process, no recycle streams were utilized in this study. Under these conditions, a methane feed of 2500 kg/h successfully yielded 2122 kg/h of ethylene and 955 kg/h of ammonia, both achieving a 99.9% purity level. The rigorous design specifications for the entire four-column separation train are summarized in Table 1.

Table 1: Feed and Column Design Specifications.

	C_01	C_02	C_03	C_04
Purpose	Distillation	Distillation	Distillation	Distillation
No. of stages	18	24	17	51
Feed stage	12	15	10	20
Pressure (bar)	15	15	8	8
Reflux ratio	0.317	0.358	0.220	2.690
Condenser duty (MW)	-1.850	-0.416	-0.670	-0.650
Condenser temperature (ºC)	-97.77	38.53	-131.62	-58.40
Distillate rate (kmol/h)	1610.71	55.98	1511.16	75.56
Reboiler duty (MW)	0.201	0.418	0.490	0.900
Reboiler temperature (ºC)	39.13	177.34	-48.45	-29.38
Bottom rate (kmol/h)	57.30	1.31	99.53	23.93
Tray spacing (m)	0.61	0.61	0.61	0.61
Tray type	Sieve	Sieve	Sieve	Sieve
Column height (m)	9.75	13.41	9.14	23.77
Diameter (m)	1.28	0.28	1.33	0.68

A dynamic model was developed to account for the fluctuations in the reactor outlet stream. In this simulation, the physical dimensions of the unit operations were established using industry-scale design principles. Distillation column heights were calculated through established heuristics and relevant correlations, while diameters were determined based on estimated flooding velocities to ensure operational stability [8]. Following the completion of equipment sizing, the validated steady-state simulation model was transitioned into a dynamic framework using Aspen Plus Dynamics V14 [8]. To ensure operational stability and robust disturbance rejection, a plant-wide control structure was implemented. This strategy integrates conventional regulatory loops with advanced regulatory schemes, where all controller parameters were optimized using the Tyreus–Luyben auto-tune variation (ATV) method adjusting the controller's gain (K_C) integral time constant (τ_I) and derivative time constant (τ_D) [9]. As illustrated in Figure 3, each distillation column implements a standard control configuration, including top pressure control, reflux drum level control, and sump level control. For column C_01, condenser temperature is regulated through utility flow adjustment, the reflux drum level is controlled via reflux flow manipulation, and the bottom-stage temperature is maintained by varying the reboiler duty. Additionally, a sump level is controlled by applying a sump level controller at the bottom. An identical control philosophy is implemented for the remaining columns (C_02, C_03, and C_04).

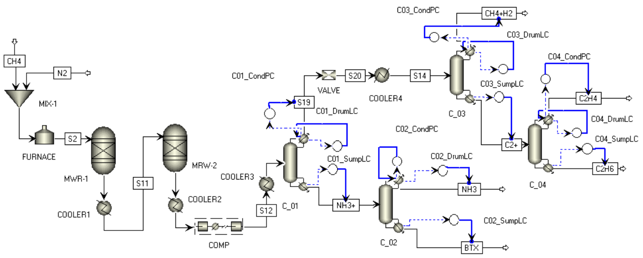


Figure 3. Plant wide control strategy for simultaneous ammonia and ethylene production.

RESULTS AND DISCUSSION

This study evaluates the dynamic performance and controller structure of the ammonia (C_02) and ethylene (C_04) purification columns to ensure process stability during simultaneous production. Given the fluctuating nature of the upstream pulsed microwave reactor effluent, achieving a robust control response in the separation train is critical for maintaining product purity and operational safety.

The initial controller response of column C_02 & C_04 exhibited significant oscillations and sluggish stabilization in both the reflux drum (C_02DrumLC) and the column sump (C_02SumpLC) levels. As shown in Figure 4, the mass flow rates and temperatures showed substantial deviations before reaching a settled state. To mitigate process instability, a rigorous tuning phase was conducted. The controller outputs were optimized to aggressively dampen oscillations, primarily utilizing Proportional-Integral (PI) control structures. The controller gains K_C and τ_I were evaluated using the Tyreus–Luyben ATV method. Table 2 summarizes the optimized parameters for the reflux drum and sump level controllers for both purification columns.

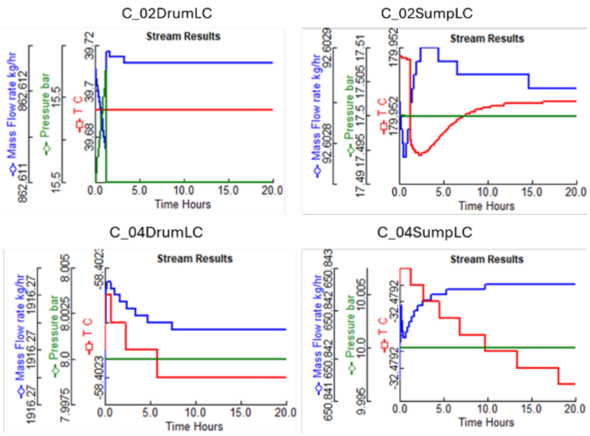


Figure 4. Controller response of ammonia and ethylene purification column (C_02 & C_04) before tuning.

Table 2. Optimized Controller's Gain and Integral Time Constant

Column	Controller Location	Gain (K_C)	Integral Time (τ_I)
C_02 (Ammonia)	Reflux Drum	32.55	10.56
	Sump	24.73	3.96
C_04 (Ethylene)	Reflux Drum	46.35	3.96
	Sump	25.67	2.64

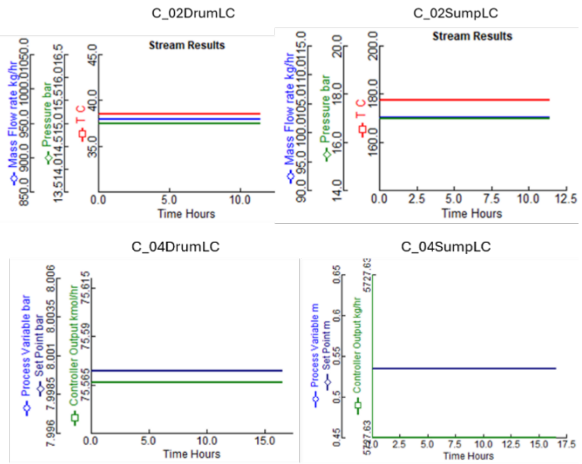


Figure 5. Controller response of ammonia and ethylene purification column (C_02 & C_04) after tuning.

Figure 5 shows that both the drum and sump levels reached their respective setpoints with minimal overshoot and zero steady-state error. This stable baseline is essential for the intensified process, as it ensures that the energy-intensive separation of ammonia and ethylene from the hydrocarbon matrix remains consistent despite the inherent transients of the microwave-assisted route

Controller Performance Analysis

To validate the reliability of the established control strategy, the column was subjected to step changes of ±10% in the feed flow rate. As illustrated in Figure 6, the C_02 control system effectively rejected these disturbances with minimal deviation. Upon a +10% increase in feed, the condenser and reboiler duties adjusted dynamically to stabilize inventory levels, reaching a new steady state shortly after the onset of the disturbance. This transition resulted in a proportional impact on the top and bottom product flow rates, directly reflecting the change in feed throughput. Similarly, the -10% feed variation demonstrated a smooth transition in both the top and bottom product flow rates. This robustness confirms that the integrated control layer can effectively manage process fluctuations while maintaining product quality and system stability.

To verify the reliability of the control strategy for the C_04 column, the system was tested with ±10% step changes in the feed flow rate. As shown in Figure 7, the control loops successfully rejected these disturbances, keeping all process variables stable. When the feed increased by 10%, the reboiler and condenser duties adjusted automatically to maintain the liquid inventory in the sump and reflux drum. This allowed the system to reach a new steady state shortly after the change, with the top and bottom product flow rates increasing to match the higher throughput. A similarly smooth transition occurred during the -10% feed reduction, where the system adjusted to a lower-load operating point without any instability. These results confirm that the integrated control layer effectively preserves column stability and product purity, even under the fluctuating conditions typical of electrified processes.

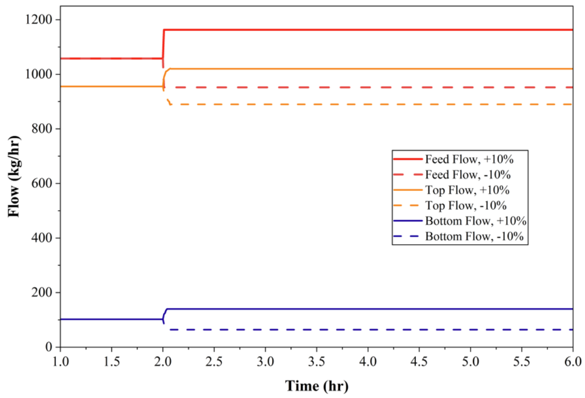


Figure 6. Controller response of the ammonia purification column (C_02) to a ±10% step change in feed flow rate

Controller Sensitivity Matrix for Ammonia Purification Column

Table 3 and corresponding Figure 8 illustrate a parametric sensitivity analysis of the ammonia (C_02) purification column, where Case-1 (Optimized TL-ATV) serves as the performance benchmark. In the dynamic simulation graphs, Case-1 is presented as the most efficient response, that returns the reflux and sump levels to their setpoints with the shortest settling time and minimal overshoot. In contrast, the low-gain scenarios (Case-2 and Case-5) appear on the graph as sluggish, long-duration curves; because a K_C of 5 lacks the proportional response to move control valves decisively, these cases take significantly more time to stabilize and allow for large deviations from the setpoint. On the other hand, the high-gain scenarios (Case-3 and Case-4) demonstrate better response compared to case-2 and case-5.

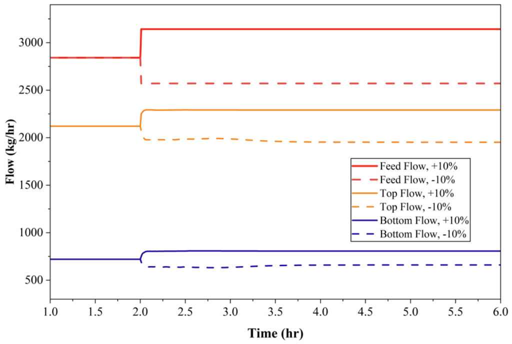
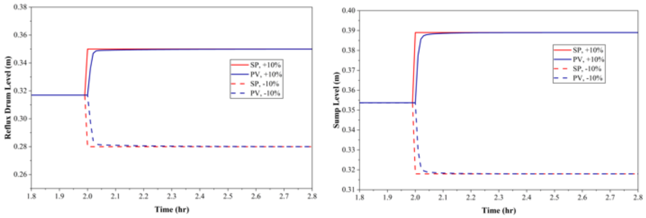


Figure 7. Controller response of the ethylene purification column (C_02) to a ±10% step change in feed flow rate

Table 3. Controller Sensitivity Matrix for Ammonia Column (C_02)

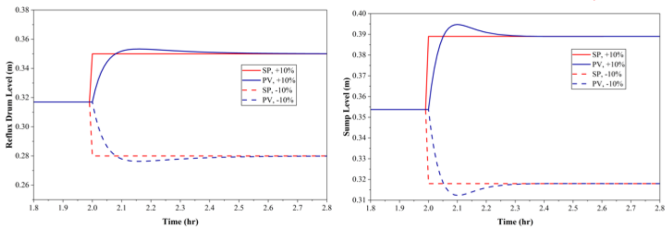
test Case	**reflux drum Level (K_C, τ_I)**	**sump level (K_C, τ_I)**
case-1: optimized (TL-ATV)	$K_C = 32.55$, $\tau_I = 10.56$	$K_C = 24.73$, $\tau_I = 3.96$
case-2: low gain / fast reset	$K_C = 5$, $\tau_I = 10.56$	$K_C = 5$, $\tau_I = 3.96$
case-3: high gain / fast reset	$K_C = 60$, $\tau_I = 10.56$	$K_C = 60$, $\tau_I = 3.96$
case-4: high gain / Slow Reset	$K_C = 60$, $\tau_I = 60$	$K_C = 60$, $\tau_I = 60$
case-5: low gain / slow reset	$K_C = 5$, $\tau_I = 60$	$K_C = 5$, $\tau_I = 60$

case-1



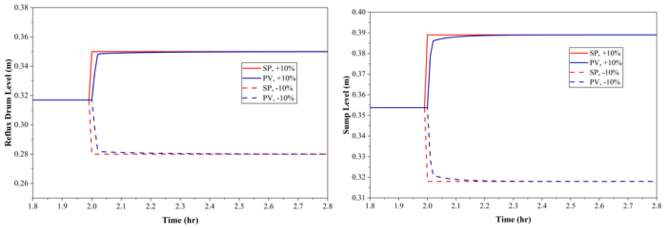
(a) Reflux drum level controller: K_c=32.55, τ_I=10.56 (b) Sump level controller: K_c=24.73, τ_I=3.96

case-2



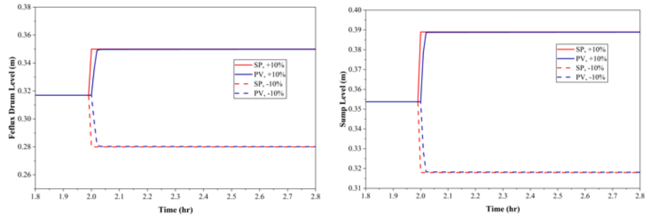
(a) Reflux drum level controller: K_c=5, τ_I=10.56 (b) Sump level controller: K_c=5, τ_I=3.96

case-3



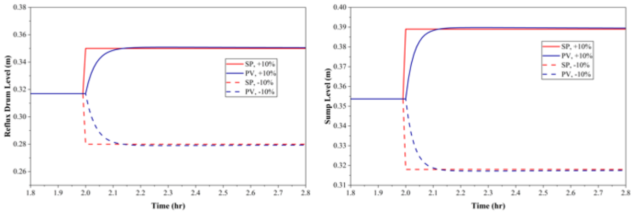
(a) Reflux drum level controller: K_c=60, τ_I=10.56 (b) Sump level controller: K_c=60, τ_I=3.96

case-4



(a) Reflux drum level controller: K_c=60, τ_I=60 (b) Sump level controller: K_c=60, τ_I=60

case-5



(a) Reflux drum level controller: K_c=5, τ_I=60 (b) Sump level controller: K_c=5, τ_I=60

Figure 8. Controller response of ammonia column (C_02)

Controller Sensitivity Matrix for Ethylene Purification Column

Table 4 and the corresponding Figure 9 illustrate the parametric sensitivity analysis for the ethylene purification column (C_04), where Case-1 (Optimized TL-ATV) consistently serves as the most efficient performance benchmark.

In the dynamic simulation graphs, Case-1 demonstrates the superior control response, characterized by high proportional gain and tight integral times (τ_I = 3.96 and 2.64) that restore the reflux and sump levels to their setpoints with the shortest settling time and minimal overshoot. In contrast, the low-gain scenarios (Case-2 and Case-5) appear as sluggish, long-duration curves; because a K_C of 5 lacks the proportional magnitude to drive the control valves effectively, these cases result in significantly prolonged stabilization periods and allow for large deviations from the setpoint. While the high-gain scenarios (Case-3 and Case-4) demonstrate a more robust initial response compared to the low-gain cases.

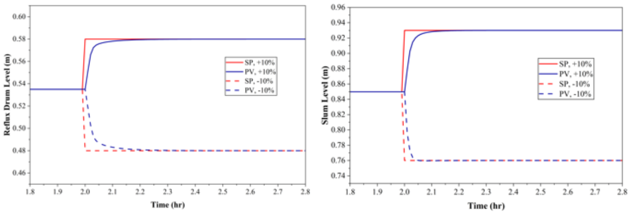
CONCLUSIONS

This study presents a comprehensive framework for the electrification of commodity chemical manufacturing by integrating steady-state and high-fidelity dynamic modeling to identify and rectify the associated challenges. The work successfully demonstrates a single-step co-production pathway that leverages microwave-assisted technology to simultaneously activate stable methane and nitrogen molecules for ethylene and ammonia synthesis. To address the inherent production fluctuations of pulsed microwave reactors, the study highlights a staggered manifold configuration to stabilize effluent flow, ensuring compatibility with industrial-scale downstream operations. Dynamic validation exemplified by the ammonia and ethylene purification column (C_02 & C_04) confirmed that a rigorously tuned control strategy can effectively reject ±10% feed disturbances while maintaining process stability and product purity. Ultimately, this systematic approach bridges the gap between laboratory-scale phenomena and industrial-scale operational reliability, establishing a robust foundation for the sustainable, electrified production of foundational chemicals

Table 4. Controller Sensitivity Matrix for Ethylene Column (C_04)

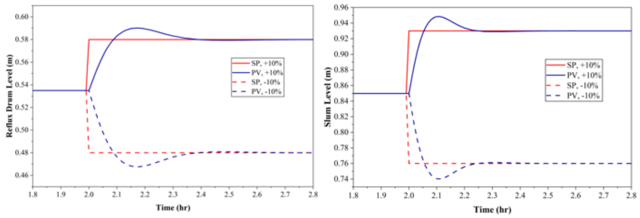
test Case	**reflux drum Level (K_C, τ_I)**	**sump level (K_C, τ_I)**
case-1: optimized (TL-ATV)	K_C = 46.35, τ_I = 3.96	K_C = 25.67, τ_I = 2.64
case-2: high gain / fast reset	K_C = 5, τ_I = 3.96	K_C = 5, τ_I = 2.64
case-3: high gain / slow reset	K_C = 80, τ_I = 3.96	K_C = 60, τ_I = 2.64
case-4: low gain / fast reset	K_C = 46.35, τ_I = 80	K_C = 25.64, τ_I = 60
case-5: low gain / slow reset	K_C = 5, τ_I = 80	K_C = 5, τ_I = 60

Case-1



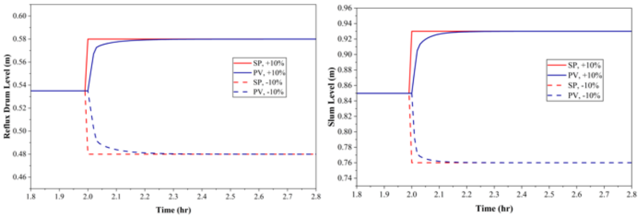
(a) Reflux drum level controller: $K_C=46.35$, $\tau_I=3.96$ (b) Sump level controller: $K_C=25.67$, $\tau_I=2.64$

case-2



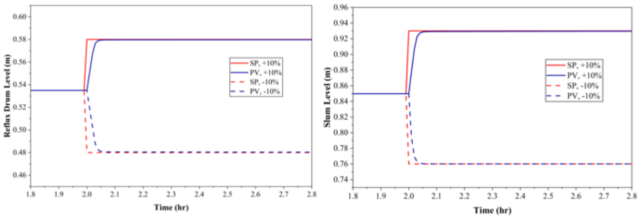
(a) Reflux drum level controller: $K_C=5$, $\tau_I=3.96$ (b) Sump level controller: $K_C=5$, $\tau_I=2.64$

case-3



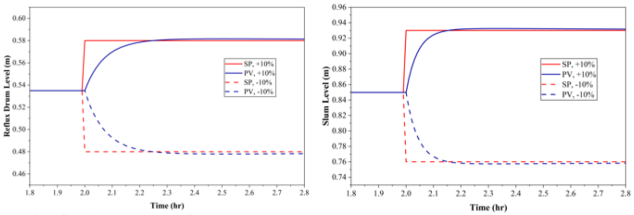
(a) Reflux drum level controller: $K_C=80$, $\tau_I=3.96$ (b) Sump level controller: $K_C=60$, $\tau_I=2.64$

case-4



(a) Reflux drum level controller: $K_C=46.35$, $\tau_I=80$ (b) Sump level controller: $K_C=25.64$, $\tau_I=60$

case-5



(a) Reflux drum level controller: $K_C=5$, $\tau_I=80$ (b) Sump level controller: $K_C=5$, $\tau_I=60$

Figure 9. Controller response of ethylene purification column (C_04) for different gains

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