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Research Article

Optimization of Parameters Using Taguchi Orthogonal Array Design for an Intensified Per-Pass Conversion of Alphabutol® Technology in Butene-1 Production

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Taguchi orthogonal design was used in this study to investigate the effects of three operational parameters (i.e., reactor temperature, reactor pressure, and catalyst ratio on per-pass conversion (PPC)). The optimal PPC was calculated from the predicted response function at 48°C of reactor temperature, 21 kg/cm^{2}g of reactor pressure, and catalyst mole ratio of 2.50. Under these conditions, the PPC was estimated to be 87.1 per cent, with a maximum SNR of 38.95. In line with the delta ranking, the decreasing order of significance of each process parameter on the average per-pass conversion (PPC) was $x_1 > x_2 > x_3$, with percentage contributions of 50.6%, 26.1%, and 23.3% for reactor temperature, temperature, and catalyst mole ratio, respectively. The optimal production condition can therefore be attained at a larger scale with the higher per-pass conversion of butene-1.

1. Introduction

The increasing demand for plastic packaging for different food products, medicines, and other sectors have led to a rise in demand for butene-1 markets [1]. This is because butene-1 is the most commonly used in the polyethene and plastic packaging industry, as it is commonly used as a comonomer in a variety of polyethene applications, including elastic and solid packaging materials [2]. Asia-Pacific is the largest market for butene-1, and the growing demand in the polyethene industry is due to the region's rapid development [3]. The demand for butene-1 is driven by the growing plastic sector in Asia-Pacific emerging markets. The butene-1 overall productivity is estimated to exceed the US $3.6 trillion in 2020, with the additional US $5.3 trillion estimated by 2027, representing a 5.9% compound annual growth rate (CAGR) for the 2020–2027 analytic era [4]. Linear low-density polyethylene (LLDPE), for example, is expected to grow at 6.4 per cent CAGR and reach $3.6 trillion by the conclusion of the study period (LLDPE). In 2020, the US market for butene-1 is anticipated to be worth $964.6 million. China, the world's second-biggest economy, is predicted to surpass US $1.1 trillion by 2027, resulting in a compound annual growth rate (CAGR) of 9.1 per cent throughout the research period of 2020 to 2027 [4]. Japan and Canada are two more noteworthy markets, with expected growth rates of 3.2 per cent and 5.4 per cent, respectively, from 2020 to 2027 [4]. Germany, on the other hand, is predicted to develop at a compound annual growth rate of around 3.7 per cent in Europe (CAGR) [4]. The activity chain in the butene-1 system adds value to the ultimate product.

Butene-1 is a monobutene that can come from a variety of sources. The most common sources for butene-1 production are natural gas, naphtha, butane, and ethylene. Multiple technologies are used to generate sources such as refineries, steam cracking of C4 hydrocarbons, ethylene dimerization, and butane dehydrogenation [5]. The technology of Alphabutol® is used to produce polymer grade butene-1 from high purity ethylene as the raw feedstock. Butene-1 is a comonomer utilized in the manufacture of various grades of polyethene. The Alphabutol® technology, which works in the liquid phase and employs a dissolved catalyst, selectively dimerizes ethylene to form butene-1. The manufacture of butene-1 polymer grade using Alphabutol® technology was created in the late 1980s by a single licensor in the globe, namely, Institut Français du Petrole (IFP) in collaboration with SABIC. It is a dimerization of ethylene to butene-1 [6]. Alphabutol® is preferred over other compounds because butene-1 is readily accessible in the C_4 fraction of naphtha crackers, but its recovery at a high purity level requires a complex strategy [7]. Among the numerous possible schemes, the most frequently used is to first extract or selectively hydrogenate butadiene, usually accompanied by the complete removal of isobutene via high-conversion MTBE synthesis, preceded by the separation of butene-1 from the other C4s via super fractionation or molecular sieves [8]. This method of processing has the significant disadvantage of being more costly in terms of investment and operational costs than the Alphabutol® approach [9]. This system requires a local outlet for butadiene and MTBE to be economically feasible. While this advantageous scenario may exist for more industrialized nations, namely, the United States of America, Japan, and Europe; it is not always the case in other countries. Coproduction of MTBE may be challenging for a petrochemical firm owing to recent MTBE bans in many places [10]. A challenging market for these related products, or a shortage of C4 cuts in the case of ethane crackers, has prompted LLDPE producers to consider ethylene dimerization by Alphabutol® for the synthesis of butene-1 [11].

The existing Alphabutol® operations have inherent challenges in terms of operating units at a high rate of production which calls for the minimization of unit production costs. Making trade-offs between these two choices has been a source of contention for a wide variety of businesses. It is critical to understand that many industries have difficulties with pump-around loop (PAL) operation owing to an inconceivable decrease in the total heat transfer coefficient (U) usually caused by pipe fouling [12]. This increases the unit maintenance cost as well production limitation occasioned by potential personal risks to the technicians while using the high-pressure water jet. The dangers associated with performing the industrial-scale experimental test are relatively high and this is large to the long residence period which usually leads to the difficulty in regulating the resulting chain reactions is critical to investigate the effects of reaction parameters on pilot plant size to mitigate this impact. The influence of operational parameters such as catalyst ratio, pressure, and temperature on the per-pass conversion (PPC) was thoroughly explored in this work utilising the orthogonal design matrix.

2. Material and Methods

2.1. Description of the Pilot Plant. The Alphabutol® pilot plant utilizes proprietary homogeneous titanium-based catalyst which demonstrates high dimerization activity coupled with excellent selectivity to butene-1 at moderate pressures and temperatures. This performance is influenced by the catalyst composition and reaction parameters. The catalytic ethylene dimerization to butene-1 also generates ethylene polymer as a side reaction. There are three main sections involved in this process which include the reaction, catalyst removal, and distillation. In the reaction section, the reactor is operating in the liquid phase at bubble point conditions. Fresh and recycled ethylene is fed to the liquid phase containing butene and hexene via a gas distributor, to make sure all ethylene is dissolved.

The homogeneous catalyst is continuously fed to the reactor section. The dimerization reaction is carried out at about 50–60°C and 20–30 atm with a reaction residence time of about 4 to 6 hours. The homogeneous catalytic reaction proceeds at an ethylene per-pass conversion of about 80–85% with a selectivity to butene-1 approaching 92%. The exothermic heat of the reaction is removed utilising an external pump-around loop (PAL). In the catalyst removal section, the active catalyst is deactivated using an amine compound and the spent catalyst is then incinerated at a higher temperature. In the distillation section, there are two columns; the first one is to recover unreacted ethylene from the top. The second column is to separate the desired product of butene-1 as overhead and the bottom is hexenes as heavies.

Data were collected at different operating conditions in the plant with mass and heat balance in mind. The data consisted of reactor parameters which consider pressure, temperature, catalyst ratio, and overall heat transfer coefficient. These parameters were collected from distributive control system (DCS) using a calibrated transmitter. The data were collected as shown in Figure 1, showing the Alphabutol® process flow diagram with parameter locations. All the data were collected from DCS using calibrated instruments before the experiment started for 300 days. These data were uploaded to Excel and analysed, excluding data that had errors in terms of power failure or runaway reaction during the experiment. Table 1 illustrated the 23 elements of the pilot plants.

2.2. Calibration of Equipment and Analyser

2.2.1. Equipment Calibration. Before the experiment, a qualified technician calibrated the instruments used. For the equipment calibration, a visual inspection of the instruments, tubing, wiring, instrument air, blowback, and connections were all performed for each installation. The "Hart Communicator" was then connected to the transmitter, and the diagnostic messages were recorded in the history form for further analysis. The transmitter was taken out of service to prevent the interference of the service fluid. The test equipment was then connected to the input and output of the transmitter. For cold junction compensation, the terminal temperature of the transmitter at stable ambient conditions was taken via the Hart Communicator. The terminal temperature value obtained was input

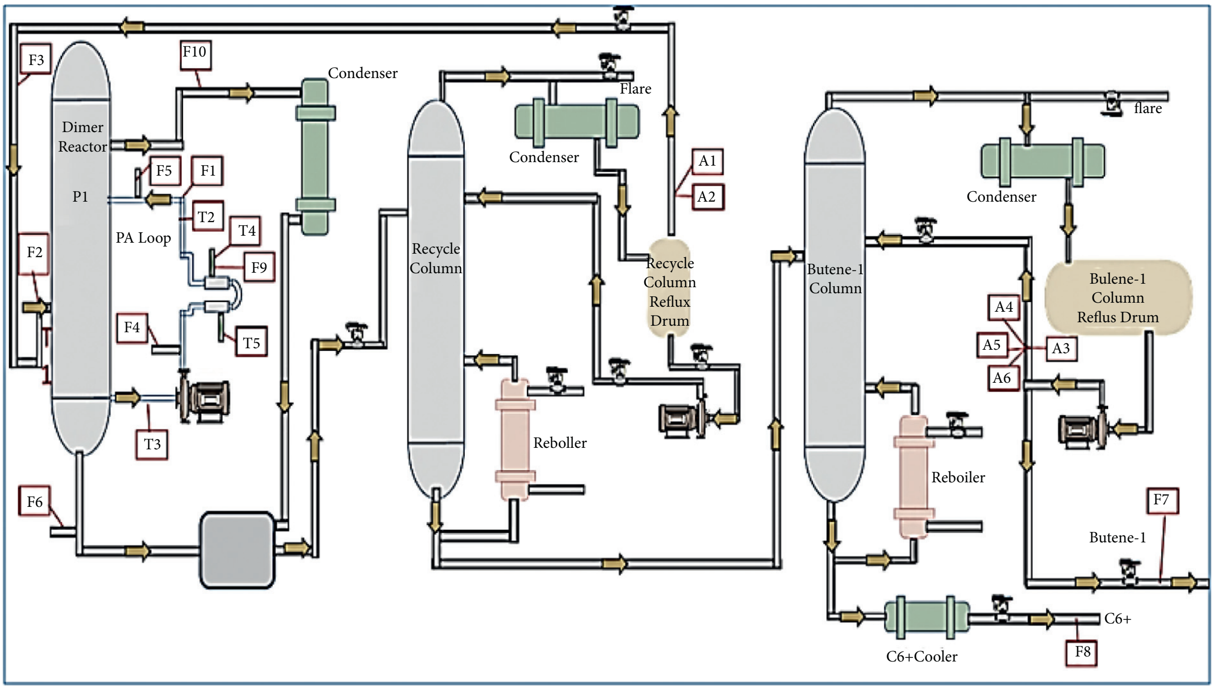

Figure 1: Alphabutol® schematic process flow diagram.

Table 1: The transmitters as well as the range and calibration frequency.

	Abbreviation	Instrument Type
Reactor temperature	T1	Transmitter
Reactor pressure	P1	Transmitter
PA loop process outlet temperature	T2	Transmitter
PA loop process inlet temperature	T3	Transmitter
PA loop CWS temperature	T4	Transmitter
PA loop CWR temperature	T5	Transmitter
PA loop flow	F1	Calculated flow at DCS from pump amperage
Ethylene feed flow	F2	DP transmitter
Recycle ethylene flow	F3	DP transmitter
Catalyst flow	F4	Coriolis
Cocatalyst flow	F5	Coriolis
Amine flow	F6	Coriolis
1-butene production flow	F7	DP transmitter
C6 + production flow	F8	DP transmitter
PA loop cooling water flow	F9	DP transmitter
Purge flow	F10	DP transmitter
Spent catalysts flow level	L1	DP transmitter
Ethane recycle	A1	Analyzer
Butene recycle	A2	Analyzer
2_BUTENE in 1-butene product	A3	Analyzer
3METH-1PENT in 1-butene product	A4	Analyzer
3METH-1PENT in 1-butene product	A5	Analyzer
1-butene purity in 1-butene product	A6	Calculated value at DCS

as the manual compensation value in the test instrument before the simulation of the thermocouple. The terminal temperature of the transmitter was stabilized throughout the calibration process. Low, mid, and high input of the calibration range to the transmitter was applied and the test equipment readings were recorded. If the AS FOUND values exceed the set tolerance, the "Found out of tolerance" box is ticked. However, if the AS FOUND values exceed 1/2 of the tolerance, the LEFT readings are recalibrated and recorded. The calibration, final readout device, and functional loop check are verified to be sure the last alarm is checked and are within the estimated accuracy. Finally, the transmitter is placed back in service after the calibration is completed.

2.2.2. Calibration of Gas Chromatograph Analyser. Firstly, the sample filter was inspected and the integrity of the sample system such as heat trace, sample pressure, sample flow, and sample cooler was checked. The oven temperature was checked and "import" is checked to transfer the current running "method" and last data and save it as the process before calibration with correct extensions. The carrier gas was also checked for proper pressure and flow settings. The process sample was closed while keeping the "run" button switched on with only the carrier gas allowed to flush out the columns completely for 3 cycles, while at the same time checking for a good baseline. The GC was then operated on the process sample using the proper pressure and flow settings, while the sample heater was monitored closely. The calibration sample container was then closed and the sample take-off and the conditioning panel was visually monitored to prevent equipment leakage.

2.3. Description of Theoretical Approach to Alphabutol® Pilot Plant Process. The focus of the ethylene dimerization process is to produce high selectivity of butene-1 by improving the catalyst system. In previous research, only a few improvements were found to improve the selection of butene-1 based on Alphabutol® technology. However, past improvement was focused on improving the advance control of the unit as reported by Jean-Marc [13], not to focus on optimizing reactor condition or addressing fouling issues. However, the researchers focused on the general hydrocarbon plant, which is a wide field that cannot be referenced for the Alphabutol® technology. Therefore, there is still room for improvement particularly research that focuses on fouling problems in industrial Alphabutol® technologies. It is not possible with an existing catalyst to prevent fouling; however, it is possible with optimizing reaction parameters to reduce fouling and increase butene-1 selectivity. The resulting findings could provide useful insights and guidelines to optimize and smoothen the process in a commercial-scale plant as more companies build new plants.

2.4. Design of L_9 (3^3) Taguchi Orthogonal Array. The set of data comprised reactor parameters such as pressure, temperature, catalyst mole ratio, and heat transfer coefficient overall. These parameters were obtained via a calibrated transmitter through the use of the distributive control system (DCS). The main objective is to study how those input characteristics affect the output response which passes per conversion (PPC). Taguchi experimental design matrix with a standard orthogonal array L_9 (3^3) was used to examine the effects of three parameters on the per-pass conversion (PPC). The lower, middle, and upper levels of optimized factors were selected based on the preliminary data gathered for 300 days under a range of operating conditions throughout the plant, with a special focus on mass and heat balance. The limits of each variable were selected since higher, middle, or lower settings resulted in a runaway or sluggish reaction, respectively. The factor ranges are presented in Table 2.

The per-pass conversion (PPC%) was estimated from the mass balance of the plant:

$$PPC = 1 - \left(\frac{((100 - A_1 - A_3/100) * (F_{10} + F_3))}{(((100 - A_1 - A_3/100) * (F_{10} + F_3) + F_2))} \right) * 100, \tag{1}$$

where A_1 is ethane recycled in PPM, A_3 is butene-2 in butene-1 product in PPM, F_2 is ethylene feed flow in kg/hr, F_3 is the recycle ethylene flow in kg/hr, and F_{10} is the purge flow in kg/hr. The results of the per-pass conversion trials were determined using the MINITAB ® statistical software package (version 18.1, United States). As shown in (2), the signal-to-noise ratio, which is the logarithmic function of the intended output (PPC), serves as the goal function for the maximization of the butene-1 production. The larger-is-better is used for the maximization.

$$\text{Maximized}_{(\text{Large-the-better})} = -10 * \log 10 \left\{ \sum \frac{(1/PPC)^2}{n} \right\}, \tag{2}$$

where "PPC" is the signal and "n" is the number of trials consisting of nine runs. Using the butene-1 manufacturing, a total of 27 experimental units were conducted using the L_9 (3^3) orthogonal design matrix in a 3×9 parametric study, totalling nine unique tests. The Taguchi technique was used to conduct an analysis of variance (ANOVA) on the response (PPC) of butene-1 production. The ratio (F) and p value ($p < 0.005$) were computed using the variables in the experimental design that were adjudged to be 5% confidence level.

3. Results and Discussion

3.1. Determination of Optimum Conditions by Taguchi Method. The Taguchi orthogonal design was used to optimize the per-pass conversion (PPC%). The optimal butene-1 production condition was established, and the statistical relevance of the process parameters on the PPC was examined appropriately. Table 3 shows the per-pass conversion (PPC%) of the butene-1 produced as well as the acquired signal-to-noise ratio (SNR) and total mean signal-to-noise (SNR_T) values. The summary of the generated experimental trials is as shown in Table 3. Based on the largest donating rule, the trial with the largest SNR ratio was estimated to be the predicted optimum condition for the production of butene-1 using the Alphabutol® technology [14]. The optimum process condition was attained at 48°C of reactor temperature, 21 kg/cm^{2}g of reactor pressure, and 2.50 of catalyst mole ratio. Under this condition, PPC was estimated to be 87.1% as obtainable in trial 4 with the highest SNR of 38.95. The results were obtained for the optimum condition at which the production parameters jointly optimized the PPC%. Hence, by using the above conditions, improved production of butene-1 was achievable at an optimized per-pass conversion.

3.2. Statistical Analysis of Means (ANOM). The mean analysis was used to determine the most effective process parameters for optimal per-pass conversion (PPC). The average

Table 2: Factor to optimize the PPC.

Variable	Symbol	Low setting (−1)	Mid setting (0)	High setting (+1)
Reactor temperature (°C)	X_1	48	51	54
Reactor pressure (kg/cm^{2}g)	X_2	18	19.2	21
T2/LC mole ratio	X_3	1.8	2.0	2.5

Table 3: Coded Taguchi L_9 $(\hat{3}3)$ orthogonal design.

Experiment number	Reactor temperature (°C) X_1	Reactor pressure (kg/cm^{2}g) X	Catalyst mole ratio X_3	Per-pass conversion (PPC) %	Signal-to-noise ratio (SNR)
1	1	1	1	85.6	38.8426
2	1	2	2	86.5	38.7179
3	2	1	2	90.3	38.8573
4	1	3	3	87.1	38.9525
5	2	2	3	86.9	38.7610
6	2	3	1	83.2	38.5609
7	3	1	3	89.4	38.7470
8	3	2	1	88.2	38.8499
9	3	3	2	85.6	38.8426

Table 4: Response matrix for per-pass conversion mean (larger is better).

Level	Reactor temperature (C)	Reactor pressure (kg/cm^{2}g)	Catalyst mole ratio
1	88.03	86.93	86.69
2	86.04	87.74	87.60
3	87.33	86.72	87.11
Delta	1.98	1.02	0.91
Rank	1st	2nd	3rd

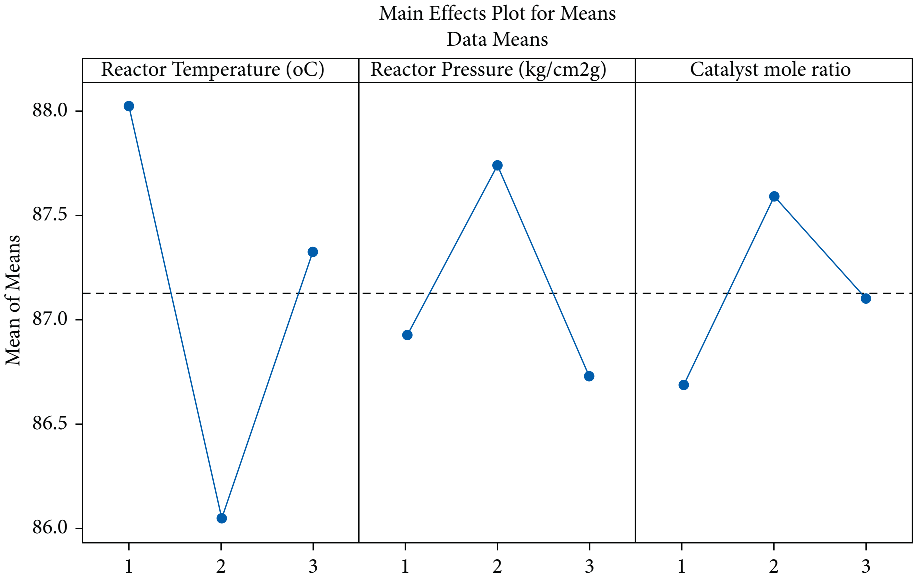

Figure 2: Relative mean of PPC% for each factor at three different levels.

response means, extremum, and delta difference (max–min) were calculated, where max and min denote the maximum and minimum average response means, respectively. The mean effects of each process factor were calculated using the value generated by the difference between their extremums [15]. Table 4 summarizes the average values for per-pass conversion optimum means.

Figure 2 shows the individualistic effects of each process variable on the per-pass conversion. The maximum average passes per conversion were obtained as 88.03% at level-1 of reactor temperature (48°C), 87.74% at level-2 of reactor pressure (19.2 kg/cm^{2}g), and 87.33% at level-1 of catalyst mole ratio (1.8). The decreasing order of significance of each process parameter on the average per-pass conversion (PPC) was $x_{1a0} > x_2 > x_3$ following the delta ranking with a corresponding percentage of 50.6%, 26.1%, and 23.3% for the reactor temperature, temperature, and catalyst mole ration, respectively.

The results obtained indicated that the reactor temperature has the most significant contribution to per-pass conversion (PPC%) with approximately half the proportion, whereas the catalyst mole ratio has the least. The temperature and pressure in the reactor may be regulated to modulate the per-pass conversion. The reactor is conducted at the bubble point, with a composition very similar to that of a binary combination of ethylene and butene-1 [16]. The pressure within the reactor is equal to the sum of each component's partial pressures. Because once the temperature is set, each component's vapour pressure is enforced. The temperature of the reactor may be regulated by altering the flow of cooling water to the pump that surrounds the heat exchanger. Controlling the pressure is accomplished by controlling the ethylene feed valve. The residence duration in the reactor is determined by the amount of product removed under level control. The catalyst flow rate is the sole remaining variable and once the pressure and temperature are set, the only method to alter the feed input is to alter the catalyst mole ratio [16].

4. Conclusion

The manufacture of polymerization grade butene-1 via Alphabutol® technology involves the utilization of high-purity ethylene as raw feedstock. Butene-1 is utilized in the manufacture of various polyethene grades as a comonomer. In the liquid phase of Alphabutol® technology, which works with a dissolved catalyst, ethylene is selectively dimerized into butene-1. This study examined the effects of the PPC on the reactor temperature, reactor pressure, and catalyst ratio using the Taguchi orthogonal design method. The optimal PPC was derived using the predicted response function at 48°C of reactor temperature, 21 kg/m^{2}g of reactor pressure, and a catalyst mole ratio of 2.50. PPC was thus estimated to be 87.1 per cent with the highest SNR of 38.95. The result was obtained for the optimum condition, which concurrently maximised the production parameters by the PPC per cent. Thus, using the above circumstances, increased synthesis of butene-1 at an optimum PPC was feasible.

Data Availability

The data used to support the findings of this study are included in the article.

Conflicts of Interest

The authors declared that there are no conflicts of interest whatsoever.

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