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The Synergic Effect of Fly Ash and High Reactivity Attapulgitite in Ternary Blended Cement

Abstract-*The main objective of this study is to study the synergic effect of fly ash (FA) and high reactivity Attapulgitite (HRA) when using them together in ternary blended cement of (OPC+FA+HRA) and evaluate the efficiency factor of FA and HRA in binary blended cement and (FA+HRA) in ternary blended cement. To achieve this objective compressive strength of binary blended cement mixes of (OPC+FA) with FA replacement percentages of (20 %, 30% and 40%), (OPC+HRA) with HRA replacement percentages of (5 and 10%) by weight of cement and ternary blended cement mixes of (OPC+FA+5% and/or 10%HRA) were tested and compared with that of reference mix at ages of (7, 28, 56 and 90) days to assess the synergic effect of FA and HRA in ternary blended cement. The results showed that using ternary blended cement of (OPC+FA+5% and/or 10%HRA) led to increase compressive strength relative to binary blended cement mixes of (OPC+FA) at the same replacement percentages by weight of cement. More significant increments in compressive strength were noticed at age of 7 days. The results showed also that the efficiency factors calculated according to modified bolmoy's equation for ternary blended cement were always higher than their corresponding binary blended cement of (OPC+.FA).*

Keywords- Fly ash, High reactivity Attapulgitite, Binary blended cement, Ternary blended cement

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1. Introduction

Concrete is the most important building materials and considered the second more material used in the world after water [1]. The global production of cement increased with fast rate. The production of cement in 1880 was less than two million tons [2], but it increased to five billion tons in 2018 [3]. The market is projected to reach about 6.2 Billion Tons in 2024 [3]. The main problem associated with cement production is increasing of harmful emissions mainly CO₂ resulting from decomposition of lime in addition to CO₂ emissions resulting from fossil fuel and electrical energy consumed. The total Co₂ emissions expected in 2025 is 3.5 billion tons/year [4]. These problems related with cement production demand serious solution to conserve both environment and natural resources. One of the best solutions is using of supplementary cementitious materials (SCM) as a partially replacement by weight of cement. SCM with high contents of aluminosilicate can react with Ca(OH)₂ produced from cement hydration process to produce compounds with cementitious properties. One of the most common SCM used in concrete is fly ash. Fly ash is a by-product material resulting from combustion coal in electric generation plants, and can be used in

concrete because of its pozzolanic and cementitious properties. Using of fly ash in concrete can improve workability [5], long term strength [6] and durability [7] as well as of its environmental benefit by reducing the cement content in concrete mixtures and conserve of thousands hectares of land which can be used as landfill for unutilized fly ash. In spite of these advantages concrete mixtures contained fly ash suffered from slow rate of strength improvement at early ages [8]. This may limit fly ash using in concrete when high early strength is required. Ternary blended cement is a method suggested to overcome low early strength for binary blended cement mixes contained fly ash [9]. Ternary blended cement uses two different types of mineral admixtures in concrete mixes, one with slow activity as fly ash and slag, and other with high reactivity like silica fume and metakaolin [8]. The main objectives of this study were studying the synergic effect of fly ash (FA) and high reactivity attapulgitite (HRA) when used them in ternary blended cement concrete mixes at different replacement percentages by weight of cement and estimate the efficiency factor of binary and ternary blended cement. To achieve these objectives compressive strength of binary blended cement of (OPC+FA) and (OPC+HRA)

mixes at multi replacement levels was estimated firstly, then compressive strength of ternary blended cement of (OPC+FA+HRA) mixes at the same total replacement percentages of binary blended cement mixes was calculated. The results of binary and ternary blended cement were compared to evaluate the synergic effect of (FA) and (HRA) in ternary system. Finally efficiency factor (k) for binary and ternary system at different ages and replacement percentages was calculated according to modified Bolomoy's equation [10].

2. Experimental work

I. Materials

a. Cement

Tables 1 and 2 show the results of the chemical analysis and physical properties of the Ordinary Portland Cement used in this research respectively. Results declared that the used cement is compatible with the Iraqi Specification No.5/1984[11].

Table1: chemical composition and main compounds of cement

Oxide composition	% by weight	Limits of Iraqi specification No.5 / 1984[11]
SiO ₂	19.6	-----
Fe ₂ O ₃	3.52	-----
Al ₂ O ₃	4.65	-----
CaO	61.56	-----
MgO	2.77	5.0(max)
SO ₃	2.71	2.8(max)
Loss on ignition	1.65	4.0(max)
Insoluble residue	0.8	1.5(max)
Lime saturation factor	0.95	0.66-1.02
Main compounds(Bogue's equation)% by weight of cement		
C ₃ S	57.65	-----
C ₂ S	12.7	-----
C ₃ A	6.37	-----
C ₄ AF	10.71	-----

b. Aggregate

Sand falls within zone two with fineness modulus of 2.95 and sulfate content of 0.07 % and crushed gravel (5-19 mm) gradation according to the requirement of the Iraqi Specification No. 45/1984[12] were used for all concrete mixes.

c. Mineral Admixtures

1-High Reactivity Attapulgit (HRA)

HRA was produced by grinding and firing of Attapulgit clays according to procedure developed by Frayyeh et. al [13]. Table 3 shows

chemical analysis and some physical properties of HRA. Table 4 indicated that (HRA) conformed to both chemical and strength activity indexes (S.A.I) requirements of the ASTM C618-05[14] Class N pozzolana.

2. Fly Ash (FA)

Hard coal (FA) from power station of Iskenderun in Turkey was used in this study. The chemical analysis and some physical properties of (FA) were shown in Table 3.

Table 2: Physical properties of cement

Physical properties	Test results	Limits of Iraqi Specification No.5/1984[11]
Specific surface area (Blaine method), m ² /kg	240	230 (min)
Setting time (Vicate's method)		
Initial setting, (minute)	105	45 (min)
Final setting, (minute)	330	600 (max)
Compressive strength, (MPa)		
3 days	19.6	15.00 (min)
7 days	28.6	23.00 (min)
Soundness using Autoclave expansion, %	0.4	0.8 (max)

Table 3: Chemical and Physical Analysis of HRA and FA

Oxide composition	Oxide content of (HRA) %	Oxide content of (FA) %
SiO ₂	60.48	65.65
Al ₂ O ₃	13.95	17.69
Fe ₂ O ₃	6.07	5.98
CaO	8.46	0.98
MgO	5.92	0.72
Na ₂ O	1.2	1.35
SO ₃	0.45	0.19
K ₂ O	2.47	2.99
L.O.I	0.1	3.1
Physical properties		
Specific Surface Area m ² /kg	2010	773
Specific Gravity	2.2	2.35
Density kg/m ³	2193	2343

The result contained in Table 4 shows that the FA used in this study was (Class F) because the summation of (SiO₂, Al₂O₃, and Fe₂O₃) is to be greater than 70%, and it identified with both chemical and strength activity indexes (S.A.I) requirements of the [14] Class F pozzolana

II. Mixing and Preparation of Concrete Specimen

Mixing of concrete components was achieved by

Table 4: Requirements of mineral admixture according to [14]

Requirement of [14]	HRA	FA	Limits of class F pozzolana [14]
(SiO ₂ +Al ₂ O ₃ +Fe ₂ O ₃) (%)	80.5	89.32	70% min.
SO ₃ (%)	0.45	0.19	5% max.
Loss on ignition (%)	0.10	3.1	6% max.
S.A.I at 7 days (%)	101.4	78	75% min.
S.A.I at 28 days (%)	108.8	89	75% min.

using a pan mixer. All the constituents were placed in the mixer and mixed for 2 min to ensure uniformity of the mix. Cement and mineral admixtures were mixed well to guarantee high homogeneity before adding them to the mixer. Then the mixing water was added gradually during mixing. After mixing, the mixture was cast into 100 mm cubic moulds at two equal layers; each layer was compacted by mechanical vibrator table. After the moulds had been filled of concrete, the surface of concrete in moulds was levelled and they were kept in the laboratory conditions for 24 h while the surfaces of moulds were covered by plastic sheets. Then, the specimens were de-moulded and kept in water for the specified period of curing in the laboratory. To prevent adhesion with concrete after hardening, these mentioned moulds have been cleaned and their internal surfaces oiled. The details of mixes used throughout this research are shown in Table 5.

3. Results and Discussion

I. Slump of binary blended cement

The results of binary blended cement slump are included in Figure 1. The results showed that using of FA as a partial replacement by weight of cement increased slump relative to ref. mix. The increment in slump when FA is used proportioned directly with the increment of percentage replacement of cement with FA. For mixes contained 20% FA the slump was 170 mm, but it increased to 190 mm and 205 mm for mixes contained 30% and 40% respectively. The increasing of slump can be attributed to spherical shape of FA particles, which they acted as a lubrication agent for concrete ingredients due to ball bearing effect of its particles [15], and electric charge when enough FA fine particles are absorbed by cement particles surfaces can cause electric repulsion and this will prevent flocculation accordingly slump or workability

will increase [16]. For binary blended cement of HRA Figure 1 shows that mixes contained HRA had a lower slump results than both Ref. mix and binary blended cement contained FA. These reductions in slump results because a like plate morphology of HRA particles increase inter-particle friction and prevent cement particles to slide over each other easily [17]. In addition to high surface area and the lower specific gravity of HRA led to increase paste volume accordingly, water demand increased.

II. Slump of ternary blended cement

The results of ternary blended cement slump are included in Figure 1. The results showed that the replacement of FA with HRA led to reduce slump. The reduction in slump relative to binary blended cement mixes increased with increasing of the percentages of FA replacement with HRA. For binary blended of 20 % FA the slump was 170mm but using of HRA reduced it to 150 mm and 75 mm for ternary blended cement mixes of (15%FA+5%HRA) and (10%FA+10%HRA) respectively. The reasons beyond reduction in slump when FA was replaced with HRA, is due to increase paste volume when FA with specific gravity of 2.35 was replaced with HRA with specific gravity of 2.2 in addition to reasons mentioned in previous section.

III. Compressive strength of binary blended cement

The results of compressive strength for binary blended cement of FA mixes are plotted in Figure 2. The results indicate that when cement was replaced with FA, compressive strength decreased with respect to Ref. mix at all ages and replacement levels. The reduction in compressive strength was more clearly at early age, because of slow nature of reactivity and little pozzolanic activity of class F fly ash at early age, which can be attributed to the higher content of crystalline phases, which are considered chemically inert in concrete [18]. So, FA acts as a filler and provides a nuclei for precipitation of Ca(OH)₂ and cement hydration products [19]. The activity of FA depends mainly on the alkalinity of pores solutions [16], and CaO (lime) content [20]. The main source of alkalinity in concrete contained low lime FA (as FA used in this study) is Ca(OH)₂ resulting from cement hydration process. At early age the amount of Ca(OH)₂ formed is low, so the alkalinity of pore solution is not high enough to ensure high reactivity of FA with Ca(OH)₂. At 28 days compressive strength results exhibit higher strength than those at 7 days and continue in increasing with increasing of age. Because with increasing of age, cement hydration

Table 5: The details of mixes by weight (kg/m³)

Mixes designation	Cement kg/m ³	FA kg/m ³	HRA kg/m ³	Sand kg/m ³	Gravel kg/m ³	Water kg/m ³
Ref. mix	500	-	-	635	990	200
20% FA	400	100	-	635	990	200
30% FA	350	150	-	635	990	200
40%	300	200	-	635	990	200
15%FA + 5% HRA	400	75	25	635	990	200
10%FA+10%HRA	400	50	50	635	990	200
25%FA+5%HRA	350	125	25	635	990	200
20%FA+10%HRA	350	100	50	635	990	200
35%FA+5%HRA	300	175	25	635	990	200
30%FA+10%HRA	300	150	50	635	990	200
5% HRA	475	-	25	635	990	200
10% HRA	450	-	50	635	990	200

process be in progress and the amount of Ca(OH)₂ increases and provides high alkalinity necessary to break down the glassy structure of FA oxides (SiO₂ and Al₂O₃) and increasing their ability to react with Ca(OH)₂ resulting from cement hydration [16]. The results of compressive strength of binary blended cement of HRA are included in Figure 3. The results show that using of HRA as a partial replacement by weight of cement led to increase compressive strength at all ages, because using of mineral admixtures as HRA with high pozzolanic reactivity and surface area can increase strength at all ages [21]. Higher increment in compressive strength was noticed with mix contained (10%HRA). The percentages of increment were (6%, 7%, 12% and 9%) relative to Ref.mix at ages of (7, 28, 56 and 90) days respectively.

Figure 4, 5 and 6 contained the results of ternary blended cement of mix contained total replacement of cement with FA and HRA of 20%, 30% and 40% respectively. The results show that using ternary blended cement led to increase compressive strength relative to binary blended cement contained FA. Tables 6 and 7 included the percentages of strength gain for ternary blended cement mixes at all ages relative to binary blended cement mixes of (OPC+FA). The results show that using ternary blended cement led to gain in strength relative to binary blended cement of (OPC+FA). The gain in strength seems to be more significant at age of 7 days and using ternary blended cement tends to be lower with progress of age. The strength gain at 7 days for mixes contained total replacement of 20 % by weight of cement were (15.4% and 23.0%) , but the strength gain reduced to (1,8% and 5.5%) at age of 90 days for ternary mixes contained (FA+5% HRA) and (FA+10% HRA) respectively. The same behavior was noticed for total replacement of 30% and 40% by weight cement but with lower percentages of strength gain. The increasing in strength gain can be attributed to synergic effect when ternary blended cement was used; due to replacement of FA with low activity and surface area with HRA which it had more activity and surface area. These

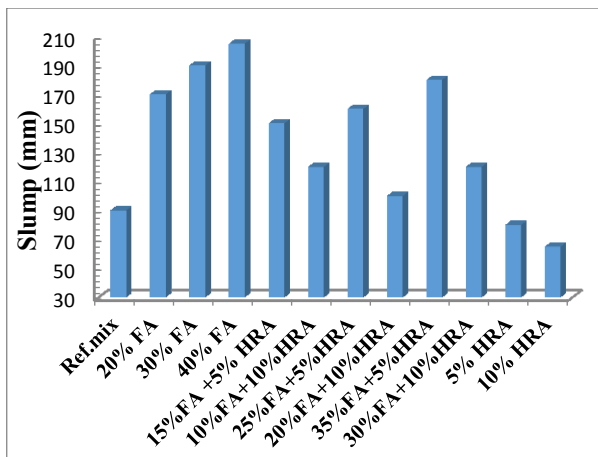


Figure 1: Slump result for binary and ternary blended cement

VI. Compressive strength of ternary blended cement

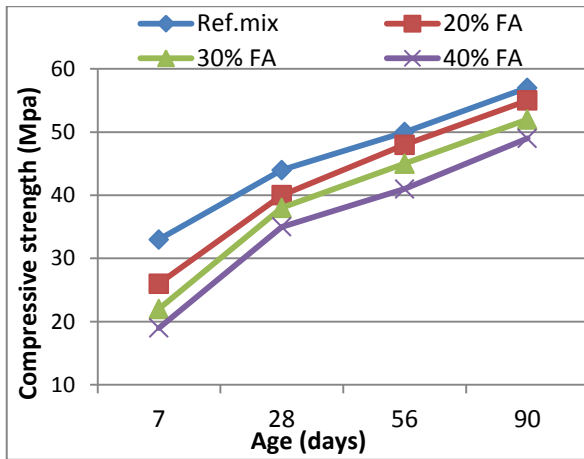


Figure 2: Compressive strength for binary blended cement of (OPC+FA)

replacements of FA with HRA led to condensate of microstructure by reducing the porosity due to increasing of CSH formation especially at early age in addition to filling effect of fine HRA particles.

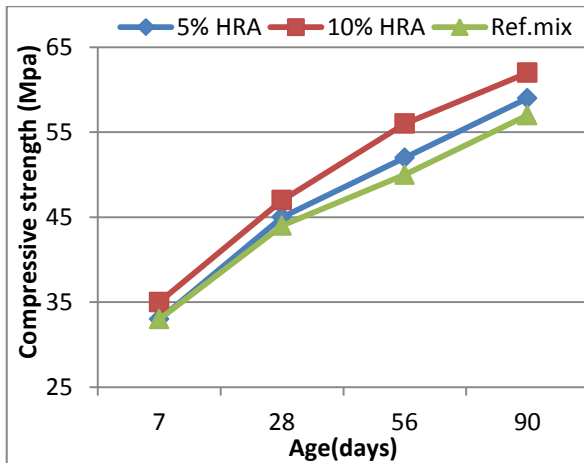


Figure 3: Compressive strength for binary blended cement of (OPC+HRA)

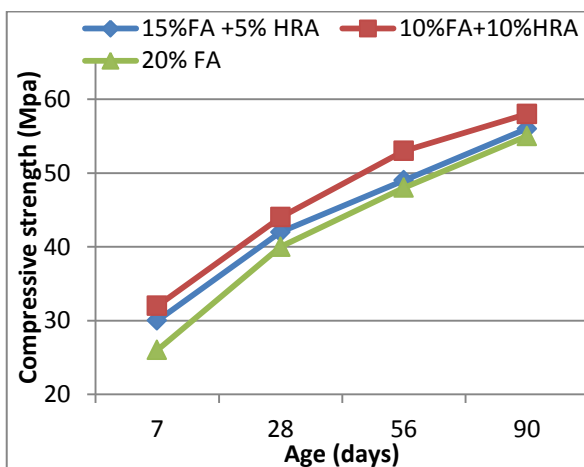


Figure 4: Compressive strength for ternary blended cement at total replacement percentage of 20%.

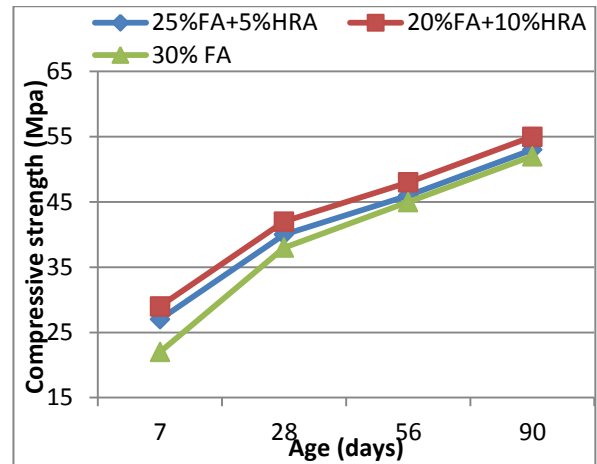


Figure 5: Compressive strength for ternary blended cement at total replacement percentage of 30%.

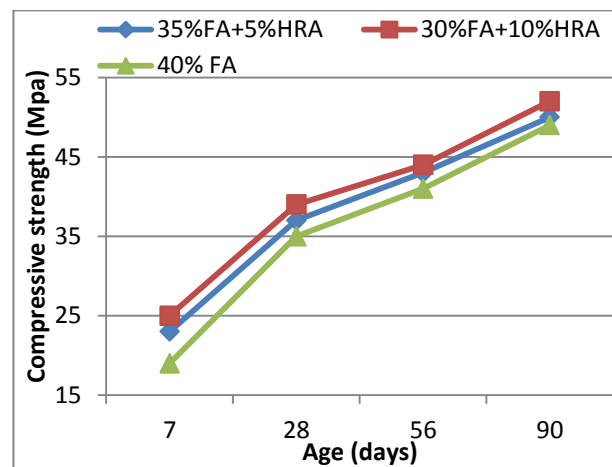


Figure 6: Compressive strength for ternary blended cement at total replacement percentage of 40%.

V. Efficiency factor (*k*)

The efficiency factor of a pozzolana can be defined as the part of pozzolanic materials which can be considered equivalent to Portland cement or the percentage of cement in concrete mixture which can be replaced by a pozzolanic material without effect on concrete strength [8]. For Portland cement $k=1$. When k is more than 1, it indicates the pozzolanic materials used are more efficient than cement, and hydration process is fast compared to cement. In such a case using of lower cement content is possible to obtain economic mix design of concrete. When k is less than 1, it means the pozzolanic materials used is less efficient than cement as hydration process is slow compared to cement [22]. The quantity of the pozzolanic materials can be multiplied by the k value to evaluate the equivalent cement content in the mixture, and by calculating k values the mix design for the preparation of the building product can be easier and more accurate [23]. In this study Bolomoy's equation [24] below was adopted to estimate efficiency factor:

$$f_c = K\left(\frac{C}{W} - 0.5\right) \quad (1)$$

To calculate efficiency factor for binary and ternary blended cement the above equation modified by Khokhar et. al[10] can be used:

$$f_c = K\left(\frac{C + kP}{W} - 0.5\right) \quad (2)$$

Where f_c is compressive strength for concrete mixes in (MPa), K is Bolomoy's constant which can be calculated from equation (1) for cement only mixes and assumed to be constant for a given type of cement and aggregate but varies with age. C is cement content (kg/m^3), W is water content in (kg/m^3), P is pozzolanic material

Table 6: Gain in strength for ternary blended cement of FA+5% HRA relative to (OPC+FA)

% Total replacement by wt. of cement	7 days	28 days	56 days	90 days
	% Gain in strength relative to OPC+FA for:	% Gain in strength relative to OPC+FA for:	% Gain in strength relative to OPC+FA for:	% Gain in strength relative to OPC+FA for:
	FA+5% HRA	FA+5%HRA	FA+5% HRA	FA+5% HRA
20%	15.4	5	2.1	1.8
30%	12.5	5.3	2.2	1.9
40%	9.5	5.7	4.9	2.0

Table 7: Gain in strength for ternary blended cement of FA+10% HRA relative to (OPC+FA)

% total replacement by wt. of cement	7 days	28 days	56 days	90 days
	%Gain in strength relative to OPC+FA for:	% Gain in strength relative to OPC+FA for:	% Gain in strength relative to OPC+FA for:	%Gain in strength relative to OPC+FA for:
	FA+10%HRA	FA+10%HRA	FA+10%HRA	FA+10%HRA
20%	23.0	10	10.4	5.5
30%	20.8	10.5	6.7	5.8
40%	19.0	11.4	7.3	6.1

Table 8: Efficiency factor (k) for binary and ternary blended cement

Mixes designation	7 days	28 days	56 days	90 days
20% FA	0.15	0.64	0.84	0.86
30% FA	0.11	0.63	0.733	0.77
40% FA	0.1	0.6	0.64	0.72
5% HRA	1.0	1.36	1.64	1.56
10% HRA	1.48	1.55	1.96	1.7
15%FA + 5% HRA	0.63	0.81	0.92	0.9
10%FA+10%HRA	0.88	1.0	1.24	1.1
25%FA+5%HRA	0.51	0.75	0.78	0.8
20%FA+10%HRA	0.67	0.87	0.9	0.9
35%FA+5%HRA	0.4	0.68	0.72	0.75
30%FA+10%HRA	0.51	0.7	0.76	0.8

content in concrete (kg/m^3) and k is efficiency factor which is be different with changing of type of mineral admixture used, age and percentages of replacement with cement. Table 8 above included the results of efficiency factor. For binary blended cement of (OPC+FA) the results shows that k values is lower at early age and

reduced with increasing of replacement percentage of cement with FA due to dilution of cement with low activity material and slow nature of FA reactivity at early age. The results show also that k values increased with increasing of concrete age. Ultimate k value was noticed at replacement percentage of 20% FA. For binary

blended cement of HRA, the results show that k values for mixes contained 5% and 10% HRA is more than 1 because of high pozzolanic reactivity of HRA. The results show that k values increased with increasing of concrete age and higher k value was noticed at concrete age of 56 days. For ternary blended cement the results show that for the same replacement percentages of cement with mineral admixtures k values for ternary blended are always higher than the corresponding binary blended cement due to synergic or combined effect when FA and HRA were used together, high efficiency of HRA as a pozzolanic material and high fineness of HRA particles could cause densifying of concrete microstructure by producing additional CSH gel and reducing intermediate transition zone porosity by filling and strengthening effects.

4. Conclusions

1- Using of ternary blended cement (ordinary Portland cement + fly ash + high reactivity Attapulgite) led to reduce slump relative to Ref. mix and binary blended cement of (OPC+FA) due to the morphology of like – plate HRA particles and increasing of paste volume.

2- Binary blended cement of (OPC+FA) exhibited lower compressive strength relative to reference mix. The reductions in compressive strength were more significant at early age and increased directly with increasing of replacement percentages of FA by weight of cement. The percentages of reduction in compressive strength were (21%, 9%, 4% and 3.5%), (33%, 14%, 10% and 9%) and (42%, 20%, 18% and 14%) relative to reference mix for replacement percentages of 20%, 30% and 40% by weight of cement at ages of (7, 28, 56 and 90) days respectively. For binary blended cement of (OPC+HRA) the results demonstrated that the using of HRA increased compressive strength at all ages and replacement levels due to its high surface area and pozzolanic activity. Ultimate compressive strength obtained with replacement percentage of 10% HRA by weight of cement, and the percentages of increment were (6%, 7%, 12% and 9%) relative to reference mix at ages of (7, 28, 56 and 90) days respectively.

3- Compressive strength of ternary blended cement is always higher than that for binary blended cement of (OPC+FA) at the same replacement percentages by weight of cement due to the synergic effect of FA and HRA. The increments in compressive strength were more significant at early age. (OPC+FA+10%HRA) mixes showed more synergic effect than (OPC+FA+5% HRA) mixes. Ultimate synergy

was noticed with mixes of (OPC+10 %FA+10% HRA).

4- Efficiency factors for binary blended cement of (OPC+FA) are lower than 1 at all ages and replacement percentages and decreased with increasing of FA replacement percentages by weight of cement. While for (OPC+HRA) efficiency factor is always more than 1 due to high reactivity and surface area of HRA. Ternary blended cement exhibited increments in efficiency factor values when compared with the corresponding binary blended cement of (OPC+FA) and the increment in k values seem to be more intrinsic at age of 7 days.

5- Using of ternary blended cement of (OPC+FA+HRA) considered an important opportunity to proceed forward in sustainable concept, by increasing the ability to replace cement causing dangerous environmental harmfulness with high replacement levels of a by- product FA, and exceeding the problems associated with FA using in concrete especially low strength improvement at early age.

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