Effects of Water-to-Cement Ratios on the Properties of Magnesium Potassium Phosphate Cement Prepared with Lithium-Extracted Magnesium Residue

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Abstract: Salt lake magnesium phosphate cement (SLMKPC) was prepared by mixing potassium dihydrogen phosphate (KH₂PO₄) with lithium-extracted magnesium residue (MR). The hydration-hardening process and the variation in the phase composition and microstructure of hydration products with the change of the water-cement-ratio (W/C) were studied by measuring the setting time, hydration exothermic temperature, and compressive strength of the hardened slurry. It was found that W/C ratios had significant effects on the setting time, hydration exothermal temperature curve, and compressive strength of SLMKPC. With the increase of W/C, the setting time was prolonged significantly. The exothermic hydration peak temperature first increased and then decreased, and the number of exothermic peaks gradually changed from one to two. The optimal compressive strength was obtained when W/C = 0.2–0.3. Hydration products changed significantly with the increase of W/C, the MgKPO₄·6H₂O (MKP) in the matrix was more stable when W/C = 0.2 and 0.3 than when W/C = 0.25, 0.35, and 0.4. When W/C = 0.3, SLMKPC had a compact structure with a certain operating space and better mechanical properties.

Keywords: magnesium residue; magnesium potassium phosphate cement; water-to-cement ratio; preparation; property

1. Introduction

Lithium, as an energy material, plays an important role in mitigation problems caused by fossil fuels. The application of lithium-ion batteries in electric vehicles can reduce environmental pollution caused by traditional vehicles [1,2]. Lithium is abundantly found in salt lake brine [3]. Lithium is mainly extracted from salt lake brine by chemical precipitation for industrial applications [4,5]. In order to remove high Mg²⁺ concentration, hydroxide or carbonate are generally added, producing a large amount of magnesium residue (MR) [6]. The accumulation of MR in lithium production areas affects the development environment of lithium resources and also causes a serious waste of magnesium resources. It is of great significance to use MR as a raw material to develop other materials and promote the effective utilization of by-products produced by lithium extraction.

It is generally found that MR contains a large amount of magnesium hydrate (Mg(OH)₂) and magnesium oxide (MgO). These magnesium-containing compounds are favourable raw materials for the synthesis of magnesium phosphate cement (MKPC). MKPC is a chemically bonded ceramic with early strength and fast hardness. MKPC is obtained by the reaction between MgO and potassium dihydrogen phosphate (KH₂PO₄) in an aqueous solution [7].

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\text{MgO} + \text{KH}_2\text{PO}_4 + 5\text{H}_2\text{O} \rightarrow \text{MgKPO}_4\cdot6\text{H}_2\text{O (MKP)}
\]
Similar to periclase MgO-based binding materials, MKPC is capable of yielding quick hardness and early strength [8–10]; thus, it is extensively used in rapid repairing and biological bone repairing applications, structural coatings, porous ceramics, and waste curing [11–13]. MgO used in MKPC is dead-burned magnesia produced from magnesite ore calcined at 1500–1700 °C [14]. The demand for high-quality magnesite has increased greatly in recent years [15], and the production of dead-burned magnesia requires higher energy. The high cost of dead-burned MgO and the shortage of raw ore make MKPC more expensive and limit its practical applications [16].

Moreover, boric acid, alkali, and alkaline earth metal plasma are generally found in salt lake brine [17]; however, it is difficult to completely extract boron [18]. Boric acid (H₃BO₃) or borax (Na₂B₄O₇·10H₂O) [19,20] can significantly regulate acid-base reactions to improve the strength or workability of an MKPC system [21]; therefore, boric acid or borax are used as reaction retarders for MKPC [22].

When MR is used as a magnesium oxide raw material of MKPC, no additional retarder is required, and the retarded time can be regulated by changing the calcination temperature of MR [23–25]. The preparation of MKPC using MR as a raw material consumes a large amount of salt lake by-products to alleviate environmental pollution and reduce the product cost and also makes full use of the characteristics of boron-rich by-products.

Numerous studies have been performed to explore the effects of water-to-cement ratios (W/C) [26,27], magnesia-to-phosphate ratios (Mg/P) [9,28,29], retarders and additives on the properties [21,30–33], and thermal Stability [34,35] of MKPC. It is generally noticed that W/C has important effects on the mechanical properties, hydration products, and workability of MKPC [36,37]. The MgO raw material used in these studies was usually commercially available MgO, which has higher purity and stability, and relatively higher price. Tan Y S and Chen W H et al. [23–25] reported that salt lake Magnesium Phosphate Cement (SLMKPC) was prepared by MR and good mechanical properties were obtained. The above studies indicate that it was feasible to use MR instead of Normal MgO for the formation of MKPC materials and the utilization of raw materials. Meanwhile, the economic cost of materials will be greatly reduced due to the reuse of waste resources. Moreover, the MgO/KH₂PO₄ mass ratio also has a significant influence on the hydration heat curve, compressive strength, and porosity of SLMKPC. However, the effects of W/C on the properties of SLMKPC are still not clear.

Therefore, in the present work, the influences of W/C ratios on the condensation time, hydration exothermal temperature, and strength performance of SLMKPC were discussed. X-ray diffraction (XRD) and scanning electron microscopy (SEM) were used to analyze the microstructures of hydration products and SLMKPC.

2. Materials and Experimental Procedure

2.1. Raw Materials

Potassium dihydrogen phosphate (KH₂PO₄; ≥99%) was purchased from Sinopharm Chemical Reagents Co Ltd. Distilled water was used as a curing liquid and prepared in the laboratory. Magnesium slag with a milky white appearance was obtained as a waste residue after lithium extraction. The magnesium slag was analyzed by chemical titration and XRD, and the main chemical components of MR were Mg²⁺ and B₂O₃ (Table 1). XRD results (Figure 1) revealed that Mg²⁺ in MR was a mixture of MgO and Mg(OH)₂. The quantitative analysis in Topas 4.2 software revealed that MR was chemically composed of Mg(OH)₂ (53%), MgO (21%), Mg₃B₂O₆ (23%), CaCO₃ (2%), and NaCl (1%). A high MgO is beneficial for the mechanical properties and economy of magnesium-based materials [38]. Therefore, it was necessary to conduct heat treatments on MR to completely convert Mg(OH)₂ into MgO.
Table 1. Chemical composition of the magnesium residue.

| Component   | Mg$^{2+}$ | Ca$^{2+}$ | B$_2$O$_3$ | SiO$_2$ | Al$^{3+}$ | Fe$^{2+}$ | K$^+$ | Li$^+$ | Na$^+$ | Cl$^-$ | SO$_4^{2-}$ | Loss on Ignition |
|-------------|-----------|-----------|------------|--------|-----------|----------|-------|-------|--------|-------|-------------|------------------|
| Content (%) | 39.55     | 0.58      | 4.22       | 0.029  | 0.022     | 0.21     | 0.044 | 0.019 | 0.12   | 0.88  | 0.32        | 25.68            |

Figure 1. XRD spectrum of MR.

2.2. Raw Material Handling

Calcined magnesium residue (CMR) with different MgO contents was obtained after calcining MR at different temperatures situation, and MgO in CMR had different active MgO contents (MgO$_a$) at different calcination temperatures. It is observable from Figure 2 that when the calcination temperature increased from 400 °C to 1200 °C, the activity of CMR first increased and then decreased and the peak temperature of MgO$_a$ reached 700 °C. For MPC materials, the activity of magnesium oxide is proportional to the reaction rate, and a fast reaction rate is not effective for sample forming and preparation [39,40]; therefore, it was necessary to find an appropriate temperature condition to maximize the physical properties of SLMKPC.

Figure 2. MgO$_a$ content and compressive strength (curing time = 1 day) changing curve with calcination temperature.
It is also noticeable from Figure 2 that when the calcination temperature was less than 800 °C, the setting time was less than 3 min and molded SLMKPC was not obtained when the reaction speed was too fast. However, when the calcination temperature was greater than 900 °C, high-strength SLMKPC was obtained. SLMKPC samples prepared by CMR at the calcination temperatures of 1000 °C and 1200 °C had the best mechanical properties (Figure 2). In previous studies [23–25], it was found that MKPC produced by calcined B-MgO (1000 °C) had good mechanical properties and a long setting time.

According to the above analysis, MR was calcined in advance in this experiment. MR was calcined in a box resistance furnace at 1000 °C for 1 h to obtain CMR. After natural cooling, the CMR was crushed to 10 mm particles, ground in a fast grinding mill for 1.5 min, and then passed through a 120-mesh sieve. The specific surface area and the average particle size were 30.118 m²/g and 10.527 µm, respectively.

2.3. Mixing Process

In order to prepare SLMKPC samples, KH₂PO₄ was first dried for 180 min in a vacuum oven at 60 °C, then mixed with CMR, and stirred evenly. The mass ratio of CMR to KH₂PO₄ was 1.5:1, which is typical for MKPC pastes and mortars [25,26]. The mixed powder was then ground in a ball mill for 40 min and passed through a 200-mesh sieve. Distilled water was mixed with the resultant particles to obtain an SLMKPC slurry.

The slurry was cast in a 20 mm × 20 mm × 20 mm mold. The specimens were cured at room temperature for 30–60 min, then removed from the mold, and cured in an environment of 20 ± 3 °C and 50% relative humidity. The micro-morphology and mechanical performance of the samples obtained after 60 min, 180 min, 1 d (day), 3 d, 7 d, 28 d, 60 d, and 90 d of curing were determined.

2.4. Test Methods

The setting time of the SLMKPC slurry was determined by a Vicat needle according to the ASTM standard C191 [16]. As SLMKPC could set very fast and its initial setting time was close to the final one; only the final setting time was measured. The hydration exothermic characteristics of the SLMKPC slurry were measured by an automatic temperature recorder. At an ambient temperature of 22 ± 2 °C, 30 g of SLMKPC dry powder was mixed evenly for standby applications. A certain amount of distilled water was taken according to the W/C, and its minimum temperature was measured. After 1 min, distilled water was poured into the dry powder, mixed, and stirred evenly, and put into an insulated container. The mechanical performance of SLMKPC was investigated on a universal testing machine (CMT6104; loading speed = 1 ± 0.5 mm/min).

When SLMKPC reached the critical test period, six parallel samples were tested and the average value was taken as the final compressive strength. The mineralogical compositions of the SLMKPC samples were measured by X-ray (XRD, X’pert Pro PANalytical X’PROPert, Almelo, The Netherlands). XRD spectra were fitted by Bruker AXS (2006) TOPAS 4.2 software (Bruker Company, Billerica, MA, USA), and the quantitative analysis of each phase was carried out by the Rietveld method [41]. SEM (JSM-5610LV, JEOL, Tokyo, Japan) was performed on dried gilded specimens [42].

3. Results

3.1. Effect of W/C Ratios on the Setting Time of SLMKPC Slurry

Figure 3 presents the relationship between the setting time and W/C ratios of SLMKPC. It is noticeable that W/C ratios had a great influence on setting time. With the increase of W/C, the setting time of SLMKPC was prolonged. When W/C was less than 0.3, the setting time increased greatly. When W/C = 0.25, the setting time increased by 31% as compared to that with W/C = 0.2. When W/C = 0.3, the setting time was 24% higher than that with W/C = 0.25. However, the increment of setting time became slow after W/C = 0.3, and the setting time increased by 15% and 8%, respectively, as compared to those with W/C between 0.35 and 0.3, 0.4, and 0.35.
Figure 3. Influence of W/C ratios on the setting time of SLMKPC slurry.

The SLMKPC slurry reacted violently with a relatively fast reaction rate when W/C was less than 0.35. When W/C < 0.3, the setting time was below 10 min, whereas when W/C = 0.3, the setting time was above 10 min. Therefore, a high W/C ratio was helpful to prolong the hydration reaction time of the SLMKPC slurry, and W/C = 0.3 was the optimal value for the SLMKPC slurry setting time.

3.2. Effects of W/C Ratios on the Hydration Temperature Curve of SLMKPC Slurry

Figure 4 displays the variation trend of the hydration exothermic curve of SLMKPC with W/C in 400 min. The hydration exothermal curve of the SLMKPC slurry had an acceleration period in the beginning, and the hydration heat reached the peak and decreased slowly with the prolonged time. The exothermic peak of hydration manifested different characteristics with the changing W/C ratio.

Figure 4. Influence of W/C ratios on the hydration heat release characteristics of SLMKPC slurry.

When W/C < 0.3, only one exothermic peak was detected and the maximum exothermic peak temperatures corresponding to W/C = 0.2, 0.25, and 0.3 were 63.2 °C, 68.5 °C, and 69.7 °C, respectively. It implies that the hydration reaction of SLMKPC became more violent in the W/C range of 0.2–0.3. When W/C = 0.2, the moisture content in the SLMKPC slurry was insufficient to allow all CMR and KH$_2$PO$_4$ to react. When the W/C ratio increased from 0.2 to 0.3, CMR and KH$_2$PO$_4$ completely reacted with water, and subsequently, the highest exothermic temperature of hydration was obtained at W/C = 0.3.
When the W/C ratio increased from 0.35 to 0.4, the number of hydration exothermic peaks changed from one to two, and the maximum temperature of the first exothermic peak was lower than that of the second one. When W/C = 0.4, the exothermic peak temperature of hydration was less than that with W/C = 0.35, indicating that the excessive liquid played a certain blocking effect on the reaction of the slurry and reduced the exothermic reaction temperature.

3.3. Effects of W/C Ratios on the Compressive Strength of SLMKPC

Figure 5 expresses the effects of W/C ratios on the compressive strength of SLMKPC. The pre-compressive strength (≤1 d) of SLMKPC decreased with the increase of W/C, whereas the long-term strength (≥3 d) first increased and then decreased with the increase of W/C. When W/C = 0.25, the compressive strength of SLMKPC after each aging period was high. When W/C ≤ 0.25, the compressive strength of the hardening body was the highest, whereas the compressive strength after 1 h of aging was 70% higher than that after 28 d of aging. The pre-compressive strength of SLMKPC was relatively low when W/C was greater than 0.30, whereas the long-term strength was significantly high.

The compressive strength differences of different W/C samples after 3 d and 28 d of curing were ΔW/C = 0.2 = 1.7 MPa, ΔW/C = 0.25 = 3.1 MPa, ΔW/C = 0.3 = 2.1 MPa, ΔW/C = 0.35 = 20.4 MPa, and ΔW/C = 0.4 = 10.6 MPa. Therefore, higher W/C ratios significantly affected the compressive strength of SLMKPC. When W/C = 0.35–0.4, the compressive strength of SLMKPC was noticeably lower than that at W/C = 0.2–0.3.

3.4. Effect of W/C Ratios on the Phase Composition of SLMKPC

Figure 6a,b displays the phase compositions of SLMKPC after 3 d and 28 d of aging at different W/C ratios, respectively. The main hydration products of SLMKPC were MKP, MgO, and Mg₃B₂O₆.

A little variation was detected in the XRD patterns of SLMKPC samples prepared at different W/C ratios, indicating that the change of W/C had no effect on the phase composition of hydration products. The quantitative analysis of SLMKPC samples with different W/C ratios after 3 d and 28 d of aging was carried out in Topas 4.2 software (Figure 6c), and a significant difference was noticed in the phase composition of the samples.

The quantity of MKP in the samples with different W/C ratios after 3 d of aging was relatively small and increased to different degrees after 28 d. The growth rate of MKP at W/C = 0.35–0.4 was greater than that at W/C = 0.2–0.3, and it is consistent with the development rule of compressive strength. When W/C = 0.2–0.35, the amount of MKP was relatively high, whereas when W/C = 0.35–0.4, the amount of MKP was relatively
small. Moreover, the amount of MKP changed with the increase of the W/C ratio and the critical period.

![Figure 6](image-url)  
**Figure 6.** Influence of W/C ratios on the phase composition of SLMKPC: (a) After 3 d of curing, (b) after 28 d of curing, (c) TOPAS quantitative analysis of the phase compositions of SLMKPC at different W/C ratios.

### 3.5. Effects of W/C Ratios on the Microstructure of SLMKPC

Figure 7 exhibits the micro-morphologies of SLMKPC samples with different W/C ratios after 28 d of curing. When W/C = 0.2, the SLMKPC sample was composed of a large number of rod-like crystals, which were closely spliced together to form a thin and dense sheet structure consisting of a few micro-cracks (Figure 7a). When the W/C ratio increased to 0.25, a large number of columnar hydration products existed in the hardened SLMKPC sample with a high degree of crystallization (Figure 7b). These hydration products were tightly packed with each other, no cracks were detected in the microstructure, and the highest compressive strength was obtained. When W/C = 0.3–0.4, the fracture and crystallization degrees of hydration products were the worst; hence, the overall structure of the sample was loose due to the existence of small cracks and voids (Figure 7c–e). When W/C = 0.35–0.4, crystals ruptured into granular ones (Figure 7d,e); hence, the compressive strength was low due to the loose crystal structure (Figure 5).
4. Discussion

It is evident from Figure 3 that a large W/C ratio resulted in a longer setting time. In addition to a large W/C ratio, an optimal W/C range enabled SLMKPC to obtain the best mechanical properties with a better operating space. The performance of MKPC was limited by a reasonable W/C range.

It is noticeable from Figure 8a that when W/C > 0.4, the slurry was very watery. Due to the presence of excess water in the solidification process, the porosity of the final sample was too large (Figure 7e); thus, the SLMKPC sample had a loose structure and low mechanical properties (Figure 5). The SLMKPC slurry was very dry at W/C < 0.2 (Figure 8c) and could not achieve operational capability due to excessive moisture loss. The optimal value Mg/P was dependent on the W/C ratio [22]. A W/C ratio of greater than 0.2 was sufficient to guarantee a complete reaction of KH$_2$PO$_4$ [43]. Therefore, the optimal W/C range for the best setting performance of SLMKPC was 0.2–0.4.

When W/C was between 0.2–0.3, only one exothermic peak appeared in the initial hydration stage (Figure 4). When W/C = 0.2, the moisture content in the SLMKPC slurry was insufficient to allow all CMR and KH$_2$PO$_4$ to react. Due to the residual part of MgO in the CMR, the hydration heat release rate and total amount were relatively low under the condition of the same raw materials. When the W/C ratio increased from 0.2 to 0.3, CMR and KH$_2$PO$_4$ completely reacted with water and reached a critical point,
resulting in the highest exothermic hydration temperature. The formation of MKPC was accompanied by a typical heat evolution curve consisting of an endothermic valley and two exothermic peaks [16]. The results show that the purity and stability of the raw material can be guaranteed because the raw material of MKPC is commercial MgO. Therefore, two exothermic peaks can be obtained in the W/C of MKPC at different stages. Two exothermic peaks only appeared when W/C was adjusted to a certain proportion in the SLMKPC slurry. When the W/C increased from 0.35 to 0.4, the number of exothermic peaks in the SLMKPC slurry changed from one to two (Figure 4).

![Figure 8](image)

**Figure 8.** Influence of W/C ratios on the setting performance of SLMKPC.

In the SLMKPC system with only one exothermic peak, a large amount of heat concentrated to release, resulting in a partial performance loss in the later period. Moreover, this exothermic peak had no significant effect on the mechanical properties and microstructure of the material. In contrast, the SLMKPC system with two exothermic peaks (W/C = 0.35–0.4) reduced the exothermic temperature and slowed down the rate of hydration (Figure 5); however, a large W/C ratio resulted in inferior mechanical properties (Figure 5) and a loose microstructure (Figure 7).

The typical setting time of an MPC system without a retarder is less than 3 min [16], and it can be extended to above 20 min after the addition of boric acid, mineral ingredients, and other retarders [44]. However, the setting time of SLMKPC without any retarder could not reach above 20 min [34]. When W/C > 0.3, the setting time reached above 10 min. Therefore, in order to convert a large quantity of MR feedstock into usable SLMKPC, the setting time corresponding to W/C = 0.2–0.3 should be considered.

According to Equation (1), the amount of MgO in the system decreased with the increase of the MKP content during the reaction. The quantitative analysis of SLMKPC (Figure 6c) revealed that the quantities of MgO and MKP in all SLMKPC samples after 3 d of curing were higher than those after 28 d of curing, and this phenomenon is different from the MKP generation process. Mg$_3$B$_2$O$_6$ played a regulating role in the absorption and release of MgO in the system because the boron compound precipitated around MgO grains in the reaction or formed a magnesium borate complex [45,46]. With the prolongation of the critical period, the precipitated magnesium borate complex was decomposed and released a small number of MgO grains.

Hydration products were the strength of the MKPC system [26–30]. It is noticeable from Figure 6c that although the amount of MKP in SLMKPC after 3 d was smaller than that after 28 days, the difference was not very large; however, the intensity difference was obvious. The compressive strength differences of different W/C samples after 3 d and 28 d of curing were ΔW/C = 0.2 = 1.7 MPa, ΔW/C = 0.25 = 3.1 MPa, ΔW/C = 0.3 = 2.1 MPa, ΔW/C = 0.35 = 20.4 MPa, and ΔW/C = 0.4 = 10.6 MPa (ΔW/C = 0.2 < ΔW/C = 0.3 < ΔW/C = 0.25 < ΔW/C = 0.35 < ΔW/C = 0.35).
It is clear that the stability of SLMKPC samples with different W/C ratios was different. The SLMKPC system with W/C = 0.2 and 0.3 was more stable than those with W/C = 0.25, 0.35, and 0.4. When W/C = 0.3, SLMKPC had the best mechanical properties (Figure 5) and a compact structure (Figure 7) [47]. Generally, the W/C of MKPC prepared with commercially available MgO is between 0.2–0.25, in which MKPC has the highest mechanical properties [46]. The optimal W/C = 0.3 for SLMKPC is slightly higher than when W/C = 0.25, which may be caused by the fact that MR has more impurities and a larger specific surface than commercially available MgO.

5. Conclusions

(1) As MR contains a large amount of Mg(OH)$_2$, when the calcination temperature was less than 800 °C, its setting time was less than 3 min; hence, SLMKPC samples could not be obtained at a too fast reaction speed. When the reaction between raw materials occurred at a temperature greater than 900 °C, strong SLMKPC samples were obtained.

(2) W/C ratios had strong effects on the condensation time, hydration exothermic temperature, and compressive strength of SLMKPC. The setting time increased from 7 min to 12 min with the increase of W/C from 0.2 to 0.4. The exothermic hydration peak temperature first increased and then decreased, and the number of exothermic peaks gradually changed from one to two. The compressive strength of SLMKPC first increased and then decreased with the increase of W/C, and the optimal compressive strength was obtained when W/C = 0.25.

(3) The SLMKPC system without any retarder had a longer setting time than the MPC system without any retarder. When W/C > 0.2, the setting time reached more than 7 min. In order to convert a large amount of MR into applicable SLMKPC, the setting time corresponding to W/C = 0.2–0.3 should be considered.

(4) With the increase of W/C, the morphology and structure of hydration products in SLMKPC gradually became loose and the number of cracks simultaneously increased. When W/C = 0.2–0.3, hydrated products of SLMKPC had a high degree of crystallization and a regular columnar arrangement and the matrix had a dense structure with few microcracks.

(5) B$^{3+}$ ions played a retarding role in the SLMKPC system. The content of Mg$_3$B$_2$O$_6$ in SLMKPC after 28 d of curing was higher than that after 3 d. It indicates that B$^{3+}$ ions involved in the deposition of magnesia grains in SLMKPC were released slowly with the increasing curing time.

(6) The main hydration product of SLMKPC at different ages was MKP. The compressive strength differences of different W/C samples after 3 d and 28 d of curing were $\Delta_{W/C=0.2}=1.7$ MPa, $\Delta_{W/C=0.25}=3.1$ MPa, $\Delta_{W/C=0.3}=2.1$ MPa, $\Delta_{W/C=0.35}=20.4$ MPa, and $\Delta_{W/C=0.4}=10.6$ MPa ($\Delta_{W/C=0.2}<\Delta_{W/C=0.3}<\Delta_{W/C=0.25}<\Delta_{W/C=0.4}<\Delta_{W/C=0.35}$). The SLMKPC systems with W/C = 0.2 and 0.3 were more stable than those with W/C = 0.25, 0.35, and 0.4. Therefore, MR calcined at 1000 °C could produce excellent SLMKPC with a compact structure, a certain operating space, and better mechanical properties when W/C = 0.3.

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