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Natural Pozzolan as a Partial Substitute for Cement in Concrete

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Abstract: In this paper, the use of natural pozzolan as a partial cement substitute in concrete materials is investigated. By means of a test series, four mixes using three types of natural pozzolan, as well as a Class F fly ash, are evaluated. The effectiveness of each pozzolan in controlling alkali-silica reactions has been studied. Correlations have been revealed between the mechanical properties of the proposed mixes and a Portland cement control mix. The results are also compared with industry standards for mortars made with fly ash and silica fume. The paper's findings indicate that one type of pozzolan may be used as a substitute for fly ash, but not for silica fume.

Keywords: Natural pozzolan, cement replacement, concrete, mortar, fly ash, mechanical properties, silica fume.

1. INTRODUCTION

Under the strong contemporary demand for modern and environmentally friendly materials, natural pozzolan can be proved to be such a material and several researchers have focused their research efforts in using it as a partial substitute in the manufacture of concrete and mortar [1-10].

Pozzolans are divided into two categories, namely the natural and the artificial, as metakaoline and silica fume (SF). In recent years, the industry has shifted to using natural pozzolans because of their lower cost and accessibility. A pozzolan is a siliceous material that can be used as an inexpensive substitute for cement in mortar mixtures. Pozzolans participate in a cementitious reaction with calcium hydroxide (i.e., lime) and other alkalis. Pozzolans are effective at lowering the mortar's heat of hydration, which improves its workability and durability. They can also improve concrete and mortar resistance to both sulfate attack and the alkalisilica reaction (ASR), which makes it beneficial to use in large concrete projects such as bridges and dams (Gibbons 1997 [11], Tsimas et al. 2005 [12], Hossain and Mol 2011 [8]), as well as in the restoration of historic and monumental masonry structures as a restoration mortar (Moropoulou et al. 1998 [13], 2004 [14] and 2005 [15], Asteris et al. 2005 [16]).

Major sources of natural and manmade pozzolans include volcanic mineral deposits, fired and crushed clay, and fly ash, which is created during the burning of coal for power generation (Gibbons 1997 [11]). Most pozzolans are profuse, but current uses for them are limited. Consequently, they represent a potential source of inexpensive construction material.

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This paper investigates the feasibility and effectiveness of using naturally occurring pozzolan materials as an additive or substitute for cement in concrete mixtures. This investigation has been carried out through a series of experiments using a Class F fly ash and three types of natural pozzolans under the following laboratory tests: chemical composition, moisture content, loss on ignition, compressive strength, strength activity index, expansion from alkali-silica reaction (ASR), heat of hydration, slump, drying shrinkage, and autoclave expansion.

2. LITERATURE REVIEW

Alkalis in cement (typically calcium hydroxide and sodium and potassium oxide) can react with aggregates containing reactive silica or carbonate mineral constituents. In cured cement, these reactions produce different types of expansion that can cause concrete deterioration through internal cracking, surface cracking, and/or aggregate pop-out (Dolar-Mantuani 1983 [17]). Classic research by Stanton (1940) [18] ascribed ASR degradation to an expansive chemical reaction between the alkalis contained in the cement paste and certain reactive forms of silica within aggregate. Stanton identified opal and chert as common forms of reactive silica.

Pozzolans as an additive reduce cracking and expansion that results from ASR. The ASR is known to be normally occurs from the following combination of conditions (Hobbs 1988 [19]):

- 1. significant quantity of reactive silica.
- 2. reactive silica within a critical range of proportions.
- 3. available alkalis above a critical level.
- 4. water from an external source.

Suitable precautions must be taken to minimize or eliminate harmful expansion and cracking, such as:

decreasing the amount of reactive aggregate (Björk 2002 [20]).

- using low-alkali cement as per ASTM C150-09 [21].
- using mineral admixtures such as pozzolans or blast furnace slag (Lane *et al.* [22]; Wang and Gillott 1993 [23]; Thomas and Innis 1998 [24]).
- reducing the availability of water such that the relative humidity in concrete pores falls below about 85–90%.

In mass and high-strength concrete, the production of heat during the hydration reaction creates thermal gradients which can induce cracking in hardening concrete (Yeon *et al.* 2012 [25], De Schutter 2002 [26]). Thermal cracking can be mitigated by reducing the heat of hydration, a goal which can be achieved through the use of pozzolans or ground, granulated blast furnace slag (Tokyay *et al.* 2012 [27], Nili and Salehi 2010 [28]).

3. CHARACTERISTICS OF CONCRETE FORMU-LATED WITH POZZOLAN ADDITIVES

3.1. Concrete Formulation Components

3.1.1. Cementitious Materials

For this investigation, pozzolanic materials were selected which conformed to ASTM C618-00 [29]. Natural pozzolan was used from Saudi Arabia (Pozzolan S1), South Africa (Pozzolan S2), and Jordan (Pozzolan J). This phase of testing also included a Class F fly ash, whose properties are specified in ASTM C618-00 [29].

The natural pozzolan samples were crushed, ground, and tested. A Cilas Granulometre 715 device was used to determine the grain size distributions of the materials. Type I Portland cement was used in the production of concrete mixes. Chemical properties of the cement were tested periodically in the laboratory. (Table 1) shows chemical composition, loss on ignition, density, and fineness of all cementi-



tious materials, while (Fig. 1) shows their particle size distri-

Fig. (1). Particle size distribution of cementitious materials.

3.1.2. Aggregate Materials

butions.

Washed, surface-dry basalt aggregates were used in this study. Fine aggregate, consisting of crushed basalt rocks, was sifted, washed over a #100 (150 μ m) sieve with a water spray, and re-graded to the final, desirable particle size distribution. Grain size distributions for all aggregates are shown in (Fig. 2). Coarse aggregates were used for slump testing, only.

3.2. Compressive Strength Testing

This investigation used test procedures outlined by ASTM C109/C109M-08 [30] to evaluate the effect of replacing Portland cement with different pozzolanic materials on

Determinate	Portland cement	Fly Ash	Pozzolan J	Pozzolan S1	Pozzolan S2
SiO ₂ (%)	20	51.06	42.56	48.44	44.7
Al ₂ O ₃ (%)	5.68	32.23	13.55	15.97	14.5
CaO (%)	64.94	4.18	9.75	8.53	9
Fe ₂ O ₃ (%)	3.98	3.42	13.24	13.2	11.8
MgO (%)	0.7	1.06	11.15	7.89	7.8
SO ₃ (%)	2.81	0.42	0.35	0.1	0.15
Other (%)	1.89	7.63	9.4	5.87	12.05
Water Requirement (%)	-	90	100	100	100
Free Water (%)	-	0.1	0.1	0.1	0.1
Loss on Ignition (%)	1.16	0.81	2.56	1.26	1.5
Insoluble Residue (%)	0.32	-	-	-	-
Blaine Fineness (m ² /kg)	320	238	461	320	564
Density (g/cm ³)	-	2.2	2.86	2.8	2.8

Table 1. Analysis of Cementitious Materials

the compressive strength of test specimens (50 mm cubes). Cement and sand paste was selected instead of concrete mix to eliminate the variability exhibited by the presence of aggregate. In the test mixtures, mineral admixtures were used to replace 10, 15, 20, 25, 30, and 40% of the mass of the Portland cement used in the control mixture. The results are presented in (Fig. 3) through (Fig. 6).



Fig. (2). Particle size distribution of aggregates.



Fig. (3). Effect of cement replacement with fly ash on strength development.



Fig. (4). Effect of cement replacement with *Pozzolan J* on strength development.



Fig. (5). Effect of cement replacement with *Pozzolan S1* on strength development.



Fig. (6). Effect of cement replacement with *Pozzolan S2* on strength development.

The control mixture consisted of:

- 500 g of Portland cement.
- 1375 g of graded standard sand (ASTM C778-06 [31]).
- 242 mL of water.

Test mixtures consisted of:

- 450, 425, 400, 375, 350, 300 g of Portland cement.
- 50, 75, 100, 125, 150, 200 g of mineral admixtures.
- 1375 g of graded standard sand.
- 242 mL of water.

The test mortars contained 1 part cement to 2.75 parts graded aggregate (by mass) and a water-to-cement ratio equal to 0.485 (by mass).

Mixtures of the dry ingredients were prepared, water was added, and the specimens were mechanically mixed in accordance with ASTM C305-06 [32]. After mixing, cubes were poured in lifts and tamped as required by ASTM C109/109M-08 [30]. Immediately after molding, the specimens were placed in a moist room at 23.0 ± 2.0 °C. After 24 hours, the cubes were removed from the moist room, de-

molded, and placed in water storage tanks until testing. The compressive strengths of two control specimens and two specimens of each test mixture were determined at 7, 28, 90, and 180 days. (Fig. 3) through (Fig. 6) show the results for the mixtures' compressive strength, which demonstrate that the natural pozzolans produce strengths similar to the fly ash.

Pozzolan S2 showed the best overall strength performance, with solid strength gain, even at 7 and 28 days. Many test mixes did not match the strength of the control mix at 7 or 28 days, but the strength of the majority of test mixes surpassed the control mix at 180 days. The distinguish performance of the S2 is maybe due to its purity.

3.3. Strength Activity Index Testing

The purpose of this phase of the study was to determine the strength activity index for concrete samples blended with fly ash, Pozzolan J, and Pozzolan S1. Specimens from a control mixture and a test mixture were prepared in accordance with ASTM C109/C109M-08 [30]. In the test mixture, 20% of the mass of Portland cement used in the control mixture was replaced by the same mass of the test pozzolan. The batches of the test cube were produced in accordance with ASTM C311-00 [33]. The following formulas were used:

Control Mixtures

- 500 g of Portland cement.
- 1375 g of graded standard sand (Ottawa sand, ASTM C778-06 [31]).
- 242 mL of water.

Test Mixtures

- 400 g of Portland cement.
- 100 g of test sample (pozzolan).
- 1375 g of graded standard sand.
- 242 mL of water required for flow ±5 of control mixture.

After molding, the specimens were placed in a moist room at 23.0 ± 2.0 °C. After 24 hours, they were removed from the moist room and stored until testing in saturated lime water as specified in ASTM C109/C109M-08 [30]. Compressive strength was determined for two control specimens and two specimens of each test mixture at 7 and 28 days. Pozzolanic activity indexes were calculated as follows:

Strength Activity Index (SAI)= $(A/B)\times100$, Strengthactivityindex-withPortlandcement. =A/Bxdwhere A= average compressive strength of test mix cubes, and B= average compressive strength of control mix cubes.

ASTM C618-00 [29] requires a minimum strength activity index of 75% at both 7 and 28 days. As shown in (Fig. 7), the Class F fly ash and Pozzolans J and S1 meet or exceed this requirement.

Also from (Fig. 7) can be concluded that at the early age of 7 days, the strength activity index of fly ash concrete due to packing effect is higher than due to pozzolanic reaction. Such detailed information could be very useful for the concrete technologist to select a suitable fly ash for use in concrete (Tangpagasit *et al.* 2005 [34]).



Fig. (7). Effect of various pozzolans on strength activity index.

3.4. Alkali-Silica Reactivity Testing

The purpose of this test series was to evaluate the effectiveness of the three test pozzolans in controlling ASR when used as a cement substitute. The investigation was carried out under laboratory conditions using procedures described in ASTM C1260-07 [35], Standard Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method). This method enables the detection of the potential of a concrete mixture to undergo ASR, which produces potentially harmful expansion.

Four levels of cement replacement were investigated: 10, 20, 30 and 40%. Basalt aggregates were crushed, sieved, washed, and re-graded as summarized in Section 2.1.2. The dry materials for the test mortar were prepared using a mixture of 1 part cement to 2.25 parts graded aggregate (by mass) to cast 25 x 25 x 286 mm bars. A water-to-cement ratio equal to 0.47 (by mass) was used. Each mold was placed in a moist cabinet or room immediately after it was filled. The specimens remained in the molds for 24 ± 2 hours, after which they were de-molded and placed in a storage container with enough tap water to totally immerse them for 24 hours at 80 ± 2.0 °C.

The initial length of each test specimen was then measured using a length comparator in accordance with ASTM C490-08 [36], Standard Practice for Use of Apparatus for the Determination of Length Change of Hardened Cement Paste, Mortar, and Concrete. Subsequently, test specimens were stored in a container at 80 ± 2.0 °C with enough NaOH solution for the samples to be totally immersed. Expansion data were recorded periodically, with at least three intermediate readings between the start and finish of the test.

A 14-day expansion greater than 0.20% indicates a potentially reactive aggregate in the field, while a 14 day expansion smaller than 0.10% indicates a non-reactive aggregate. (Fig. **8** – Fig. **10**) show the expansion test results. These results show that, although fly ash is slightly more effective than the natural pozzolans, both pozzolans significantly reduce the 14-day expansions and control ASR.

3.5. Heat of Hydration Testing

Heat of hydration is the quantity of heat (in joules per gram of unhydrated cement) that develops over the course of hydration at a given temperature. The most common method of determining the heat of hydration is described in ASTM C186-98 [37], Standard Test Method for Heat of Hydration of Hydraulic Cement. It is accomplished by measuring the heat of solutions of unhydrated and hydrated cement in a mixture of nitric and hydrofluoric acids. The difference between the two values represents the heat of hydration.



Fig. (8). Effect of cement replacement with fly ash on ASR expansion.



Fig. (9). Effect of cement replacement with Pozzolan J on ASR expansion.

This method was used to calculate the heat of hydration for one Portland cement control mixture and two test mixtures incorporating fly ash and Pozzolan S1. The results at curing durations of 1, 3, 7, 14, and 28 days are shown in (Fig. 11). This test series determined that the Pozzolan S1 mixture produced about 15% less heat of hydration than the fly ash mixture, where the fly ash mixture produced about 30% less heat of hydration than the control mixture.

3.6. Effect of Pozzolan and Water-to-Cement Ratio on Slump

Ten 2-inch cube specimens were prepared with varying amounts of Pozzolan S1 (between 0 and 30%), and water-tocement (W/C) ratios varying between 0.456 and 0.5. (Table 2) displays two slump testing mixture examples: a control mix and a 10% pozzolan mix. The slumps of nine specimens at 0, 30, and 45 minutes are shown in (Fig. 12). The results in (Fig. 12) show that increasing the amount of pozzolan generally increased the slump and workability of these specimens. In five specimens, the amount of pozzolan also varied between 10% and 30%, but the W/C ratios were decreased. As expected, reducing the W/C ratio reduced the slump of the specimens.



Fig. (10). Effect of cement replacement with Pozzolan S1 on ASR expansion.



Fig. (11). Effect of various pozzolans on heat of hydration.

3.7. Effect of Pozzolan and Water-to-Cement Ratio on Strength

Specimens with varying amounts of Pozzolan S1 and W/C ratios were tested in compression after 7 and 28 days to determine the effect of the pozzolan and W/C ratio on the early compressive strength. The results of these tests are shown in (Fig. 13).

Activity indexes are calculated by dividing the specimen strength by the strength of a control specimen at the same age and multiplying by 100%. The results in (Fig. 14) show that increasing the percentage of pozzolan generally decreases the activity index.

Three mixes were designed with 20% pozzolan, but each used pozzolan of a different fineness. As shown in (Fig. 15),

the early compressive strengths of these specimens increased with increasing pozzolan fineness.

Table 2. Mixture for Control Specimens

Test Mixes					
Pozzolan replacem	Pozzolan replacement		10%		
Total Volume (m ³)	0.03	0.03		
W/c		0.5	0.45		
	Ingredient	s			
<i>O. P. C.</i>	kg	10.5	8.4		
Pozzolan	"	0	2.1		
3/4"	"	22.35	22.92		
3/8"	"	13.08	13.41		
F. Sand	"	19.08	19.56		
Free water	L	5.25	4.719		
Total water	"	5.652	5.139		
Pr - 150	"	0.063	0.063		
	Slump (mn	n)			
Initial		170	50		
30 min		120	-		
45 min		105	-		
60 min	60 min		-		
Temperature (°C)					
Water		16	16		
Ambient		15	16.8		
Concrete(i)		20.5	20		
Concrete(f)		19.5	-		



Fig. (12). Slump versus W/C-ratio and cement replacement with Pozzolan S1.



Fig. (13). Strength versus W/C-ratio and cement replacement with Pozzolan S1.



Fig. (14). Activity index versus W/C and cement replacement with Pozzolan S1.

4. COMPARISON OF POZZOLAN S1 WITH ASTM SPECIFICATIONS FOR FLY ASH AND SILICA FUME

The properties of 20-micron fine natural Pozzolan S1 were investigated and compared with the ASTM specifications for fly ash and silica fume to determine the extent of their similarity. The following ASTM specifications were used as reference standards:

 ASTM C618-00 [29], Standard Specification for Coal Fly Ash and Raw or Calcined Natural Pozzolan for Use as a Mineral Admixture in Concrete. ASTM C1240-00 [38], Standard Specification for use of Silica Fume as a Mineral Admixture in Hydraulic Cement Concrete Mortar and Grout.



Fig. (15). Effect of Pozzolan S1 fineness on activity index (20% replacement).

4.1. Physical Tests

The following physical tests were conducted on the pozzolan specimens:

- strength activity index (ASTM C311-00 [33]).
- density (ASTM C188-95 [39]).
- fineness per sieve analysis (ASTM C311-00 [33]).
- Blaine fineness (ASTM C204-00 [40]).
- increase in drying shrinkage (ASTM C157 [41]).
- expansion stability using autoclave (ASTM C157-08 [42]).

Two types of specimens were used: (1) a control specimen with 100% Portland cement and sand and (2) an activity specimen with 80% cement, 20% natural Pozzolan S1, and sand. The chemical composition and physical properties of the control specimen were used for comparison with the activity specimen. The results of the physical tests are shown in (Table 3 and Table 4).

Table 3.	Strength	Activity	Index	Results
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Property	Control specimen	Control Activity specimen specimen		
Shear Strength at 7 days (N/mm ²)	29.5 22.6			
Strength Activity Index at 7days,%	76.6			
Shear Strength (N/mm ²) at 28 days	36.0 28.0			
Strength Activity Index at 28 days,%	77.8			

 Table 4.
 Density, Fineness, Dry Shrinkage, and Volume Stability

Physical Property		Results	
Density		2.83 g/cm ³	
F .	Sieve analysis	10.8%	
Fineness	Blaine	359 m²/kg	
Increase in drying shr	-0.009% *		
Volume stability (autoclave expansion / contraction)		0.2%	

 \ast The ASTM standard specifies to add (-) sign if the dry shrinkage rate of control specimens is greater than the tested specimens

4.2. Chemical Tests

Chemical analysis of the specimens was conducted to determine the effectiveness of the natural pozzolan in contributing to sulfate resistance. The results of the chemical analysis are given in (Table 5 and Table 6).

Table 5. Chemical Composition

Chemical Composition,%	Control Cement	Natural Pozzolan S1
SiO ₂	19.26	48.44
CaO	63.34	8.53
Fe ₂ O ₃	3.6	13.3
Al ₂ O ₃	5.35	15.97
MgO	2.36	7.98
SO ₃	2.48	0.1
K ₂ O	0.51	1.37
Cl	0.011	0.012

Table 6.Chemical Properties

Property	Control Cement	Natural Pozzolan S1
Loss on Ignition,%	2.35	1.26
Moisture Content,%	-	0.34
РН	-	9.0
Carbon Content,%	-	0.33
Insoluble Residue,%	0.74	-

4.3. Physical Analysis

The physical properties of the natural pozzolan specimens are compared with the ASTM requirements for fly ash and silica fume in (Table 7). As shown in this table, the natural pozzolan specimens met the requirements for fly ash but

Test	Results for Pozzolan S1	ASTM C618-00 Class N Pozzolan, Class F and C Fly Ash [29]	ASTM C1240-00 Silica Fume [38]
Blaine Fineness (m ² /kg)	359	-	15 (min.)
Pct. wt. remaining on 45-micron sieve	10.8	34 (max.)	10 (min.)
Strength Activity Index at 7 days,%	76.6	75 (min.)	85 (min.)
Strength Activity Index at 28 days,%	77.8	75 (min.)	-
Autoclave expansion/contraction,%	0.02	0.8 (max.)	-
Pct. Drying Shrinkage at 28 days	-0.009	0.03 (max.)	-

Table 7. Comparison Between Pozzolan Physical Properties and ASTM Requirements

not for silica fume. The fineness values were less than 34%, and the strength activity indexes were greater than 75%. Their volume stabilities were less than 0.8%, and the increase in drying shrinkage at 28 days was less than 0.03%. While the natural pozzolan met the silica fume requirements for Blaine fineness and the percentage remaining on the 45 micron sieve, it is significant that it did not meet the 7-day strength activity index of 85%. The reason may be attributed to the Silica fume ultrafine material with spherical particles less than 1 μ m in diameter being much less than the size of natural pozzolan.

4.4. Chemical Analysis

The chemical test results for the natural pozzolan specimen were compared with the ASTM requirements, as shown in (Table 8). The natural pozzolan specimens met all requirements for fly ash but did not meet the 85% silicon dioxide requirement for silica fume. It is worth noting that the ASTM specification for class F fly ash requires the sum of Fe2O3, Al2O3, and SiO2 to be at least 70%. It also specifies SO3 less than 5%, moisture content less than 3%, and loss on ignition less than 6%. Therefore, natural pozzolan behaves more as a fly ash than a silica fume.

5. CONCLUDING REMARKS

From the previous results we can conclude that:

 At the early ages, the strength activity index of fly ash due to packing effect is higher than due to pozzolanic reaction.

- Pozzolan S1 can provide a satisfactory substitute for fly ash and other natural pozzolans as tested against ASTM C618-00 [29]. It was clearly found to be effective in controlling ASR. It also produces about 15% less heat of hydration than Class F fly ash, whereas Class F fly ash produces about 30% less heat of hydration than Portland cement only.
- Pozzolan specimens ground to a size of 20 microns meet the chemical and physical requirements for fly ash. The fineness values were less than 34%, and the strength activity indexes at 7 and 28 days were greater than 75%. Volume stabilities were less than 0.8%, and the increase in drying shrinkage at 28 days was less than 0.03%. The percentage of total iron oxide, aluminum oxide, and silicon dioxide was greater than 70%. The percentage of sulfur trioxide was less than 5%. Moisture content was less that 3%. Loss on ignition was less than 6%.
- The chemical and physical properties of Pozzolan S1 are comparable to fly ash, and the one can be substituted for the other.

It should be mentioned that Pozzolan S1 did not meet the ASTM C1240-00 [38] chemical and physical requirements for silica fume. Its strength activity index at 7 days was less than 85%, and the percentage of silicon dioxide was not met. However, it met the requirements for the percentage remaining on the 45 micron sieve at greater than 10%, moisture content less than 3%, loss on ignition less than 6%, and Blaine fineness greater than 15 m2/kg.

		ASTM C618-00 [29]			
Test	Results for Pozzolan S1	Natural Daggalan Class N	Fly ash		Silica fume
		Naturai rozzoian Ciass N	Class F	Class C	
$Fe_2O_3 + Al_2O_3 + SiO_2$ (%)	77.61	70 (min.)	70 (min.)	50 (min.)	-
SO ₃ (%)	0.34	4 (max.)	5 (max.)	5 (max.)	-
Moisture content (%)	0.1	3 (max.)	3 (max.)	3 (max.)	-
Loss on ignition (%)	1.26	10 (max.)	6 (max.)	6 (max.)	6 (max.)
SiO ₂ (%)	48.44	-	-	-	85 (min.)

 Table 8.
 Comparison Between Pozzolan Properties and ASTM Requirements

Based on the above findings, it is concluded that Pozzolan S1 is not a suitable replacement for ASTM C1240-00 [38] silica fume despite meeting the requirements for fineness, moisture content, and loss on ignition.

CONFLICT OF INTEREST

The author(s) confirm that this article content has no conflicts of interest.

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