Effects of Temperature, Alkali Source, and Mg/Al Ratio on Phase Purity and Yield of Hydrothermal Synthesized Smectites

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Abstract: Smectites are a major type of clay minerals. Hydrothermally synthesized smectites have become a major research topic because of the unstable quality or excessive impurity of natural smectites. The high-phase purity and yield of hydrothermally synthesized smectites are vital in advanced industries and materials applications. In this study, a Taguchi orthogonal array was integrated with eight factors to avoid biased experimental results, thus creating relatively robust factor portfolios to investigate the effects of temperature, alkali sources, and the magnesium (Mg)/aluminum (Al) ratio on the phase purity and yield of hydrothermally synthesized smectites. The synthesized environment was mainly established using trioctahedral smectites based on the formula Na$_{2x}$[(Al$_{2(1-x)}$Mg$_{2x}$)Si$_4$O$_{10}$(OH)$_2$]. X-ray diffraction and Rietveld refinement were used for the quantitative analysis of the products’ mineral facies and calculating the synthesized smectites’ phase purity and yield. The Taguchi method was employed to calculate each factor’s effect on the product quality. The results indicated that among the numerous factor portfolios, a relatively high temperature, ammonia solution as the alkali source, and a relatively high Mg/Al ratio were conducive to enhanced phase purity and yield of synthesized smectites. The optimized products of the synthesized smectites achieved a phase purity of 92.5% and a yield of 88.3%.

Keywords: hydrothermally synthesized smectite; Taguchi method; temperature; alkali sources; Mg/Al ratio; phase purity; yield

1. Introduction

Smectites are a major type of clay minerals commonly found on the Earth’s surface. Smectites are widely applied in various fields, including use as drug carriers in biomedicine [1], as well as ceramics, metal casting clarifiers, drilling fluid materials, fillers, and supports for semiconductor nanoparticles in materials science because of their high cation exchange capability, large specific surface area, and swelling [2–5]. They have also been used in environmental rehabilitation as carriers for adsorbents and catalysts, hazardous material adsorbents, and barriers for highly radioactive waste [3,6–9].

Natural smectite clays vary in their structural composition, mineral facies, and impurities, all of which may limit their potential use without pretreatment purification because of differences in the geological environment of mineral deposits. However, the purification of natural smectites is often quite complex. According to the authors’ experience, natural smectites may still contain a large number of impurities even after pretreatment. This can be remedied by smectites synthesized under controlled conditions [10].

Although high-phase purity and yield are critical for advanced applications of synthetic smectites, synthesis of high-purity smectites remains difficult [11]. To the authors’ knowledge, nearly no studies...
have quantitatively examined the phase purity and yield of synthesized smectites. Only the author had explored the effects of eight main factors on the phase purity and yield of hydrothermally synthesized smectites, namely, source of silicon (Si), aluminum (Al), and magnesium (Mg), mineralizer type, mineralizer amount, proportion of excess sodium (Na), reaction time, and initial pH value [12]. However, other studies have suggested that reaction temperature, alkaline solution type, and Mg/Al ratio could also affect hydrothermally synthesized smectites.

Temperatures used in research on the hydrothermal synthesis of smectites have varied. Reinholdt et al. [10,13] employed experimental conditions of 220 °C for 72 h to synthesize smectites with the molecular formulas MgO-Al2O3-SiO2 and ZnO-Al2O3-SiO2, adding an appropriate amount of fluorine as a mineralizer. Zhang et al. [14] reviewed the experimental environment and results for smectite and saponite synthesis. Only a few studies have attempted the synthesis at room temperature, whereas the majority have employed a temperature range of 200–350 °C. Unlike trioctahedral smectite, dioctahedral saponite could be successfully synthesized at a relatively low temperature. Andrieux and Petit [15] noted that although samples containing a few smectites could be synthesized at temperatures of 150–200 °C, raising the temperature to 220 °C enhances synthesis and the pure phase of minerals. Xie et al. [16] performed the hydrothermal synthesis of Cu-saponite in a temperature range of 60–230 °C. The intensities of the (001) reflections of the final samples increased with temperature, with the highest peak value of the (001) reflection at 230 °C.

According to the majority of studies, smectite mainly forms in neutral to alkaline natural geological environments, and the hydrothermal synthesis of smectite generally occurs in similar environments. Therefore, the precursors used in the gel solution preparation are primarily alkaline. Although several alkaline chemicals have been used to adjust the precursors’ pH value, there have been no systematic studies exploring whether different alkaline solutions could affect the outcome of synthesis.

Although the Mg/Al ratio in precursors is another critical factor affecting synthesis, it has been variously reported in the literature. Based on the molecular formula R2x(Al2(1−x)M2x□)Si4O10(OH)2, Reinholdt et al. [10,13] performed smectite synthesis with X = 0.1–1.0. The X-ray diffraction (XRD) result indicated that when the Mg content was 0.9–1.0, a mixed phase of trioctahedral smectite and talc was produced. Li et al. [17] observed that when synthesizing smectites in an alkaline environment, high Mg content tends to produce the purest even site facies. Golubeva [18] synthesized Al-Mg–based smectites at temperatures of 200–350 °C for 5–228 h with an autogenous pressure ranging from 20 to 70 MPa. The results indicated that samples with high substitution of magnesium for aluminum could successfully synthesize a product within this relatively wide temperature range, whereas those with a relatively low degree of magnesium substitution required a temperature above 350 °C for products with a smectitic phase.

Research has generally employed a one-factor-at-a-time experiment method that alters one factor at a time while the others remain fixed. For example, when factors A to F are fixed at level 1 for experiments 1 and 2, changing factor G to level 2 could produce the optimal result (Table 1). However, insufficient consideration of various factor portfolios during the experimental design could lead to biased results. In experiments 3 and 4 in Table 1, when factors A to F are fixed at level 2, factor G at level 2 yields a relatively unfavorable result. The combination of various factors may provide a more comprehensive view of the actual effects of the factors at different levels.

Most of the aforementioned studies on the effects of temperature, alkali source, Mg/Al ratio, and other factors on the hydrothermal synthesis of smectites adopted research methods similar to the one-factor-at-a-time approach. The factor types and portfolios of hydrothermal smectite synthesis are extremely complex; therefore, this study considered the comprehensive effects of 12 factors. Given the large number of factors; however, if seven are at level 3 and five at level 2, all possible factor portfolios would require at least 69,984 (37 × 25) experiments. To avoid this, the Taguchi method was adopted in the experimental design to maintain the same quality of results while effectively reducing the number of experiments.
This study employed the Taguchi method in the experimental design to perform the hydrothermal synthesis of trioctahedral smectites. XRD and the BGMN Rietveld method [19] were adopted for the quantitative phase analysis of the smectite products. Through the analysis of variations, the role and effects of temperature, alkali sources, and Mg/Al ratio factors on the phase purity and yield of hydrothermally synthesized smectites were discussed, and the phase purity and yield of hydrothermally synthesized smectites were also optimized.

### Table 1.

| Experiment No. | Factors and Levels | Results |
|----------------|--------------------|---------|
|                | A  | B  | C  | D  | E  | F  | G  |     |
| 1              | 1  | 1  | 1  | 1  | 1  | 1  | 1  | 1.4 |
| 2              | 1  | 1  | 1  | 1  | 1  | 1  | 2  | 1.8 |
| 3              | 2  | 2  | 2  | 2  | 2  | 2  | 1  | 2.0 |
| 4              | 2  | 2  | 2  | 2  | 2  | 2  | 2  | 1.2 |

2. Materials and Methods

2.1. Experimental Design of the Taguchi Method

This study used the Taguchi method’s L18 orthogonal array to minimize the interaction between experimental factors. This orthogonal array contains eight factors, namely, one factor at level 2 and seven at level 3 (i.e., factors A–H in Figure 1). The eight factors comprised the sources of Si, Al, and Mg, mineralizer types, amount of added mineralizer, excess added Na, reaction time, and initial pH value, with the level of each factor indicated in Table 2. Through various orthogonal combinations of these factors, this study aimed to objectively examine the effects of the reaction temperature, alkali solution type, and Mg/Al ratio on the hydrothermal synthesis of smectites.

![Figure 1](image-url)
The reaction temperature, alkali solution type, and the Mg/Al ratio (i.e., $\alpha$, $\beta$, and $\gamma$ in Figure 1) were arranged in an L4 orthogonal array as an external orthogonal array of L18, with the factor levels given in Table 3. This design corresponded to various orthogonal combinations of the other eight factors to avoid biased experimental results produced by unsuitable combinations.

Table 3. Three factors and levels of hydrothermally synthesized smectites in the external orthogonal array.

| No. | Factor          | Level 1       | Level 2       |
|-----|-----------------|---------------|---------------|
| $\alpha$ | Reaction temperature | 200 °C | 220 °C  |
| $\beta$ | Alkaline chemicals | NH$_4$OH | NaOH         |
| $\gamma$ | Mg/Al ratio | 8:2 | 9:1          |

Based on the combined number of levels for each factor in the L18 orthogonal array, 18 experimental combinations were required, whereas L4 required four experiments for each combination. A total of 72 (18 × 4) hydrothermal synthesis experiments were thus performed in this study.

2.2. Synthesis of Smectites

According to the factors of the Taguchi method (Figure 1 and Tables 2 and 3), after all the initial materials were formulated and left to age for 1 h, a steel autoclave with a Teflon container under hydrothermal conditions was used to perform the smectite synthesis. The synthesized product was washed three times with deionized water, dried at 60 °C for 24 h, then weighed and ground for subsequent measurement.

2.3. Analytical Method and Quantitative Phase Analysis

The dried smectite products were evenly divided into two parts. One part was scanned without modification through XRD. The resulting XRD patterns are shown in this paper. A total of 10 wt% Al$_2$O$_3$ powder as an internal standard was added to the other part, and the XRD results were used for quantitative analysis.

XRD analysis of the synthesized product was performed using a Bruker D2-Phaser powder diffractometer (Bruker AXS, Germany) (Cu–K$_\alpha$ radiation; 2$\theta$: 5°–70°, step: 0.02°, scanning rate: 3°/min).

The integrated Profex interface and BGMN Rietveld program were capable of formulating a model for the turbostratic disorder of smectites [20], and the mineral composition of impurities could also be calculated using this program. This study employed the Rietveld refinement of this program.
to perform quantitative phase analysis for the calculation of the phase purity and the yield of the synthesized smectite samples.

The experimental data were subsequently used to calculate the average (AVG) phase purity and yield, and the effectiveness and weight of each factor were calculated using the Taguchi method. The optimized factors were also calculated.

3. Results and Discussion

3.1. Results for Each Experimental Combination (Yield and Phase Purity)

The theoretical yield of the synthesized smectite products was calculated using the equation for the ideal formula of smectite and the added initial chemicals. The yield of each synthesized smectite product may be calculated using the following equation:

\[
\text{Yield} = \frac{\text{weight of product}}{\text{theoretical weight of product}} \times 100\% \tag{1}
\]

The products may contain impure facies (e.g., analcime, gibbsite, boehmite, and brucite), as indicated by the XRD results (Figure 2). Furthermore, amorphous materials without XRD peak characteristics are often found in such products. Therefore, this study employed an equation to define phase purity. Facies content (including amorphous material) data obtained by applying the BGMN Rietveld refinement program to the XRD data were substituted into Equation (2). All other impurities in Equation (2) include amorphous material.

\[
\text{Purity} = \frac{\text{weight of smectite product}}{\text{weight of (smectite + all other impurities) in product}} \times 100\% \tag{2}
\]

![XRD analysis indicating numerous impurities in the products of experiments 7 and 11 in the L18 array.](image)

Figure 2. XRD analysis indicating numerous impurities in the products of experiments 7 and 11 in the L18 array.

Tables 4 and 5 show the phase purity and yield for all 72 products and the average (AVG) for each experimental combination in the L18 and L4 orthogonal arrays.
Table 4. Phase purity (%), average (%), and SD (standard deviation) of the synthesized smectites in the experimental combinations.

| No. | L4-1 | L4-2 | L4-3 | L4-4 | Average |
|-----|------|------|------|------|---------|
| 1   | 21.0 | 38.6 | 68.3 | 44.3 | 43.05   |
| 2   | 72.4 | 62.0 | 80.4 | 72.4 | 71.80   |
| 3   | 46.8 | 51.7 | 54.4 | 88.1 | 60.25   |
| 4   | 20.5 | 43.6 | 44.7 | 45.3 | 38.53   |
| 5   | 74.6 | 63.8 | 66.9 | 42.3 | 61.90   |
| 6   | 77.5 | 49.9 | 44.9 | 87.6 | 64.98   |
| 7   | 30.1 | 40.9 | 40.8 | 29.3 | 35.28   |
| 8   | 64.4 | 40.3 | 57.9 | 30.7 | 48.33   |
| 9   | 81.6 | 90.2 | 56.7 | 41.5 | 67.50   |
| 10  | 47.5 | 50.1 | 64.3 | 43.7 | 51.40   |
| 11  | 70.7 | 35.7 | 90.9 | 89.2 | 71.63   |
| 12  | 48.8 | 43.4 | 49.4 | 49.5 | 47.78   |
| 13  | 74.4 | 71.8 | 72.3 | 70.8 | 72.33   |
| 14  | 79.1 | 57.9 | 48.5 | 45.1 | 57.65   |
| 15  | 43.6 | 48.7 | 64.4 | 19.7 | 44.10   |
| 16  | 85.5 | 93.4 | 90.1 | 66.5 | 83.88   |
| 17  | 29.3 | 19.6 | 54.7 | 45.0 | 37.15   |
| 18  | 57.6 | 52.3 | 62.1 | 37.8 | 52.45   |
| Ave | 56.97| 52.99| 61.76| 52.71| 56.11   |

The phase purity of all 72 samples ranged from 19.6% to 93.4%, with the average phase purity for each experimental combination in the L4 ranging from 52.71% to 61.76%, thus resulting in an average (AVG) of only 56.11% for all 72 samples. The standard deviation (SD) of the four experimental combinations in the L4 array ranged from 14.72 to 21.66, indicating substantial variations in the results.

Table 5. Yield (%), average, and SD of each synthesized smectite in the experimental combinations.

| No. | L4-1 | L4-2 | L4-3 | L4-4 | Average |
|-----|------|------|------|------|---------|
| 1   | 13.7 | 25.7 | 26.0 | 17.4 | 20.69   |
| 2   | 43.1 | 39.4 | 62.1 | 49.3 | 48.49   |
| 3   | 19.1 | 21.1 | 26.4 | 73.3 | 34.99   |
| 4   | 13.1 | 29.2 | 30.1 | 28.3 | 25.19   |
| 5   | 26.9 | 19.6 | 28.1 | 19.2 | 23.43   |
| 6   | 47.8 | 33.4 | 23.0 | 69.3 | 43.38   |
| 7   | 17.8 | 13.0 | 25.9 | 19.0 | 18.93   |
| 8   | 29.9 | 22.4 | 23.0 | 18.0 | 23.31   |
| 9   | 51.6 | 63.2 | 55.0 | 27.3 | 49.26   |
| 10  | 38.0 | 41.2 | 41.7 | 32.6 | 38.38   |
| 11  | 51.2 | 24.2 | 74.5 | 60.4 | 52.56   |
| 12  | 29.9 | 32.8 | 40.9 | 32.6 | 34.05   |
| 13  | 68.6 | 64.2 | 68.0 | 67.2 | 67.00   |
| 14  | 60.5 | 39.7 | 33.0 | 28.5 | 40.40   |
| 15  | 25.4 | 32.2 | 37.3 | 13.4 | 27.05   |
| 16  | 70.8 | 75.3 | 81.5 | 49.9 | 69.34   |
| 17  | 21.4 | 12.8 | 45.0 | 27.6 | 26.71   |
| 18  | 35.5 | 34.2 | 37.5 | 18.4 | 31.40   |
| Ave | 36.91| 34.64| 42.17| 36.21| 37.48   |

The yield of all 72 samples ranged from 13.0% to 81.5%, with the AVG yield of the L4 experimental combinations ranging from 34.64% to 42.17%, for an overall AVG yield for all 72 samples of only 37.48%, indicating that the majority of chemicals added in the synthesis process were wasted. The SD for all
the L4 experimental combinations ranged from 18.44 to 19.98, thus demonstrating a large variation in the yield.

3.2. Effects of Factors on Synthesis Results

Through data integration and analysis of variance using the Taguchi method, the experimental factors, namely, reaction temperature, alkaline solution types, and Mg/Al ratio, could be further specified. The effects of these factors on the phase purity and yield of hydrothermally synthesized samples are shown in Figure 3.

![Figure 3](image-url)

**Figure 3.** The factor response graphs for the average phase purity (left) and yield (right) of smectite synthesis.

The factor response graphs illustrate the effect of factor variations at different levels on the experimental results for various orthogonal combinations of factors. In Figure 3 (left), the average phase purity of the synthesized smectites was 54.98% with the temperature (factor $\alpha$) at level 1 ($\alpha_1$, 200 °C) and 57.24% with the temperature at level 2 ($\alpha_2$, 220 °C). This indicated that a reaction temperature of 220 °C more effectively increased the phase purity of the hydrothermally synthesized smectites than a temperature of 200 °C.

The average phase purity of alkaline chemicals (factor $\beta$) was 59.36% at level 1 ($\beta_1$, NH$_4$OH) but only 52.85% at level 2 ($\beta_2$, NaOH), indicating that NH$_4$OH as the alkali source was more effective than NaOH. The overall phase purity was 57.38% with the Mg/Al ratio (factor $\gamma$) at level 2 ($\gamma_2$, 9:1), and 54.84% at level 1 ($\gamma_1$, 8:2), indicating that a high Mg/Al ratio produced superior results.

Figure 3 (right) shows the reaction trend of each factor for the yield of hydrothermally synthesized smectites was similar to that for the phase purity, with both demonstrating superior results under conditions of $\alpha_2$, $\beta_1$, and $\gamma_2$.

Using analysis of variance, the weight of each factor variation affecting the results was obtained. Table 6 and Figure 4 indicate that the weights of the alkali source on the phase purity and yield are 6.60% and 3.57%, respectively. The effect of alkali source was more significant than that of the temperature change and Mg/Al ratio. The reaction temperature had the least effect on the phase purity (1.78%), whereas the Mg/Al ratio had the least effect on yield (1.93%).
Table 6. Analysis of variations in phase purity (left) and yield (right) of hydrothermally synthesized smectites.

| Factor | Analysis of Variance of Phase Purity | Analysis of Variance of Yield |
|--------|--------------------------------------|------------------------------|
|        | SS DOF Var Contribution | SS DOF Var Contribution |
| α      | 29.5 1 29.49 1.78% | 37.0 1 36.98 2.47% |
| β      | 109.5 1 109.52 6.60% | 53.5 1 53.46 3.57% |
| γ      | 48.8 1 48.81 2.94% | 28.9 1 28.88 1.93% |
| Error  | 1471.7 4 367.94 88.68% | 1377.2 4 344.29 92.03% |
| Total  | 1659.6 7 237.08 100.00% | 1496.5 7 213.78 100.00% |

(SS: sum of squares; DOF: degrees of freedom; Var: variance).

Figure 4. Weighted effect of each factor on the phase purity (left) and yield (right) of hydrothermally synthesized smectites.

Nonetheless, the absolute weights of all three factors for the phase purity and yield were lower than 10%. One possible reason is that the range of variation range for each factor was not wide enough, or that other factors had a stronger effect. Analysis of variance for phase purity and yield (Table 6) indicated that the error terms might have resulted from neglect of other significant factors or errors in the experiments. However, other calculations demonstrated that this study’s experimental results were highly reliable, with experimental error having little effect. Therefore, the error terms in the analysis represented other, neglected factors in hydrothermal smectite synthesis that exerted a stronger effect than reaction temperature, alkali sources, and Mg/Al ratio.

The factor response trend chart (Figure 3) indicates that an increased exothermic reaction temperature or Mg/Al ratio could result in improved synthesis. However, these factors could only be increased to a certain level, beyond which a decrease in synthesis may occur.

3.3. Optimized Samples and Their Properties

The optimal factor condition obtained using the Taguchi method was applied to hydrothermal smectite synthesis. The reaction temperature was set to 220 °C, NH₄OH was employed as the alkali source, and the Mg/Al ratio was 9:1. The other parameters were as follows: TEOS (tetraethoxysilane), Al(NO₃)₃, and Mg(CH₃COO)₂·4H₂O were used as sources of Si, Al, and Mg ions, respectively; 0.02 mL of NH₄F was added as the mineralizer; the stoichiometry of Na ions was 110%; the precursor’s initial pH value was 11; and the reaction time was 72 h.

Using the optimal factors, four optimized samples were created, with XRD analysis results presented in Figure 5, indicating no evident impurity phases. XRD data for the four samples were
analyzed using the Rietveld refinement method coupled with BGMN. These results similarly indicated no impurity phases except for a small amount of amorphous material (Table 7).

![Graph showing XRD analysis results for smectite samples employing optimal factors based on the Taguchi method.](image)

**Figure 5.** XRD analysis results for smectite samples employing optimal factors based on the Taguchi method.

**Table 7.** Phase purity and yield of smectites synthesized under optimized conditions (unit: %).

| Sample No. | Purity | Amorphous | Yield | Rwp * |
|------------|--------|-----------|-------|-------|
| Optimized 1| 90.3   | 9.7       | 87.6  | 5.4   |
| Optimized 2| 96.0   | 4.4       | 92.9  | 5.7   |
| Optimized 3| 88.0   | 12.0      | 82.5  | 5.6   |
| Optimized 4| 95.8   | 4.2       | 90.3  | 5.2   |
| **Average**| **92.5** | **7.5**  | **88.3** | **-** |
| **SD**     | 4.0    | -         | 4.4   | -     |

*Rwp: The weighted residual squared sum.*

Optimized product 2 exhibited the optimal results, with a phase purity of 96% and a yield of 92.9% (Table 7). The AVG phase purity and yield of the four optimized products increased to 92.5% and 88.3%, respectively. The content of amorphous material in the four optimized products ranged from 4.2% to 12.0%, resulting in an AVG of only 7.5%. The SDs of the phase purity and yield of the four optimized products decreased to 4.0 and 4.4, respectively, indicating the significantly enhanced reproducibility of such high-quality synthesized products.

Smectite is capable of absorbing water or organic chemicals, and its structure changes with the interlayer space. The partially optimized samples were placed in an oven at 70 °C for adsorption of ethylene glycol vapor, heated to 500 °C for 1 h, and then evaluated using XRD.

The results (Figure 6) show that the synthesized product’s d\(_{001}\) peak shifted from \(2\theta = 6.683^\circ\) to \(4.838^\circ\) after the ethylene glycol vapor process, indicating that d\(_{001}\) expanded from 13.216 to 18.238 Å. After heat treatment at 500 °C for 1 h, the d\(_{001}\) peak shifted from \(2\theta = 6.683^\circ\) to 8.859°, indicating that d\(_{001}\) diminished from 13.216 to 9.973 Å. This is consistent with the adsorption and desorption properties of smectite minerals, thus verifying that the synthesized products were smectite phase based.
Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) results for the optimized synthesized smectites are shown in Figure 7. TGA showed that the first mass loss occurred at a temperature of approximately 87.8 °C, less than 100 °C, and was induced by water adsorption on the smectite surface during the heating process, resulting in a weight loss of about 8.3%. The second substantial mass loss occurred after reaching 800 °C and was caused by dehydrogenation of the constitution water in the smectite layer.

The DTA results exhibited a first endothermic peak at a temperature of 100–200 °C, which was induced by adsorption of water on the smectite surface during heat treatment. The second major endothermic peak occurred at 837 °C, caused by the dehydrogenation of the constitution water, and an exothermic peak appeared at 852 °C, representing a phase transition induced by the destruction of the Mg smectite crystal structures. These results were extremely similar to the smectite TGA curve proposed by Takahashi et al. [21].

Table 8 lists the AVG phase purity and yield of the 72 regular samples prior to optimization and those of the four optimized samples for comparison. The AVG phase purity and yield of the
preoptimization products were only 56.1% and 37.5%, respectively. After these were synthesized using the Taguchi method parameter optimization, the AVG phase purity and yield increased to 92% (1.65 times higher than initially) and 88.3% (2.35 times higher than initially), respectively. In addition, the optimized impurity phase decreased from an average of 11.2% to <0.1% and the amorphous material from an average of 32.7% to 7.5%, indicating that such parameter optimization could substantially enhance the phase purity and yield of hydrothermally synthesized smectites.

Table 8. Comparison of the AVGs of the purity and yield of 72 regular and four optimized synthesized smectite products (unit: %).

| Synthetic Products       | Avg. of 72 Regular Samples | Avg. of Four Optimized Samples |
|--------------------------|----------------------------|--------------------------------|
| Purity of smectite       | 56.1                       | 92.5                           |
| Impurity                 | 11.2                       | <0.1                           |
| Amorphous material       | 32.7                       | 7.5                            |
| Yield of smectite        | 37.5                       | 88.3                           |

4. Conclusions

This study was designed using the Taguchi method and developed an orthogonal array of eight factors, namely, sources of Si, Al, and Mg, mineralizer type, amount of added mineralizer, excess Na, initial pH value, and reaction time. By using such a design, this study avoided biased results and comprehensively evaluated the effects of reaction temperature, alkali sources, and Mg/Al ratio on the phase purity and yield of hydrothermally synthesized smectites. These factors were also optimized, as may be seen from the phase purity and yield of the hydrothermally synthesized smectites, which indicated that a reaction temperature of 220 °C (compared with 200 °C), NH₄OH as the alkali source (compared with NaOH), and an Mg/Al ratio of 9:1 (compared with 8:2) could produce superior results. This combination of optimized conditions was able to increase the phase purity of the synthesized smectites to 92.5%, with no other mineral impurity content except for amorphous materials. Similarly, the product yield could be increased to 88.3%, enabling more efficient conversion from chemicals to products. The overall SDs for phase purity and yield of the smectite product after multiple syntheses both decreased substantially, indicating a considerable enhancement of the synthesized product’s reproducibility.

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