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Effect of solution concentrations and replacement levels of metakaolin on the resistance of mortars exposed to magnesium sulfate solutions

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Abstract

This paper reports an experimental study on the magnesium sulfate resistance of mortar and paste specimens incorporating 0%, 5%, 10% and 15% metakaolin (MK). The resistance of mortar specimens was evaluated using visual examination, reduction in compressive strength and expansion measurements.

Results confirmed that mortar specimens with a high replacement level of metakaolin showed lower resistance to a higher sulfate concentration of magnesium solution. However, in a lower concentration, there were no visibly remarkable differences in the deterioration of mortar specimens, even up to 360 days of exposure, regardless of replacement levels of metakaolin.

The negative effect of metakaolin on the magnesium sulfate resistance is partially attributed to the formation of gypsum but not ettringite and thaumasite. In addition, the reduction of calcium hydroxide and the increase of secondary C–S–H in the cement matrix due to pozzolanic reaction of metakaolin provided an opportunity to lead to the conversion of primary and secondary C–S–H gel into the M–S–H gel.

It is concluded that it is necessary to pay special attention when using metakaolin in concrete exposed to highly concentrated magnesium sulfate solution.

Keywords: Metakaolin; Sulfate attack; Reduction in compressive strength; Expansion; Gypsum

1. Introduction

In recent years, metakaolin (MK) has been studied because of its high pozzolanic properties [1–4]. It is well known that MK is a reactive aluminosilicate, which is formed by the dehydroxylation of kaolin precursor upon heating in the temperature range of 700–800 °C [5,6]. The effects of MK on the durability and mechanical properties of mortar or concrete have been widely reported [7–9].

The performance of concrete incorporating MK, at appropriate replacement levels, is similar to that of concrete containing silica fume. Recent works have shown that MK is effective as a supplementary cementitious material on improving the durability of concrete, for example, alkali–silica reaction [10] and resistance to chloride ingress [11]. In particular, concerning sulfate resistance of MK, Khatib and Wild [7] found that mortars containing higher replacement level of MK exhibited a superior resistance against sodium sulfate attack during test period of 520 days, because of significant consumption of calcium hydroxide and refinement of pore structure due to the pozzolanic reaction of MK. Ramlochan and Thomas [12] also showed that high-reactivity metakaolin has a beneficial effect on the resistance to sodium sulfate attack of mortar at the replacement level of 15% or more. However, few studies have been reported on the resistance of MK to magnesium sulfate attack, although there have been reported works related to the magnesium sulfate resistance of silica fume in the literature [13–15]. In fact, magnesium sulfate is known to be particularly corrosive in sulfate attacks [16], and magnesium ions are not uncom-

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mon in groundwater [17]. Mainly, magnesium sulfate attack on the cement matrix is a disintegration of the C–S–H gel to M–S–H (or M–C–S–H) gel, which is a noncementtitious material [18–21]. Thus, the characteristics of C–S–H gel formed by pozzolanic reaction of MK may need to be considered when concrete containing MK is exposed to magnesium sulfate environment.

The objective of this study is to investigate the resistance against sulfate attack, in particular, magnesium sulfate attack, of mortar specimens containing MK exposed to different solution concentrations. Experimental results include visual examination, reduction in compressive strength and expansion. In addition, to identify the products formed by the sulfate attack, X-ray diffraction was performed on paste samples with or without MK, which has been subjected to magnesium sulfate attack for 360 days.

2. Experimental details

2.1. Materials

The cement used in this study was a commercial grade ASTM C150 Type I portland cement, which had been produced by S Cement in South Korea. The physical properties and chemical composition data, as provided by the cement manufacturer, are presented in Table 1. The C_3S, C_2S, C_3A and C_4AF contents of the cement by Bogue calculation were 54.9%, 16.6%, 10.3% and 9.1%, respectively. The chemical composition of MK used as supplementary cementitious material is also given in Table 1. The MK was supplied by K Chemical.

River sand fine aggregate, which is immune to most chemical agents and has little organic material, was employed for manufacturing mortar specimens. The fine aggregate was less than 5 mm. The specific gravity, absorption and fineness modulus of the fine aggregate were 2.60, 0.80% and 2.80, respectively. To obtain a suitable workability, a polycarbonic acid-based superplasticizing chemical admixture was used in all mortar mixtures.

Table 1
Chemical composition and physical properties of cement and MK

	Cement	MK
Chemical composition		
SiO_2 (%)	20.2	56.0
Al_2O_3 (%)	5.8	37.0
Fe_2O_3 (%)	3.0	2.4
CaO (%)	63.3	2.4
MgO (%)	3.4	0.3
SO_3 (%)	2.1	–
TiO_2 (%)	–	0.2
Na_2O (%)	0.2	0.3
K_2O (%)	0.3	0.6
Loss on ignition (%)	1.2	0.6
Physical properties		
Specific gravity	3.13	2.63
Specific surface area (m^2/kg)	312	12,000

Table 2
Mortar mixture proportions

Mixture	Replacement levels of MK (%)	Cement (g)	MK (g)	Water (g)	Sand (g)
MK0	0	1000	0	450	2000
MK5	5	950	50	450	2000
MK10	10	900	100	450	2000
MK15	15	850	150	450	2000

2.2. Source of sulfate

All mortar and paste samples were immersed in magnesium sulfate solution for 360 days. Sulfate solution concentrations used were 0.42% (3380 ppm of SO_4^{2-}), 1.27% (10,140 ppm of SO_4^{2-}) and 4.24% (33,800 ppm of SO_4^{2-}), respectively. The test solutions were made by dissolving regent-grade chemicals in tap water. These sulfate solutions were renewed every 4 weeks. Tap water was used as a control for the comparison solution. All test solutions, including tap water, were maintained at 20±1 °C during the test period. The test solution and specimens were kept in plastic containers, with space between the mortar specimens.

2.3. Mixtures

Four different mortar mixtures were prepared. Cement was partially replaced with 0%, 5%, 10% and 15% of MK by mass. The water–cementitious material ratio (w/cm) was fixed at 0.45 by mass. The cementitious material/fine aggregate was invariant at 2.0 in all mortar mixtures. The details of mortar mixtures used in this study are given in Table 2.

All mortar specimens were demoulded after 24 h of casting and then cured in tap water for an additional 6 days. Thereafter, some of the specimens were moved to magnesium sulfate solutions and continuously immersed in the solutions.

Paste samples were made using 100 g of total cementitious materials (cement+MK) and 45 g of deionized water with no superplasticiser. After hand mixing, the paste samples were caste and rotated in their moulds for 8 h, to avoid segregation, and demoulded after 1 day. All paste samples were cast into plastic cylinders 13 mm in diameter. After demoulding, the paste samples were cured in tap water for 6 days and then coated with epoxy on all surfaces except the upper side.

2.4. Test techniques

The mortar specimens tested in this study are divided into cubes (50×50×50 mm) for compressive strength

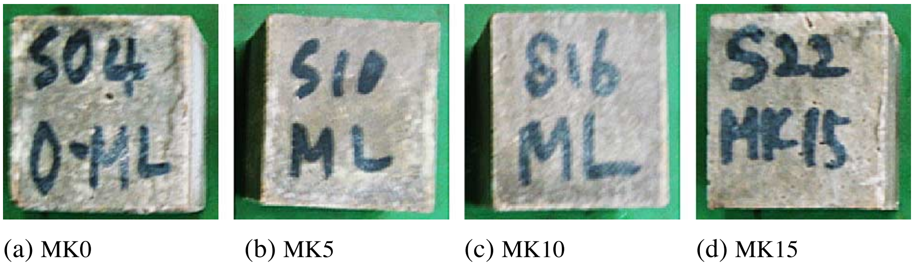
(a) MK0 (b) MK5 (c) MK10 (d) MK15

Fig. 1. Fifty-millimeter mortar cubes exposed to 0.42% magnesium sulfate solution for 360 days.

measurement, and prism (25×25×285 mm) for expansion measurement.

Prior to compressive strength tests, cubic mortar specimens were thoroughly visually investigated, and signs of deterioration, e.g., cracking, spalling and delamination, were checked.

The compressive strength of the mortar cubes was measured after 0, 28, 91, 180, 270 and 360 days of magnesium sulfate exposure. Companion mortar specimens cured in tap water for the same period were also tested. The strength deterioration was evaluated by the reduction in compressive strength, as expressed in the following equation:

$$\text{Reduction in compressive strength }(\%) = (f'_{\text{cw}} - f'_{\text{cs}})/f'_{\text{cw}} \times 100$$

where, f'_{cw} is the average strength of the specimens cured in tap water and f'_{cs} means the average strength of the specimens immersed in magnesium sulfate solutions. Three mortar specimens from tap water and magnesium sulfate solutions were tested at each test period, respectively, and then the values were averaged.

Based on ASTM C1012, expansion measurements of mortar prisms exposed to magnesium sulfate solutions were carried out, and the values of three specimens were averaged.

Paste samples immersed in sulfate solutions for 360 days were used for XRD analysis. The upper side portions exposed to the magnesium sulfate solutions were sliced 5–10 mm in thickness. Thereafter, the selected paste portions were ground by hand using acetone. The XRD was conducted using the RINT D/max 2500 (Rigaku, Japan) X-ray diffractometer. For the XRD tests, CuKα radiation, with a wavelength of l.5405 Å, at a voltage of 30 kV and a current of 30 mA, was used. The analysis was performed between 5° and 40° 2θ, at a scanning speed of 2°/min.

3. Test results

3.1. Visual examination

The photographs of the mortar cubes with different replacement levels of MK (0, 5, 10 and 15 mass%) exposed to 0.42%, 1.27% and 4.24% magnesium sulfate solutions for 360 days are shown in Figs. 1–3, respectively.

In low concentration of magnesium sulfate solution (0.42%), none of the mortar specimens with or without MK showed any significant visible changes for up to 360 days (Fig. 1), although it was observed that a small amount of white substance was deposited on the faces of them. Thus, it may be concluded that the storage for 360 days in 0.42% magnesium sulfate solution did not considerably affect the deterioration of mortar specimen, regardless of the replacement level of MK. However, for the mortar mixtures exposed to 1.27% magnesium sulfate solution for 360 days (Fig. 2), different results of visual examination were observed. The MK5 and MK10 mortar specimens exhibited visible deterioration spalling at the corners, whereas the MK0 mortar specimen showed little deterioration, even up to 360 days of exposure. Furthermore, the MK15 mortar specimen that was affected the worst in this solution exhibited relatively severe spalling and softening, especially at the corners and edges of the sample. The signs of deterioration in the MK15 mortar were first observed at the corners of the specimen as earlier as about 91 days. However, it seemed that the


(a) MK0 (b) MK5 (c) MK10 (d) MK15

Fig. 2. Fifty-millimeter mortar cubes exposed to 1.27% magnesium sulfate solution for 360 days.

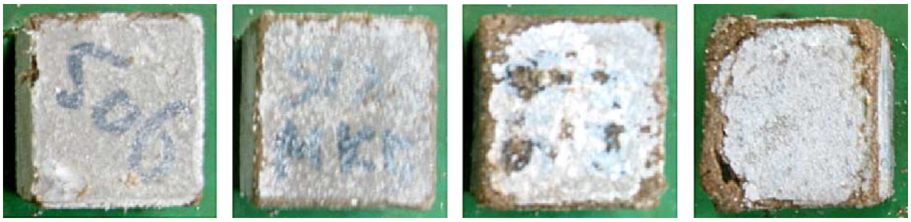
(a) MK0 (b) MK5 (c) MK10 (d) MK15

Fig. 3. Fifty-millimeter mortar cubes exposed to 4.24% magnesium sulfate solution for 360 days.

visual examination of mortar specimens containing MK exposed to 1.27% solution did not suggest the obvious relationship between visible deterioration and replacement level of MK.

The visual examination of mortar specimens stored in the high-concentration solution (4.24% $MgSO_4$) for 360 days (Fig. 3) indicated that the intensity of the damage by magnesium sulfate attack was greater as the replacement level of MK increased. Unlike the exposure to 0.42% and 1.27% magnesium sulfate solutions, the remarkable deterioration showing spalling and visible mass loss was observed at the corners and edges of MK0 mortar specimen exposed to 4.24% magnesium sulfate solution. MK15 mortar specimens had shown the first signs of deterioration at 56 days of exposure, and further exposure to the solution caused the maximum degradation, including very severe spalling and wide cracking throughout the specimen. The surface white-materials, which are soft and mushy, had significantly peeled off, and a cavity in the vicinity of edge of the specimen was also observed. Moreover, a lot of cracks along the edge characterized the most pronounced deterioration of the MK15 mortar specimen in this sulfate environment. It can therefore be concluded, through visual examination, that a higher replacement level of MK negatively affects the resistance against magnesium sulfate attack of mortar specimens.

3.2. Reduction in compressive strength

Table 3 presents the data on strength development of mortar specimens cured in tap water. These data indicated that MK15 mortar specimens showed a better compressive strength development compared to MK0 mortar specimens during the entire period of tap water curing.

The data on the reduction in the compressive strength of MK0, MK5, MK10 and MK15 mortar specimens exposed to magnesium sulfate solutions with different concentrations are shown in Figs. 4–6, respectively. Fig. 4 related to the low concentration (0.42% $MgSO_4$) indicated that there was no noticeable difference in reduction in compressive strength, regardless of replacement level of MK, up to an exposure period of 360 days. For MK5 and MK10 mortar specimens, the negative values of reduction in compressive strength were observed at 28 days of exposure. However, the reduction in compressive strength of all mortar specimens immersed in this sulfate solution was relatively small, even after 360 days of exposure.

Table 3
Strength development of mortar specimens cured in tap water

Mixture	Replacement levels of MK (%)	Compressive strength (MPa)			
		7 days	28 days	91 days	360 days
MK0	0	37.0	48.6	56.8	59.2
MK5	5	33.8	46.3	55.5	60.4
MK10	10	43.2	50.4	56.9	62.2
MK15	15	41.8	51.2	63.0	66.8

Fig. 5 showed the data on the reduction in compressive strength of mortar specimens immersed in magnesium sulfate solution of 1.27% and on the more pronounced deterioration of all mortar specimens with MK compared to the MK0 mortar specimen after 270 days of exposure. For the MK0 mortar specimen, the reduction in compressive strength was almost linear after 28 days of exposure, and it was observed to be a smaller value among all mortar specimens after 360 days of exposure. On the other hand, MK10 mortar specimen showed relatively small values in the reduction in compressive strength up to 180 days of exposure. Thereafter, the values sharply increased with exposure period. In this solution, the detrimental effect of MK replacement level on the reduction in compressive strength was not clear.

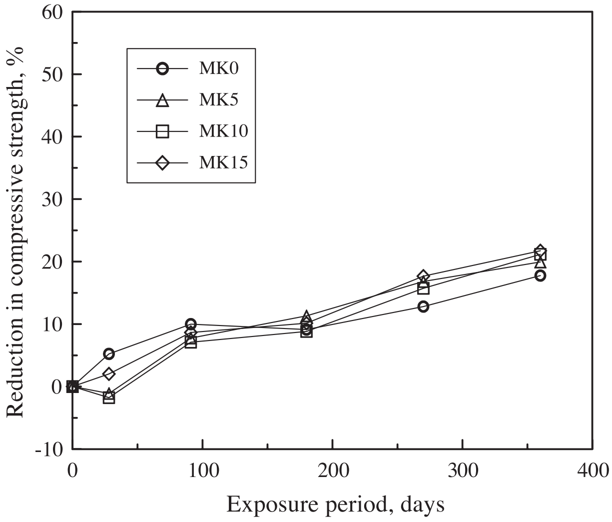

Fig. 4. Reduction in the compressive strength of mortar specimens exposed to 0.42% magnesium sulfate solution.

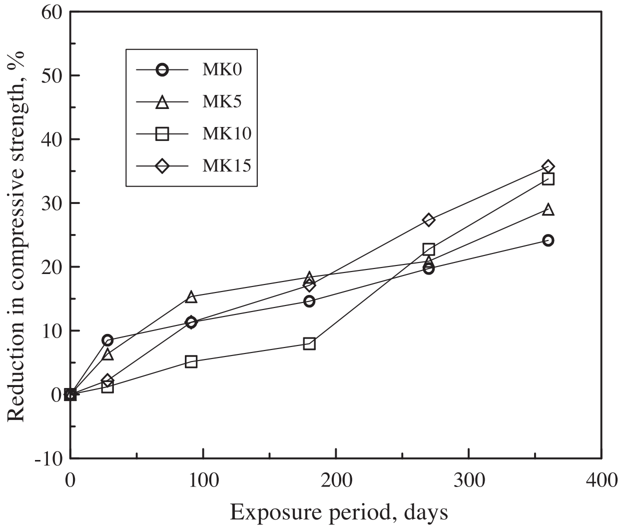

Fig. 5. Reduction the in compressive strength of mortar specimens exposed to 1.27% magnesium sulfate solution.

The data on the reduction in the compressive strength of mortar specimens exposed to 4.24% magnesium sulfate solution were periodically plotted in Fig. 6. Up to 91 days of exposure, MK0 mortar specimens exhibited the most remarkable deterioration with respect to reduction in compressive strength. However, the rate of strength loss was somewhat reduced after the exposure period. At longer exposures, e.g., after 270 days, an obvious trend, which showed more strength deterioration with a higher replacement level of MK, was observed. The value of reduction in the compressive strength of MK0 mortar specimen in this solution (4.24%) after 360 days of exposure was about 35.3%, whereas it was about 17.7% in 0.42% sulfate solution (Fig. 4) and about 24.1% in 1.27% sulfate solution (Fig. 5) after the same exposure period. Comparably, MK15 mortar specimens, after 360 days of exposure, reduced about 21.7% for 0.42% $MgSO_4$, 35.7% for 1.27% $MgSO_4$ and 54.0% for 4.24% $MgSO_4$, respectively.

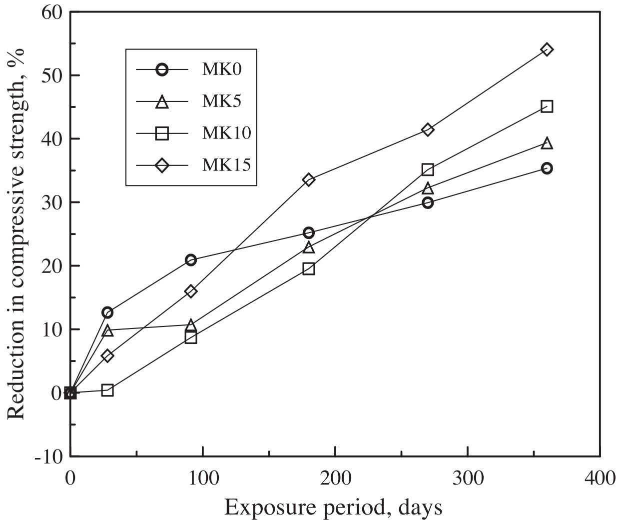

Fig. 6. Reduction in the compressive strength of mortar specimens exposed to 4.24% magnesium sulfate solution.

3.3. Expansion

The results on the effects of magnesium sulfate solution concentrations and replacement levels of MK on the expansion of mortar specimens are shown in Figs. 7–9, respectively. The expansion of the mortar bars stored in the magnesium sulfate solutions was measured at predetermined times of exposure up to 1 year.

The data obtained from expansion measurement indicated the different characteristics of mortar specimens with or without MK with various magnesium sulfate solution concentrations.

The results on the effect of MK replacement levels exposed to a low concentration of sulfate solution (0.42% $MgSO_4$) are shown in Fig. 7. The mortar specimens, regardless of replacement levels of MK, showed similar expansion up to 270 days of exposure. After 360 days of exposure, only the MK5 mortar specimen showed an expansion value of over 0.1%. These results had a good agreement with visual examination, indicating no deterioration, as shown in Fig. 1. The expansion results of mortar specimens with different replacement levels of MK exposed to a medium solution concentration of magnesium sulfate (1.27% $MgSO_4$) are presented in Fig. 8. From the expansion test results, the mortar specimens with 5% to 15% replacement levels of MK exhibited poor resistance to magnesium sulfate attack. After 360 days of exposure, expansion values were 0.343%, 0.339% and 0.469% for MK5, MK10 and MK15 mortar specimens, respectively, whereas the MK0 mortar specimens exhibited about 0.185% expansion after the same period of exposure. During the test period, the expansion values of MK5 mortar specimens were very similar to those of MK10 mortar specimens.

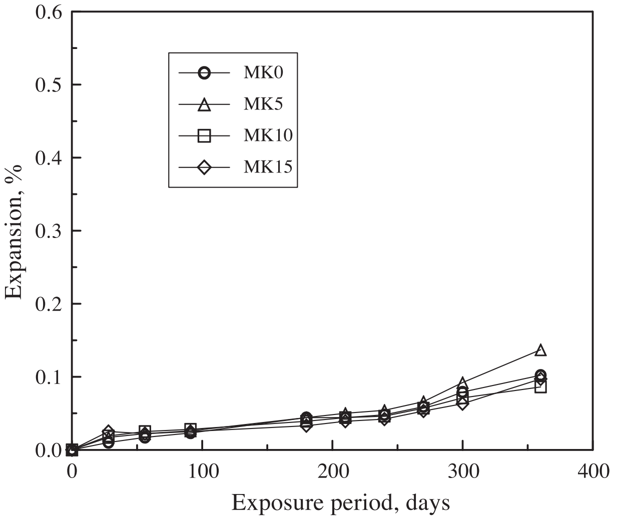

Fig. 7. Expansion of mortar specimens exposed to 0.42% magnesium sulfate solution.

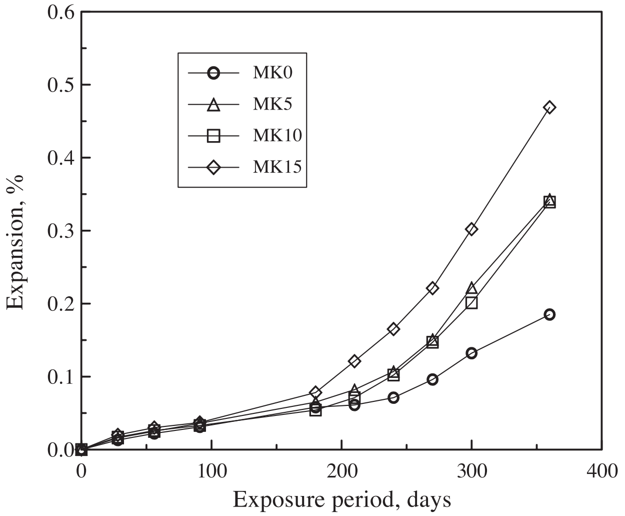

Fig. 8. Expansion of mortar specimens exposed to 1.27% magnesium sulfate solution.

Fig. 9 presents the expansion data for the mortar specimens with or without MK exposed to a high solution concentration of magnesium sulfate (4.24% $MgSO_4$). The expansion trend of the MK0 mortar was similar to that of MK5 and MK10 mortars up to 240 days of exposure. The final measured expansion values for the MK0 mortar was about 0.244% after 360 days of exposure, whereas mortar specimens with partial replacement levels of MK showed expansions of 0.360%, 0.410% and 0.556% for MK5, MK10 and MK15 mortar mixtures, respectively. In summary, it was found that the mortar specimens exposed to this high concentration of sulfate solution showed significant increase in expansion values as the partial replacement level of MK increased. This expansion trend implied that the MK mortar specimen, when exposed to magnesium sulfate solution, shows poor sulfate resistance.

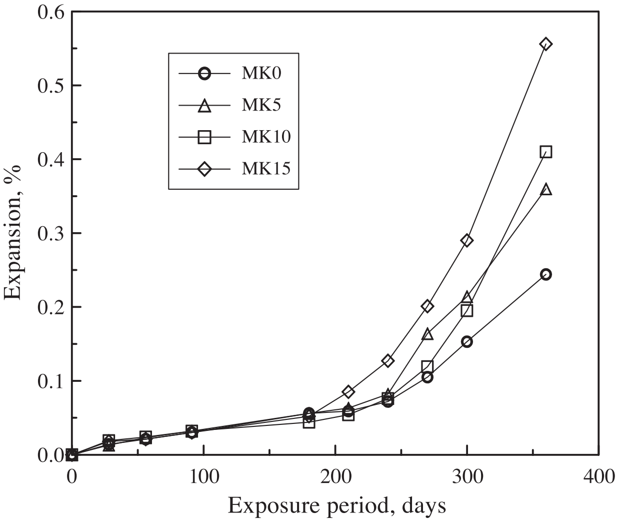

Fig. 9. Expansion of mortar specimens exposed to 4.24% magnesium sulfate solution.

3.4. Minerology

From the above experiments, the obvious trend of deterioration of mortar specimens was observed in both 1.27% and 4.24% magnesium sulfate solutions. The XRD study indicates the main cause of deterioration with the solution concentrations of magnesium sulfate in Figs. 10–13. Paste samples with different replacement levels of MK were examined by XRD after 360 days of exposure to magnesium sulfate solutions with different concentration levels.

Fig. 10 shows the XRD patterns for the powdered samples from MK0 paste specimens. These patterns indicated the presence of ettringite (E), thaumasite (T), gypsum (G), portlandite (P), calcite (C) and brucite (B), which are the expected products formed by cement hydration and magnesium sulfate attack. The XRD pattern for the paste sample immersed in a low solution concentration of magnesium sulfate indicated maximum intensity peaks for portlandite (18.1° and 34.0° 2θ). Similarly, the intensity peaks for portlandite in the MK0 paste sample subjected to 1.27% were also relatively strong (Fig. 10 (b)). On the contrary, significant reduction in the intensity of portlandite peaks was observed in the sample exposed to a high solution concentration of magnesium sulfate (Fig. 10 (c)). Another observation is the variation of gypsum peaks at around 11.7° and 20.7° 2θ with solution concentrations of exposure; the gypsum peaks were greatly increased with increased sulfate solution concentration. The peaks for

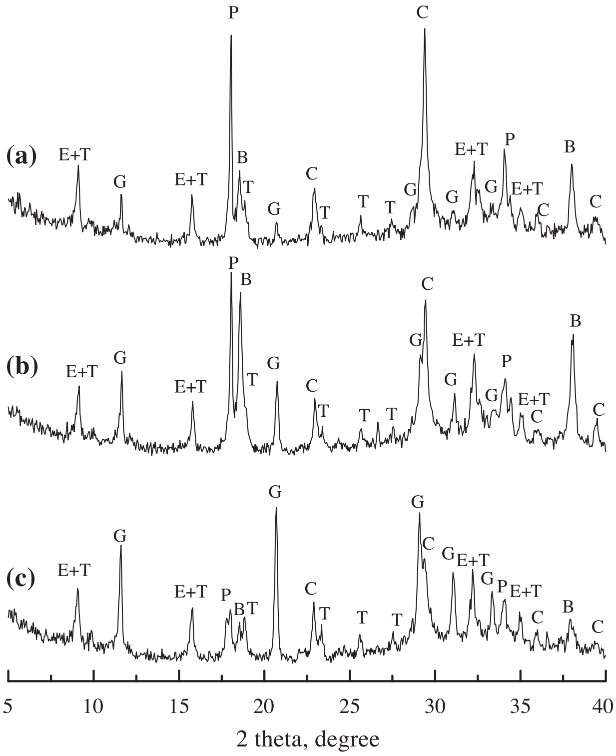

Fig. 10. XRD patterns of MK0 pastes exposed to magnesium sulfate solutions for 360 days: (a) 0.42%, (b) 1.27% and (c) 4.24% $MgSO_4$.

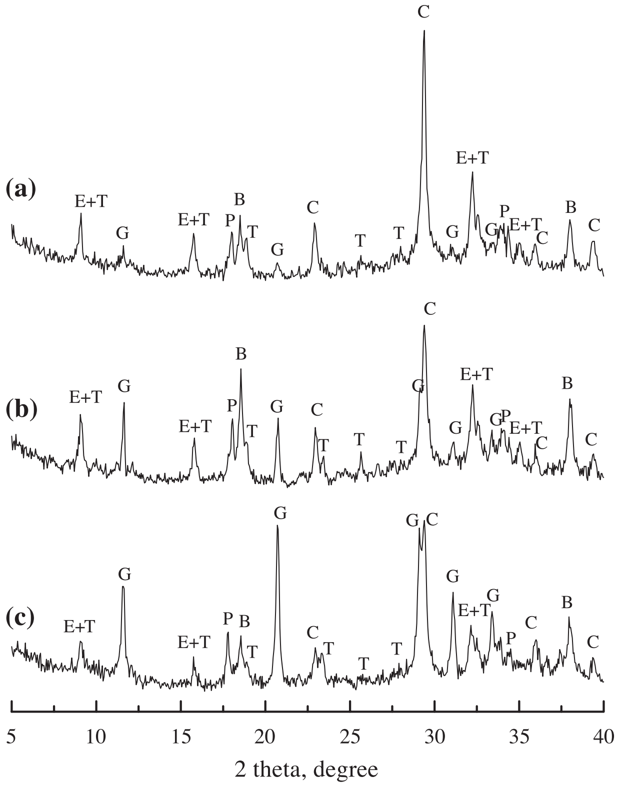

Fig. 11. XRD patterns of MK5 pastes exposed to magnesium sulfate solutions for 360 days: (a) 0.42%, (b) 1.27% and (c) 4.24% $MgSO_4$.

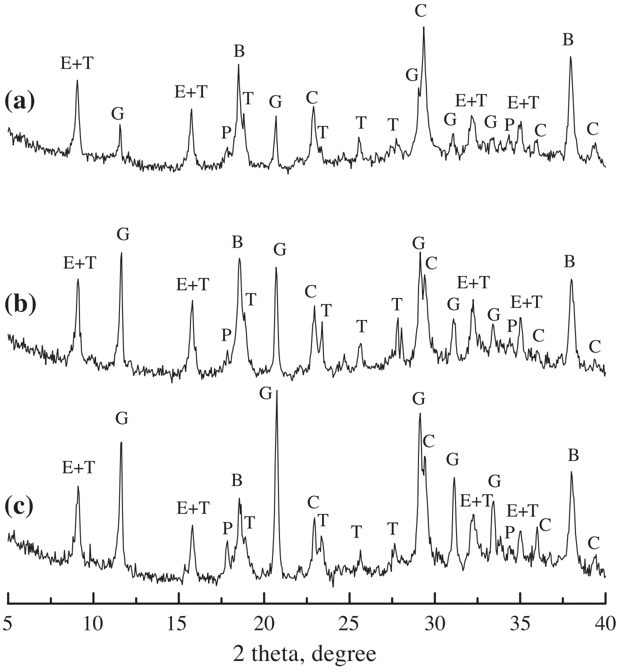

Fig. 12. XRD patterns of MK10 pastes exposed to magnesium sulfate solutions for 360 days: (a) 0.42%, (b) 1.27% and (c) 4.24% $MgSO_4$.

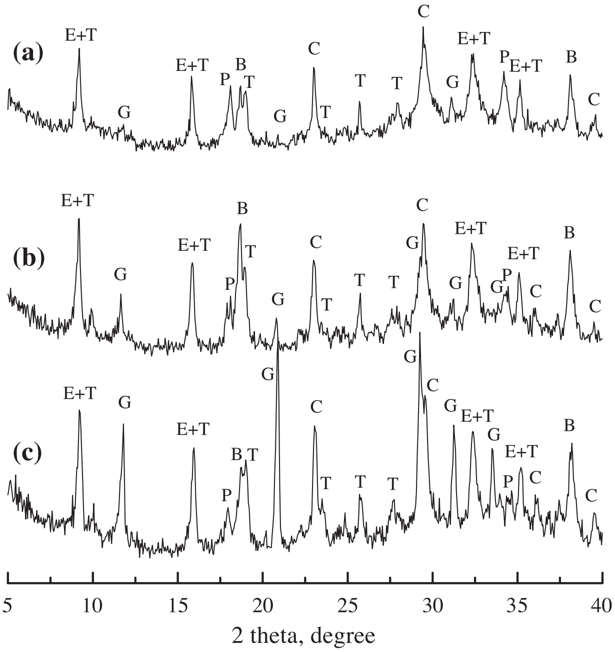

Fig. 13. XRD patterns of MK15 pastes exposed to magnesium sulfate solutions for 360 days: (a) 0.42%, (b) 1.27% and (c) 4.24% $MgSO_4$.

ettringite and/or thaumasite, however, were similar to one another, regardless of magnesium sulfate solution concentrations. Although thaumasite formation is frequently regarded as a low-temperature phenomenon, it can also be detected in the paste specimens exposed at ambient conditions [22,23]. For the XRD trace of the paste sample exposed to a medium solution concentration of magnesium sulfate (1.27%), a great amount of brucite was detected from its peaks at 18.5° and 38.0° 2θ. However, there was only height of small peaks for brucite in the MK0 paste sample exposed to a high concentration solution, because a considerable amount of brucite layer was peeled off from the cement matrix due to the loss of cohesiveness.

XRD patterns of MK5 paste samples exposed to magnesium sulfate solutions are shown in Fig. 11. Similarly, the peak intensity for gypsum was significantly increased with the increase of sulfate solution concentrations. However, the peak intensity for ettringite and thaumasite at around 9.1° 2θ in 4.24% magnesium sulfate solution was somewhat lower compared to that in 0.42% or 1.27% solutions. For the 10% replacement level of MK, the XRD patterns (Fig. 12) were similar to those of MK5 paste samples, irrespective of the solution concentrations of magnesium sulfate. In addition, there was no significant difference in the peak intensity at around 9.1° 2θ, indicating the coexistence of ettringite and thaumasite [17,24].

XRD traces of paste samples incorporating 15% MK showed clearly different patterns with solution concentrations of magnesium sulfate exposure (Fig. 13). Although the MK15 mortar specimen exposed to 0.42% magnesium sulfate solution visibly exhibited a relatively sound condition, a strong peak intensity at around 9.1° 2θ (ettringite+thaumasite) was detected in the corresponding paste sample (Fig. 13 (a)), but the peaks for gypsum were very weak in intensity. With increasing solution concentrations for the MK15 paste sample, drastic increases in peak intensity for gypsum at around 11.7° and 20.7° 2θ were observed, whereas there was only little or negligible change at around 9.1° 2θ.

Subsequently, it seems that there is a strong relationship between the deterioration of mortar specimens incorporating MK and gypsum formation but not with ettringite and thaumasite formations, especially in high solution concentration of magnesium sulfate. Thus, it appeared that the XRD intensity peaks for gypsum correlated quite well with visual examinations.

4. Discussion

Sulfate attack on concrete is primarily attributed to sodium, magnesium and calcium salts. Investigations on the effects of cations accompanying sulfate ions were reported elsewhere [14,25,26]. The different behavior of samples subjected to the sodium sulfate and magnesium sulfate environments indicates that the mechanisms of deterioration in these environments are different from each other.

As mentioned earlier, there are negative effects on the resistance to sulfate attack, in particular, to magnesium sulfate attack, of the hardened cement matrix incorporating MK, although it has been reported that MK significantly improves other durability aspects, such as the resistance to chloride ingress, AAR and carbonation. Actually, the mortar specimens incorporating MK levels greater than 15% exhibited good resistance to 5% sodium sulfate solution [7,12]. However, experimental results shown in the present study confirmed that the incorporation of MK in 1.27% and 4.24% magnesium sulfate solutions negatively affected the performance of mortar specimens as the replacement levels increased. Similar study on mortar specimens with silica fume in a similar sulfate environment was carried out by Moon et al. [14]. They confirmed that the conversion of secondary C–S–H gel into M–S–H (or M–C–S–H) gel, which is a noncementitious product and results in the softening of cement matrix, eventually led to the pronounced deterioration of silica fume blended mortar specimens exposed to severe magnesium sulfate environment.

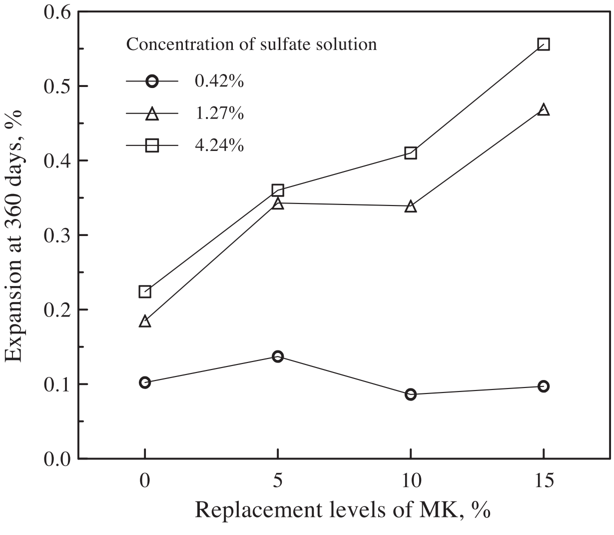

Fig. 14. Effect of solution concentrations and MK replacement levels on the expansion of mortar specimens exposed to magnesium sulfate solutions.

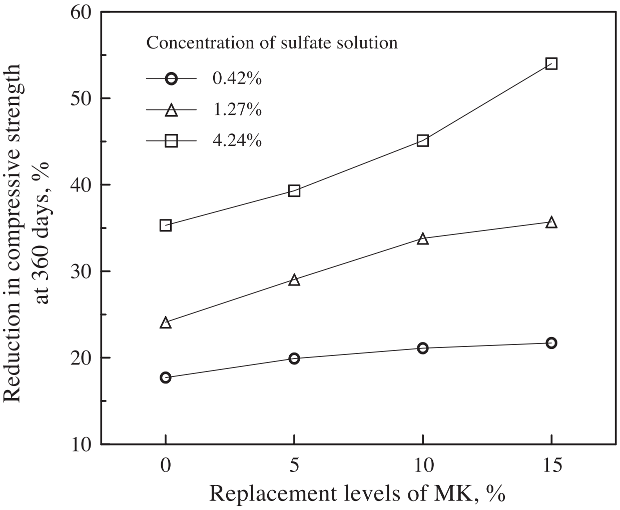

Fig. 15. Effect of solution concentrations and MK replacement levels on the reduction in the compressive strength of mortar specimens exposed to magnesium sulfate solutions.

Some investigators suggested that, to evaluate the deterioration degree of concrete due to sulfate attack, it is necessary to understand the influence of calcium hydroxide content on the severity levels of attack [27,28]. In other words, the role of calcium hydroxide in hardened concrete is important because it contributes substantially to the poor sulfate resistance of the concrete. For further improving the durability of concrete, particularly in relation to sulfate resistance, it has been emphasized that calcium hydroxide content must be reduced in the cement matrix. However, MK is less effective in reducing the calcium hydroxide content in OPC concrete compared to silica fume. This may be due to the lower SiO_2 content of MK relative to silica fume [29]. On the contrary, relatively more Al_2O_3 content of MK may produce the alumina-bearing products.

Using thermogravimetric analysis, Wild and Khatib [29] demonstrated that the calcium hydroxide content reduces in OPC systems with increased replacement level of MK. However, from visual examination in the present study (Fig. 3), it was clearly observed that the worst deterioration in 4.24% magnesium sulfate solution took place in mortar specimens with a high replacement level of MK (MK15). Probably, this phenomenon is significantly related to the formation of M–S–H in the cement–MK system, as in a study [14] carried out previously.

The effect of varying concentrations of magnesium sulfate solution was evaluated by Turker et al. [13], and Santhanam et al. [30] emphasized the importance in that field structures made with the same concrete may be attacked in different regions by different concentrations of sulfate, due to the development of drying and wetting fronts. Furthermore, as far as the concentration of magnesium sulfate solution is concerned [30,31], ettringite formation is commonly observed at a low concentration (>4000 ppm of SO_4^{2-}), a combined ettringite and gypsum formation is observed at a medium concentration (<4000 and <7500 ppm of SO_4^{2-}), and M–S–H formation accelerates the deterioration at a high concentration (>7500 ppm of SO_4^{2-}).

The visual examination of mortar specimens (Fig. 1) and the data on the reduction in compressive strength (Fig. 4) and expansion (Fig. 7) indicate that the deterioration in 0.42% magnesium sulfate solution (3380 ppm of SO_4^{2-}) was little or negligible, regardless of the replacement level of MK, although more ettringite was formed in the system than expected (Figs. 10 (a), 11 (a), 12 (a) and 13 (a)). However, the results of the expansion of mortar specimen and XRD of the paste specimen in 4.24% magnesium sulfate solution confirm that gypsum formation has a significant relation with the considerable expansion, especially in mortar specimens with a high replacement level of MK. One reason is that ettringite becomes unstable at the lower alkalinity provided by brucite formation during exposure to a high concentration of magnesium sulfate solution and will eventually decompose to gypsum [21,32].

In Fig. 14, it was found that raising the solution concentration from 0.42% to 1.27% resulted in a large increasing expansion, with increasing the solution concentration to 4.24% had little effect on expansion. This implies the possible presence of the critical concentration influencing the expansion of mortar specimens with or without MK. However, with respect to the reduction in the compressive strength of mortar specimens attacked by magnesium sulfate solution, a general trend indicating an almost linear increase of the values in the reduction in compressive strength at 360 days with the increase of concentration of the solutions was observed (Fig. 15).

5. Conclusions

In this paper, results are reported which demonstrate that increased metakaolin replacement levels play a negative role on the performance of mortar or paste specimens when exposed to magnesium sulfate solution. The degree of deterioration of mortar specimens incorporating MK was more severe with an increase of the solution concentration of magnesium sulfate. This likely significantly contributed to gypsum formation as magnesium sulfate attack. Tensile stresses developed during gypsum formation may lead to significant expansion [33]. Another possible reason may be the decalcification of primary and secondary C–S–H gel following the formation of M–S–H gel. However, this cannot be detected through XRD observation because of its poor crystallinity.

For mortar specimens incorporating MK exposed to 1.27% $MgSO_4$ solution, the expansion performance was different from the reduction in compressive strength (Figs. 14 and 15). Thus, a further detailed investigation is needed for the evaluation of the resistance to magnesium sulfate attack.

While metakaolin has been reported elsewhere to improve resistance to sodium sulfate attack, from this study, it does not provide benefit when exposed to magnesium sulfate attack.

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