Application of the Taguchi method to optimize the strength level of dispersed-filled polymers with reduced combustibility

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Abstract. The article considers optimization of strength level of dispersed-filled polymer materials based on polyurethane BASF with addition of perlite powder and aluminum hydroxide of different dispersion degree. The Taguchi experiment was chosen as a method of investigation. The experiment plan includes 7 controlled factors and 2 interference factors. An optimal combination of factors is determined to provide a maximum signal-to-noise ratio and a minimum variation coefficient. Controlled factors are ranked by degree of influence on sensitivity of tensile strength to interference. The applicability of the Taguchi method to optimize the strength of dispersed-filled polymers has been confirmed.

1. Introduction

At present, it is a pressing task to develop and research functional materials that have a unique level of not one, but several service characteristics. Similar objects include light filled polymers [1] with reduced combustible properties used in various industries. Practical realization of these materials is possible in the form of two-layer systems, at the same time the first layer (substrate) has low density, and the second layer prevents ignition and development of material combustion process. Specialized powder fillers are added to the polymers to obtain these properties. The main characteristic determining the quality and reliability of filled polymers is mechanical strength [2].

The tensile strength of any type of filled polymer depends on a variety of design and technological factors. If it is necessary to take into account all the influencing values, the task of obtaining an adequate mathematical model using classical methods of experiment planning becomes difficult to implement in practice due to the need to carry out a large number of experiments. For such cases, it is more effective to use a special optimization tool.

The aim of the study is to evaluate the possibility of using the Taguchi experiment planning method [3] to investigate and optimize the mechanical strength level of filled polymers. The main objectives of the study are: selection of the investigated factors, preparation of the plan of the experimental study and its implementation, processing of the results of the experiment and determining the optimal combination of factor levels at which the strength level of materials is close to maximum with a minimum spread of values.

2. Materials and methods

Samples of filled polymers based on BASF base-component polyurethane (the main component of the Elasturan 6010/EX1 brand) with the addition of perlite powders and aluminum hydroxide are the objects
of this study. Perlite provides the substrate facilitation, and aluminum hydroxide introduced into the outer layer of the material belongs to the class of flame retardant substances. Tensile strength is chosen as the studied characteristic [4]. Tests for strength evaluation were carried out on an electromechanical breaking machine. Statistical planning of the Taguchi experiment is chosen as a study method.

The main stages of the Taguchi experiment [5, 6] are:

- Selection of design and interference factors.
- Selection of suitable design and interference matrices.
- Selection of output statistics.
- Carrying out an experiment according to the plan.
- Processing the results of the experiment:
  - Calculation of output statistics.
  - Calculation of main effects of factors.
  - Ranking and determination of optimal levels of factors.

The difference of Taguchi approach to experiment planning is that the result is not a mathematical model of dependence of the investigated parameter on the considered influencing factors, but a set of levels of controlled factors, at which the parameter tends to the optimal value (specific value, minimum or maximum) and at the same time the variation of its values is minimal. This is achieved by considering the second group of interference factors and analyzing the specific signal-to-noise ratio parameter.

After analysis of filled polymers production technology, 7 factors were determined, which have most influence on strength. They are selected as control factors, their names with variation levels are summarized in the list:

- (X1) Filler type - 2 levels (perlite and aluminum hydroxide)
- (X4) Curing agent fraction, % by weight - 2 levels (30 and 36);
- (X5) Mechanical-chemical processing – 2 levels (yes/no);
- (X6) Presence of plasticizer – 2 levels (yes/no);
- (X7) Mixing time, min - 2 levels (5 and 10 minutes)
- (X2) Filler ratio, % by weight – 3 levels (50, 60 and 70);
- (X3) Filler dispersion, μm - 3 levels (groups I, II and III);
  - I group – 1-5 μm;
  - II group – 10-40 μm;
  - III group – 50-100 μm.

Two interference factors were chosen: number of samples group (1 or 2), in each of which average value of strength of materials was determined, and deviation of mass percentage of content of filler in mixture from nominal value (-5% and +5%).

The experiment plan based on the controlled factors highlighted above can be designated by 32x25 because there are 3 two-level and 5 two-level factors. Taking into account recommendations [6] we select orthogonal matrix L12 given in table 1. In all columns of the matrix with the same number of factor levels, each level is repeated the same number of times (4 times for two-level and 3 times for 3-level).

| № of experiment | X1 | X2 | X3 | X4 | X5 | X6 | X7 |
|-----------------|----|----|----|----|----|----|----|
| 1               | 1  | 1  | 1  | 1  | 1  | 1  | 1  |
| 2               | 1  | 1  | 1  | 2  | 1  | 1  | 2  |
| 3               | 1  | 1  | 2  | 1  | 2  | 1  | 3  |
The interference matrix contains only 2 two-level factors and is given in table 2. The complete set of combinations of factor levels consists of 4 combinations.

### Table 2. Interference matrix.

| № of experiment | X1 | X2 |
|------------------|----|----|
| 1                | 1  | 1  |
| 2                | 1  | 2  |
| 3                | 2  | 1  |
| 4                | 2  | 2  |

Before carrying out the experiment according to the formed plan, it is necessary to determine the compositions of the samples. In this case, it is necessary to provide a required condition for combining each of the 12 rows of the planning matrix with each row of the interference matrix. Therefore, the total number of combinations is 48. The view of the table with the compositions of samples № 1–24 is given in table 3.

### Table 3. Samples compositions for Taguchi experiment.

| №  | X1  | X2 | X3 | X4 | X5 | X6 | X7 | Samples group |
|----|-----|----|----|----|----|----|----|---------------|
| 1  | perlite | 30  | Yes | yes | 5  | 45 | I  | 1            |
| 2  | perlite | 30  | Yes | yes | 5  | 55 | I  | 1            |
| 3  | perlite | 30  | Yes | yes | 5  | 45 | I  | 2            |
| 4  | perlite | 30  | Yes | yes | 5  | 55 | I  | 2            |
| 5  | perlite | 30  | Yes | no  | 5  | 45 | II | 1            |
| 6  | perlite | 30  | Yes | no  | 5  | 55 | II | 1            |
| 7  | perlite | 30  | Yes | no  | 5  | 45 | II | 2            |
| 8  | perlite | 30  | Yes | no  | 5  | 55 | II | 2            |
| 9  | perlite | 30  | No  | yes | 10 | 45 | III| 1            |
| 10 | perlite | 30  | No  | yes | 10 | 55 | III| 1            |
| 11 | perlite | 30  | No  | yes | 10 | 45 | III| 2            |
| 12 | perlite | 30  | No  | yes | 10 | 55 | III| 2            |
| 13 | perlite | 36  | No  | no  | 10 | 45 | I  | 1            |
| 14 | perlite | 36  | No  | no  | 10 | 55 | I  | 1            |
| 15 | perlite | 36  | No  | no  | 10 | 45 | I  | 2            |
| 16 | perlite | 36  | No  | no  | 10 | 55 | I  | 2            |
| 17 | perlite | 36  | Yes | yes | 5  | 54 | II | 1            |
| 18 | perlite | 36  | Yes | yes | 5  | 66 | II | 1            |
| 19 | perlite | 36  | Yes | yes | 5  | 54 | II | 2            |
| 20 | perlite | 36  | Yes | yes | 5  | 66 | II | 2            |
| 21 | perlite | 36  | Yes | no  | 10 | 54 | III| 1            |
3. Results and discussion

In accordance with table 3, samples of filled tensile polymers were prepared to evaluate the tensile strength of TS as defined by equation (1):

$$TS = \frac{F_{\text{max}}}{A},$$

where $F_{\text{max}}$ – force value at break, H; 
$A$ – cross-sectional area of non-deformed sample, m$^2$.

The results of calculating the strength of the samples are listed in table 4. In this table, the data are presented in a format convenient for further processing - for each of 12 combinations of levels of design factors, 4 combinations of levels of interference factors are considered. Each of the 48 values is the arithmetic average of strength assessments for three samples forming a group of the same composition.

**Table 4. The results of strength assessments of samples of filled polymers.**

| №  | Combination of levels of main factors | Average tensile strength TS, MPa |
|----|---------------------------------------|----------------------------------|
|    |                                       | Interference factors levels      |
|    |                                       | 11  | 12  | 21  | 22  |
| 1  | 1111111                               | 1.84 | 1.42 | 1.87 | 1.69 |
| 2  | 1112112                               | 1.67 | 1.92 | 2.00 | 1.70 |
| 3  | 1121121                               | 1.38 | 1.50 | 1.18 | 1.07 |
| 4  | 1222111                               | 0.70 | 0.75 | 1.19 | 0.45 |
| 5  | 1211122                               | 1.10 | 0.86 | 1.11 | 0.89 |
| 6  | 1212223                               | 1.43 | 0.96 | 1.23 | 1.19 |
| 7  | 2121221                               | 2.97 | 3.85 | 3.68 | 4.44 |
| 8  | 2122122                               | 2.64 | 3.21 | 3.06 | 3.43 |
| 9  | 2111233                               | 3.73 | 3.06 | 3.78 | 3.35 |
| 10 | 2212131                               | 4.45 | 6.96 | 6.01 | 5.71 |
| 11 | 2221232                               | 3.65 | 3.00 | 3.09 | 3.43 |
| 12 | 2222133                               | 5.05 | 0.97 | 4.72 | 1.36 |

Before calculating the output statistics, it is necessary to calculate the following parameters [7, 8]:

- average of the response $m_i$,

$$m_i = \frac{1}{n} \sum_{j,k} y_{ijk}^i,$$

where $y_{ijk}^i$ - response function value the i-th line for noise levels j and k.

- response dispersion $V_i$,

$$V_i = \frac{1}{n-1} \left( \sum_{j,k} y_{ijk}^i - m_i \right)^2$$

- squared coefficient of variation $C_{vi}$:

$$C_{vi} = \frac{V_i}{(m_i)^2}$$

In the Taguchi method, the signal-to-noise ratio $\eta$ is used as the output statistics. For the case when the largest given value of y is the best, this ratio is determined by the equation:
\[ \eta = -10 \log \frac{1}{n \sum_{j=1}^{n} y_i^2} \]  

Estimates of the values of the listed parameters are summarized in table 5.

| № of experiment | Combination of factors levels | \( m_i \) | \( V_i \) | \( C_{vi}, 10^2 \) | \( \eta, \text{db} \) |
|-----------------|-------------------------------|-----------|--------|-----------------|----------------|
| 1               | Y1111111                       | 1,71      | 0,042  | 1,46            | 4,48           |
| 2               | Y1112112                       | 1,82      | 0,026  | 0,80            | 5,14           |
| 3               | Y11211213                      | 1,28      | 0,037  | 2,28            | 1,94           |
| 4               | Y1222211                      | 0,77      | 0,094  | 15,78           | -3,72          |
| 5               | Y1211122                      | 0,99      | 0,019  | 1,90            | -0,29          |
| 6               | Y1212223                      | 1,20      | 0,038  | 2,63            | 1,32           |
| 7               | Y2121221                      | 3,74      | 0,366  | 2,62            | 11,17          |
| 8               | Y2122212                      | 3,09      | 0,111  | 1,17            | 9,66           |
| 9               | Y2111233                      | 3,48      | 0,115  | 0,95            | 10,73          |
| 10              | Y2212131                      | 5,78      | 1,073  | 3,21            | 14,90          |
| 11              | Y2221232                      | 3,29      | 0,091  | 0,84            | 10,27          |
| 12              | Y2222133                      | 3,03      | 4,655  | 50,86           | 3,75           |

A row with an optimal combination of levels of controlled factors is determined by the following criterion: maximum ratio \( \eta \) with a minimum coefficient of variation. According to the first condition, we have a maximum for 3 rows: 10 (14.9 dB), 7 (11.17 dB) and 9 (10.73 dB). Herewith, for the same rows, the squared variation coefficient is significantly lower for row 9 (0.95 versus 2.62 and 3.21). Therefore, row 9 is optimal according to the indicated criterion.

The final stage of experimental results processing is to calculate the main effects of the controlled factors and determine the combination of their levels that ensure the maximum value of the ratio \( \eta \). The main effects for each \( k \)-th of two (three) levels of factors \( x_j \) are defined as the average values of the ratios \( \eta_i \) over those \( n_k \) rows, in which the factor in question has a given level \( k \) (6):

\[
(\eta)_{x,k} = \frac{1}{n_k} \sum_{i} \eta_i (x_j k) \]  

The final main effect (ME) for two- and three-level factors is determined by formulas (7) and (8):

\[
\eta_{x_j} = |\eta_{x_1} - \eta_{x_2}| \]  

\[
\eta_{x_j} = \max \left\{ |\eta_{x_1} - \eta_{x_2}|, |\eta_{x_1} - \eta_{x_3}|, |\eta_{x_2} - \eta_{x_3}| \right\} \]  

The calculation results of the main effects of the seven controlled factors are summarized in table 6. The ranks of the factors indicate the degree of influence of the factor on the ratio \( \eta \) and increase with decreasing value of the main effect of the factor. Figure 1 shows examples of graphs of the main effects for factors X1 and X7.
Table 6. Main effects of controlled factors.

| Level | X1 | X2 | X3 | X4 | X5 | X6 | X7 |
|-------|----|----|----|----|----|----|----|
| 1     | 1.48 | 7.19 | 5.17 | 6.38 | 6.27 | 1.96 | 6.71 |
| 2     | 10.08 | 6.56 | 5.51 | 5.17 | 5.28 | 5.46 | 6.19 |
| 3     | - | - | - | - | - | 9.91 | 4.44 |
| η \(\eta_{X_j}\), dB | 8.61 | 0.63 | 0.34 | 1.21 | 0.99 | 7.96 | 2.27 |
| Rank | 1 | 6 | 7 | 4 | 5 | 2 | 3 |

From the data in table 6 it follows that the maximum value of \(\eta\) is achieved for processing factors \(X_1\), \(X_2\), \(X_3\), \(X_4\), \(X_5\), \(X_6\), \(X_7\). The greatest influence on the \(\eta\) value is exerted by such controlled factors as the type of filler, its mass fraction in the mixture, and the degree of dispersion of the powders introduced into the polymer.

![Graphs of the main effects of two-level (X1) and three-level (X7) factors.](image)

**Figure 1.** Graphs of the main effects of two-level (X1) and three-level (X7) factors.

### 4. Conclusion
The results of processing the data obtained during the Taguchi experiment determined optimal combinations of levels of seven controlled factors from the point of view of the composite criterion and maximization of the ratio \(\eta\). Three parameters associated with the filler (type, mass fraction, and degree of dispersion) have the greatest influence on the value of \(\eta\) among the controlled factors. This result is fully consistent with previous studies of filled polymers of this type. Thus, the Taguchi technique can be successfully and effectively applied to study the strength of the filled polymers of the discussed type. Further research will aim to assess the applicability of Taguchi experiment planning to optimize other quality indicators of filled polymers and their manufacturing technology.

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