Effect of the MgO/Silica Fume Ratio on the Reaction Process of the MgO–SiO$_2$–H$_2$O System

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Received: 6 November 2018; Accepted: 21 December 2018; Published: 26 December 2018

Abstract: In order to clarify the effect of the MgO–silica fume (SF) ratio on the reaction process of the MgO–SiO$_2$–H$_2$O system, the reaction products and degree of reaction were characterized. Furthermore, the parameters of the reaction thermodynamics were calculated and the reaction kinetics were deduced. The results indicate that a large amount of Mg(OH)$_2$ and small quantities of magnesium silicate hydrate (M–S–H) gels were generated upon dissolution of MgO. However, the M–S–H gels were continuously generated until the SF or Mg(OH)$_2$ was consumed completely. For a MgO dosage less than 50% of the total MgO–SiO$_2$–H$_2$O system, the main product was M–S–H gel, while for a MgO dosage greater than 50%, the main product was Mg(OH)$_2$. The results indicate that M–S–H gels have greater stability than Mg(OH)$_2$, and the final reaction product was prone to be M–S–H gels. Based on the experimental values, an equation is proposed for the reaction kinetics of MgO.

Keywords: MgO; silica fume; reaction process; thermodynamics; kinetics

1. Introduction

The MgO–SiO$_2$–H$_2$O system was developed as a novel cementitious material [1–5]. Numerous applications have been realized, including usage in thermal insulation material [6], waste sealing material [7], refractory material [8], and soil stabilization [9]. Magnesium silicate hydrate (M–S–H) gel is the typical reaction product formed in the MgO–SiO$_2$–H$_2$O system [7–14]. Cole [15] found a crystalline M–S–H phase, which was identified as 4MgO·SiO$_2$·8.5H$_2$O. Gollop [16] found that M–S–H gel forms in Portland cement pastes by magnesium sulfate attack, and characterized the gel as a poorly crystallized serpentine (3MgO·2SiO$_2$·2H$_2$O). Brew [17,18] characterized chemically synthesized M–S–H gels and investigated the incorporation of cesium and potassium. Vandeperre et al. [19] discovered that brucite (Mg(OH)$_2$) could react with the amorphous silica present in pulverized fuel ash to form M–S–H gels. In fact, the reaction products containing Mg were found to be Mg(OH)$_2$ and hydrotalcite-like phases, rather than M–S–H gels [20]. In addition, because magnesium is frequently associated with calcium in carbonate rocks (the raw material for cement production) [21], the existence of MgO in cement-based materials is inevitable. Overall, the reaction process of MgO in the MgO–SiO$_2$–H$_2$O system and the effects of the MgO/SF ratio on the reaction process are not clearly understood, and further research is necessary to clarify these reactions.

In this study, the reaction processes and reaction products of MgO/silica fume (SF) pastes were characterized. Further, thermodynamic calculations and kinetic deductions were conducted, and the
effects of the MgO/SF ratio on the reaction process of the MgO–SiO₂–H₂O system clarified. The results contribute to a better understanding of the reaction processes of the MgO–SiO₂–H₂O system and the development of a novel cementitious material.

2. Materials and Methods

2.1. Raw Materials

The chemical compositions of MgO and SF [3], as determined by X-ray fluorescence spectroscopy (XRF, PANalytical, Almelo, The Netherlands), are listed in Table 1. The particle size distributions of MgO and SF powders [3], as measured by laser diffraction (Partica LA-950V2, HORIBA, Kyoto, Japan) are presented in Figure 1.

| Material | Composition (wt.%) | Specific Density (g/cm³) | BET Specific Surface Area (m²/g) |
|----------|-------------------|--------------------------|-------------------------------|
| SiO₂     | MgO   | CaO | K₂O | Na₂O | Al₂O₃ | P₂O₅ | SO₃ |
| MgO      | 0.29  | 97.22 | 1.45 | –    | 0.21  | 0.06 | 0.20 | 2.96 | 51.60 |
| SF       | 95.74 | 0.63  | 1.25 | 1.07 | 0.33  | 0.50 | 0.20 | 0.15 | 2.03 | 16.04 |

Note: –, undetected.

Figure 1. Particle size distributions of MgO and SF powders. Reprinted with permission from [3]; Copyright 2014 Elsevier.

2.2. Sample Preparation

2.2.1. Preparation of MgO/SF Pastes

According to the proportions of the mixture listed in Table 2, MgO/SF pastes were prepared by mixing reactive MgO, SF, and water homogeneously, followed by sealing of the mixture in polyethylene bags and curing at room temperature (25 ± 1 °C) [3].

| Sample ID | MgO (wt.%) | SF (wt.%) | W/B Ratio |
|-----------|------------|-----------|-----------|
| M₀.₂S₀.₈  | 20         | 80        | 1.0       |
| M₀.₄S₀.₆  | 40         | 60        | 1.0       |
| M₀.₅S₀.₅  | 50         | 50        | 1.0       |
| M₀.₆S₀.₄  | 60         | 40        | 1.0       |
| M₀.₈S₀.₂  | 80         | 20        | 1.0       |

Note: W/B ratio is the water to binder (MgO and SF) ratio by mass.
2.2.2. Preparation of M–S–H Gel

M–S–H gel with a Mg/Si ratio of 1.0 was synthesized as reported in [3]. In accordance with [17], Mg(NO$_3$)$_2$·6H$_2$O solution was slowly added to a Na$_2$SiO$_3$·5H$_2$O solution by stirring in a flask at 0 °C. The precipitated composite was filtered and washed using ultrapure water [3].

2.3. Testing Methods

2.3.1. Measurement of Mechanical Strength

The mechanical strength of MgO/SF mortar specimens (40 × 40 × 160 mm$^3$) were measured according to ASTM C 349 at each curing age (days 3, 7, 28, and 90) [22].

2.3.2. Measurement of Heat Evolution

The heat evolution of the MgO/SF pastes [3] was measured up to day 10 at 25 °C using a TAM-Air isothermal calorimeter (Thermometric AB, Jarfalla, Sweden) according to ASTM C 1702-09 [23].

2.3.3. Characterization of the Reaction Products

MgO/SF pastes cured at day 1, 3, 7, 28, and 90 were frozen (−195 °C) by immersion in liquid nitrogen and then dried via the freeze-drying technique [24]. The reaction products were characterized by using X-ray diffraction (X’Pert Pro diffractometer, Cu K$_\alpha$, 40 kV and 40 mA; Philips, Amsterdam, The Netherlands) and thermal analysis (Netzsch STA 449C, 10 °C/min in N$_2$ atmosphere; Netzsch, Bavaria, Germany) [3].

2.3.4. Measurement of MgO Content

The residual MgO content in MgO/SF pastes was determined by quantitative X-ray diffraction (Q-XRD, Cu K$_\alpha$, 40 kV and 40 mA; PANalytical B.V., Almelo, The Netherlands) using the K-value method [25], in which ZnO was used as the internal standard material.

2.3.5. Calculation of Mg(OH)$_2$ and M–S–H Gel Contents

The contents of Mg(OH)$_2$ and M–S–H gel were calculated according to the weight losses displayed in the DTG/TG curves, as shown in Equations (1)–(3) [3,4].

\[
C_{[\text{Mg(OH)}_2]} = \frac{\Delta M_3 \times M_{[\text{Mg(OH)}_2]}}{M_{[\text{H}_2\text{O}]}} 
\]

\[
C_{[\text{PAW}]}_{\text{M–S–H gel}} = \Delta M_1 
\]

\[
C_{[\text{CBW}]}_{\text{M–S–H gel}} = \Delta M_2 - \Delta M_3 - \Delta M_4 
\]

where \(C_{[\text{Mg(OH)}_2]}\) is the Mg(OH)$_2$ content; \(C_{[\text{PAW}]}_{\text{M–S–H gel}}\) and \(C_{[\text{CBW}]}_{\text{M–S–H gel}}\) are the contents of the M–S–H gel (wt.%), characterized by the physically absorbed water (PAW) and chemically bound water (CBW), respectively; \(M_{[\text{Mg(OH)}_2]}\) and \(M_{[\text{H}_2\text{O}]}\) are the molar masses of Mg(OH)$_2$ (g/mol) and H$_2$O (g/mol), respectively; \(\Delta M_1\) and \(\Delta M_2\) are the weight losses in the range of 50–200 °C and 200–1000 °C in TG curves, respectively; \(\Delta M_3\) and \(\Delta M_4\) are the weight losses due to the dehydration of Mg(OH)$_2$ and decomposition of MgCO$_3$, as shown in Section “Mg(OH)$_2$ Content”.

2.3.6. Degree of Reaction

The degree of reaction (\(\alpha\)) of MgO was calculated using Equation (4).

\[
\alpha = \frac{C_0(\text{MgO}) - C_f(\text{MgO})}{C_0(\text{MgO})} 
\]
where $C_0(\text{MgO})$ and $C_t(\text{MgO})$ are the mass ratios of MgO in MgO/SF pastes cured at 0 and $t$ days, respectively, which are normalized using the residual mass ratio of the samples following calcining at 1000 °C.

3. Results and Discussion

3.1. Reaction Process

3.1.1. Compressive Strength and Flexural Strength

Figure 2 shows the compressive strength and flexural strength development of MgO/SF mortar up to 90 days. The compressive strength and flexural strength increased with curing age. However, because MgO hydrated too slowly, it was not able to gain sufficient strength after day 28 and strength retrogression occurred even for $M_{0.8}S_{0.2}$ 90 days later. The main reason for this is that MgO produces a large amount of Mg(OH)$_2$ in the later stage of the reaction. The gelling ability of Mg(OH)$_2$ is poor and volume expansion occurs simultaneously, which leads to a decrease in strength. The incorporation of SF with MgO resulted in higher strength because of the reaction between them, and strength increased with the increase in SF amount. $M_{0.5}S_{0.5}$ mortar has the highest strength; its compressive strength at days 3, 7, 28, and 90 is 18.4, 40.6, 64.7 and 75.6 MPa, and its flexural strength is 3.2, 5.6, 6.1 and 9.4 MPa, respectively. This phenomenon can be attributed to the fact that when the MgO content is low, the amount of M–S–H gel generated is small and the strength is low. When the MgO content is high, the reaction products are mainly Mg(OH)$_2$, and the mechanical properties are poor. In general, the strength increased with curing time up to 90 days. However, it was found that further curing did not lead to any increase in strength; on the contrary, the strength sometimes decreased.

![Figure 2. Mechanical strength of MgO/SF mortar: (a) compressive strength; (b) flexural strength.](image)

3.1.2. Heat Evolution

Three peaks (designated as I, II, and III, at approximately 0.5, 12, and 48 h) [5] were observed in the rate of heat evolution curves of the MgO/SF pastes (Figure 3a) [3]: the initial peak (I) is related to the exothermic wetting of the mixtures [5]; the second peak (II) corresponds to the formation of Mg(OH)$_2$; the third peak (III) is attributed to the formation of the M–S–H gel [5]. As the MgO content decreased, the second peak became less distinct, while the third peak broadened (Figure 3a), resulting in a decrease in the early cumulative heat. The cumulative heat of the $M_{0.4}S_{0.6}$ paste (255.88 J/g) was lower than that of the $M_{0.6}S_{0.4}$ paste (299.06 J/g) and $M_{0.5}S_{0.1}$ paste (302.94 J/g) at an early curing age (100 h), while at an advanced curing age (300 h), the cumulative heats were 351.83, 370.32 and 400.05 J/g, respectively. The results indicate that the formation of Mg(OH)$_2$ is restrained by increasing the content of SF, which also confirms that the formation of the M–S–H gel is a continuous process [5].
3.1.3. Variation in the Composition of MGO/SF Pastes

The XRD patterns of synthetic M–S–H gel and SF (Figure 4) contain broad peaks in the range of 23°–28°, 32°–38°, and 58°–62° [6], all of which correspond to the M–S–H gel. Another broad peak exists at 15°–25° and is attributed to amorphous SiO₂ [3,26]. In addition, the sharp peaks at 36.9°, 42.9°, 62.3°, 74.7°, and 78.6° are attributed to MgO [5] and the peaks at 18.6°, 38.0°, 50.8°, and 58.6° correspond to Mg(OH)₂ [5]. The Mg(OH)₂ peaks appeared in the XRD patterns of all MgO/SF pastes at an early age (day 3). However, no Mg(OH)₂ was found in the M₀.₂S₀.₈ and M₀.₄S₀.₆ pastes after day 90 (Figure 4b), and the formation of the M–S–H gel was confirmed. Notably, all MgO fully reacted in the M₀.₂S₀.₈ paste after day 90.

Figure 4. XRD patterns of MgO/SF pastes, MgO, SF and synthetic M–S–H gel: (a) M–S–H gel and SF, cured for 3 days; (b) M–S–H gel and SF, cured for 90 days.

MgO Content

The MgO content was determined via the Q-XRD method; the results are shown in Figure 5. The MgO content rapidly decreased within the first 7 days, after which the rate of decrease lowered. After 90 days, no MgO remained in the M₀.₂S₀.₈ or M₀.₄S₀.₆ pastes, while the content of MgO in the M₀.₅S₀.₅, M₀.₆S₀.₄, and M₀.₈S₀.₂ pastes was 1.34, 2.91, and 3.07%, respectively.

Figure 3. Heat evolution of MgO/SF pastes: (a) rate of heat evolution; (b) cumulative heat.
Mg(OH)$_2$ Content

Figures 6 and 7 show the DSC/DTG curves of the MgO/SF pastes. The first endothermic valley ($\Delta M_1$, ranging from 50–200 °C) corresponds to the weight loss of the PAW of reaction products (mainly products of the M–S–H gel) [3,8]; the second endothermic valley ($\Delta M_2$, at approximately 400 °C) occurs due to the removal of the hydroxyl from Mg(OH)$_2$ [4–8]; and, the exothermic effect at approximately 850 °C is associated with the recrystallization of the M–S–H gel [3,12–16]. In addition, $\Delta M_2$ (which is in the range 200–1000 °C) is the weight loss of the CBW from the reaction products (including Mg(OH)$_2$, MgCO$_3$ and M–S–H gel) [3]. Here, $\Delta M_4$ is the weight loss due to the decomposition of MgCO$_3$ [3,5], and the initial and final points are determined by the virtual baseline of the DSC curves (as shown in Figure 7a).
The Mg(OH)$_2$ content was calculated using Equation (1), the results are shown in Figure 8. The Mg(OH)$_2$ content in the M$_{0.2}$S$_{0.8}$, M$_{0.4}$S$_{0.6}$, M$_{0.5}$S$_{0.5}$, M$_{0.6}$S$_{0.4}$, and M$_{0.8}$S$_{0.2}$ pastes increased to 13.29\%, 22.85\%, 25.27\%, 35.52\%, and 59.34\% in the first 14 days, respectively, and then slowly decreased. No Mg(OH)$_2$ was found in the M$_{0.2}$S$_{0.8}$ paste after 90 days, while a small amount of Mg(OH)$_2$ (approximately 5\%) was found in the M$_{0.4}$S$_{0.6}$ paste.

PAW and CBW Contents

Owing to the formation of M–S–H gel, the first endothermic valley gradually became prominent (Figure 6). Therefore, the amounts of PAW and CBW associated with the M–S–H gel (shown in Figure 9) are considered to be indicative of the M–S–H gel content. The amounts of both PAW and CBW continuously increased, which is related to the gradual formation of the M–S–H gel. The amounts of PAW and CBW in the M$_{0.4}$S$_{0.6}$ paste were higher than those in other MgO/SF pastes, which indicated that the amount of M–S–H gel in the M$_{0.4}$S$_{0.6}$ paste is higher than that in the other MgO/SF pastes.

\[ \text{The Mg(OH)}_2 \text{ content was calculated using Equation (1), the results are shown in Figure 8. The Mg(OH)}_2 \text{ content in the M}_{0.2}\text{S}_{0.8}, \text{M}_{0.4}\text{S}_{0.6}, \text{M}_{0.5}\text{S}_{0.5}, \text{M}_{0.6}\text{S}_{0.4}, \text{and M}_{0.8}\text{S}_{0.2} \text{ pastes increased to 13.29\%, 22.85\%, 25.27\%, 35.52\%, and 59.34\% in the first 14 days, respectively, and then slowly decreased. No Mg(OH)}_2 \text{ was found in the M}_{0.2}\text{S}_{0.8} \text{ paste after 90 days, while a small amount of Mg(OH)}_2 \text{ (approximately 5\%) was found in the M}_{0.4}\text{S}_{0.6} \text{ paste.}

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\[ \text{The Mg(OH)}_2 \text{ content was calculated using Equation (1), the results are shown in Figure 8. The Mg(OH)}_2 \text{ content in the M}_{0.2}\text{S}_{0.8}, \text{M}_{0.4}\text{S}_{0.6}, \text{M}_{0.5}\text{S}_{0.5}, \text{M}_{0.6}\text{S}_{0.4}, \text{and M}_{0.8}\text{S}_{0.2} \text{ pastes increased to 13.29\%, 22.85\%, 25.27\%, 35.52\%, and 59.34\% in the first 14 days, respectively, and then slowly decreased. No Mg(OH)}_2 \text{ was found in the M}_{0.2}\text{S}_{0.8} \text{ paste after 90 days, while a small amount of Mg(OH)}_2 \text{ (approximately 5\%) was found in the M}_{0.4}\text{S}_{0.6} \text{ paste.}

3.2. Reaction Thermodynamics

The laws of thermodynamics can predict the probability of a reaction, as well as the final state achieved when the reaction is completed. These laws are often used in analyzing cement-based materials [27–29]. Consequently, they were employed in the analysis of the reaction of the MgO–SiO$_2$–H$_2$O system in this study. The thermodynamic coefficients for the minerals (or species) of the MgO–SiO$_2$–H$_2$O system are shown in Table 3.
3.3. Reaction Kinetics

3.3.1. Theoretical Deduction of the Reaction Kinetics Equation

The reaction process for the MgO–SiO$_2$–H$_2$O system was divided into two stages, as in references [3,14]. First, MgO reacted with water to form Mg(OH)$_2$ and then reacted with dissolved

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**Table 3.** Thermodynamic coefficients of the minerals (or species) in the MgO-SiO$_2$–H$_2$O system at 25 °C, 1 bar [30].

| Minerals or Species | $S_m^0$ (kJ·mol$^{-1}$) | $\Delta_f H_m^0$ (kJ·mol$^{-1}$) | $\Delta_f G_m^0$ (kJ·mol$^{-1}$) |
|---------------------|--------------------------|-------------------------------|-------------------------------|
| Mg$^{2+}$ (aq)       | −138.00                  | −466.85                       | −454.89                       |
| OH$^-$ (aq)          | −10.71                   | −230.02                       | −157.34                       |
| H$^+$ (aq)           | 0.00                     | 0.00                          | 0.00                          |
| H$_2$O (aq)          | 69.95                    | −285.83                       | −237.19                       |
| MgO (s)              | 26.95                    | −601.50                       | −569.23                       |
| SiO$_2$ (Amorphous)  | 47.40                    | −903.20                       | −850.59                       |
| H$_3$SiO$_4$$^-$ (aq)| 112.55                   | −1426.16                      | −1253.98                      |
| H$_2$SiO$_4$$^2-$ (aq)| −12.97                  | −1396.62                      | −1187.02                      |
| Mg(OH)$_2$ (s)       | 63.14                    | −924.54                       | −833.56                       |
| Mg$_3$Si$_2$H$_4$ (Chrysotile)| 221.30             | −4361.66                      | −4034.02                      |
| Mg$_3$Si$_4$H (Talc) | 260.80                   | −5915.90                      | −5536.27                      |

The thermodynamic calculations (standard molar entropy $\Delta_f S^0$, standard molar enthalpy of formation $\Delta_f H^0$, and standard molar Gibbs free energy $\Delta_f G^0$) for the potential reactions of the MgO–SiO$_2$–H$_2$O system were computed using Equations (5)–(7):

\[
\Delta_f S^0 = \sum_i v_i S_m^0
\]
\[
\Delta_f H^0 = \sum_i v_i \Delta_f H_m^0
\]
\[
\Delta_f G^0 = \sum_i v_i \Delta_f G_m^0 = -RT \ln K
\]

where, $v_i$ is the stoichiometric reaction coefficient, $R = 8.314$ J/(mol·K), and $T$ is the temperature in K [3]. Here, $\Delta_f S^0 > 0$ indicates that the reaction tends to be more uniform, and $\Delta_f H^0 < 0$ indicates that it is an exothermic reaction (reactions occur spontaneously). A chemical reaction occurs spontaneously when $\Delta_f G^0 < 0$, and under an isothermal constant pressure condition [3]. Note that a higher $K$ value signifies that the degree of reactant conversion is higher and the tendency for a positive reaction to occur is greater [3].

Some of the potential reactions of the MgO–SiO$_2$–H$_2$O system are listed in Table 4. Based on the $\Delta_f G_m^0$ and $\Delta_f G^0$ values, M–S–H phases are more stable than Mg(OH)$_2$. Furthermore, the formation of the M–S–H phases is much easier than Mg(OH)$_2$, as the $\Delta_f G^0$ values of reactions (4) (−63.99 kJ·mol$^{-1}$) and (5) (−261.32 kJ·mol$^{-1}$) are much higher than reaction (6) (−299.58 kJ·mol$^{-1}$).

**Table 4.** Thermodynamic calculations for potential reactions.

| ID | Minerals or Species | Reaction Equation | $\Delta_f S^0$ (kJ·mol$^{-1}$) | $\Delta_f H^0$ (kJ·mol$^{-1}$) | $\Delta_f G^0$ (kJ·mol$^{-1}$) | logK |
|----|---------------------|-------------------|-----------------------------|-----------------------------|-----------------------------|------|
| 1  | MgO                 | MgO + 2H$^+$ → Mg$^{2+}$ + H$_2$O | −95.00                      | −151.18                     | −122.85                     | 21.52 |
| 2  | H$_2$SiO$_4$$^2-$   | SiO$_2$ + 2OH$^-$ → H$_2$SiO$_4$$^2-$ | −38.95                      | −33.38                      | −21.75                      | 3.81  |
| 3  | H$_3$SiO$_4$$^-$    | SiO$_2$ + OH$^-$ + H$_2$O → H$_3$SiO$_4$$^-$ | 5.91                        | −7.11                       | −8.86                       | 1.55  |
| 4  | Mg(OH)$_2$          | Mg$^{2+}$ + 2OH$^-$ → Mg(OH)$_2$ | 222.56                      | 2.35                        | −63.99                      | 11.21 |
| 5  | Chrysotile          | 3Mg$^{2+}$ + 6OH$^-$ + 5SiO$_2$ ↔ 3MgO 5SiO$_2$ 2H$_2$O + H$_2$O | 674.71                      | −60.42                      | −261.32                     | 45.78 |
| 6  | Talc                | 3Mg$^{2+}$ + 6OH$^-$ + 4SiO$_2$ ↔ 3MgO 4SiO$_2$ 2H$_2$O + 2H$_2$O | 689.36                      | −94.09                      | −299.58                     | 52.48 |
silica to form M−S−H gel [5]. Wei et al. [2,13] calculated the thermodynamics of the MgO−SiO$_2$−H$_2$O system and confirmed that the formation of M−S−H gel and Mg(OH)$_2$ occurred simultaneously. The dissolution of MgO is represented by Equation (8):

$$\text{MgO} + \text{H}_2\text{O} \rightarrow \text{Mg}^{2+} + 2\text{OH}^-$$

(8)

The amount of MgO rapidly decreased at an early age, when the content of MgO was high, whereas the content of MgO decreased slowly when the amount of MgO was low. The reaction rate of MgO in the MgO−SiO$_2$−H$_2$O system is proportional to the first power of the MgO content, as shown in Equation (9):

$$-\frac{d[C_t(\text{MgO})]}{dt} = k \times C_t(\text{MgO})$$

(9)

where $k$ is the reaction rate constant.

By integrating both sides, Equation (9) can be modified to Equation (10):

$$\int_{C_0(\text{MgO})}^{C_t(\text{MgO})} \frac{d[C_t(\text{MgO})]}{C_t(\text{MgO})} = k \int_0^t dt$$

(10)

Equation (10) can also be written as follows:

$$C_t(\text{MgO}) = C_0(\text{MgO}) \times e^{-kt}$$

(11)

The relationship between $\alpha$ and $t$ is shown in Equation (12) (formulated by combining Equations (7) and (11)):

$$\alpha = 1 - e^{-kt}$$

(12)

3.3.2. Verification of the Reaction Kinetics Equation

The estimated values of $\alpha$ for varying MgO/SF pastes were fitted using the above reaction kinetics equation (Equation (12)). The fitted curves and their parameters are shown in Figure 10 and Table 5, respectively. The results demonstrate that the reaction kinetics follow the well-defined kinetics equation and the correlation is obvious ($R^2 > 0.97$), which confirms the theoretical deduction process.
Table 5. Fitted parameters.

| Sample ID | K (10^{-6} s^{-1}) | R^2  |
|-----------|---------------------|------|
| M_{0.2}S_{0.8} | 2.748               | 0.973|
| M_{0.4}S_{0.6} | 2.040               | 0.988|
| M_{0.5}S_{0.5} | 1.542               | 0.984|
| M_{0.6}S_{0.4} | 1.577               | 0.971|
| M_{0.8}S_{0.2} | 3.018               | 0.977|

Note: R^2 is the correlation coefficient.

The reaction kinetics for MgO in different MgO/SF pastes were calculated using the relation between the degree of reaction (\(\alpha\)) and curing age (\(t\)). The rate constant decreased with increasing MgO content when the dosage of MgO was lower than 50%, and increased with increasing MgO content when the dosage of MgO was higher than 50%.

4. Conclusions

When mixed with SF and water, MgO hydrated to form brucite, which immediately reacted with dissolved silica to produce M–S–H gel. The formation of amorphous M–S–H gel at room temperature was confirmed via XRD, TG, and microstructure analyses and the effect of the MgO/SF ratio on the reaction process of the MgO–SiO\(_2\)–H\(_2\)O system was discussed. XRD revealed the components of the reaction products, and TG was found to be a useful tool for determining the quantity of the reaction products. The following conclusions can be drawn from the present study:

- Mg(OH)\(_2\) results from the dissolution of MgO. M–S–H gels generates from the reaction between Mg\(^{2+}\) and hydrated silica, and consequently the dissolution of Mg(OH)\(_2\) and SiO\(_2\) is promoted.
- The formation reaction for the M–S–H gel is the main reaction in the MgO–SiO\(_2\)–H\(_2\)O system when the dosage of MgO is lower than 50%, while the formation reaction of Mg(OH)\(_2\) is the main reaction when the dosage of MgO is higher than 50%.
- Based on the thermodynamic calculations, M–S–H gels are more stable than Mg(OH)\(_2\). Furthermore, the formation reactions for the M–S–H gels occurred more completely than those for Mg(OH)\(_2\).
- The reaction kinetics of MgO in the MgO–SiO\(_2\)–H\(_2\)O system conforms to \(\alpha = 1 - e^{-kt}\) (\(R^2 > 0.97\)). Because of the decrease in the SF dosage, the rate constant decreased with decreasing SF content when the dosage of MgO was lower than 50%. As a result of the formation rate of the M–S–H gels being lower than Mg(OH)\(_2\), the rate constant increased with increasing MgO content when the dosage of MgO was higher than 50%.

Author Contributions: Conceptualization, Q.Y. and J.W.; Methodology, Z.L. and J.W.; Software, Y.X. and J.Z.; Validation, Z.L., Y.X. and J.Z.; Formal Analysis, Z.L. and H.L.; Investigation, H.L.; Resources, Q.Y.; Data Curation, J.W.; Writing—Original Draft Preparation, Z.L.; Writing—Review & Editing, Y.X. and J.Z.; Visualization, H.L.; Supervision, Q.Y.; Project Administration, J.W.; Funding Acquisition, J.W.

Funding: This work was funded by the Natural Science Foundation of China (No. 51802099 and 51172075), the Natural Science Foundation of Guangdong Province China (2018A030310389), and the Fundamental Research Funds for the Central Universities.

Acknowledgments: The authors wish to express their gratitude and sincere appreciation to the National Natural Science Foundation of China, Guangdong Low Carbon Technologies Engineering Center for Building Materials (South China University of Technology), and Guangdong Hydrodynamics Application Research Key Laboratory (Guangdong Research Institute of Water Resources and Hydropower).

Conflicts of Interest: The authors declare no conflict of interest.
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