ARTICLE

Improving the Quality of Response Surface Analysis of an Experiment for Coffee-Supplemented Milk Beverage: II. Heterogeneous Third-Order Models and Multi-response Optimization

Sungsue Rheem[1*], Insoo Rheem[2], and Sejong Oh[3*]

[1]Graduate School of Public Administration, Korea University, Sejong 30019, South Korea
[2]Department of Laboratory Medicine, Dankook University Hospital, Cheonan 31116, South Korea
[3]Division of Animal Science, Chonnam National University, Gwangju 61186, South Korea

*Corresponding authors:

Sungsue Rheem
Graduate School of Public Administration, Korea University, Sejong 30019, South Korea
E-mail: rheem@korea.ac.kr
Phone: +82-10-8725-7124

Sejong Oh
Division of Animal Science, Chonnam National University, Gwangju 61186, South Korea
E-mail: soh@jnu.ac.kr
Phone: +82-62-530-2116

ABSTRACT

This research was motivated by our encounter with the situation where the quality of response surface analysis of an experiment was so poor that seemingly unreliable modeling and optimization results were presented. Such a situation took place in a research to optimize manufacturing conditions for improving storage stability of coffee-supplemented milk beverage by using response surface methodology, where two responses are Y_1 = particle size and Y_2 = zeta-potential, two factors are F_1 = speed of primary homogenization (rpm) and F_2 = concentration of emulsifier (%), and the optimization objective is to simultaneously minimize Y_1 and maximize Y_2. For response surface analysis, practically, the second-order polynomial regression model is almost solely used. But, there exists the cases in which the second-order model fails to provide a good fit, to which remedies are seldom known to researchers. Thus, as an alternative to a failed second-order model, we present the heterogeneous third-order model, which can be used when the experimental plan is a two-factor central composite design having -1, 0, and 1 as the coded levels of factors. And, for multi-response optimization, we suggest the modified desirability function technique. Using these two methods, we have obtained response surface models with improved fits and multi-response optimization results with predictions better than those in the previous research; our predicted minimum of Y_1 is 183.4, which is lower than the previous observation, and our predicted maximum of Y_2 is 30.93, which is higher than the previous observation. Our predicted optimum combination of conditions are found to be (F_1, F_2) = (5,000, 0.295), which is different from the previous combination. This research is expected to help improve the quality of response surface analysis in experimental sciences including food science of animal resources.

Keywords: Response surface methodology, central composite design, heterogeneous third-order model, multi-response optimization, desirability

INTRODUCTION

Response surface methodology (RSM) is a set of statistical techniques for modeling and optimizing responses through the design and analysis of experiments (Myers et al, 2009), which has been widely used in engineering, agriculture, life science, microbiology and food sciences. A search by Google Scholar revealed that the number of scientific articles whose titles mentioned ‘response surface’ was 157 in 2000 but it became 1,860 in 2018, which is an 11.8-fold increase during recent 18 years. This indicates that RSM has been established as an important tool for modeling and optimization in experimental sciences including food sciences of animal resources.

In RSM, the central composite designs (CCD, Box and Wilson, 1951) have been most frequently used as experimental plans, and the second-order polynomial regression models have been usually employed for data analysis. And, the response surface model can be said to be well fitted and reliable when it satisfies the following criteria: (1) the model is significant (the model p-value $\leqq$ 0.05), (2) the lack of fit is non-significant (the lack-of-fit p-value > 0.05), (3) the R-square $\geqq$ 0.9 (Giunta, 1997), and (4) the adjusted R-square $\geqq$ 0.8 (Myers et al, 2009).

However, in reality, it is observed that, for some data, the analysis models, which are the second-order models in most cases, do not satisfy the above criteria. A remedy in this case is to use a third-order model that consists of linear, quadratic, cubic, and relevant interaction terms (Rheem and Rheem, 2012). For example, when there are two factors, letting X_1 and X_2 denote coded factors, the third-order model has the following terms: linear terms X_1 and X_2, quadratic terms X_1^2 and X_2^2, cubic terms X_1^3 and X_2^3, and the two-factor interaction term X_1X_2. There exist the cases where this method improves the model. But, this method is applicable to a CCD that uses five values, which are denoted by $-\alpha$, -1, 0, 1, and α, as the levels of coded factors.

When the experimental design is a CCD in which -1, 0, and 1 are the levels of coded factors, as in Tables 1, cubic terms cannot be added to the model, since $(-1)^3 = -1$, $(0)^3 = 0$, and $(1)^3 = 1$, which makes the cubic terms equal to the linear terms. For example, in Table

1B, we can see that $X_1=X_1^3$ and $X_2=X_2^3$, and thus the X_1^3 and X_2^3 terms cannot be chosen from among the candidates of additional model terms for augmenting the second-order model..

This problem can be solved by adding the terms of the interaction between the linear term of one factor and the quadratic term of another factor. For example, in Table 1B, $X_1^2X_2$ and $X_1X_2^2$ are such interaction terms. The model that contains such interaction terms can be named a *heterogeneous third-order model*, since the sum of the exponents in each of such interaction terms is three. Thus, a remedy in this case is to augment the second-order model to the heterogeneous third-order model by adding the $X_1^2X_2$ and $X_1X_2^2$ terms, which are chosen from among the candidates of additional model terms in Table 1B, to the second-order model.

A dataset, which is obtained through the screening of the data in Ahn et al (2017), will be re-analyzed for the illustration of the remedy suggested in this research note. Since Ahn et al (2017) has two responses and a purpose of it is the multi-response optimization of them, this research note, which is a continuation of Rheem and Oh (2019), will model both responses by using heterogeneous third-order models, and optimize them simultaneously by employing the desirability function technique.

MATERIALS AND METHODS

Dataset to be re-analyzed. Data analysis should include data screening, which is necessary for accurate modeling. The original data to be used for re-analysis is the data described in Ahn et al. (2017), in which they tried to optimize manufacturing conditions for improving storage stability of coffee-supplemented milk beverage by using response surface methodology. Through data screening, one outlier was deleted from their data (Rheem and Oh, 2019). The response variables, Y_1 and Y_2, and the factors in this experiment are described in Table 1A. The dataset from which an outlier is eliminated is given in Table 1B. Here, the experimental design is a CCD for two factors with the coded levels of -1, 0, and 1. Using this data, we will fit to the data second-order models and heterogeneous third-order models.

Statistical analysis. Data were analyzed by the use of SAS software. SAS/STAT (2013) was employed for the statistical modeling of data. Graphs were produced by SAS/GRAPH (2013).

RESULTS AND DISCUSSION

Fitting the second-order model to the data. First, for each of Y_1 and Y_2, the second-order polynomial regression model containing 2 linear, 2 quadratic, and 1 interaction terms was fitted to the data by using RSREG procedure of SAS/STAT.

For both Y_1 and Y_2, the second-order models are unsatisfactory. For Y_1, the model is non-significant ($p=0.5962 > 0.05$), the lack of fit is significant ($p=0.0131 < 0.05$), the R-square = 0.40 < 0.9, and the adjusted R-square = -0.11 < 0.8. Also, for Y_2, the model is non-significant ($p=0.2924 > 0.05$), the lack of fit is significant ($p=0.0203 < 0.05$), and the R-square = 0.57 < 0.9, and the adjusted R-square = 0.21 < 0.8. None of the four criteria are met for both Y_1 and Y_2. Thus, we will augment the analysis models for their improvement.

Fitting the heterogeneous third-order model to the data. For each of Y_1 and Y_2, since the second-order model has a poor fit for the data, next we will fit to the data a heterogeneous third-order model that consists of the X_1, X_2, X_1^2, X_2^2, X_1X_2, $X_1^2X_2$, and $X_1X_2^2$ terms, by adding the $X_1^2X_2$ and $X_1X_2^2$ terms to the second-order model, in the anticipation of a possible improvement in modeling.

For both Y_1 and Y_2, the heterogeneous third-order models are satisfactory. For Y_1, the model is significant ($p=0.0243 < 0.05$), the lack of fit is non-significant ($p=0.1276 > 0.05$), the R-square = 0.94 > 0.9, and the adjusted R-square = 0.84 = 0.8. Also, for Y_2, the model is significant ($p=0.0371 < 0.05$), the lack of fit is non-significant ($p=0.0820 > 0.05$), and the R-square = 0.93 > 0.9, and the adjusted R-square = 0.80 $\geqq$ 0.8. All of the four criteria are satisfied for both Y_1 and Y_2.

Thus, we accept these models as our final models. Letting $\hat{Y}_1$ and $\hat{Y}_2$ denote the predicted values of Y_1, and Y_2, we specify our heterogeneous third-order models as

$$\hat{Y}_1 = b_0 + b_1 X_1 + b_2 X_2 + b_{11} X_1^2 + b_{22} X_2^2 + b_{12} X_1 X_2 + b_{112} X_1^2 X_2 + b_{122} X_1 X_2^2$$

and

$$\hat{Y}_2 = c_0 + c_1 X_1 + c_2 X_2 + c_{11} X_1^2 + c_{22} X_2^2 + c_{12} X_1 X_2 + c_{112} X_1^2 X_2 + c_{122} X_1 X_2^2$$

where the coefficients b_1, b_2, …, b_{122} and c_1, c_2, …, c_{122} are given in Table 2A and Table 2B.

Drawing the 3D plots of the response surface. Each of the three-dimensional (3D) response surface plot was drawn with the vertical axis representing the predicted response and two horizontal axes indicating the two explanatory factors. Plots A and B in Figure 1 are the 3D response surface plots for the effects of the two actual factors on the two predicted responses.

Multi-response optimization of two responses. In Ahn et al (2017), the optimization objective was to minimize Y_1 (particle size) and maximize Y_2 (zeta-potential) simultaneously. For this multi-response optimization, we modified the desirability function technique of Derringer and Suich (1980). In this modified technique, first, we define the desirability function for the minimization of Y_1 as

$$D_1 = [\text{Maximum}(\hat{Y}_1) - \hat{Y}_1)] / [\text{Maximum}(\hat{Y}_1) - \text{Minimum}(\hat{Y}_1)],$$

and define the desirability function for the maximization of Y_1 as

$$D_2 = [\hat{Y}_2 - \text{Minimum}(\hat{Y}_2)] / [\text{Maximum}(\hat{Y}_2) - \text{Minimum}(\hat{Y}_2)].$$

Here, for $\hat{Y}_1$, when $\hat{Y}_1$ is minimized, D_1 becomes 1; otherwise $0 \leqq D_1 < 1$, and for $\hat{Y}_2$, when $\hat{Y}_2$ is maximized, D_2 becomes 1; otherwise $0 \leqq D_2 < 1$. Now, we define CD, which means the composite desirability, as

$$CD = (D_1 D_2)^{(1/2)}$$

which is the geometric mean of D_1 and D_2. Then, we find the combination of the values of X_1 and X_2 that maximizes CD. This combination is the optimum point of (X_1, X_2). Now, by converting this optimum point to the combination of the levels of the actual factors, we achieve the multi-response optimization of minimizing Y_1 and maximizing Y_2 simultaneously.

For the minimization and maximization of Y_1 and Y_2 and the maximization of CD, we performed the searches on a grid (Oh et el, 1995). First, we obtained Minimum $(\hat{Y}_1)$ = 170.8131135, Maximum $(\hat{Y}_1)$=221.6698750, Minimum $(\hat{Y}_2)$=24.7334750, and Maximum $(\hat{Y}_2)$=35.2957228, and using these values, we implemented our modified desirability function technique that maximizes the composite desirability defined above. Plot C in Figure 1 shows the 3D surface plot of the composite desirability function for our multi-response optimization. Table 2C presents the results of our multi-response optimization.

In Ahn et al (2017), at their optimum point, their Y_1 value was 190.1 and their Y_2 value was -25.94±0.06, whereas, at our predicted optimum point, the predicted Y_1 was 183.4 and the predicted Y_2 was 30.93. We can see that our predicted minimum of Y_1 is smaller than their observed Y_1, and our predicted maximum of Y_2 is greater than their observed Y_2. Their optimum conditions for their multi-response optimization were F_1 = speed of primary homogenization (rpm) = 5,000 and F_2 = concentration of emulsifier (%) = 0.2071, whereas our optimum conditions are F_1 = 5,000 and F_2 = 0.295. We can see that our predicted combination of optimum factor levels is different from theirs. A validation experiment will be needed to verify the result of multi-response optimization obtained by the method proposed in this article.

CONCLUSION

This article suggests the use of the heterogeneous third-order model for better modeling and a modified desirability function technique for multi-response optimization. The heterogeneous third-order model can be used when (1) the experimental design is a two-factor central composite design having -1, 0, and 1 as the coded levels of factors, and (2) a second-order model fails to provide a good fit for the data. How to construct the heterogeneous third-order model is to use X_1, X_2, X_1^2, X_2^2, X_1X_2, $X_1^2X_2$, and $X_1X_2^2$ as model terms. A modified desirability function technique first defines a desirability function for each response according to each optimization objective, and then finds out the combination of factor levels that maximizes the geometric mean of the values from desirability functions for multiple responses. An illustrative new analysis of the data from a previous research has produced statistical models with better fits and optimization results with better predictions. This suggestion is expected to help enhance the quality of response surface analyses of the experiments in food science of animal resources.

ACKNOWLEFGMENTS

This work was financially supported by the Graduate School of Public Administration at Korea University in 2018. And also this work was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Minister of Education, Science, and Technology (NRF-2016R1A2B4007519).

REFERENCES

Ahn, S; Park, J; Kim, J; Oh, D; Kim, M; Chung, D; Jhoo, J; and Kim, G. (2017). Optimization of Manufacturing Conditions for Improving Storage Stability of Coffee-

Supplemented Milk Beverage Using Response Surface Methodology. *Korean Journal for Food Science of Animal Resources.* 37(1): 87-97.

Box, G. E. P. and Wilson, K. B. (1951), On the Experimental Attainment of Optimum Conditions, *Journal of the Royal Statistical Society, Series B*, 13: 1–45.

Derringer, D. and Suich, R. (1980), "Simultaneous Optimization of Several Response Variables," *Journal of Quality Technology*, Vol. 12, pp. 214-219.

Giunta, A. A. (1997). “Aircraft multidisciplinary design optimization using design of experiments theory and response surface modeling methods.” Ph.D. thesis, Virginia Polytechnic Institute and State University.

Myers, R. H., Montgomery, D. C., and Anderson-Cook, C. M. (2009). *Response Surface Methodology: Process and Product Optimization Using Designed Experiments*. 3rd edition, John Wiley & Sons.

Oh, S., Rheem, S., Sim, J., Kim, S. and Baek, Y. (1995). Optimizing conditions for the growth of *Lactobacillus casei* YIT 9018 in tryptone-glucose medium by using response surface methodology. *Applied Environmental Microbiology*, 61(11): 3809-3814.

Rheem, S and Oh, S. (2019). Improving the Quality of Response Surface Analysis of an Experiment for Coffee-Supplemented Milk Beverage: I. Data Screening at the Center Point and Maximum Possible R-Square. *Korean Journal for Food Science of Animal Resources.* 39(1): 653-659.

Rheem, I. and Rheem, S. (2012). Response Surface Analysis in the Presence of the Lack of Fit of the Second-Order Polynomial Regression Model. *Journal of the Korean Data Analysis Society*, 14(6): 2995–3002. (in Korean)

SAS Institute Inc. (2013). *SAS/STAT user's guide, release 6.04*. SAS Institute, Inc., Cary, NC, USA.

SAS Institute Inc. (2013). *SAS/GRAPH user's guide, release 6.04*. SAS Institute, Inc., Cary, NC, USA.

Table 1. Response variables, actual and coded factors, experimental design, and response data

A. Response variables, actual and coded factors, and the levels of the factors

Response variables = Y_1, Y_2	Actual factor	Coded factor	Actual factor level corresponding to the coded factor level of		
			-1	0	1
Y_1 = Particle size	F_1 = Speed of primary homogenization (rpm)	X_1	5,000	10,000	15,000
Y_2 = Zeta-potential	F_2 = Concentration of emulsifier (%)	X_2	0.1	0.2	0.3

B. Experimental design and response data with candidates of additional model terms

Experimental design in coded levels and response data						Candidates of additional model terms for augmenting the 2nd-order model			
Standard order	Design point	X_1	X_2	Y_1	Y_2	X_1^3	X_2^3	$X_1^2X_2$	$X_1X_2^2$
1	1	-1	-1	179.900	27.5000	-1	-1	-1	-1
2	2	-1	1	178.267	29.9667	-1	1	1	-1
3	3	1	-1	179.533	24.3000	1	-1	-1	1
4	4	1	1	219.767	32.5666	1	1	1	1
5	5	-1	0	217.867	36.1000	-1	0	0	0
6	6	1	0	178.367	28.2667	1	0	0	0
7	7	0	-1	185.333	29.1000	0	-1	0	0
8	8	0	1	182.167	28.2000	0	1	0	0
9	9	0	0	186.433	30.8300	0	0	0	0
10	9	0	0	181.933	29.0667	0	0	0	0
11	9	0	0	175.633	29.6000	0	0	0	0
12	9	0	0	180.333	29.1000	0	0	0	0

Table 2. Results of modeling and optimization

A. Coefficient estimates in the heterogeneous 3rd-order model on Y_1

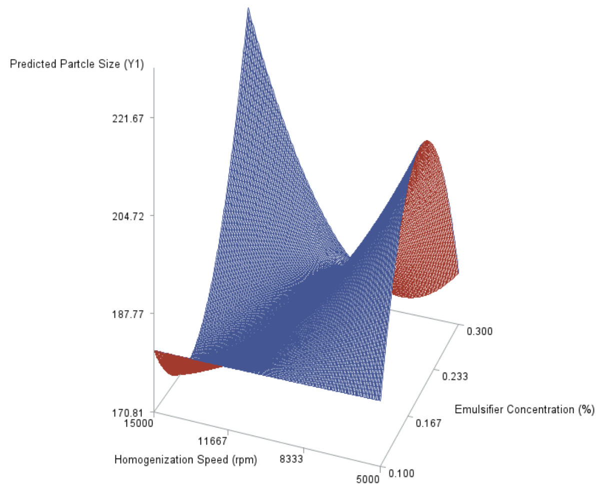
Term	Parameter estimate	Standard error	t-value	p-value
Intercept	b_0=182.99	2.76	66.24	<0.0001
X_1	b_1=-19.75	4.28	-4.62	0.0099
X_2	b_2=-1.58	4.28	-0.37	0.7302
X_1^2	b_{11}=-11.33	3.71	3.06	0.0378
X_2^2	b_{22}=-3.04	3.71	-0.82	0.4579
X_1X_2	b_{12}=10.47	3.03	3.46	0.0258
$X_1^2X_2$	b_{112}=11.23	5.24	2.14	0.0987
$X_1X_2^2$	b_{122}=30.03	5.24	5.73	0.0046

B. Coefficient estimates in the heterogeneous 3rd-order model on Y_2

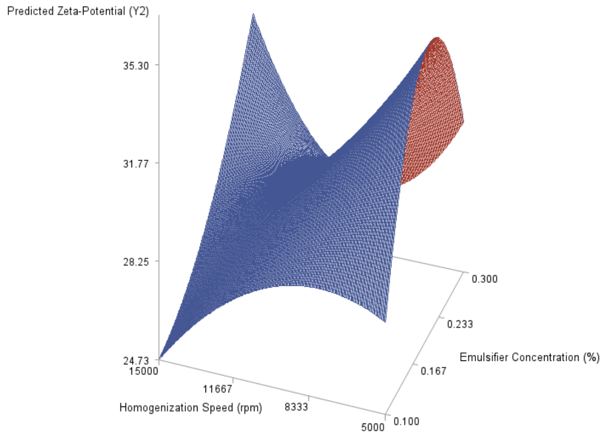
Term	Parameter estimate	Standard error	t-value	p-value
Intercept	c_0=30.08	0.58	51.52	<0.0001
X_1	c_1=-3.92	0.90	-4.33	0.0124
X_2	c_2=-0.45	0.90	-0.50	0.6450
X_1^2	c_{11}=1.23	0.78	1.57	0.1904
X_2^2	c_{22}=-2.30	0.78	-2.94	0.0426
X_1X_2	c_{12}=1.45	0.64	2.27	0.0860
$X_1^2X_2$	c_{112}=3.13	1.11	2.83	0.0474
$X_1X_2^2$	c_{122}=3.77	1.11	3.40	0.0273

C. Results of multi-response optimization

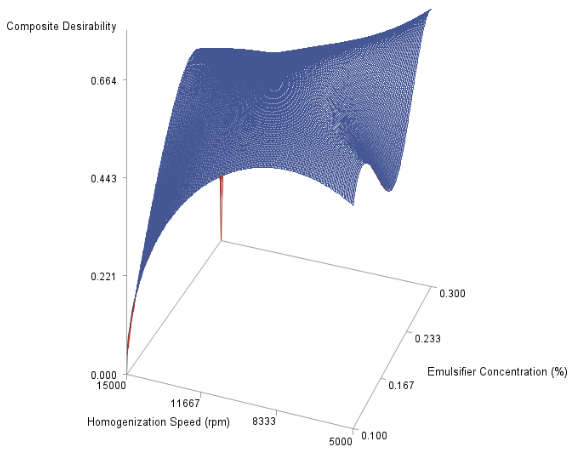
X_1	X_2	F_1 = Speed of primary homogenization (rpm)	F_2 = Concentration of emulsifier (%)	Predicted Minimum of Y_1 = Particle size	Predicted Maximum of Y_2 = Zeta-potential	D_1	D_2	CD = Composite Desirability
-1	0.95	5,000	0.295	183.4	30.93	0.752	0.587	0.664



Plot A. 3D surface plot of the predicted response Y_1



Plot B. 3D surface plot of the predicted response Y_2



Plot C. 3D surface plot of the composite desirability function

Fig. 1. 3D surface plots of predicted responses and the composite desirability function