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Strength and workability of magnesium silicate hydrate binder systems

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HIGHLIGHTS

- Strength and workability of magnesium silicate hydrate binder systems were studied.
- Effect of superplasticizers, MgO/SiO₂ ratio and crushed quartz filler is reported.
- The optimal binary system for strength and workability includes 60% MgO and 40% silica fume.
- Crushed quartz filler reduces water demand and improves the compressive strength of mortars.

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ABSTRACT

This paper discusses the optimization of magnesium silicate hydrate (M-S-H) binder systems for cement paste and mortar. The compressive strength and workability (water demand, flow) of M-S-H binder systems were studied. The binary systems contain magnesia and silica fume in which each constituent accounts for 40–60% by mass of the binder. The ternary systems include 10–40% very fine crushed quartz which is used as a fine filler for the partial replacement for the cementitious materials. It was found that the pure M-S-H binder system has a much higher water demand compared to Portland cement due to the very high silica fume content. The use of polymer-based superplasticizer effectively reduced water to cementitious materials ratio (w/c) for M-S-H binder system. The optimal binary binder composition for strength and workability was found to contain 60% magnesium and 40% silica fume (Mg/Si molar ratio = 2.25). The addition of very fine crushed quartz filler from 10 to 40%, as a partial replacement for the magnesia and silica fume, further improved the rheology of the mixture allowing for a workable mix with a w/c = 0.30. The highest compressive strength of mortar samples achieved with ternary M-S-H system is up to 87 MPa.

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1. Introduction

The formation of the magnesium silicate hydrate (M-S-H) was first discovered in 1953 during an investigation on the deterioration mechanism of concrete in sea water [1]. M-S-H formation since then had been generally considered a detrimental phenomenon associated with sulphate attack on concrete [2–5] and attracted little research interest as cementitious materials. Based on microstructure analysis, potential chemical reactions forming M-S-H gel at room temperature were suggested [6,7]:

 $3Mg^{2+} + 6OH^- + 4SiO_2 \rightarrow Mg_3Si_4O_{10}(OH)_2 + 2H_2O \tag{1}$

 $3Mg^{2+} + 60H^- + 2SiO_2 \rightarrow Mg_3Si_2O_5(OH)_4 + H_2O \tag{2}$

 $8MgO + 8SiO_2 + 16H_2O \rightarrow M_8Si_8O_{20}(OH)_8 \cdot 12H_2O \eqno(3)$

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http://dx.doi.org/10.1016/j.conbuildmat.2016.11.109 0950-0618/© 2016 Elsevier Ltd. All rights reserved. The microstructure and chemical composition of M-S-H are related to the ratio of reactants and hydrothermal curing regimes. The M-S-H formed in sulphate attack of concrete was reported to have high Mg/Si molar ratio of between 1.0 and 4.0 [1–3]. However, most of synthesized M-S-H gels in paste samples have lower Mg/Si ratio of between 0.67 and 1.5 [7–12] which suggests that the optimal formation of M-S-H gel without residual brucite can be obtained when the MgO content accounts for 30–50% of the MgO-SiO₂ mixtures. Furthermore, higher Mg/Si ratio of M-S-H gel has been found at elevated temperature [13]. The M-S-H microstructure consists of disordered intermediate phases and resembles the formation of talc (Mg/Si = 0.75), serpentine or chrysotile (Mg/Si = 1.5), or deweylite at Mg/Si ratios between 0.75 and 1.50 [9].

Results from studies on the mechanical properties of M-S-H system in cementitious materials are quite varied and dependent on mix proportions and materials sources [7,11,14–17]. Wei et al [11] prepared M-S-H binder mortars containing 60–90% reactive MgO (10–40% silica fume) whereby the mortar mix with M-S-H







binder containing 70% MgO obtained the highest 28 day compressive strength result of 57.4 MPa. In such case, the MgO/SiO₂ ratio for optimal M-S-H gel formation did not result in the highest strength. In contrast to the work of Wei et al [11], Jin and Al-Tabbaa [16] have shown that compressive strength increases with increasing silica content in the range of 0-50% and this was despite an increase in w/c ratio at the higher silica replacement levels. The highest compressive strength of 70 MPa at 28 days for a M-S-H binder mixture was reported by Zhang et al. [15] in which the M-S-H binder system contained 20% MgO, 5% MgCO₃, 25% silica fume (SF) and 50% quart sand filler. Apart from these encouraging experimental results, other studies report poor compressive strengths for magnesium silicate hydrate binder system [17]. In addition to the limited strength, one of the greatest challenges faced in the development of a M-S-H binder system compared to Portland cement (PC) is the very high water demand and poor workability of the binder, primarily due to fineness of SF and cell lattice structure of reactive magnesia [18,19].

The primary aim of this investigation is to develop an optimal composition based on the workability and compressive strength characteristics of a M-S-H binder systems. In the first stage, the effects of superplasticizers type, dosage and MgO/SiO₂ ratios (in the range of 40:60–60:40) on water demand and strength were determined for a binary system of magnesia and SF. The MgO/SiO₂ range used in this study was chosen based on the variability of results found in the available literature and the high compressive strength in excess of 60 MPa achieved for the 40:60 MgO/SiO₂ mix [7]. The second stage involved replacing a portion of the Mg/SiO₂ binder with crushed quartz filler to produce a ternary system (MgO-SF-QF) with a low water demand for high compressive strength.

2. Materials and testing methods

2.1. Materials

Light burnt magnesium oxide (Calix MgO 150) was supplied by Calix Ltd. (Australia). SF was supplied by Sika NZ as condensed silica fume. Crushed quartz filler (QF) was ground in a ring mill from local quartz sand until obtaining desired particle size. Quartz sand (S) is a local fine aggregate. Two superplasticizers were used in this investigation; Viscocrete-5-555 (liquid), a third generation polymer-based superplasticizer was provided by Sika NZ and sodium hexametaphosphate (SHMP), an inorganic superplasticizer supplied by Ajax Chemicals (Australia) with chemical formula Na (PO₃)_n·Na₂O.

The particle size distribution of the binder constituents was determined by Laser diffraction method (Fig. 1). Material morphology, discussed in Section 3, was examined by Scanning Electron Microscopy (SEM) to characterize particle size, shape and texture.

2.2. Optimization of superplasticizer dosage

The use of superplasticizers is crucial to reduce water demand and improve both the strength and workability of the mix. For these reasons, tests with superplasticizers were performed to optimize superplasticizer dosages for M-S-H binder. Viscocrete-5-555, a polymer-based superplasticizer was selected and compared to an inorganic superplasticizer (Sodium hexametaphosphate). The superplasticizer dosage optimization procedure is described as follows:

- Step 1: Prepare a set of containers with each container contained 50 g binder (40% MgO + 60% SF) dry mixed until homogeneous.
- Step 2: Place superplasticizer into each container. The superplasticizer dosage was increased from 0 to 5% by 1% unit increments.
- Step 3: Gradually add water in each container while slightly stirring until obtaining a flowable/self-levelling paste that has a specified flow of 150±5 mm as per ASTM C1437 without tamping. The amount of water poured into each container was recorded with an accuracy of ±1 ml. The superplasticizer efficiency was calculated based on the amount of added water including the water content contained in the superplasticizer. The mixture containing the lowest water content indicated optimal superplasticizer dosage.

2.3. Mix proportions

Three MgO/SiO₂ ratios (by mass) were selected and listed in Table 1. The low MgO/SiO₂ system (MgO/SiO₂ = 40/60) provided a high SF content binder to facilitate the formation of M-S-H gel [7,8]. The high MgO/SiO₂ system (MgO/SiO₂ = 60/40) provided a greater portion of MgO particles, which are coarser than SF, with the intention of improving the packing density of the binder despite the possible reduction in the formation of M-S-H gel. In cases of higher MgO content, the hydration products might contain residual brucite (Mg(OH)₂) and in such conditions, both brucite and M-S-H gel would co-exist in the hardened concrete as cementing materials. The selection of the high MgO, low SF mix was also intended to reduce the detrimental workability effects associated with a high SF content.

Five series resulting in a total of 21 mixtures were prepared for compressive strength testing. The mix proportions, listed in Table 2 are described as follows:

• Series 1 included three M-S-H paste mixtures in with a water/ cement (w/c) ratio of 0.40 and using a 3% polymer-based superplasticizer to achieve adequate workability. Mixes were labeled S1.40–S1.60 in accordance with silica fume content (%) by mass in the binder.

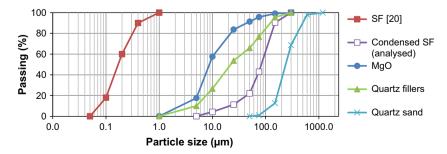


Fig. 1. Particle size distribution of materials. (See above-mentioned reference for further information.)

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