



Investigation into the synergistic effects in ternary cementitious systems containing portland cement, fly ash and silica fume

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ABSTRACT

This research was primarily conducted to verify the presence of synergistic effects in ternary cementitious systems containing portland cement (OPC), class C fly ash (FA) and silica fume (SF). A subsequent objective of the study was to quantify the magnitude of the synergy and to determine its source. For a ternary mixture containing 20% FA and 5% SF by mass, the synergistic effect was observed mostly at later ages (7 days onward) and it resulted in an increased compressive strength and resistance to chloride ion penetration as well as a reduced rate of water absorption (sorptivity) compared to predictions based on individual effects of FA and SF in respective binary systems. The observed synergy was attributed to both chemical and physical effects. The chemical effect manifested itself in the form of an increased amount of hydration products. The physical effect associated with packing density was, somewhat contrary to general belief, not due to an optimized particle size distribution of the binder components of the ternary cementitious system. Instead, it was the result of smaller initial inter-particle spacing caused by lower specific gravities of both FA and SF which, in turn, led to a lower volumetric w/cm. If the mixture design was adjusted to account for these differences, the physical effect would be diminished.

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1. Introduction

An increasing interest in the utilization of ternary OPC/FA/SF mixtures is a direct result of their excellent performance, which has been frequently attributed to synergistic effects taking place between fly ash and silica fume. The synergy can be described in two different ways. The first type of synergy is rather intuitive and its basic principle is that, when combined in a ternary system, FA compensates for deficiencies of SF, and vice versa, SF compensates for deficiencies of FA [1]. However, the “classical” definition of the term “synergy” describes it as the “interaction of two or more agents or forces so that their combined effect is greater than the sum of their individual effects” [2].

Some of the previously published data suggests the existence of synergistic effects (according to the above classical definition) in ternary systems with respect to compressive strength [3,4], permeation-related properties [5], plastic shrinkage cracking and chloride ion penetrability [6], sulfate resistance [7] and alkali-silica reaction [8,9]. However, there seems to be a disagreement regarding the source of this synergy, i.e., whether it is associated with

chemical effects, physical effects or both. Lilkov et al. [10] showed that, due to chemical interaction of fly ash and silica fume in the ternary system, the synergy manifested itself in the increased amount of hydration products and in the decreased amount of calcium hydroxide observed at an early age (within the first 24 h). Further, Weng et al. [11] reported a significant increase in the formation of hydration products between 3 and 7 days in a ternary system containing 10% SF and 30% FA.

On the other hand, Kwan and Wong [12] reported that ternary OPC/FA/SF mixtures possess a higher packing density than binary mixtures, thereby providing an indirect proof to synergy arising from the physical effects. Similarly, Popovics [4] suggested that the improvement in workability observed for the ternary mixtures studied was a result of a more optimal (i.e., less “gap-graded”) particle size distribution. Yet, the data reported by Isaia [3] indicated that both the physical and the chemical synergistic effects were present in the investigated ternary systems incorporating fly ash and silica fume. The physical effect was presumably due to a higher content of particles smaller than 5 μm , whereas the chemical effect was associated with higher pozzolanic activity of the ternary system, both of which were well reflected by an increased compressive strength.

It was clear from the literature review that the previous research dealing with the issues concerning synergistic effects in ternary systems was not “holistic”. Generally, the focus was either on the

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physical or on the chemical aspect, or on evaluation of the impact of synergy on certain properties of ternary mixtures. Therefore, the present study was undertaken with three distinct objectives. The first objective was to verify whether synergistic effects (according to the “classical” definition) exist in a ternary cementitious system, in which silica fume and moderate amounts of class C fly ash are incorporated. Such moderate (15–25% by the total mass of the binder) fly ash contents have been frequently used in ternary mixtures evaluated by researchers and are common in field applications. Although differences exist between class C and class F fly ash (most notably higher reactivity typically exhibited by class C fly ash due to both pozzolanic and hydraulic properties) this study was limited to class C fly ash. This is, in part, due to local fly ash availability, and the fact that in majority of the above mentioned literature studies class F fly ash was usually incorporated. If the existence of the synergy was successfully confirmed, the subsequent objectives of this research were to quantify the magnitude of the synergy, and to examine whether the synergy is of a physical nature, a chemical nature, or both. To accomplish the above goals, a series of experiments was conducted, as broadly described in the following sections.

2. Experimental details

2.1. Materials

Ordinary ASTM C 150 Type I portland cement (OPC), class C fly ash (FA) meeting the requirements of ASTM C 618 and dry silica fume (SF) were used. In field applications, typically densified silica fume is used due to more economical transportation stemming from increased bulk density compared to undensified silica fume, and due to health concerns related to handling fine undensified silica fume particles. However, it was reported that densified SF particles undergo agglomeration [13,14], and thus they cannot be easily dispersed, particularly in paste mixtures, unless an ultrasound technique is applied [15]. Accordingly, in this study it was decided to use densified SF in mortar mixtures and undensified silica fume in pastes. Table 1 summarizes the chemical composition

and physical properties of the cementitious materials used in the study as provided by their suppliers.

Mortar mixtures incorporated siliceous sand with a fineness modulus of 3.7. To ensure satisfactory workability of mortar mixtures, approximately 2340 ml/m³ of polycarboxylate high range water reducing admixture (HRWRA) was used in each mixture. HRWRA was not used in paste mixtures.

2.2. Mixture proportions and preparation

The following four types of binder systems were used in this study: plain cement mixture (OPC), binary mixture with 20% fly ash (20FA), binary mixture containing 5% silica fume (5SF) and ternary mixture with 20% fly ash and 5% silica fume (20FA/5SF). In each case, the cementitious materials (FA and SF) were used to replace a given percentage of cement (based on the total mass of the binder). These binder systems were used to prepare both the paste and mortar mixtures, each at a constant water-to-cementitious materials ratio (w/cm) of 0.41 by mass. All mortar mixtures were prepared with a paste content of 35.5% by volume. The detailed proportions of mortar mixtures are provided in Table 2.

Both the paste and the mortar mixtures were prepared using a laboratory Hobart-type mixer following the procedures of ASTM C 305. Paste specimens were made for compressive strength, thermogravimetric analysis (TGA) and isothermal calorimetry. Initially, paste specimens were also prepared for use in the rapid chloride permeability (RCP) and sorptivity tests. However, since paste sorptivity specimens developed extensive shrinkage cracking during conditioning, it was decided to conduct both the RCP and the sorptivity tests using mortar specimens.

2.3. Test procedures

For each part of this study, a distinct set of tests, each believed to be most suitable for achieving the previously stated objectives, was performed. The entire test matrix is summarized in Table 3.

Table 1
Physical properties and chemical composition of cementitious materials used in study on synergistic effect (as reported by the suppliers).

Property	Cement	Class C fly ash	Silica fume
<i>Physical</i>			
Specific gravity	3.15	2.56	2.20
Fineness			
Retained on #325 mesh	–	20.4	–
Blaine's surface area (cm ² /g)	3470	–	–
Compressive strength tested on mortar cubes (MPa)			
1 day	16.5	–	–
3 day	26.0	–	–
7 day	32.2	–	–
28 day	–	–	–
Strength activity index with portland cement at 7 days, % of cement control	–	97	–
<i>Chemical</i>			
Silicon dioxide SiO ₂ (%)	20.04	41.40	93.07
Aluminum oxide Al ₂ O ₃ (%)	5.84	19.98	0.62
Ferric oxide Fe ₂ O ₃ (%)	2.28	5.95	0.41
Calcium oxide CaO (%)	64.87	16.23	0.66
Magnesium oxide MgO (%)	1.63	3.72	1.16
Sulfur trioxide SO ₃ (%)	3.28	0.99	<0.01
Loss on ignition (%)	1.13	1.05	2.71
Sodium oxide Na ₂ O (%)	0.14	–	–
Potassium oxide K ₂ O (%)	0.88	–	–
Total alkali as sodium oxide Na ₂ O (%)	0.72	2.46	0.67
Insoluble residue (%)	0.47	–	–
<i>Potential compound composition (Bogue)</i>			
Tricalcium silicate C ₃ S (%)	60	–	–
Dicalcium silicate C ₂ S (%)	12	–	–
Tricalcium aluminate C ₃ A (%)	12	–	–
Tetracalcium aluminoferrite C ₄ AF (%)	7	–	–

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