**Article**

**Effect of Mixing Light-Burned MgO with Different Activity on the Expansion of Cement Paste**

Yang Wang 1, Caoning Zhang 2 and Lingling Xu 1,*

1 College of Materials Science and Engineering, Nanjing Tech University, Nanjing 210009, China; 18362080050@163.com
2 Hangzhou Silan Integrated Circuits Co., Ltd., Hangzhou 310023, China; 18013006534@163.com
* Correspondence: xulingling1964@163.com; Tel.: +86-13951846173

**Abstract:** In mass concrete, shrinkage resulting from temperature drop and drying leads to cracking, which can seriously affect the strength and durability of cement-based materials. Fortunately, expansion agents can deter or prevent these effects, especially MgO expansion agents (MEAs). In this study, the effects of four MEAs of different activity on the expansion properties, strength, and hydration of cement paste were explored. The different expansion phenomena between the high activity and low activity MgO was especially explained by the hydration model and dynamic theory. The results indicate that when the other conditions were the same, higher curing temperature and dosage could improve the expansion to some extent. Moreover, the hydration of high activity MgO and the expansion behavior occurred mainly in the early hydration stage, while the hydration of low activity MgO and the expansion behavior had a high contribution rate in the later stage, and the final expansion of cement mixed with low activity MgO was larger.

**Keywords:** MgO expansion agent; expansion properties; hydration model

---

**1. Introduction**

In the nineteenth and early twentieth centuries, a lot of structural damage to bridges and dams took place not long after being put in service due to the high content of MgO contained in cement used in Europe [1,2]. Since then, much more research has been undertaken to investigate the soundness of dead-burned MgO in cement. Additionally, standards for limiting the content of MgO in cement have been established in many countries. For example, the content of MgO should be below 5% in cement and can be 6% when passing soundness testing under autoclaving according to Chinese standards [3].

It has been found that the expansion produced by MgO could be utilized to compensate for the shrinkage of cementitious material under certain conditions. Mehta [4] proposed in 1980 that when 5% of the periclase calcined at 900–950 °C was added to the concrete, the expansion stress generated during the hydration process could compensate for the shrinkage and cracks of the concrete. Since then, many more efforts [5–8] have been put into controlling cracks in concrete by taking advantage of the delayed expansion of MgO. Meanwhile, some new problems should be solved, the most significant of which is the relatively little available source of the high MgO cement. Hence, in order to solve the limitation of receiving source and to better control the quality of MgO, magnesite was separately calcined to be an MgO expansion agent. In addition, dolomite, serpentine, and other magnesium-containing minerals were also considered to prepare MgO expansion agent [9–15].

Some work has been performed on the hydration mechanism of periclase in water. P Liu [16,17] believed that firstly, H2O molecules chemically adsorb on the surface of MgO defects in water, and then the surface of MgO produces hydroxilation. Mejias J A [18] thought that this hydroxilation reaction rearranges the atoms at the interface between MgO and H2O. Then, a layer of Mg(OH)2 directly forms on the surface; or the dissociated...
Mg$^{2+}$ intermediate forms a supersaturated solution with OH$^-$, finally crystallizing to Mg(OH)$_2$. However, Wogelius [19] considered that hydroxylation takes place in the near-surface region of MgO crystals. In water systems, the surface of MgO reacts with H$^+$ to form a surface layer similar to brucite structure. Then, Mg$^{2+}$ are replaced by H$^+$ to form dissociated Mg$^{2+}$ intermediate, finally spreading to the surface and forming Mg(OH)$_2$ crystal. Meanwhile Fengze Cao [20] found that the shape of Mg(OH)$_2$ crystal depends on the reactivity of MgO. Low reactivity of MgO leads to a formation of an octahedron, while high reactivity of MgO leads to a formation of a hexagonal prism.

Numerous studies have shown that the hydration degree of periclase is affected by many factors. Hongwu Zheng [21] considered that the factors affecting the degree of hydration of periclase includes the number of MgO crystals, the size of the MgO crystal, and the curing environment of cement paste. The hydration kinetics and hydration degree should be determined with certain conditions, such as the number, size, and defects of MgO crystal. Haiyan Qian [22] studied the relationship between hydration degree and time of light-burned MgO at different temperatures. She found that the hydration reaction of light-burned MgO is consistent with the first-order reaction kinetics; that is, the hydration process is controlled by chemical reactions. Jiaping Liu [23] and Changwen Miao [24] found that the effect of increasing temperature on hydration of high activity magnesium oxide expansion agent is more significant. The expansion of high activity magnesium oxide expansion agent at normal temperature is lower than that of low activity magnesium oxide expansion agent, but higher than that of the low activity magnesium oxide expansion agent at high temperature. Therefore, Lanqing Yu [25] considered that MEAs with a low hydration reactivity ranked better in cracking resistance than MEAs with a high hydration reactivity when the light-burned MEA is applied to field concrete poured at a high temperature.

It is well known that the reaction of periclase with water produces magnesium hydroxide (Mg(OH)$_2$) with a volume expansion of about 148% [26,27], resulting in uneven deformation of the paste, eventually causing poor soundness of cement. However, the increase of the volume of hydration product is not exactly equal to the macroscopic expansion produced by MgO in cement paste. There were many explanations for the hydration mechanism of periclase, and most experts agreed with the theory of the swelling of absorbing water proposed by Deng Min [3] and the theory of crystal growth pressure proposed by Chatterji [28]. Based on these, combined with the hydration mechanism of MgO single crystals [28] and polycrystals [29] (Figure 1), Liwu Mo [3] proposed the hydration model of two types of MgO expansion agent in cement paste, including porous MEAs with high activity and less porous MEAs with low activity. He believed that there were three reasons for the different expansions of the two expansion agents. First, the activity of the hydration reaction was different, the second was the internal structure of the MEA, and the third was the difference in the matrix performance of the cement hydration product [3]. Fengze Cao’s research [30,31] on the hydration process of MEAs in mortar showed that the expansion is driven by the recrystallization of Mg(OH)$_2$. The sufficient water supply facilitates the immigration of Mg$^{2+}$ and the recrystallization of Mg(OH)$_2$, which generates a high crystal growth pressure and promotes the expansion of mortars.

Light-burned MgO expansion agent can effectively compensate the shrinkage of cement-based materials, ensure the quality of the project, and have broad prospects for development because of the good expansion properties of periclase in the hydration stage. Due to the complex engineering conditions, the activity and dosage of light-burned MgO expansion agent will change accordingly to achieve the best effect. In this study, the relationship between various conditions and hydration expansion was discussed, with the aim of serving as a guide for the application of MgO expansion agents.
2. Materials and Methods

2.1. Materials

Clinker with low magnesium content (DM1#) (45–80 μm particle size) produced by Jiangnan Onoda Cement Co., Ltd (Nanjing, China) was used in this experiment. The chemical composition, the mineral composition, and the X-ray diffraction (XRD) pattern of DM1# are presented in Tables 1 and 2, and Figure 1, respectively. Gypsum was mixed into the cement system at a concentration of 5% weight of cement to adjust the cementation time of cement clinker, and the whole system was called C1#.

Table 1. The chemical composition of cement clinker (wt.%).

| Material | Loss | SiO₂ | Fe₂O₃ | Al₂O₃ | SO₃ | CaO | K₂O | Na₂O | MgO | Total |
|----------|------|------|-------|-------|-----|-----|-----|------|-----|-------|
| DM1#     | 1.16 | 20.51| 4.47  | 3.89  | 0.38| 65.53| 0.54| 0.23 | 1.81| 98.52 |

Table 2. The mineral composition of cement clinker (wt.%).

| Material | C₃S | C₂S | C₄AF | C₃A | t-CaO | t-MgO | Total   |
|----------|-----|-----|------|-----|-------|-------|---------|
| DM1#     | 60.00| 22.18| 13.19| 3.38| 0.56  | 1.25  | 100.46  |

Herein, four types of light-burned MgO manufactured in Jiangsu Bote New Materials CO., Ltd. (Nanjing, China) were used in the experiments. Hydration activity that was obtained from the citric acid method and other basic physical properties of these samples are summarized in Table 3, and its chemical composition are shown in Table 4. The XRD patterns of light-burned MgO are shown in Figure 2.

Table 3. The basic properties of four MgO.

| Samples | Active Reaction Time (s) | Density (kg/m³) | Specific Surface Area (m²/g) |
|---------|-------------------------|-----------------|-----------------------------|
| A₁      | 65                      | 3790            | 17.99                       |
| A₂      | 110                     | 3500            | 12.25                       |
| A₃      | 143                     | 3430            | 13.13                       |
| A₄      | 190                     | 3730            | 11.69                       |

Figure 1. The XRD pattern of DM1# (A—C₃S, B—C₂S, C—C₃A, F—C₄AF, D—CaO, P—MgO).
Table 4. The chemical composition of four MgO (wt.%).

| Material | SiO₂ | Fe₂O₃ | Al₂O₃ | CaO  | MgO  | K₂O | Na₂O | SO₃ | Loss | Total  |
|----------|------|-------|-------|------|------|-----|------|-----|------|--------|
| A₁       | 2.76 | 0.26  | 0.42  | 3.18 | 88.52| 0.03| 0.06 | 0.61| 3.62 | 99.46  |
| A₂       | 2.46 | 0.29  | 0.43  | 3.15 | 90.02| 0.02| 0.05 | 0.68| 2.54 | 99.64  |
| A₃       | 4.28 | 0.32  | 0.45  | 3.22 | 87.82| 0.02| 0.05 | 0.18| 2.16 | 99.1   |
| A₄       | 3.98 | 0.3   | 0.4   | 3.01 | 89.32| 0.01| 0.04 | 0.63| 2.04 | 99.73  |

Figure 2. The XRD patterns of four light-burned MgO.

2.2. Sample Preparation

C1₈ cement slurry samples were prepared in strict accordance with GB/T 1346-2011 (in China). Then, these materials, including C1₈ cement and four different types of light-burned MgO, were respectively blended well. After being mixed well, cement slurry samples were put in 20 mm × 20 mm × 20 mm cubes for compressive strength test. For measuring the expansion degree of the cement specimens, the specimens were cast into metallic molds, which could separate the cement paste into 20 mm × 20 mm cubes for compressive strength test. For measuring the expansion degree of the cement specimens, the specimens were cast into metallic molds, which could separate the cement paste into 20 mm × 20 mm × 80 mm cuboid specimens. After being solidified (approximately 24h), the cement specimen cubes were stripped and maintained for 1, 3, 7, 14, 28, 60, 90 days respectively at different temperature in water.

2.3. Methods

2.3.1. Expansion Rate Test

The initial length of the solidified (L₀) specimens mixed with different kinds and different mixing amounts of light-burned MgO should be measured immediately. Next, the specimens were cured at different temperature in water until the specified ages for measuring the length of different ages (L₁). The expansions of specimens may be determined as follows:

\[ \text{Expansion (\%)} = (L₁ - L₀) \times 100\% / L₀ \]  (1)

2.3.2. Compressive Strength Test

Cement strength was determined in accordance with the plastic mortar strength test (Chinese Standard GB17671, or ISO 679). Through the curing of the specified period, three samples were selected for each condition and then investigated for compressive strength. Finally, the average value was obtained as the compressive strength of the cement specimens.
2.3.3. Scanning Electron Microscopy (SEM) Characterization

Scanning electron microscopy (SEM) was applied to observe the hydration morphology of these specimens at different ages at 20 °C.

2.3.4. Measurement of Hydration Degree of Periclase in Cement Paste

A quantitative X-ray diffractometric method (XRD internal standard method) using ZnO as an internal standard substance and high-purity MgO, Mg(OH)₂ as admixture has been conducted for determining the content of periclase in above-mentioned cement paste specimens. The measurement data were obtained within the 35–45° range for the specimens at a speed of 0.25°/min with Cu Kα radiation. According to the content of periclase in different hydration ages, the corresponding relationship among the hydration degree of periclase in the cement paste, the curing temperature and the curing time was determined.

3. Results and Discussion

3.1. Expansion Property of Specimens

3.1.1. The Effects of Added MgO Content on the Expansion of Cement Pastes Mixed with Light-Burned MgO

Figure 3 shows the expansion curves of cement pastes containing different hydration activities and different contents of MgO cured at 20 °C in water. It was obvious that any specimens without active MgO expand slightly (about only 0.037%) in the first 7 days, but after 7 days the expansion curve levels off or even falls—namely, cement specimens no longer expand or even begin to shrink. This phenomenon may result from partial irreversible stress expansion caused by the early production (AFt). The mixing of MgO resulted in continuous expansion on cement clinker after 7 days. Additionally, the expansion of cement pastes clearly increased with added content of MgO in cement.

![Graphs showing expansion curves](image)

**Figure 3.** The expansion curves of cement paste mixed with MgO of different activities and amounts at 20 °C curing: (a) M65 s; (b) M110 s; (c) M143 s; (d) M190 s.
It was apparent that the content of MgO did not have a linear relationship with its corresponding expansion rate. The reason may be that specimens were in the plastic expansion stage before cement paste reached full hardening. In this stage, the platy brucite produced by the hydration of MgO interspersed in gel pores generated by cement hardening, resulting in the condition that the enlarged volume from the hydration of MgO did not fully contribute to the expansion value of cement [28], ultimately leading to a nonlinear relationship between the content of MgO and its corresponding expansion rate.

In summary, within a certain range, the greater the MgO content, the greater and faster the expansion rate. In addition, the MgO content did not have a linear relationship with its corresponding expansion rate.

3.1.2. The Effects of Temperature on the Expansion of Cement Paste Mixed with Light-Burned MgO

For observing more obvious expansion phenomena, 10% MgO of different activities was mixed in the cement pastes. The expansion curves of samples mixed 10% different activity MgO at different temperature were presented in Figure 4. As shown in Figure 4, it caused some obvious expansions as the curing temperature increases. At the temperature of 20 °C, 30 °C and 38 °C, these expansion curves continued to increase slowly, namely, the expansion rate grew gradually with the growth of time. Furthermore, it continued to grow after 90 days. When the curing temperature is 60 °C, the expansion of specimens is greatly accelerated, especially for the specimens mixed with MgO (110 s), MgO (143 s) and MgO (190 s), and these corresponding curves all rise slowly or even level off after 14 days. On the condition that the curing temperature is 80 °C, the expansion curve sharply increases before 14 days, while it tends to flatten without growth after 14 days. That is to say, specimens almost reach the “ultimate” expansion through 14 days of maintenance.

![Figure 4](image-url) The expansion curves of cement paste mixed with MgO of different activities at different temperatures: (a) M65 s; (b) M110 s; (c) M143 s; (d) M190 s.
The above reflect that the higher the curing temperature is, the faster the expansion of cement mixed with light-burned MgO is, and the shorter time reaching stable expansion is.

3.1.3. The Effects of MgO Activity on the Expansion of Cement Paste Mixed with Light-Burned MgO

It can be observed in Figure 5a,b that different hydration activities of MgO (9%) have some influences on cement paste. Evidently, the specimens mixed with MgO (65 s) expand rapidly in the early stages at these temperatures, such as an expansion value of 0.079% after 3 days, 0.120% after 7 days, and 0.188% after 14 days at 20 °C. From that point, the growth of the expansion curve levels off. The specimens mixed with MgO (110 s) were similar to the specimens mixed with MgO (65 s). Nevertheless, there was a trend for the specimens mixed with MgO (143 s) and MgO (190 s), except rapid growth in the first three days, that the expansion in the early period was not large, and the increase in the later period became larger, especially curing at 38 °C. Under curing at 60 °C and 80 °C, these samples, with low activity or high activity, reached maximum expansion on about the 28th day (Figure 5c,d).

![Figure 5](image_url)

**Figure 5.** The expansion curves of cement paste mixed with MgO (10%) of different activities at different temperatures: (a) 20 °C; (b) 38 °C; (c) 60 °C; (d) 80 °C.

Notably, in Figure 5a–d, no matter for the expansion speed rate (the slope of the expansion rate curve) or the expansion rate, specimens mixed with low activity MgO were initially smaller than specimens mixed with high activity MgO, but the former exceeded the latter as the curing time increases. The difference was that the increase in temperature accelerated these two surpassing times. Generally speaking, specimens mixed with low activity MgO generated greater expansion.

These demonstrated that at the same curing temperature, the contribution of high activity MgO to the expansion of cement paste was mainly reflected in the early hydration stage, while the compensated supply of MgO with low hydration activity to cement paste was mainly reflected in the later period of hydration, which is consistent with Lanqing
Yu’s research [25]. Moreover, the final expansion produced by the cement mixed with low activity MgO was larger.

3.2. Measurement of Hydration Degree of Periclase in Cement Paste

Figures 6 and 7 and Tables 5 and 6 show at different curing temperatures, the XRD patterns, and corresponding hydration degree of cement paste specimens mixed with 10% MgO of different activities, including 65 s and 190 s. Figures 6 and 7 demonstrate that at the same curing temperature, the intensity of the diffraction peaks of periclase was gradually reduced with the increase of the curing age, instead, the intensity of the diffraction peaks of brucite gradually increased. It meant that with the increase in curing age, the content of periclase in cement paste specimens decreased, the amount of brucite increased, and the hydration degree of periclase increased. These results are also reflected in Tables 5 and 6, which additionally reflect that high activity MgO (65 s) was mainly hydrated in the early hydration stage, while low activity MgO (190 s) was mainly hydrated in the later period of hydration. Curing temperature could speed up the whole hydration process. These phenomena coincided with the law of specimen expansion. It should be pointed out that in the later period of hydration, especially under the curing temperature of 38 °C, 60 °C, and 80 °C, although the hydration degree of low activity MgO was always lower than that of high activity MgO, the expansion rate of specimens mixed with low activity MgO was much higher than that of specimens mixed with high activity MgO. It further explained that the expansion of cement paste mixed with light-burned MgO was not a simple MgO hydration process.

![Figure 6. XRD pattern of hydration slurry mixed with MgO (65 s) at different temperatures: (a) 20 °C; (b) 38 °C; (c) 60 °C; (d) 80 °C.](image_url)
Figure 6. XRD pattern of hydration slurry mixed with MgO (65 s) at different temperatures: (a) 20 °C; (b) 38 °C; (c) 60 °C; (d) 80 °C.

(a)  (b)  
(c)  (d)

Figure 7. XRD pattern of hydration slurry mixed with MgO (190 s) at different temperatures: (a) 20 °C; (b) 38 °C; (c) 60 °C; (d) 80 °C.

Table 5. The hydration degree of MgO (65 s) in cement paste.

| Curing Temperature/°C | Curing Age/Day | Loss/% | Content of Brucite/% | Content of Periclase/% | Hydration Degree/% |
|-----------------------|----------------|--------|----------------------|-----------------------|-------------------|
| 20                    | 3              | 16.82  | 6.23                 | 4.65                  | 45.88             |
|                       | 28             | 23.47  | 6.48                 | 3.95                  | 50.03             |
|                       | 90             | 23.93  | 7.18                 | 3.25                  | 68.64             |
| 30                    | 3              | 17.93  | 7.16                 | 3.76                  | 55.64             |
|                       | 28             | 22.24  | 7.13                 | 3.45                  | 57.05             |
|                       | 90             | 23.29  | 9.35                 | 1.41                  | 82.20             |
| 38                    | 3              | 18.91  | 8.50                 | 2.51                  | 70.01             |
|                       | 28             | 22.6   | 8.83                 | 1.85                  | 77.06             |
|                       | 90             | 24.04  | 9.62                 | 0.95                  | 87.89             |
| 60                    | 3              | 18.97  | 7.52                 | 3.37                  | 59.73             |
|                       | 28             | 21.93  | 9.82                 | 1.01                  | 90.48             |
|                       | 90             | 23.97  | 10.52                | 0.38                  | 95.16             |
| 80                    | 3              | 18.78  | 9.44                 | 1.70                  | 79.73             |
|                       | 28             | 20.46  | 10.62                | 0.68                  | 91.73             |
|                       | 90             | 20.37  | 10.77                | 0.34                  | 96.27             |
Table 6. The hydration degree of MgO (190 s) in cement paste.

| Curing Temperature/°C | Curing Age/Day | Loss/% | Content of Brucite/% | Content of Periclase/% | Hydration Degree/% |
|----------------------|----------------|--------|----------------------|-----------------------|-------------------|
| 20                   | 3              | 18.28  | 4.94                 | 5.68                  | 30.28             |
|                      | 28             | 18.30  | 6.68                 | 4.16                  | 32.59             |
|                      | 90             | 23.23  | 6.38                 | 4.06                  | 58.92             |
| 30                   | 3              | 14.36  | 5.59                 | 5.34                  | 41.94             |
|                      | 28             | 20.50  | 6.04                 | 4.57                  | 46.47             |
|                      | 90             | 22.15  | 7.47                 | 3.14                  | 65.24             |
| 38                   | 3              | 14.07  | 6.61                 | 4.95                  | 43.34             |
|                      | 28             | 16.52  | 6.67                 | 4.29                  | 48.15             |
|                      | 90             | 23.59  | 8.48                 | 2.07                  | 71.77             |
| 60                   | 3              | 16.85  | 6.44                 | 4.47                  | 47.06             |
|                      | 28             | 17.01  | 11.39                | 0.19                  | 83.92             |
|                      | 90             | 23.18  | 9.93                 | 0.76                  | 87.78             |
| 80                   | 3              | 14.30  | 11.42                | 0.48                  | 84.78             |
|                      | 28             | 14.49  | 11.43                | 0.45                  | 85.10             |
|                      | 90             | 22.40  | 10.45                | 0.38                  | 89.44             |

In a word, the increase of temperature and hydration age can improve the hydration degree of periclase. The increase of temperature can promote the compensation of magnesia to cement-based materials in a short time. For cement-based materials with early shrinkage, MgO expansion agents with relatively high hydration activity should be used. On the contrary, for cement-based materials with relatively obvious late shrinkage, MgO expansion agents with lower hydration activity were more suitable. Furthermore, the expansion of cement paste mixed with light-burned MgO was not a simple MgO hydration process.

3.3. Microstructure of Specimens

Figure 8a,b are SEM images of MgO of hydration activity 65 s and SEM images of Mg(OH)\(_2\) after hydration. It is easy to see from the figure that magnesium oxide was clustered together, and these single particles were made up of a large number of magnesium oxide crystallites. After MgO hydrated into Mg(OH)\(_2\), short pieces appeared, and the phenomenon of superimposed flakes was observed.

In Figure 8c,e–g, it can be seen that with the growth of hydration age, MgO gradually hydrated to Mg(OH)\(_2\), diffusing and growing into the surrounding pores in a dispersed state. From Figure 8f, it was distinct that there were a small amount of clear flakes of Mg(OH)\(_2\) crystals at 28 days, while a large number of flaky Mg(OH)\(_2\) crystals appeared under 90d curing (Figure 8g).
Figure 8. Cont.
There are three possible reasons for the different expansion phenomena between the high activity and the low activity MgO. Firstly, the hydration reaction activity was different. Due to the high hydration rate of high activity MgO, a large amount of Mg(OH)₂ can be produced in the early stage of hydration, resulting in rapid expansion in the early stage. Therefore, the amount of MgO supplied in the late hydration was relatively small, and the resulting expansion was also small. Instead, there were more expansion components for the relatively low activity MgO in the late hydration stage. Secondly, the internal structure was different. The hydration of MgO occurred at the grain surface, internal pores [16], and MgO grain boundaries [3] at the same time. On the one hand, the high activity MgO was porous and had a large specific surface area, which provided a certain growing space for the hydration products on the inner pore surface [16]. Hence, the effective expansion of these hydration was small. On the other hand, the atom order at the grain boundary with many defects was irregular, so the atoms here easily reacted with water [29]. Moreover, there was little room left for growth of the hydration product at the grain boundary, resulting in greater expansion here. For high activity MgO (Figure 9), due to the fact that the internal surface area was much larger than the grain boundary area [28], the hydration of internal surface area played a major role. For low activity MgO (Figure 10), contrary to the former, more hydration products were formed at grain boundaries [28], while the growth space at grain boundaries was smaller. Therefore, high activity MgO produced less effective expansion than low activity MgO. Thirdly, the change in properties of cement product matrix was also a considerable reason. Cement paste had viscoelasticity [32] and this viscoelasticity was time-efficient [33]. The cement paste generated viscous flow under the expansion force generated by the hydration of MgO, thereby absorbing part of the expansion energy, and the viscous flow had a relaxing effect on the expansion force, ultimately reducing the expansion [32]. The hydration of high activity MgO occurred mainly in early stages when the viscosity of cement paste is small and easy to produce viscous flow [33]. Therefore, a large amount of expansion energy was consumed in viscous flow, leading to less expansion [29]. However, for the low activity MgO, hydration was concentrated in the late period when the viscous flow was slow, consuming less energy of expansion, finally resulting in more expansion than the former.
Conclusions

In this study, the expansion behavior and hydration degree of cement paste mixed with light-burned MgO were mainly discussed, providing some guidance in designing new cement systems later. The conclusions can be shown as follows.

(1) At the same temperature, the hydration of high activity MgO and the expansion behavior occurred mainly in the early stage of hydration, while the hydration of low activity MgO and the expansion behavior had a high contribution rate in the later stage of hydration. Through hydration model analysis, the reasons found for the differences included three aspects: different hydration reaction activities, distinct internal structure of mixed MgO, and the change in properties of cement product matrix.

For cement-based materials with early shrinkage, MgO expansion agents with relatively high hydration activity should be used. On the contrary, for cement-based materials with relatively obvious late shrinkage, MgO expansion agents with lower hydration activity were more suitable.

(2) The curing temperature greatly influences the hydration speed of MgO. When the other conditions are the same, the higher the curing temperature, the greater the hydration degree of MgO, the greater the expansion rate, the faster the expansion of cement mixed with light-burned MgO.

(3) The increase of MgO content can promote the expansion of cement. The greater the dosage, the greater the expansion rate and the faster the expansion rate, especially when the dosage is more than 3.6%, the growing rate of expansion curve (expansion speed rate) enhanced. Moreover, the content of MgO did not have a linear relationship with its corresponding expansion rate.

(4) The addition of MgO had little effect on the strength of cement paste, only a small reduction, as it counteracted the content of the other components which can provide strength.
**Author Contributions:** Conceptualization, L.X.; tests and data interpretation, Y.W. and C.Z.; writing—original draft preparation, Y.W.; review and editing, Y.W. and L.X. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was financially supported by the National Key Research and Development Plan of China (2016YFB0303601-2), and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

**Data Availability Statement:** The data supporting the findings of this paper are available from the corresponding authors upon reasonable request.

**Acknowledgments:** The authors would like to acknowledge the assistance from Tao Feng, Ruijie Jiang, Xin Shi, Nan Wu and Jian Han from Nanjing Tech University.

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

**References**

1. Lea, F.M. *The Chemistry of Cement and Concrete*; Chemical Publishing Company: New York, NY, USA, 1971.
2. Mehta, P. *History and Status of Performance Tests for Evaluation of Soundness of Cements*; ASTM International: West Conshohocken, PA, USA, 1978.
3. Mo, L.W. Microstructure and Property of MgO-Type Expansive Agent. Ph.D. Thesis, Nanjing Tech University, Nanjing, China, 2008.
4. Mehta, P.; Pirtz, D. Magnesium oxide additive for producing selfstressed mass concrete. In Proceedings of the 7th International Congress on the Chemistry of Cement, Paris, France, 30 June–4 July 1980.
5. Li, C. Review of quick damming technology of MgO concrete. *Adv. Sci. Technol. Water Resour.* 2013, 33, 82–87. [CrossRef]
6. Gao, P.W.; Wu, S.X.; Lu, X.L.; Deng, M.; Lin, P.H.; Wu, Z.R.; Tang, M.S. Soundness evaluation of concrete with MgO. *Constr. Build. Mater.* 2007, 21, 132–138. [CrossRef]
7. Zhu, B. On construction of dams by concrete with gentle volume expansion. *J. Hydroelectr. Eng.* 2000, 1–13. [CrossRef]
8. Ye, Q.; Chen, H.X.; Wang, Y.Q. Effect of MgO and gypsum content on long-term expansion of low heat Portland slag cement with slight expansion. *Cem. Concr. Compos.* 2004, 26, 331–337. [CrossRef]
9. Gao, P.; Lu, X.; Geng, F.; Li, X.; Hou, J.; Lin, H.; Shi, N. Production of MgO-type expansive agent in dam concrete by use of industrial by-products. *Build. Environ.* 2008, 43, 453–457. [CrossRef]
10. Xu, L.L.; Deng, M. Dolomite used as raw material to produce MgO-based expansive agent. *Cem. Concr. Res.* 2005, 35, 1480–1485. [CrossRef]
11. Xu, L.L.; Deng, M.; Wang, X.; Zhao, X. Studies on preparation of a new expansive agent based on MgO. *J. Mater. Sci. Eng.* 2004, 22, 249–253. [CrossRef]
12. Xu, L.L.; Deng, M.; Zhao, X. Compensating shrinkage of cement paste by new MgO-based expansive material. *J. Build. Mater.* 2005, 8, 67–70. [CrossRef]
13. Zhao, X.; Deng, M. Preparation and Performance of MgO-type Expansive Agent. *J. East China Inst. Technol.* 2005, 28, 71–75. [CrossRef]
14. Cao, F.; Liu, Y.; Yan, P. Properties and mechanism of the compound MgO expansive agent (CMEA) produced by calcining the mixture of dolomite and serpentine tailings. *Constr. Build. Mater.* 2021, 277, 122331. [CrossRef]
15. Liu, P.C.; Deng, M. Regulating the Expansion Characteristics of Cementitious Materials Using Blended MgO-Type Expansive Agent. *Materials* 2019, 12, 976. [CrossRef] [PubMed]
16. Liu, P.; Kendelevicz, T.; Brown, G.E.; Parks, G.A. Reaction of water with MgO (100) surfaces. Part I: Synchrotron X-ray photoemission studies of low-defect surfaces. *Surf. Sci.* 1998, 412, 287–314. [CrossRef]
17. Liu, P.; Kendelevicz, T.; Brown, G.E.; Parks, G.A. Reaction of water with MgO (100) surfaces. Part II: Synchrotron photoemission studies of defective surfaces. *Surf. Sci.* 1998, 412, 315–332. [CrossRef]
18. Mejias, J.A.; Berry, A.J.; Refson, K.; Fraser, D.G. The kinetics and mechanism of MgO dissolution. *Chem. Phys. Lett.* 1999, 314, 558–563. [CrossRef]
19. Xu, Y.M.; Xu, L.L.; Li, W.W. Research Progress of the Periclase and Quantitative Method in Cement Clinker. *Mater. Rep.* 2013, 27, 355–358.
20. Cao, F.; Miao, M.; Yan, P. Effects of reactivity of MgO expansive agent on its performance in cement-based materials and an improvement of the evaluating method of MEA reactivity. *Constr. Build. Mater.* 2018, 175, 257–266. [CrossRef]
21. Zheng, H.W. Magnesium Oxide in Cement. *Fujian Build. Mater.* 2000, 3, 9–11.
22. Qian, H.Y.; Li, S.Y.; Deng, M. Hydrochemical Kinetics of Light-Burned Magnesia. *Ind. Miner. Process.* 2007, 12, 1–4.
23. Liu, J.P.; Wang, Y.J.; Tian, Q.; Zhang, S.Z. Temperature sensitivity of light calcined magnesia expansion agent and its mechanism analysis. *J. Southeast Univ.* 2011, 41, 359–364.
24. Yu, L.Q.; Deng, M.; Mo, L.W. Effects of Lightly Burnt MgO Expansive Agent on the Deformation and Microstructure of Reinforced Concrete Wall. *Adv. Mater. Sci. Eng.* 2019, 4, 1948123. [CrossRef]
25. Tian, Q.; Tu, Y.J.; Liu, J.P.; Miao, C.W. Temperature Sensitivity Analysis on Expansive Property of MgO Composite Expansion Agent. *Water Power* 2010, 36, 49–51, 86. [CrossRef]

26. Ye, Q.; Yu, K.; Zhang, Z. Expansion of ordinary Portland cement paste varied with nano-MgO. *Constr. Build. Mater.* 2015, 78, 189–193. [CrossRef]

27. Chatterji, S. Mechanism of expansion of concrete due to the presence of dead-burnt CaO and MgO. *Cem. Concr. Res.* 1995, 25, 51–56. [CrossRef]

28. Kitamura, A.; Oniduka, K.; Tanaka, K. Hydration characteristics of magnesia. *Taikabutsu Overseas* 1996, 16, 3–11.

29. Cao, F.; Yan, P. The influence of the hydration procedure of MgO expansive agent on the expansive behavior of shrinkage-compensating mortar. *Constr. Build. Mater.* 2019, 202, 162–168. [CrossRef]

30. Cao, F.; Miao, M.; Yan, P. Hydration characteristics and expansive mechanism of MgO expansive agents. *Constr. Build. Mater.* 2018, 183, 234–242. [CrossRef]

31. Salomao, R.; Bittencourt, L.; Pandolfelli, V. A novel approach for magnesia hydration assessment in refractory castables. *Ceram. Int.* 2007, 33, 803–810. [CrossRef]

32. Jennings, H.M.; Bullard, J.W.; Thomas, J.J.; Andrade, J.E.; Chen, J.J.; Scherer, G.W. Characterization and modeling of pores and surfaces in cement paste. *J. Adv. Concr. Technol.* 2008, 6, 5–29. [CrossRef]

33. Thomas, J.J.; Jennings, H.M. A colloidal interpretation of chemical aging of the C-S-H gel and its effects on the properties of cement paste. *Cem. Concr. Res.* 2006, 36, 30–38. [CrossRef]

34. Mo, L.W.; Deng, M.; Tang, M.S. Effects of calcination condition on expansion property of MgO-type expansive agent used in cement-based materials. *Cem. Concr. Res.* 2010, 40, 437–446. [CrossRef]