Robust Assessment of Controllable Operating Parameters of Entrained Flow Cogasification of Petcoke with Coal: Considering Some Uncertainties

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ABSTRACT: This paper is focused on the effects of some controllable operating parameters on the robustness of the coke/coal entrained flow cogasification process considering some uncertainties in it. In the present work, the operating variables were categorized into controllable parameters (CPs) (oxygen and steam concentrations, OC and SC) and hard-to-control parameters (temperature and coal/coke blending ratio) according to the actual modes during the cogasification process. Then, some robust response surface methodology (RSM) models, that is, mean RSM model and variance RSM model, for some important performance indexes \([H_2, CO, and (H_2 + CO)]\) production] with the CPs as independent variables, were found using combined array methodology. Then, the effects of OC and SC not only on the mean but also on the variance of each performance index were systematically investigated. Finally, the cogasification process was robustly optimized using the mean square criterion and desirability function. The result shows that the average production of \(H_2\) and that of \((H_2 + CO)\) increases with increasing OC but decreases with increasing SC. Additionally, higher OC suppresses the fluctuations in \(H_2\) and \((H_2 + CO)\) production, while higher SC enlarges the fluctuations in \(H_2\) production. Assuming that the variance of temperature in a gasifier is 20 °C and the variance of the coal/coke blending ratio is 5%, the multiobjective robust optimization solutions of OC and SC are 1.56 and 50%, respectively, and a satisfactory performance for high syngas production with low fluctuation can be gained.

1. INTRODUCTION

Petcoke is an important byproduct delivered from the oil-refining process. However, high sulfur contents have imposed some environmental limitations on their utilizations as combustion fuels. Cogasification is regarded as a promising and green method to convert them into syngas \((H_2 + CO)\) to produce some valuable chemicals or generate electricity.\(^1\,^2\) Cogasification is a well-established technology. Compared to petcoke, coal has higher ash content, and the metal elements in the ash can promote the gasification process.\(^2\,^3\) Therefore, cogasification of petcoke with coal is an attractive method for petcoke utilization. Blending petcoke with coal for gasification also has some other advantages such as being helpful to increase syngas yield, to improve the slag flow, to reduce some harmful elements’ environmental impacts, and so on.\(^3\)

In the past years, various researchers had carried out many experiments about the cogasification of petcoke with coal to investigate the influences of some operating variables on the gasification performance with the expectation of some valuable guidance for industrial operating decisions. Murthy et al.\(^1\) and Mondal et al.\(^2\) and Furimsky\(^5\) had well-reviewed on the early experimental studies. Cogasification of coke/coal in fixed-bed,\(^6\) fluidized-bed,\(^6\) and entrained-bed gasifiers\(^6\,^8\,^9\) has been reported in these literature studies. It is believed that an entrained-flow gasifier is the suitable choice because this type of gasifier can provide a higher temperature than other types that can greatly enhance the reactivities of petcoke for gasification.\(^1,^5\) Recently, quite a few scholars have reported their studies on this subject. For instance, Vejahati et al.\(^4\) performed some cogasification experiments of oil sand coke with sub-bituminous coal in an entrained-flow gasifier to assess the combined effects of the operating variables (temperature and oxygen and steam concentrations, i.e., \(T\) and \(O_2\) and \(H_2O\)) on the gasification performance. Shen et al.\(^8\) experimentally studied cogasification performance of coal and petroleum coke.
Experimental factors were assumed to be equally well-controlled in the laboratory and treated as deterministic variables, it may differ from the actually production greatly. For the scenario of an actual gasification process with an entrained-flow gasifier, oxygen and steam are spouted into the gasifier in the gaseous phase, thus they are relatively stable and easy to control. On the contrary, the fuel feedstock such as coke or coal is introduced into the gasifier in the solid phase, so they are relatively unstable and hard to control. In addition, temperature in the gasifier is not an independent variable and it distributes unevenly in the gasifier, so it is relatively unstable and hard to control too. Also, in turn, the gasification performance usually fluctuates rather than being constant because of the uncertainties in the hard-to-control factors and other factors. Therefore, when analyzing the operating parameters’ effects on gasification performance, the easy-to-control operating parameters (controllable parameters, CPs) should be paid more attention and their effects should be focused on. Additionally, not only the “position effects” but also the “dispersion effects” of CPs on gasification performance, that is, not only on the mean but also on the variance of some gasification performance indexes, should be investigated critically. This is very important for optimizing and stabilizing the gasification process and downstream applications. Only then the experimental study can provide a more scientific understanding of the total process and a more practical guidance for commercial operations.

In reviewing current studies, to our best knowledge, treating some hard-to-control parameters as uncertainty variables and focusing on analyzing the “position effects” and the “dispersion effects” of CPs in the coke/coal cogasification process remain absent. This paper presents our study on this subject based on the work of Vejahati. In their work, all experimental factors were assumed to be equally well-controllable and some deterministic RSM models were found to assess the effects of operating variables. Although in this paper, we treated oxygen concentration (OC, % O₂) and steam concentration (SC, % H₂O) as CPs with certainty forms but assumed temperature and blending ratio (BR) as hard-to-control variables with uncertainty forms. Then, using a robust RSM (RRSM) based on combined array design, proposed by Montgomery,13,14 we separately found the mean and variance of some critical performance indexes [H₂, CO, and (H₂ + CO) production] of the coke/coal cogasification process based on the experimental datasets in Vejahati’s work. The objectives of this paper are threefold: (a) to present the application of RRSM modeling with a combined array in entrained-flow cogasification of petcoke with coal when considering some

uncertainties; (b) to systematically assess both the mean and variance effects of CPs on the cogasification process of coke/coal; (c) and then to ascertain the optimal solutions of CPs that lead to satisfactory values of the response and little variance.

2. EXPERIMENTAL SECTION

In the current case,4 coke/coal cogasification experiments had been conducted by Vejahati et al. to quantify the relationships between the gasification performance and vital operating factors. The fluid coke and sub-bituminous Genesee coal, respectively, supplied by Syncrude Co. and Sherritt Co., were cogasified in an entrained-flow coal gasifier. The proximate and ultimate analyses and specific energy of the fuels are presented in Table 1. The entrained-flow gasification system is shown in Figure 1. The details about this gasification system can be seen in ref 4.

Table 1. Proximate and Ultimate Analyses of Fuels

| sample           | proximate analysis (wt %, AR) | ultimate analysis (wt %, daf) | ultimate analysis (wt %, daf) |
|------------------|-------------------------------|------------------------------|------------------------------|
| Genesee coal     | FC 50.26 VM 29.89 ash 15.4 moisture 4.45 | C 78.98 H 4.33 N 1.33 O 14.69 S 0.67 | SE (MJ/kg) 25.83 |
| fluid coke       | FC 85.81 VM 6.6 ash 6.22 moisture 1.37 | C 86.38 H 1.98 N 2.09 O 17.8 S 7.77 | SE (MJ/kg) 30.80 |

*: AR, as received; daf, dry and ash-free basis; FC, fixed carbon; VM, volatile matter; and SE, specific energy. bBy difference.

Figure 1. Schematic diagram of the entrained-flow gasification system (reprinted from ref 4) Copyright [2011] [ACS Publications].

According to the principle of central composite design methodology,15 the experiments were conducted over a temperature range of 1000–1400 °C, using steam and oxygen to carbon weight ratios of 0.9–4.3 and 0–0.4, respectively, equivalent to 15–50 vol % steam and 0–3 vol % oxygen in N₂ carrier gas. The bulk samples were crushed and sieved to a particle cut size of 45–75 μm. A fixed 60 g/h nominal flow rate of carbon (not coal) was maintained for all runs. The response variables used in Vejahati’s work were H₂, CO, H₂/CO ratio in...
variables. The common polynomial model used in RSM is

deterministic ones. That is, no

should be noted that all the independent variables involved in

this polynomial model are deterministic ones. That is, no

factors in the experiments are regarded as hard-to-control

considering the noise or uncertainties in the uncontrollable

factors are considered. The early solution for this problem was the well-known
crossed array experimental strategy and signal-to-noise ratio (SNR) statistic, which was first proposed by Taguchi. However, the crossed array usually results in excessive

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The common polynomial model used in RSM is the second-degree model

\[ y(x) = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_i^2 x_i^2 + \sum_{i<j}^{k} \beta_{ij} x_i x_j + \epsilon \]

where \( \beta^* \) is the regression coefficient and \( \epsilon \) is the random experimental error assumed to have a zero mean.

Then, the polynomial surface \( y(x) \) could be estimated using the so-called ordinary least-squares (OLS) algorithm. It should be noted that all the independent variables involved in this polynomial model are deterministic ones. That is, no factors in the experiments are regarded as hard-to-control variables. Thus, this type of RSM model is suitable for certainly analyzing the factors’ influences on the response without considering the noise or uncertainties in the uncontrollable factors.

3. METHODOLOGY

In most cases, general RSM modeling establishes a relationship between a response of interest, \( y \), and a number of associated control (or input) variables denoted by \( x_1, x_2, ..., x_k \) that can be used to predict response values for given settings of the control variables. The common polynomial model used in RSM is the second-degree model

\[ y(x) = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_i^2 x_i^2 + \sum_{i<j}^{k} \beta_{ij} x_i x_j + \epsilon \]

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crossed array experimental strategy and signal-to-noise ratio (SNR) statistic, which was first proposed by Taguchi. However, the crossed array usually results in excessive experiments but less effective information, and the SNR statistic was also viewed unfavorably by many researchers for lack of statistical foundation. The RRSM approach, first introduced by Myers and Carter and revitalized by Vining and Myers, suggests that the process characteristic and its process variability form a dual response system, and two separate models are established for the mean and variance of the response to analyze the “location effect” and "dispersion

| Table 2. Design Space and Experimental Values of Response Variables; Copyright [2011][ACS Publications] |
|---|
| factors | response variables |
| run | \( A, O_2 \) (vol %) | \( B, H_2O \) (vol %) | \( C, T \) (°C) | \( D, \) coke (wt %) | \( H_2 \) (mol/kg, daf) | \( CO \) (mol/kg, daf) | \( H_2 + CO \) (mol/kg, daf) |
| 1 | 3 (1) | 15 (−1) | 1000 (−1) | 0 (−1) | 38.40 | 24.20 | 62.6 |
| 2 | 0 (−1) | 18.5 (−0.8) | 1400 (1) | 100 (1) | 30.40 | 12.40 | 42.8 |
| 3 | 0 (−1) | 50 (1) | 1000 (−1) | 100 (1) | 15.17 | 3.23 | 18.4 |
| 4 | 1.49 (−0.01) | 15 (−1) | 1400 (1) | 0 (−1) | 43.67 | 40.10 | 83.77 |
| 5 | 1.59 (0.06) | 31.28 (−0.07) | 1200 (0) | 100 (1) | 23.19 | 7.22 | 30.41 |
| 6 | 0 (−1) | 50 (1) | 1400 (1) | 0 (−1) | 63.10 | 39.02 | 102.12 |
| 7 | 2.1 (0.4) | 15 (−1) | 1200 (0) | 50 (0) | 28.73 | 21.25 | 49.98 |
| 8 | 0.06 (−0.96) | 15 (−1) | 1000 (−1) | 100 (1) | 11.00 | 4.51 | 15.51 |
| 9 | 0 (−1) | 50 (1) | 1400 (1) | 0 (−1) | 37.97 | 23.38 | 61.17 |
| 10 | 2.6 (0.73) | 50 (1) | 1400 (1) | 100 (1) | 39.79 | 17.22 | 57.01 |
| 11 | 0 (−1) | 15 (−1) | 1400 (1) | 100 (1) | 19.45 | 13.61 | 33.06 |
| 12 | 0 (−1) | 50 (1) | 1400 (1) | 0 (−1) | 63.74 | 39.19 | 102.93 |
| 13 | 0 (−1) | 15 (−1) | 1200 (0) | 33 (−0.34) | 42.73 | 28.61 | 71.34 |
| 14 | 0 (−1) | 18.5 (−0.8) | 1400 (1) | 100 (1) | 34.99 | 14.39 | 49.38 |
| 15 | 0.86 (−0.43) | 50 (1) | 1400 (1) | 67 (0.34) | 47.51 | 20.00 | 67.51 |
| 16 | 3 (1) | 15 (−1) | 1400 (1) | 100 (1) | 22.04 | 15.41 | 37.45 |
| 17 | 3 (1) | 50 (1) | 1000 (−1) | 100 (1) | 13.84 | 2.31 | 16.15 |
| 18 | 0 (−1) | 31.63 (−0.05) | 1000 (−1) | 0 (−1) | 50.80 | 19.20 | 70.0 |
| 19 | 0.06 (−0.96) | 15 (−1) | 1000 (−1) | 100 (1) | 10.66 | 3.13 | 13.79 |
| 20 | 1.22 (−0.19) | 38.63 (0.35) | 1200 (0) | 0 (−1) | 54.37 | 35.22 | 89.59 |
| 21 | 0 (−1) | 38.45 (0.34) | 1200 (0) | 67 (0.34) | 42.74 | 14.51 | 57.25 |
| 22 | 1.53 (0.02) | 50 (1) | 1000 (−1) | 33 (−0.34) | 45.77 | 12.81 | 58.58 |
| 23 | 3 (1) | 50 (1) | 1200 (0) | 0 (−1) | 55.56 | 33.35 | 88.91 |
| 24 | 3 (1) | 33.22 (0.04) | 1400 (1) | 33 (−0.34) | 42.70 | 26.31 | 69.01 |
| 25 | 3 (1) | 27.95 (−0.26) | 1000 (−1) | 67 (0.34) | 21.00 | 6.85 | 27.85 |
| 26 | 1.5 (0) | 32.5 (0) | 1200 (0) | 50 (0) | 37.10 | 18.62 | 55.72 |
| 27 | 1.5 (0) | 32.5 (0) | 1200 (0) | 50 (0) | 36.10 | 16.65 | 52.75 |
| 28 | 1.5 (0) | 32.5 (0) | 1200 (0) | 50 (0) | 38.10 | 20.59 | 58.69 |
effect”, respectively. In practice, the two separate models give the analyst a more scientific understanding of the total process and thus allow them to see what levels of the controllable factors can lead to satisfactory values of the response and the variance. However, Myers’ RRSM approach is also based on the crossed array strategy, and the interactions between controllable and hard-to-control parameters cannot be estimated by this approach. To overcome these shortcomings, Montgomery suggested that the controllable and hard-to-control variables should be organized in a combined array rather than a crossed array. Thus, the RSM model should be developed including two parts: one is the part that contains controllable factors and the other is the part that contains the main effect of hard-to-control factors and the interactions between the controllable and hard-to-control factors. Then, the general second-degree RSM model based on Montgomery’s combined array strategy could be written as

\[ y(x, z) = f(x) + h(x, z) + \epsilon \]

\[ = \left[ \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_i^2 x_i^2 + \sum_{i<j}^{k} \beta_i \beta_j x_i x_j \right] \\
+ \left[ \sum_{i=1}^{r} \delta_i z_i + \sum_{i=1}^{r} \sum_{j=1}^{r} \delta_i \delta_j z_i z_j \right] + \epsilon \]

where \( z = (z_1, z_2, ..., z_r) \) is the hard-to-control variables, \( \gamma \) and \( \delta \) are the coefficients, and \( k \) and \( r \) are the number of controllable and hard-to-control variables, respectively.

Assume independent hard-to-control variables with zero mean and variances \( \sigma^2_\delta \). Furthermore, consider that the hard-to-control variables and the random error are uncorrelated. With these assumptions, the RRSM model including the mean model and variance model based on a combined array can be written as

\[
\begin{align*}
\mu_j(x) &= E[y(x, z)] = f(x) \\
\sigma_j^2(x, z) &= V[y(x, z)] = \sum_{i=1}^{r} \left( \frac{\partial y(x, z)}{\partial z_i} \right)^2 \sigma_{\delta i}^2 + \sigma^2_\epsilon
\end{align*}
\]

where \( \sigma^2_\epsilon \) is the variances of random experimental error \( \epsilon \).

4. RESULTS AND DISCUSSION

4.1. Model Development. As mentioned in Section 1, the OC and the SC should be regarded as controllable factors and certainty inputs (denoted as \( x_c \) and \( x_{dp} \), respectively) for modeling the present coke/coal cogasification process, while the temperature and the coal/ coke BR should be treated as hard-to-control factors with uncertainty forms (denoted as \( z_{ci} \) and \( z_{dp} \), respectively), although they are controllable during the experiments. All of the abovementioned input variables participate in modeling with coded forms. Then, according to the philosophy of Montgomery’s RSM method (eq 3), three RSM regression models for \( H_2 \), \( CO \) and \( H_2 + CO \), respectively, were developed with the OLS algorithm as follows

\[
\begin{align*}
\hat{y}_{H_2} &= (38.17 - 2.51 x_A + 5.38 x_B + 0.37 x_A^2 \\
&- 1.98 x_B^2 + 1.4 x_B x_B) + (7.19 - 0.97 x_A \\
&+ 1.09 x_B) z_C + (-13.93 + 0.04 x_A \\
&- 0.43 x_B) z_D \\
\hat{y}_{CO} &= (18.27 - 0.39 x_A - 0.36 x_B - 0.68 x_A^2 \\
&+ 2.79 x_B^2 + 0.95 x_A x_B) \\
&+ (6.86 + 0.28 x_A + 0.47 x_B) z_C \\
&+ (-11.76 + 0.35 x_A + 0.25 x_B) z_D \\
\hat{y}_{H_2+CO} &= (56.44 - 2.9 x_A + 5.02 x_B - 0.31 x_A^2 \\
&+ 0.81 x_B^2 + 2.34 x_B x_B) \\
&+ (14.05 - 0.69 x_A + 1.51 x_B) z_C \\
&+ (-25.67 + 0.39 x_A - 0.19 x_B) z_D
\end{align*}
\]

Then, according to eq 4, the RRSM models, that is, the mean and variance RSM model, for \( H_2 \), \( CO \), and \( H_2 + CO \) can be calculated, respectively, as follows

\[
\begin{align*}
\mu(y_{H_2}) &= 38.17 - 2.51 x_A + 5.38 x_B + 0.37 x_A^2 - 1.98 x_B^2 \\
&+ 1.4 x_B x_B \\
\sigma^2(y_{H_2}) &= (7.19 - 0.97 x_A + 1.09 x_B) z_C^2 \\
&+ (-13.93 + 0.04 x_A - 0.43 x_B) z_D^2 + \sigma_{e-H_2}^2 \\
\mu(y_{CO}) &= 18.27 - 0.39 x_A - 0.36 x_B - 0.68 x_A^2 + 2.79 x_B^2 \\
&+ 0.95 x_A x_B \\
\sigma^2(y_{CO}) &= (6.86 + 0.28 x_A + 0.47 x_B) z_C^2 \\
&+ (-11.76 + 0.35 x_A + 0.25 x_B) z_D^2 + \sigma_{e-CO}^2 \\
\mu(y_{H_2+CO}) &= 56.44 - 2.9 x_A + 5.02 x_B - 0.31 x_A^2 \\
&+ 0.81 x_B^2 + 2.34 x_B x_B \\
\sigma^2(y_{H_2+CO}) &= (14.05 - 0.69 x_A + 1.51 x_B) z_C^2 \\
&+ (-25.67 + 0.39 x_A - 0.19 x_B) z_D^2
\end{align*}
\]

4.2. Model Test and Comparison. Analysis of variance (ANOVA) is usually used to determine the adequacy of the RSM model. In ANOVA, if the probability associated with the F-ratio is small (usually the p-value less 0.05), then the model is considered a better statistical fit for the data than the response mean alone. Additionally, statistical indicators including RMSE, \( R^2 \), and adjusted \( R^2 \) are usually employed to evaluate the fitness of the RSM model. Compared with \( R^2 \), which always becomes inflated by increasing the number of terms, adjusted \( R^2 \) takes the numbers of the model parameter into account, so it is a better fitness measurement of the RSM model. A smaller RMSE and an adjusted \( R^2 \) closer to 1 indicate that the RSM model provides a better fit to the experimental data. Equations 7-2—8-2 are used to compute the indicators.

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RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (\hat{y}_i - y_i)^2}

R^2 = 1 - \frac{\sum_{i=1}^{n} (\hat{y}_i - y_i)^2}{\sum_{i=1}^{n} (y_i - \bar{y})^2}

\text{adjusted } R^2 = 1 - \left[ (1 - R^2) \times \frac{n - 1}{n - p - 1} \right]

where \( n \) is the number of experimental data, \( y_i \) is the experimental values, \( \hat{y}_i \) is the predicted values, \( \bar{y} \) is the average experimental values, and \( p \) is the number of input variables.

In Vejahati’s work, all the experimental factors were assumed to be equally well-controllable and treated as certainty variables. Based on this assumption, they found some certainty RSM models for syngas production analysis. Different from Vejahati’s certainty RSM model, we treat OC and SC as certainty inputs, while temperature and coal/coke BR as uncertainty inputs; thus, the RSM models in our study are uncertainty regression models. The fitness of our uncertainty RSM models were evaluated by RMSE, \( R^2 \), and adjusted \( R^2 \).

For comparison, Vejahati’s certainty RSM models were also evaluated by these statistical indicators. Additionally, ANOVA results, especially the variances of residual (\( \sigma^2 \)), of our models were compared with those of Vejahati’s certainty models to discuss the benefits of our models.

Results of ANOVA are summed up in Table 3 in terms of the sum and means of squares of residuals, corresponding degree of freedom, F-ratio, and p-value. Overall, all the p-values are very small (<0.0001), this manifests that both our uncertainty models and Vejahati’s certainty models have high effectiveness.

Figure 2 shows the parity plots of the predicted and the experimental values for \( \text{H}_2 \), \( \text{CO} \), and \( \text{H}_2 + \text{CO} \),...
respectively. The corresponding values of RMSE, $R^2$, and adjusted $R^2$ are also listed in Figure 2. Intuitively, it can be observed from Figure 2 that the points, whether calculated by our uncertainty model or by Vejahati’s certainty model, locate closely to the diagonal. The values of RMSE are all small compared with their respective scale. $R^2$ and adjusted $R^2$ are all greater than or equal to 0.9, which are very close to 1. All of the abovementioned results demonstrate that both our uncertainty models and Vejahati’s certainty models have enough prediction accuracy for further analysis and optimization.

Although a closer comparison of RMSE, $R^2$, and adjusted $R^2$ between our uncertainty models and Vejahati’s certainty models may show that the fitness of our models is slightly weaker than that of Vejahati’s models, and this does not deny the sufficient precision of our uncertainty models to approximate the cogasification process. The reason for the decrease in the fitness of our uncertainty models lies in the fact that temperature and BR were treated as uncertainty inputs in our models, and part of their variance that is originally in the fitted model was brought into the random error or residuals because of the special polynomial form of the regression model. Figure 3 shows the changes in the residuals’ variances ($\sigma^2$) because of the change in role in temperature and BR that form certainty to uncertainty variables as regression inputs, which also can be founded in the ANOVA results of the two types of the found models (Table 3). Although our uncertainty models are compromising on the fitness slightly, they are obviously in line with the actual situations of the industrial gasification process that temperature and BR are hard-to-control factors and should be regarded as uncertainty variables. In addition, most importantly, the variances of the residuals ($\sigma^2$) will be fully used in the variance RSM model (eq 4) as part of turbulence information for “dispersion effect” analysis of operation parameters, which differs from the general certainty RSM model greatly that these turbulence information is completely ignored. This just shows the advantages of Montgomery’s RRSM approach and confirms the conformance of our uncertainty models to the actual production of the industrial gasification process.

4.3. Robust Assessment of the Effect of Controllable Operating Parameters. For the robustness study, both the “location effect” and the “dispersion effect” should be analyzed. In this study, the perturbation plot was used for global sensitivity assessment of the CPs’ effects on the means and the variances of the cogasification performance indexes [H$_2$, CO, and (H$_2$ + CO) production]. Perturbation plots help to compare the effect of all factors in a single plot at a particular point in the design space. This makes the comparison of the factors much more effective than the traditional “one-plot-per-factor”.

4.3.1. H$_2$ Production Analysis. Figure 4 shows the CPs’ effects on the mean ($\mu_{H_2}$) and the variance ($\sigma_{H_2}$) of H$_2$ production.
production. As shown in this figure, the average production of \( \text{H}_2 \) increases with increasing SC but it decreases with increasing OC. This is consistent with Vejahati’s conclusion. The reason obviously lies in the fact that the prime reaction for \( \text{H}_2 \) production during the gasification process is the steam gasification reaction (SGR) \((\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2)\). High SC will be favorable for \( \text{H}_2 \) production. However, the SGR is a typical oxygen-insufficient reaction, and excess oxygen will strongly encourage the combustion of \( \text{H}_2 \) \((2\text{H}_2 + \text{O}_2 \rightarrow 2\text{H}_2\text{O})\), consume part of the hydrogen, and reduce the proportion of hydrogen in syngas.

The variance curve reveals the influence of the CPs on the fluctuation in \( \text{H}_2 \) production. From this figure, it can be seen that, in general, higher OC suppresses the fluctuation in \( \text{H}_2 \) production. This could also be because excess oxygen increases hydrogen combustion \((2\text{H}_2 + \text{O}_2 \rightarrow 2\text{H}_2\text{O})\) and reduces its proportion in the syngas, so the fluctuation of hydrogen decreases with the increase in oxygen. Contrary to the effect of OC, higher SC enlarges the fluctuation in \( \text{H}_2 \) production. The reason may also be that hydrogen generation mainly depends on the SGR \((\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2)\). Increasing the steam will generate more \( \text{H}_2 \) increase its proportion in the syngas, and therefore may increase its fluctuation.

**4.3.2. CO Production Analysis.** Figure 5 shows the CPs’ effects on the mean \((\mu_{\text{CO}})\) and the variance \((\sigma_{\text{CO}})\) of CO production. It can be seen from this figure that the mean of CO production is mainly affected by SC rather than OC because the profile line of OC is horizontal flat, which indicates that CO production is not affected by the OC. For SC, the mean of CO production in general decreases first and then increases with an increase in it. The effects of OC and SC on CO production are in accordance with the results obtained by Vejahati et al. and also with the findings by other researchers. There are actually multiple reactions for CO production during the gasification process. Among them, the partial oxidation reaction \((\text{C} + 0.5\text{O}_2 \rightarrow \text{CO})\) and SGR \((\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2)\) are the fundamental reactions for CO production. Obviously, these two reactions will compete with each other for the carbon sites necessary. On the other hand, and most importantly, they will also be competed with the carbon combustion reaction \((\text{C} + \text{O}_2 \rightarrow \text{CO}_2)\) and WGS reaction \((\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2)\) for the carbon sites to produce more \( \text{CO}_2 \) and \( \text{H}_2 \), which will reduce CO production significantly.

From this figure, it also can be seen that, in general, the fluctuation in CO production increases not only with increasing OC but also with increasing SC. In addition, the more inclined profile line of SC shows that the effect of SC on CO production fluctuation is more significant than that of OC. This indicates that excessive steam is more likely to cause greater fluctuation of CO production. On the one hand, increasing OC will increase carbon competition, which may increase the fluctuation in CO production. On the other hand, because the main reaction in the gasification process is the SGR \((\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2)\), increasing steam will not only produce more \( \text{H}_2 \) but also improve the production of \( \text{CO} \), so it may increase the fluctuation in CO production. It is also because the SGR is the dominant reaction in the gasification process, and the effect of SC on the fluctuation of CO production is greater than that of OC.

**4.3.3. \((\text{H}_2 + \text{CO})\) Production Analysis.** As mentioned earlier in Section 2, \( \text{H}_2 \) is the main target product, and CO produced by the gasification process can be converted into \( \text{H}_2 \) through the WGS reaction, so \((\text{H}_2 + \text{CO})\) production is an important total indicator for assessing the gasification process performance. Although the robustness effects of CPs on \( \text{H}_2 \) production and CO production are systematically discussed in the previous two sections, respectively, it is necessary to discuss this total indicator as a whole. Figure 6 shows the effects of OC and SC on the mean and variance of \((\text{H}_2 + \text{CO})\) production. As seen in this figure, overall, the mean of \((\text{H}_2 + \text{CO})\) production decreases with the increase in OC but it increases with the increase in SC. Therefore, in general, to increase the production of \( \text{H}_2 \) and \( \text{CO} \), it is necessary to reduce the OC and increase the steam supply.

Figure 6 also shows the CPs’ influences on the fluctuation in \((\text{H}_2 + \text{CO})\) production. It can be seen from this figure that the fluctuation in \((\text{H}_2 + \text{CO})\) production decreases with increasing OC but it increases with increasing SC. This shows that, in general, to reduce the fluctuation of effective composition of syngas, it is necessary to increase OC and reduce steam supply. This is the opposite of the requirement of CP for increasing the mean production of \((\text{H}_2 + \text{CO})\). Therefore, it is very necessary to carry out robust optimization to coordinate this conflict and find a group of optimal operation parameter combinations so as to maximize the gasification output and minimize the fluctuation.

**4.4. Robust Optimization.** As mentioned in the Introduction section, one purpose of this study is to ascertain...
the CPs’ values to gain higher performances with lower fluctuations for coke/coal entrained-flow cogasification process. Differing from the general RSM optimization that only the response function itself is optimized, two objective functions, that is, the mean function and the variance function of the response, need to be optimized synchronously for an RRSM optimization problem. A method called mean square error (MSE)\(^{24,25}\) is usually adopted to integrate these two objective functions.

\[
\min \text{MSE}_i = -\{\bar{y}(x, z)\}^2 + \hat{\sigma}^2_i y(x, z) \quad (12)
\]

The RRSM optimization in the current study is a multiresponse problem because there are three responses that are needed to be dealt with: H\(_2\), CO, and (H\(_2\) + CO) production. The most popular approach to optimization of multiple responses is the desirability function approach, as proposed by Derringer and Suich.\(^{26}\) To optimize the MSE model of eq 12, the desirability function form of each MSE is shown in eq 13.

\[
d_i = \frac{\max(\text{MSE}_i) - \text{MSE}_i}{\max(\text{MSE}) - \min(\text{MSE})} \quad (13)
\]

where \(d_i\) is the desirability, a desirability of zero indicates that the optimization result is completely undesirable and a desirability of one indicates that the optimization result is completely desirable.

Thus, the desirabilities for all MSE (i.e., “score” values) can be combined into a single objective measure to be maximized using a geometric mean function \(D\) as the decision criterion

\[
\max D = (d_1 \times d_2 \times \ldots d_k)^{1/k} \quad (14)
\]

For the current experiments, we assumed that the variance of temperature in the gasifier is 20 °C and the variance of the BR is 5%. Then, the multiobjective robust optimization of the current cogasification process performed in JMP software is shown in Figure 7. The robust optimum solutions of \(x_A\) and \(x_B\) with coded units are 0.095 and 1, respectively. The corresponding original robust optimum solutions of CPs, OC and SC, are 1.56 and 50 vol %, respectively. That is, when the volume percentage concentration of oxygen and steam in N\(_2\) carrier gas is 1.56 and 50%, respectively, a satisfactory performance for high syngas production with low fluctuation can be gained with a desirability score of 0.734.

5. SUMMARY AND CONCLUSIONS

Taking the uncertainties into account, according to the actual modes during the actually entrained-flow coke/coal cogasification process, we distinguish the operating parameters into controllable variables (i.e., OC and SC) and hard-to-CPs (i.e., temperature and coal/coke BR). Then, based on the experiments carried out by Vejahati et al, some RRSM models, namely, mean RSM model and variance RSM model, for H\(_2\), CO, and H\(_2\) + CO production with OC and SC as independent variables were found using combined array methodology. Then, their effects not only on the mean but also on the variance of each performance indexes were systematically investigated. Finally, the MSE criterion and desirability functions were employed in robustly optimizing the cogasification process. Some conclusions are summarized as follows:

- Robust assessment of the CPs’ effects on some important indexes of the entrained-flow coal/coke cogasification process is necessary. By only taking the uncertainties in hard-to-control operating parameters and the performance fluctuations caused by these uncertainties into account the investigation of the operating parameters’ effects on the performance cogasification process based on lab-scale experiments can provide more valuable guidance for actual industrial production.
- Because of the special modeling principles, combined array design is an appropriate methodology to achieve the abovementioned robust analysis for coke/coal entrained-flow cogasification progress, which is demonstrated in the present study. A detailed ANOVA shows that our RRSM models based on combined array design methodology for the current coke/coal cogasification process had been well-developed. Moreover, compared to Vejahati’s certainty RSM models, our robust RSM models have fully used the variances of the residuals (\(\sigma^2_i\)) in the variance RSM model, which helps for the achievement of “dispersion effect” analysis of CPs.
- For the coke/coal entrained-flow cogasification process, the average production of H\(_2\) increases with increasing SC but it decreases with increasing OC. The average CO production is mainly affected by SC, and it generally decreases first and then increases with the increasing SC. The CP’s effect on the average production of (H\(_2\) + CO) is similar to the case of H\(_2\) production.
- The “dispersion effect” of CPs on coke/coke cogasification performance shows that, in general, higher OC suppresses the fluctuation in H\(_2\) production, whereas higher SC enlarges the fluctuation in H\(_2\) production. However, their effects on the fluctuation
in CO production are opposite. The effects of CPs on the fluctuation in \((H_2 + CO)\) production are similar to the case of \(H_2\) production.

- With the assumption that the variance of temperature in the gasifier is 20 °C and the variance of the BR is 5%, the multiobjective robust optimization solutions, OC and SC, are 1.56 and 50%, respectively, and a satisfactory performance for high syngas production with low fluctuation can be gained with high desirability.

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### Notes

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