OPTIMIZING THE PROCESS OF OBTAINING MELAMINE FORMALDEHYDE RESIN USING THE BOX-WILSON METHOD

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Abstract
The Box-Wilson method of mathematical planning of experiments was used in order to study the influence coefficients of the factors influencing the process of melamine synthesis on the course of the reaction.

Keywords: chemical ingeneering, melamine, formaldehyde, mathematic planning, Box-Wilson.

Introduction

Based on the a priori data, according to the results obtained for each of the factors, all the factors involved in the synthesis of melamine formaldehyde resin were selected. Also, for them, the main stages and intermediate intervals of changing stages were determined. Information on the intervals is presented in Table 1 below.

Table 1 Factors affecting the process of melamine synthesis and their intervals of change

Levels of factors	Factors			
	X_1	X_2	X_3	X_4
The top step	90,0	20,0	120	95
Middle stage	80,0	25,0	90	93
Lower step	80,0	30,0	60	91
Change intervals	10,0	5,0	30	2
Unit of measure	%	%	min	°C

Among the factors involved in the process of melamine synthesis, the following main ones were selected:

1) X_1 – amount of melamine relative to formalin, %;
2) X_2 – concentration of the sodium hydroxide solution introduced into the pitching process, %;
3) X_3 – process duration, minutes;
4) X_4 – process temperature, °C.

Four selected factors were ranked in two, ie, a full factorial experiment of type 2^4 was planned. For this, 2 split replicates were used. Using the 2^{4-1} type planning method of the replica from the full factorial experiment of type 2^4, we obtained the following generation ratios:

$$X_4 = X_1 \cdot X_2 \quad (1)$$

As a result of preliminary calculations of factors in the process of melamine synthesis, a planning matrix of experiments was developed and the calculations were continued. The results are shown in Table 2.

Table 2 Experiment planning matrix and results based on it and their values

Experiment number	Factor code					Y_1	Y_2	$Y_{ўр}$
	X_0	X_1	X_2	X_3	X_4			
1	+	+	+	+	+	83,101	79,402	81,251
2	+	–	+	+	–	65,312	60,203	62,771
3	+	+	–	+	–	61,513	64,919	65,216
4	+	–	–	+	+	59,603	62,105	60,854
5	+	+	+	–	+	68,905	65,301	67,103
6	+	–	+	–	–	38,604	43,503	41,053
7	+	+	–	–	–	45,807	49,801	47,804
8	+	–	–	–	+	48,711	46,108	47,409

It can be seen from Table 2 that a total of 8 experiments were carried out on the basis of a previously prepared matrix, in which the steps determined for each selected factor were used and coded with "+" and "–" signs in the matrix. Also, if all factors take the maximum values in the X0 state, the "+" and "-" signs in X1 are placed one after the other according to the order number of the experiments, in the X2 state, the "+" and "-" signs are in pairs of 2, one pair of signs is the second sign alternates with a pair, in X3 there are four "+" codes from above and four "–" codes, and in X3 there is a "+" sign in experiment No. 1, a pair of negative and positive signs is placed in the sequence, and at the end there is another "+" in the odd position ” is indicated. This, in turn, represents the upper and lower values of the range of changes. For example, in one experiment, the amount of melamine compared to formalin was 90%, the concentration of the sodium hydroxide solution introduced into the pitching process was 30%, the duration of the process was 2 hours, at a temperature of 94ºC, while in another experiment, the amount of melamine compared to formalin was 70%, the concentration of the sodium hydroxide solution introduced into the pitching process 20%, process duration 1 hour, performed under conditions with temperature indicators of 91ºC.

In this direction, synthesis experiments were carried out and expressed in the form of the following regression equation:

$$Y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4\,; \quad (2)$$

where b0, b1, b2, b3, b4 are the regression coefficients of the incomplete quadratic equation.

The studied chemical changes can also be expressed in the form of a direct linear relationship within the defined change intervals. Also, regression coefficients can be determined using the method of least squares. Traditionally, regression coefficients are determined by the following formula:

$$b_i = \frac{\sum_{i=1}^{n}(Y_{ij} \cdot Y_i)}{N} \; ; \quad (3)$$

here i – serial number of the experiment on the synthesis of melamine formaldehyde resin (from 1 to 8);

j – ordinal number of the factor affecting the synthesis process (1, 2, 3, 4);

X_{ij} – factor coding symbol;

N – the number of experiments on the matrix.

We calculated the significance of the coefficients of the regression equation by putting the results of the matrix construction and preliminary calculations based on it into formula 3. As a result, b_0 = 58.931; b_1 = 5.9102; b_2 = 4.1101; b_3 = 8.0902; b_4 = 5.2301 values were generated.

Table 3 Statistical analysis of the mathematical model developed for the chemical process of melamine formaldehyde resin synthesis

Y_1	Y_2	$Y_{ўр}$	ΔY_i	ΔY_i^2	S_i^2	$Y_{хис}$	$\Delta Y_i'$	$(\Delta Y_i')^2$
83,101	79,402	81,251	1,851	3,4225	6,8451	82,263	–1,01	1,025
65,312	60,203	62,771	2,552	6,5025	13,005	59,988	2,761	7,631
61,513	64,919	65,216	–1,701	2,8901	5,7803	63,588	–0,391	0,015
59,603	62,105	60,854	–1,251	1,5625	3,1251	62,213	–1,362	1,856
68,905	65,301	67,103	1,801	3,2403	6,4807	66,088	1,012	1,025
38,604	43,503	41,053	–2,452	6,0025	12,005	43,813	–2,76	7,631
45,807	49,801	47,804	–2,001	4,0008	8,0009	47,413	0,393	0,150
48,711	46,108	47,409	1,302	1,6901	3,3812	46,038	1,361	1,856

By inserting the calculated "b" values, that is, the coefficients, into formula 2, it was possible to obtain a first-order regression equation that looks like this:

$$Y = 58{,}931 + 5{,}9102\, X_1 + 4{,}1101\, X_2 + 8{,}0902\, X_3 + 5{,}2301\, X_4 \quad (3.4)$$

In this way, a mathematical model of the chemical process for the synthesis of melamine-formaldehyde resin was obtained. The data were statistically processed to determine the appropriateness of the obtained mathematical model for the chemical process and to check its validity. The results are shown in Table 3.

Mathematical variance was used to determine variation values for repeated experiments. The variance is calculated using the following formula, which is the most convenient and correct for this method of mathematical planning:

$$S_i^2 = \frac{\sum_{q=1}^{n}(Y_q - Y_{cp})}{n-1} \; ; \; (5)$$

here Y_q – the result of a separate experience;

$Y_{ўр}$ – arithmetic mean value;

(*n* – 1) and is the number corresponding to the degree of freedom, which corresponds to the number of repeated experiments minus one.

For two repeated experiments, formula 5 is modified in the prescribed manner and it looks like this:

$$S_i^2 = \frac{2\Delta Y^2}{1}\;; \qquad (6)$$

In order to make sure that the variance is primary during the calculations, it is necessary to carry out a series of calculations according to the Cochrane criterion. The Cochrane criterion is a formula that looks like this:

$$G_{\exp} = \frac{S_{\max}^2}{\sum_{i=1}^{N} S_i^2}\;; (7)$$

$G_{cri} = 0{,}679$ $\qquad$ $G_{exp} = 0{,}280$

$$G_{exp} < G_{мез}$$

The obtained results fully correspond to the requirements imposed by the formula 3.7. This, in turn, indicates that the variance is primary.

In order to check the proportionality of the mathematical model obtained on this basis for the applied chemical process, in short, to test the adequacy of the mode, first of all it is necessary to check the proportionality of the dispersion underlying it. The following formula is used for this:

$$S_{ад}^2 = \frac{\sum_{i=1}^{N}(\Delta_i^{'})}{f}\;; (8)$$

After doing this and making sure of the proportionality, Eq. comes from (Table 3).

Then, based on the results obtained,

$$\Delta Y'_i = Y_{cp} - Y_{cou};\ (9)$$

$\Delta Y'_i$ was found according to the formula.

After this action is also successfully performed, the effectiveness of the dispersion is determined. The following formula is useful for determining the effectiveness of dispersion:

$$S_y^2 = \frac{\sum_{i=1}^{N}\sum_{q=1}^{n}(Y_{iq} - Y)^2}{N(n-1)}\;; (10)$$

Here $i = 1, 2, \ldots, N$

$q = 1, 2, \ldots, n$ accepts values.

For two replicate experiments, Formula 10 above takes the following form:

$$S_y^2 = \frac{2\sum_{i-1}^{N}(Y_{iq} - Y)^2}{N} = \frac{\sum_{i-1}^{N} S_i^2}{N}\;; (11)$$

Result: $S_y^2 = \frac{58,62}{8} = 7,3275$

The variance of the ratio for the process is calculated using the following formula:

$$S_{ad}^2 = \frac{n\sum(Y_{cp} - Y_{pac})^2}{N - q}; \quad (12)$$

Here $q = K + 1$;

K – number of regression coefficients.

Result: $S_{ad}^2 = \frac{2 \cdot 21,3263}{8-(4+1)} = \frac{42,6526}{3} = 14,2175$

Fisher's test was used to test the proportionality of such models. Fisher's criterion is derived as follows:

$$F_{экс} = \frac{S_{ad}^2}{S_y^2}; \quad (13)$$

For $f_1 = 2$, $f_2 = 8$ will be equal to F_{tab} (2.11) = 4,5.

In this case, $(F_{экс} = \frac{14,2175}{6,81} = 2,088) < (F_{tab} = 4,5)$. It follows that the obtained mathematical model was proved to be proportional, that is, adequate for the process.

To check whether the regression coefficients are significant or insignificant, it is necessary to find the variance of the coefficients. For this, it is appropriate to use the following formula:

$$S_{bi}^2 = \frac{S_y^2}{N} = \frac{7,3275}{8} = 0,9159 \quad (14)$$

And from that $S_{bi} = \pm\sqrt{S_{bi}^2} = \sqrt{0,9159} = 0,95$ is caused.

Then, $\Delta b_i = t\, S_{bi}$ the confidence interval corresponding to the expression is determined. Here, t – is the tabular value of the Student's criterion, which corresponds to the number of degrees of freedom, with the help of which the selected level of significance is determined. This value is usually equal to 0.05. And Sbi is the squared error of the regression coefficient.

$$\Delta t_{cri} = 3,18$$

$$\Delta b_i = 3,18 \cdot 0,95 = 3,02$$

If the absolute value of the coefficient is greater than the confidence interval, then the coefficient developed for the factor involved in chemical processes is significant for this chemical process. The results are presented in Table 4.

Table 4 Value and significance of the regression coefficients determined for the factors involved in the process of melamine synthesis

b_i	Values	Hint	Value of Δb_i	Results
b_0	31,2412	>	3,02	The coefficient is significant
b_1	5,98008	>	3,02	The coefficient is significant
b_2	5,64011	>	3,02	The coefficient is significant
b_3	8,47102	>	3,02	The coefficient is significant
b_4	9,93051	>	3,045	The coefficient is significant

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From the table 4 above, it can be seen that all the factors involved in the synthesis of melamine formaldehyde are important, and changing the factors to one degree or another can have a positive or negative effect on the production of melamine formaldehyde.

Conclusion

Thus, the most optimal conditions for obtaining melamine-formaldehyde resin are 1:0.8 ratio of formalin and melamine, use of 25% solution of NaOH introduced into the process, setting the time in the range of 1.5-2 hours and ensuring the temperature at 92-94ºC. scheduled to arrive.

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