Optimization of the Mechanical Properties of a Hydrotreating Catalyst in the Impregnating and Drying Processes

Dongfang Wu and Jiancheng Zhou

Dept. of Chemical Engineering, School of Chemistry and Chemical Engineering, Southeast University, Jiangning District, Nanjing 211189, China

Yongdan Li

Tianjin Key Laboratory of Catalysis Science and Technology and State Key Laboratory for Chemical Engineering (Tianjin University), School of Chemical Engineering, Tianjin University, Tianjin 300072, China

DOI 10.1002/aic.11612

Published online November 3, 2008 in Wiley InterScience (www.interscience.wiley.com).

The Taguchi method, a powerful tool to the optimization for quality, is used to improve the mechanical properties of a $PCoMo/Al_2O_3$ hydrotreating catalyst in the impregnating and drying processes. An orthogonal array is selected to analyze the effects of the process factors, i.e., impregnating temperature, impregnating time, drying temperature, and drying time. Through this study, not only can the main process factors that affect the mean strength and the Weibull modulus be found, but also the optimal factor levels can be obtained. Results show that the drying temperature is the most significant process factor affecting both the mean strength and the Weibull modulus. It has been shown that the mean strength, Weibull modulus, and mechanical reliability of the hydrotreating catalyst can be improved significantly with the optimization of the impregnating and drying process factors. Experimental results are also provided to confirm the effectiveness of the Taguchi method. © 2008 American Institute of Chemical Engineers AIChE J, 54: 3116-3123, 2008

Keywords: solid catalysts, mechanical strength, reliability, Weibull distribution, Taguchi method

open literature.

Introduction

The mechanical strength of solid catalysts is one of the key parameters for the reliable and efficient performance of an industrial reactor. 1-6 Mechanical failure of the catalyst pellets results in the formation of fragments and fines, which causes maldistribution of fluid flow and increases the pressure drop across the reactor. In many industrial applications, physical breakage of the catalyst pellets is more often the cause of process shutdowns and catalyst replacements than

© 2008 American Institute of Chemical Engineers

cal materials with a brittle failure mode, and their mechanical failure is due to brittle fracture arising from a sudden catastrophic growth of a critical flaw under tensile stress induced in the catalyst bulk. 9-11 Solid catalysts are porous and full of defects, crystal edges, dislocations, or nonidentical materials enclosed, e.g., graphite and other additives. Any discontinuity that appears in the catalyst bulk may be treated as a flaw. Variations of size, shape and orientation of

their loss of catalytic activity. 7,8 However, academic institu-

tions active in catalysis research generally concentrate on the

chemistry rather than the catalyst mechanical properties. Few

articles on the catalyst strength are therefore available in the

Mixed oxides and oxide-supported metal catalysts are typi-

Correspondence concerning this article should be addressed to D. F. Wu at

these flaws result in a large scatter of the catalyst strength data. It has been proposed that the statistical variation in the catalyst strength can be modeled well with a two-parameter Weibull distribution. $^{9-13}$

$$P = 1 - \exp\left[-\left(\frac{F}{F_0}\right)^m\right] \tag{1}$$

where P is the probability of failure, F the load at failure, m the Weibull modulus, and F_0 the scale parameter. The higher the Weibull modulus, the narrower is the strength data distribution. The scale parameter corresponding to the fracture strength with a failure probability of 63.2% is closely related to the mean strength \bar{F} of the distribution. $^{13-15}$

$$\overline{F} = F_0 \Gamma \left(1 + \frac{1}{m} \right) \tag{2}$$

where Γ is the gamma function. For the Weibull modulus of 3–20, $\Gamma(1+1/m)$ takes values between 0.9 and 1.0, i.e., $\bar{F} = (0.9 \sim 1.0) \cdot F_0$.

It is well-known that the mechanical properties of solid catalysts are dependent on the material properties and microstructure of catalyst pellets. According to Griffith equation, 16 the fracture strength of brittle materials, such as solid catalysts, is a function of the surface energy, the Young's modulus, and the size of the microcracks (flaws) inside the material. Jayatilaka and Trutrum 17 concluded that the Weibull parameters, m and F_0 were governed by the flaw distribution in the material. During catalyst manufacturing, various process factors (parameters) will have effects on the physical properties of the material, and in consequence on the mechanical properties of catalyst pellets. It is therefore important for catalyst developers to optimize manufacturing process factors for improving the mechanical properties of solid catalysts.

To optimize process factors, an appropriate design of experiment is required. In general, classical experimental design methods are too complex and not easy to use. Furthermore, a large number of experiments have to be carried out when the number of process factors increases. To solve this problem, the Taguchi method was proposed by Taguchi in the late 1960s. 18-20 The method which is one of the fractional factorial designs uses a special design of orthogonal arrays to study the entire parameter space with a significantly small number of experiments, especially for the case that the interactions among process parameters are neglected. The experimental results are then transformed into a signal-to-noise (S/N) ratio. Taguchi recommended the use of the S/N ratio to measure the quality characteristics deviating from the desired values. Based on the analysis of the S/N ratio, the optimal levels of the process factors are determined. Furthermore, a statistical analysis of variance (ANOVA) is performed to see which process factors are statistically significant. Finally, a confirmation experiment is conducted to verify the optimal process factors obtained from the Taguchi method.

The Taguchi method has been shown to be a great success in industry for improving the product quality. The rapid growth of interest in the Taguchi method has led to numerous applications of the method in various fields in a worldwide range of industries and nations.^{21–23} However, it is not widely used by catalyst researchers and developers. To our

knowledge, only three articles were found to use the method to optimize the catalyst properties. In this work, the Taguchi method was used to optimize the mechanical properties of solid catalysts in the impregnating and drying processes, considering the time-consuming nature of these processes. An L_9 orthogonal array was selected for experimental layout. The PCoMo/Al₂O₃ catalyst, one of the hydrotreating catalysts widely used in petroleum refining industry, was selected as a model.

Experimental

Impregnation and drying

The PCoMo/Al₂O₃ catalyst was prepared by a coimpregnation technique using pore filling method. The support material was commercial γ -Al₂O₃ (3.82 mm cylindrical extrudates), available in China market. After drying at 200°C for 3 h, 20 g of the support was coimpregnated with 40 ml of a mixed aqueous solution of ammonium heptamolybdate, cobalt nitrate and phosphoric acid. After impregnation, the solids were left at room temperature for 2 h. Then the drying in air was carried out in a drying cupboard, by putting the samples after the temperature reached the preset value. The catalyst contains around 12 wt % of MoO₃, 4 wt % of CoO, and 3 wt % P₂O₅ after calcination.

Taguchi experimental design

Four process parameters, i.e., impregnating temperature, impregnating time, drying temperature, and drying time were selected as controllable factors. Their ranges were chosen based on the conditions often reported in the literature. ^{27–29} For each process factor, three levels were selected in this study, as shown in Table 1.

Four factors, each at three levels, are considered; therefore, an L_9 orthogonal array with four columns and nine rows can be employed if the interactions among these factors are neglected. This array has eight degrees of freedom and it can handle three-level process factors. Each process factor is assigned to a column, nine process-factor combinations being available. Therefore, only nine experiments are required to study the entire parameter space. The experimental layout for the four factors using the L_9 orthogonal array is shown in Table 2. Although this design does not have sufficient degrees of freedom to study the interactions among the factors and experimental error in four-factor systems, it was chosen on the basis that it requires the minimum possible number of experiments. Studying the interactions and error is possible only if a larger orthogonal array is used; however, this would increase the number of experiments significantly and diminish the advantage of Taguchi method.

Table 1. Factors and Their Levels Under Investigation

		Level		
Symbol	Factor	1	2	3
A	Impregnating temperature (°C)	20	50	80
В	Impregnating time (h)	4	8	12
C	Drying temperature (°C)	120	150	180
D	Drying time (h)	4	8	12

Table 2. Experimental Layout Using an $L_9(3^4)$ Orthogonal Array

	Factor and Level						
Trial No.	A	В	С	D			
1	1	1	1	1			
2	1	2	2	2			
3	1	3	3	3			
4	2	1	2	3			
5	2	2	3	1			
6	2	3	1	2			
7	3	1	3	2			
8	3	2	1	3			
9	3	3	2	1			

Determination of quality characteristics

It has been elucidated that the mean strength and Weibull modulus are the two important indices to the mechanical strength and reliability of solid catalysts; ^{5,9,12} in this study they were therefore obtained as the quality characteristics of the catalyst samples in the Taguchi method. A commercial strength tester made in Dalian, China, was used to measure the mechanical strength of the catalyst samples, described in detail elsewhere. ¹⁰ The knife-edge cutting strength was adopted, as it is more suitable for the extrudates than the crushing strength. ^{5,10} For each sample, 30 dried pellets were tested to obtain representative mechanical properties.

Mean strength was determined according to the arithmetical average of the observed strength data. Weibull parameters were obtained by the linear regression analysis. $^{14,15,30-32}$ The measured strength data are ranked in ascending order and then the probability of failure assigned to each strength F_i , is evaluated with the following probability estimator 14,15,33,34 :

$$P_i = \frac{i - 0.5}{n} \tag{3}$$

where P_i is the probability of failure for the *i*th ranked strength datum, and n is the sample size, i.e., the number of specimens tested. By taking the logarithm twice, Eq. 1 can be rewritten in a linear form.

$$\ln \ln \left(\frac{1}{1-P}\right) = m \ln F - m \ln F_0 \tag{4}$$

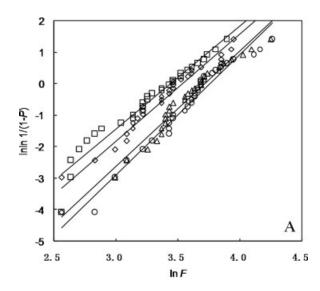
The Weibull modulus can thus be obtained directly from the slope term in Eq. 4 and the scale parameter can be deduced from the intercept term.

Results and Discussion

Figure 1(A) illustrates the fit of the measured strength data with Eq. 4. And the corresponding Weibull cumulative probability distributions are shown in Figure 1(B), where the data points are the measured strength values and the curves are drawn by the fitted Weibull distributions, Eq. 1. For the sake of good clarity, only four samples are shown in this figure. Other samples have similar behavior. It shows that the measured strength data of each sample follow Weibull distribution fairly well.

Table 3 presents the statistical results of the measured strength data of the catalyst samples and the estimated Weibull parameters. It is known that the low-strength/probability part of the catalyst strength distribution is the key domain for the mechanical reliability of the catalyst pellets. ^{5,12} Therefore, the fracture loads corresponding to three critical probabilities of failure were also computed with the fitted Weibull distributions. Clearly, the larger the predicted load, the higher the mechanical reliability, and hence the better the industrial performance of the catalyst. ^{5,12}

The predicted fracture loads with low probabilities of failure in Table 3 show remarkable differences between the samples obtained at different operating conditions. The impregnation and drying process factors, therefore, have great effects on the mechanical properties of solid catalysts. These results illustrate that there is a great possibility of improving



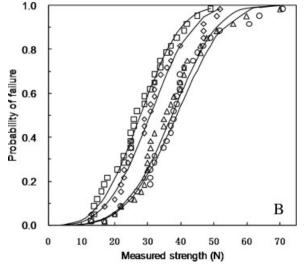


Figure 1. Weibull plots (A) and Weibull distribution curves (B) of the measured strength data of selected samples in Table 2.

(\triangle) Sample 5; (\square) Sample 6; (\bigcirc) Sample 7; (\diamondsuit) Sample 9.

Table 3. Experimental Results and Statistical Analyses

Measured Strength Data (N)		Weibull Parameters		Fracture Load with Specific Probabilities of Failure (N)			S/N Ratio (dB)		
Trial No.	Mean	Standard Deviation	m	F_0 (N)	1%	5%	10%	Mean	m
1	32.40	18.06	2.43	36.33	5.48	10.71	14.40	30.21	7.72
2	39.20	12.63	4.04	43.20	13.83	20.71	24.75	31.87	12.13
3	40.17	12.47	4.19	44.30	14.77	21.80	25.89	32.08	12.44
4	34.37	10.44	4.16	37.80	12.51	18.51	22.00	30.72	12.38
5	37.23	11.79	3.89	41.02	12.56	19.11	22.99	31.42	11.79
6	27.93	9.76	3.29	31.12	7.68	12.61	15.69	28.92	10.34
7	38.70	12.22	3.83	42.67	12.84	19.65	23.71	31.75	11.67
8	32.57	11.19	3.61	36.17	10.11	15.88	19.39	30.26	11.15
9	30.83	10.54	3.41	34.25	8.90	14.34	17.71	29.78	10.67

the mechanical strength and reliability of solid catalysts with the optimization of the manufacturing process factors.

Analysis of the S/N ratio

In the Taguchi method, the term "signal" represents the desirable value (mean) for the output characteristic and the term "noise" represents the undesirable value (S.D.) for the output characteristic. Therefore, the signal-to-noise (S/N) ratio is the ratio of the mean to the S.D. Taguchi used the S/N ratio to measure the quality characteristic deviating from the desired value. 18-20 The S/N ratio η is defined as

$$\eta = -10 \cdot \log(\text{MSD}) \tag{5}$$

where MSD is the mean square deviation for the output characteristic.

Usually, there are three categories of quality characteristics in the analysis of the S/N ratio, i.e., the-lower-the-better, the-higher-the-better, and the-nominal-the-better. 18-20 The MSD for the-higher-the-better quality characteristic can be expressed as

$$MSD = \frac{1}{r} \sum_{i=1}^{r} \frac{1}{y_i^2}$$
 (6)

and for the-lower-the-better quality characteristic

$$MSD = \frac{1}{r} \sum_{i=1}^{r} y_i^2$$
 (7)

where r is the repetition number of each experiment in the orthogonal array and y_i is the quality characteristic, i.e., the mean strength or Weibull modulus in this case.

Table 4. S/N Response Table for the Mean Strength

		Mean S/N Ratio (dB)						
Factor	Level 1	Level 2	Level 3	Max-Min				
A	31.38	30.35	30.60	1.03				
В	30.90	31.18	30.26	0.92				
C	29.80	30.79	31.75	1.95				
D	30.47	30.85	31.02	0.55				

The total mean S/N ratio $\bar{\eta} = 30.78 \text{ dB}$

Clearly, the-higher-the-better quality characteristic for the mean strength or Weibull modulus should be taken for obtaining optimal performance of the catalyst. So, the S/N ratios corresponding to the mean strength and Weibull modulus were computed with Eqs. 5 and 6, where r is equal to unity in this work, and listed in the last two columns in Table 3.

As the experimental design is orthogonal, it is then possible to separate out the effect of each process factor at different levels. For example, the mean S/N ratio for the impregnating temperature at Levels 1, 2, and 3 can be calculated by averaging the S/N ratios for the experiments 1-3, 4-6, and 7-9, respectively. The mean S/N ratio for each level of the other process factors can be computed in the similar manner. As a result, an S/N response table can be obtained, as shown in Tables 4 and 5 for the mean strength and Weibull modulus, respectively. In addition, the total mean S/N ratios $\bar{\eta}$ for the nine experiments are also calculated and listed in Tables 4 and 5. Figures 2 and 3 show the corresponding S/N response graphs for the mean strength and Weibull modulus, respectively.

Regardless of the category of the quality characteristic, a greater S/N ratio corresponds to a smaller variance of the output characteristic around the desired value, and hence to a better quality characteristic. Therefore, the optimal level of the process factors is the level with the greatest S/N ratio. 18-20 From Figure 2, it can be found that the optimal factor levels for the mean strength are the impregnating temperature at Level 1, the impregnating time at Level 2, the drying temperature at Level 3 and the drying time at Level 3, denoted as A1B2C3D3. As can be seen from Figure 3, the optimal combination of the process factor levels, however, are A2B2C3D3 for the Weibull modulus. The relative importance among the process factors for the quality characteristics

Table 5. S/N Response Table for the Weibull Modulus

		Mean S/N ratio (dB)						
Factor	Level 1	Level 2	Level 3	Max-Min				
A	10.76	11.51	11.16	0.74				
В	10.59	11.69	11.15	1.10				
C	9.74	11.72	11.97	2.23				
D	10.06	11.38	11.99	1.93				

The total mean S/N ratio $\bar{\eta} = 11.14$ dB.

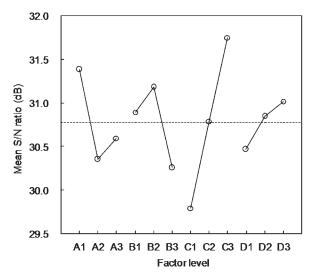


Figure 2. S/N graph for the mean strength.

still needs to be known so that the optimal combination of the process factor levels can be determined more accurately.

Analysis of variance

The purpose of the analysis of variance (ANOVA) is to investigate which process factors significantly affect the quality characteristic. This is to be accomplished by separating the total variability of the S/N ratio, which is measured by the sum of the squared deviation from the total mean S/N ratio, into contributions by each of the process factors and the error. $^{18-20}$ First, the total sum of squared deviation SS_T from the total mean S/N ratio $\bar{\eta}$ can be calculated as

$$SS_{T} = \sum_{i=1}^{k} (\eta_i - \overline{\eta})^2$$
 (8)

where k is the number of experiments in the orthogonal array and η_i is the S/N ratio for the ith experiment.

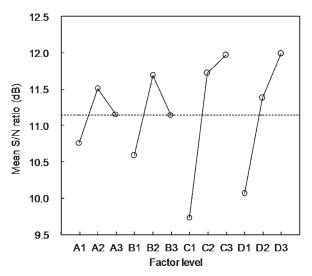


Figure 3. S/N graph for the Weibull modulus.

3120

The total sum of squared deviation is decomposed into two sources: the sum of squared deviation SS_p due to each process factor and the sum of squared error SS_e . In this study, the SS_e is equal to zero since no column in the orthogonal array is left empty for the error of experiments. The SS_p for factor p can be calculated as

$$SS_{p} = t \cdot \sum_{i=1}^{q} (\overline{\eta}_{p,i} - \overline{\eta})^{2}$$
(9)

where t is the repetition number of each level of the factor in the orthogonal array, q is the level number of the factor, and $\bar{\eta}_{\mathrm{p},i}$ is the mean S/N ratio for the factor at level i. The percentage contribution ρ_{p} by each of the factors is a ratio of the sum of squared deviation SS_p due to each factor to the total sum of squared deviation SS_T. Clearly, the larger the percentage contribution ρ_{p} , the more significant the corresponding factor.

In addition, there is a tool called an F-test^{18–20,35} to see which process factors have a significant effect on the quality characteristic. In performing the F-test, the mean of squared deviation MS_p due to each factor needs to be calculated. The mean of squared deviation MSp is equal to the sum of squared deviation SSp divided by the number of degrees of freedom associated with the factor. Then, the F-test value for each factor is simply the ratio of the mean of squared deviation MS_p to the mean of squared error. The process factor with a larger F-test value has a more significant effect on the quality characteristic. However, it should be noted that there is no degree of freedom for the experimental error in this study, so a "pooling technology" was used to estimate approximately the error variance, i.e., the mean of squared error. As a rule of thumb, the Taguchi method suggests that the pooling is generally done until the degrees of freedom of the error become close to half of the total degrees of freedom of the designed experiment. ^{18,19} Thus, the two process factors having the smallest contribution were pooled in this case.

Table 6 shows the results of ANOVA for the mean strength. It can be found that the drying temperature is the most significant process factor affecting the mean strength. The next is the impregnating temperature, followed by the impregnating time. The change of the drying time in the range examined has little effect on the mean strength. Table 7 shows the results of ANOVA for the Weibull modulus. As can be seen, the drying temperature is also the most significant process factor affecting the Weibull modulus, followed in turn by the drying time and then impregnating time. The

Table 6. Analysis of Variance for the Mean Strength

Factor	Sum of Squares	Degrees of Freedom	Mean Square	F-test	Percentage Contribution (%)
Α	1.74	2	0.87	1.93	18.78
В	1.33	2	0.67°	1.47	14.37
C	5.73	2	2.86	6.34	61.73
D	0.47	2	0.24°	_	5.12
(Error)	(1.81)	(4)	(0.45)		
Total	9.28	8			100

The variances of factors B and D (with open circle) are pooled into the error factor.

Table 7. Analysis of Variance for the Weibull Modulus

Factor	Sum of Squares	Degrees of Freedom	Mean Square	F-test	Percentage Contribution (%)
A	0.83	2	0.41°	_	4.73
В	1.81	2	0.90°	1.37	10.36
C	8.99	2	4.50	6.83	51.49
D	5.84	2	2.92	4.43	33.43
(Error)	(2.64)	(4)	(0.66)		
Total	17.47	8			100

The variances of factors A and B (with open circle) are pooled into the error factor.

change of the impregnating temperature has an insignificant effect on the Weibull modulus.

Confirmation experiments

Once the optimal levels of the process factors have been selected, the final step is to predict and verify the improvement of the quality characteristic using the optimal levels of the process factors. ^{18–20} The estimated S/N ratio $\eta_{\rm opt}$ using the optimal levels of the process factors can be calculated as

$$\eta_{\text{opt}} = \overline{\eta} + \sum_{p=1}^{o} (\overline{\eta}_{p,\text{opt}} - \overline{\eta})$$
 (10)

where $\bar{\eta}$ is the total mean S/N ratio, $\bar{\eta}_{p,\text{opt}}$ is the mean S/N ratio for factor p at its optimal level, and o is the number of the process factors that affect the quality characteristic significantly. In prediction, the two factors pooled into the error factor in the above ANOVA are not included in Eq. 10; therefore, the estimated η_{opt} values are equal to 32.36 and 12.82 for the mean strength and Weibull modulus, respectively. Based on the η_{opt} value, the quality characteristic for the optimal levels of the process factors can be predicted with Eqs. 5 and 6.

To verify these predictions, two additional experiments were conducted at the optimal levels of the process factors. Table 8 shows the comparison of the predicted and experimental values of the mean strength and Weibull modulus. It can be seen that only minor discrepancies exist between the predicted and experimental results. There may be two reasons for the discrepancies. First, in prediction only the most significant two factors are involved in Eq. 10 as the other two have been pooled into the error factor, which may overestimate the experimental error variance. Indeed, higher mean strength and Weibull modulus are predicted if the impregnating time (factor B), which makes the 14.37% and 10.36% contributions to the mean strength and Weibull modulus, respectively, is included in Eq. 10. The second reason for the discrepancies may be the interactions among the process factors. It is quite likely that the impregnating temperature and impregnating time, or the drying temperature and drying time affect the catalyst mechanical properties with a small interacting manner.

On the whole, good agreements are observed between the predicted and actual experimental values, which indicate that the overall impact of the interactions among the process factors, and the experimental error are both relatively small and that the experimental results have a satisfactory reproducibil-

ity. It can also be found from Table 8 that the mean strength and Weibull modulus are both improved by using the optimal levels of the process factors, as compared with the data in Table 3. These results confirm the "no-interaction" assumption, experimental reproducibility, and prior design and analysis for optimizing the process factors in this study.

The Taguchi method is a simple and powerful tool that is suitable for analyzing the complex systems having multivariable and high nonlinearity. ^{18–20} However, it should be noted that the meticulousness in setting the levels of the various controllable factors correctly to reduce experimental error is crucial to the success of the analysis when using the Taguchi method. Thus, the process factors investigated and other controllable factors must be strictly set to their proper levels for each experiment in the orthogonal array. Failure to do so, even for a single factor, could destroy the valuable property of orthogonalization, rendering conclusions from the experimental error.

On the improvement of the mechanical reliability

The mechanical performance of a catalyst in industry lies on the mechanical reliability of the catalyst pellets, controlled by the low-strength/probability part of the strength distribution.^{5,12} The larger the predicted load corresponding to low probability of failure, the higher the mechanical reliability, and hence the better the industrial performance of the catalyst. The ultimate purpose of optimizing the mean strength or Weibull modulus is, therefore, to improve the catalyst mechanical reliability.

As is seen from Table 8, the mechanical reliability of the catalyst pellets has been greatly improved with the optimization of either the mean strength or the Weibull modulus using the Taguchi method, as compared with the data in Table 3. Comparing the two optimal samples, it can be found that the one obtained with the optimization of the mean strength has a higher mechanical reliability and is therefore to be preferred. Nevertheless, it does not mean that the mean strength is more important than the Weibull modulus. Comparing the two optimal combinations of the factor levels, there is only one factor level differing from each other between them. This process factor is the impregnating temperature, which has a significant effect on the mean strength, but little effect on the Weibull modulus, as shown in the above ANOVA; therefore, the sample obtained with the optimization of the mean strength has not only the highest mean

Table 8. Results of the Confirmation Experiments

	Measured Strength Data (N)		n Weibull Parameters		Fracture Load with Specific Probabilities of Failure (N)		ific es of
Optimal level	Mean	Standard Deviation	m	F_0 (N)	1%	5%	10%
A1B2C3D3 Experiment Prediction	45.57 41.50	12.83	4.23	50.00	16.84	24.77	29.36
A2B2C3D3 Experiment Prediction	39.80	10.23	4.49 4.37	43.55	15.65	22.49	26.39

strength but also a very large Weibull modulus which is close to that of the sample obtained with the optimization of the Weibull modulus. This gives the reason why the optimization of the mean strength leads to a higher mechanical reliability in this work.

It should be noted that although this study is shown to be a great success, there is a possibility of failure in using single objective optimization at a time for the mean strength or Weibull modulus, for improving the catalyst mechanical reliability. The fracture loads with low probabilities of failure are dependent not only on the Weibull modulus but also on the scale parameter related to the mean strength. Therefore, single objective optimization at a time cannot fully ensure the improvement of the catalyst mechanical reliability. To solve this problem, a multiobjective optimization technique using Taguchi quality loss function has been developed to optimize the multiple quality characteristics simultaneously.³⁶ The multiobjective optimization is useful in the sense that at the same optimal factor levels one can get the optimal values of multiple quality characteristics at the same time rather than a single optimal quality characteristic. If this technique is used to optimize the mean strength and Weibull modulus simultaneously, the mechanical reliability of the catalyst pellets will be improved affirmatively.

Another issue that should be mentioned is that the optimization of a catalyst is a multidimensional task. In addition to the mechanical property, many other catalyst properties such as catalytic activity and selectivity should be optimized. However, the best values of all these desired properties may correspond to different optimal levels of the manufacturing process factors. Therefore, caution should be exercised when using the optimal factor levels obtained with the optimization of single catalyst property, for example, the mechanical property in this work, in the catalyst manufacturing process.

Conclusions

3122

This article has discussed an application of the Taguchi method for improving the mechanical properties of a hydrotreating catalyst. As shown in this study, the Taguchi method provides a systematic and efficient methodology for the optimization of the process factors with far less efforts than would be required for most optimization techniques.

It has been shown that the mean strength, Weibull modulus, and mechanical reliability of the hydrotreating catalyst can be improved significantly with the optimization of the impregnating and drying process factors. The optimal factor levels are the impregnating temperature at 20°C (for the mean strength) or 50°C (for the Weibull modulus), the impregnating time at 8 h, the drying temperature at 180°C, and the drying time at 12 h. It was also found that the drying temperature is the most significant process factor affecting the mean strength, followed in turn by the impregnating temperature and then the impregnating time. The change of the drying time in the range examined has little effect on the mean strength. And the contribution order of the process factors for the Weibull modulus is the drying temperature, drying time, then impregnating time, and then impregnating temperature. Finally, the confirmation experiments were also conducted to verify the validity of the Taguchi method.

Acknowledgments

Financial supports from Jiangsu Provincial Natural Science Foundation under Grant No. BK2007119, the Program for New Century Excellent Talents in University under Grant No. NCET-07-0185, the Science and Technology Foundation of Southeast University under Grant No. XJ0619243, and the Teaching and Research Program of Southeast University for Excellent Young Teachers are gratefully acknowledged. Y.L. thanks financial supports for this work by the Natural Science Foundation of China under Grant Nos. 20425619 and 20736007, the Program of Introducing Talents to the University Disciplines under File No. B06006, and the Program for Changjiang Scholars and Innovative Research Teams in Universities under File No. IRT0641.

Literature Cited

- Richardson JT. Principles of Catalyst Development. New York: Plenum Press, 1989.
- Satterfield CN. Heterogeneous Catalysis in Industrial Practice, 2nd ed. New York: McGraw-Hill, 1991.
- Denny PJ, Twigg MV. Factor determining the life of industrial heterogeneous catalysts. Stud Surf Sci Catal. 1980;6:589–591.
- Couroyer C, Ghadiri M, Laval P, Brunard N, Kolenda F. Methodology for investigating the mechanical strength of reforming catalyst beads. *Oil Gas Sci Technol*. 2000;55:67–85.
- Wu DF, Zhou JC, Li YD. Mechanical strength of solid catalysts: recent developments and future prospects. AIChE J. 2007;53:2618–2629.
- Wu DF, Song LY, Zhang BQ, Li YD. Effect of the mechanical failure of catalyst pellets on the pressure drop of a reactor. *Chem Eng Sci.* 2003;58:3995–4004.
- Beaver ER. Mechanical testing of extruded, tableted, and ringformed catalysts. In: Weller SW, editor. Standardization of Catalyst Test Methods, AIChE Symp Ser. Vol 70, No. 143, New York: AIChE, 1974:1–4.
- Beaver ER. Mechanical testing of catalysts. Chem Eng Prog. 1975:71:44–45.
- Li YD, Li XM, Chang L, Wu DH, Fang ZP, Shi YH. Understandings on the scattering properties of the mechanical strength data of solid catalysts: a statistical analysis of iron-based high-temperature shift catalysts. *Catal Today*. 1999;51:73–84.
- Li YD, Wu DF, Zhang JP, Chang L, Wu DH, Fang ZP, Shi YH. Measurement and statistics of single pellet mechanical strength of differently shaped catalysts. *Powder Technol*. 2000;113:176–184.
- Subero-Couroyer C, Ghadiri M, Brunard N, Kolenda F. Weibull analysis of quasi-static crushing strength of catalyst particles. *Trans IChemE A: Chem Eng Res Des.* 2003;81:953–962.
- 12. Wu DF, Zhou JC, Li YD. Distribution of the mechanical strength of solid catalysts. *Trans IChemE A: Chem Eng Res Des.* 2006;84:1152–1157.
- Weibull W. A statistical distribution function of wide applicability. J Appl Mech. 1951;18:293–297.
- Khalili A, Kromp K. Statistical properties of Weibull estimators. J Mater Sci. 1991;26:6741–6752.
- Wu DF, Zhou JC, Li YD. Methods for estimating Weibull parameters for brittle materials. J Mater Sci. 2006;41:5630–5638.
- 16. Griffith AA. The phenomena of rupture and flow in solid. *Phil Trans Roy Soc Lond A*. 1921;221:163–198.
- 17. Jayatilaka ADS, Trustrum K. Statistical approach to brittle fracture. *J Mater Sci.* 1977;12:1426–1430.
- Taguchi G. Introduction to Quality Engineering. Tokyo: Asian Productivity Organization, 1990.
- Ross PJ. Taguchi Techniques for Quality Engineering. New York: McGraw-Hill, 1988.
- Montgomery DC. Design and Analysis of Experiments. New York: Wiley, 2001.
- Bendell A, Disney J, Pridmore WA. Taguchi Methods: Applications in World Industry. UK: IFS Publications, 1989.
- Madaeni SS, Koocheki S. Application of Taguchi method in the optimization of wastewater treatment using spiral-wound reverse osmosis element. *Chem Eng J.* 2006;119:37–44.
- Mousavi SM, Yaghmaei S, Jafari A, Vossoughi M, Ghobadi Z. Optimization of ferrous biooxidation rate in a packed bed bioreactor using Taguchi approach. *Chem Eng Process*. 2007;46:935–940.

- Ozdemir C, Akin AN, Yildirim R. Low temperature CO oxidation in hydrogen rich streams on Pt-SnO₂/Al₂O₃ catalyst using Taguchi method. *Appl Catal A*. 2004;258:145–152.
- Ozdemir C, Akin AN, Yildirim R. Maximization of total surface area of Pt-SnO₂/Al₂O₃ catalyst by the Taguchi method. Korean J Chem Eng. 2003;20:840–843.
- 26. Dawson EA, Barnes PA. A new approach to the statistical optimisation of catalyst preparation. *Appl Catal A*. 1992;90:217–231.
- 27. Grange P, Vanhaeren X. Hydrotreating catalysts, an old story with new challenges. *Catal Today*. 1997;36:375–391.
- Radomyski B, Szczygiee J, Trawczynski J. Reaction of thiophene with hydrogen over CoMo/γ-Al₂O₃ catalysts. The role of cobalt. Appl Catal. 1986;25:295–302.
- Ramirez J. Hydrodesulphurization activity and characterization of sulphided molybdenum and cobalt-molybdenum catalysts. *Appl* Catal. 1989;52:211–224.
- Trustrum K, Jayatilaka ADS. On estimating the Weibull modulus for a brittle material. J Mater Sci. 1979;14:1080–1084.

- 31. Bergman B. On the estimation of the Weibull modulus. *J Mater Sci Lett.* 1984;3:689–692.
- Wu DF, Zhou JC, Li YD. Statistical analysis of pellet size variation in commercial catalysts. *Part Part Syst Charact*. 2005;22:63–68
- 33. Wu DF, Zhou JC, Li YD. Unbiased estimation of Weibull parameters with the linear regression method. *J Eur Ceram Soc.* 2006;26:1099–1105.
- 34. Wu DF, Lu GZ, Jiang H, Li YD. Improved estimation of Weibull parameters with the linear regression method. *J Am Ceram Soc.* 2004;87:1799–1802.
- 35. Fisher RA. Statistical Methods for Research Workers. London: Oliver and Boyd, 1925.
- Antony J. Simultaneous optimisation of multiple quality characteristics in manufacturing processes using Taguchi's quality loss function. *Int J Adv Manuf Tech.* 2001;17:134–138.

Manuscript received Jan. 10, 2008, and revision received July 15, 2008.