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MECHANICAL TREATMENT OF FLY ASHES PART II: PARTICLE MORPHOLOGIES IN GROUND FLY ASHES (GFA) AND WORKABILITY OF GFA-CEMENT MORTARS

J. Payá, J. Monzó, M.V. Borrachero, E. Peris-Mora and E. González-López
Departamento de Ingeniería de la Construcción
Universidad Politécnica de Valencia
Camino de Vera s/n E-46071 Valencia (Spain)

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ABSTRACT

Mechanical treatment (by grinding) effects on particle morphology and specific gravity of fly ashes, and workability of ground fly ash (GFA) cement mortars have been studied. Different shape morphologies of GFA particles have been established: shell shaped and irregular solid fragments. Real and bulk specific gravity values were measured, proving that grinding process increased the content of poor shape particles. Particle Packing Factor (PPF) for GFA decreased below 50%. Workability of GFA-cement mortars is negatively affected, but it is still greater that only cement mortar one. Good correlations between flow table spread (FTS) values per water volume unit and fly ash replacing percentage have been obtained, and a relative workability factor W_r is established. Determination of W_r value permits to compare the effect of grinding or other fly ash processing methods on workability of mortars. Finally, good linear relationships between W_r values and the inverse of mean diameter particle or calculated specific surface area were found.

Introduction

Upgrading of fly ashes for their use in mortar and concrete is the subject of several investigations in recent years. Mechanical treatment of fly ashes by milling is one of the known methods; however, few specific studies, to the best our knowledge, on their influence on ground fly ash-cement mortars have been reported (1,2).

Its generally recognized that loss on ignition, fineness, particle distribution, shape morphology and smoothness surface of fly ash particles influence decisively on rheological behavior, flowability and workability of fly ash-cement mixtures. So, for example, Uchikawa and col. (3) established a fluidity evaluation index which was a function of specific gravity, particle shape factor, Blaine specific surface area and porosity of fly ash powder packed under determined pressure; Nagataki and col. (4) found a good correlation between the viscosity of fly ash or fly ash-cement pastes and bulk specific gravity of fly ashes; Dhir and col. (5,6) studied a wide spectrum of fly ashes showing fineness as the single most significant physical parameter characterizing water-reducing ability of fly ashes in concrete, but better correlations were obtained between the product of fineness (as percentage retained on 45 μm sieve) by loss on ignition, and the reduction in water demand or the increase in slump of concrete; Hughes and col. (7) proposed the grading modulus of fly ash for predicting workability (measured as VB time). On our part, we established (8) a good correlation between workability (measured as flow table spread) of standard mortars with several substitution percentages of air classified fly ashes and mean diameter of fly ashes.

Mechanical treatment of fly ashes, obviously, crushes a lot of particles and shape morphology and smoothness surface of particles change; moreover, increasing fineness is achieved by grinding (1,2,9). Since fly ash consists largely of multi-sized solid and hollow particles of spherical shape, several authors agree that the presence of fly ash in cement mixtures improves their workability and water-demand due to ball-bearing effect (10), dispersion effect in cement particles (11), hydrophilic character (12) and increased packing density of the solid material (13).

Preliminary studies on physical, mineralogical, morphological and chemical characteristics of ground fly ashes (GFA from now on) have been recently reported (9). In this paper a more deep study on shape morphology of GFA and workability of GFA-cement mortars are presented. A useful indication of the workability of fly ash-cement mortars is proposed: relative workability factor (RWF) of fly ash.

Experimental Section

Materials, Apparatus and Procedures. A low calcium fly ash was used in cement mixture preparation. Data of cement composition, fine aggregate, water and fly ash were previously reported (9,14). A laboratory ball-mill (Gabbrielli Mill) was used for grinding original fly ash (T0); three GFA samples were obtained for varying grinding time: T10 (ten minutes), T40 (forty minutes) and T60 (sixty minutes). The scanning electron microphotographs were taken with an ISI-DS-130 scanning electron microscope (SEM) equipped with an energy dispersive X-ray (EDX) attachment (Kevex 8000 model) with 20 kV accelerating voltage. Mortars were prepared by mixing 450 g of cement, 1350 g of natural sand and different water volumes from 200 to 225 mL for control mortar, and replacing a part of cement by original fly ash T0 or GFA samples (T10, T40, T60) from 15 to 60 % in weight. Workability of mortars were determined according to the method already described (8) using a flow table. FTS are given in mm, as a mean of maximun and minimun diameters of the spread cone, with an accuracy of ±1 mm. Real specific gravity was determined using standard Le Chatelier flask as ASTM C-188 indicates and toluene (reagent grade) was used in the determination.

Results and Discussion

Shape morphology of GFA. External and internal particle morphology of fly ashes have been studied by Diamond (15), by Lauf (16), and by Fisher and col. (17), finding and characterizing several types of particles; spherical and spheroidal shapes: hollow particles (cenospheres and plerospheres) and solid particles; irregular shapes: vesicular, porous and highly irregular particles. In a recent paper, new shape morphologies have been identified for GFA (9). Now, a more detailed study on "new" morphologies is presented. Figure 1 shows a wide range of morphologies examinated under the scanning electron microscope.

As a whole, GFA showed two types of general morphologies: a) a large proportion of the

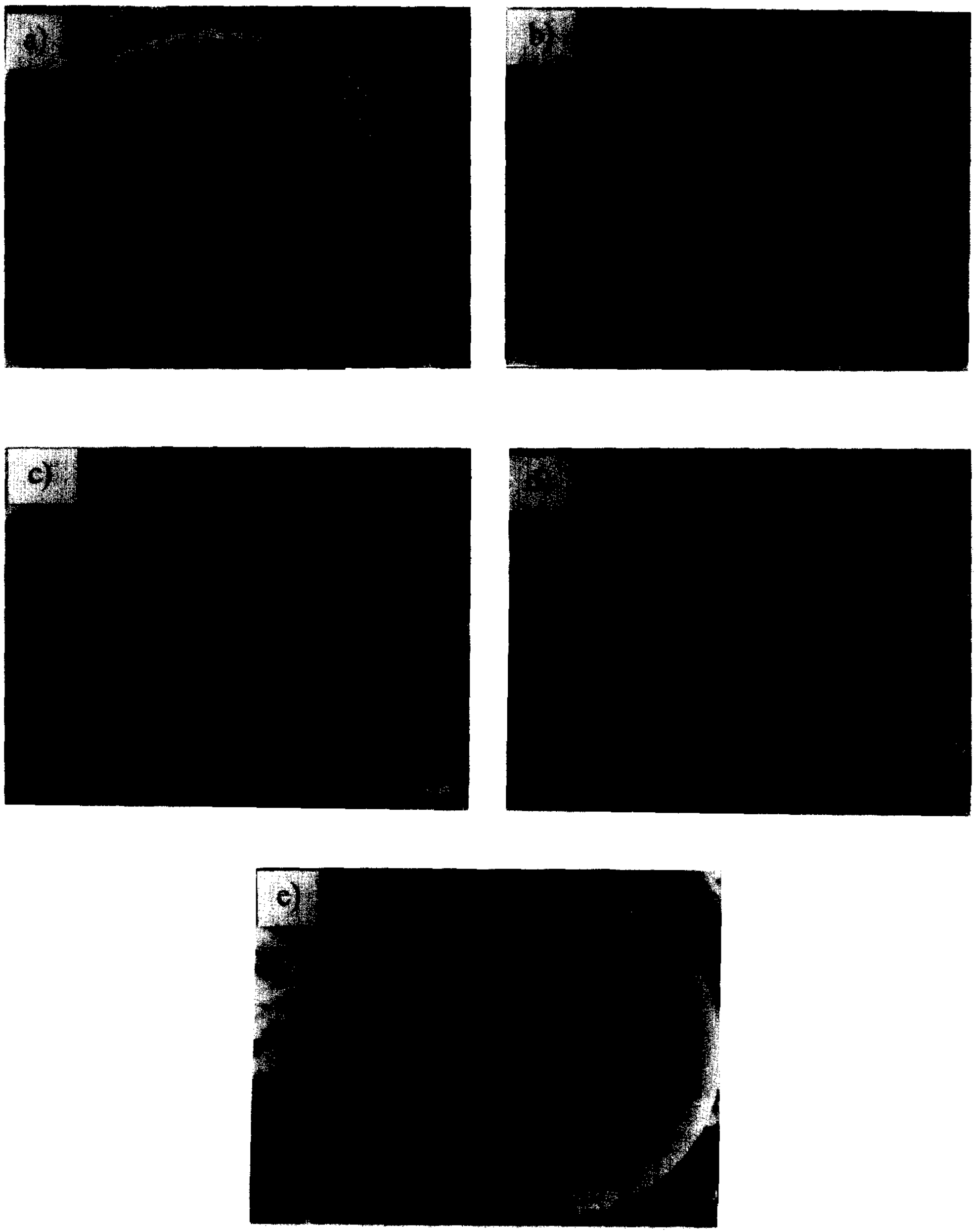

FIG. 1.
SEM microphotographs: a–e) Shell–shaped fragments.

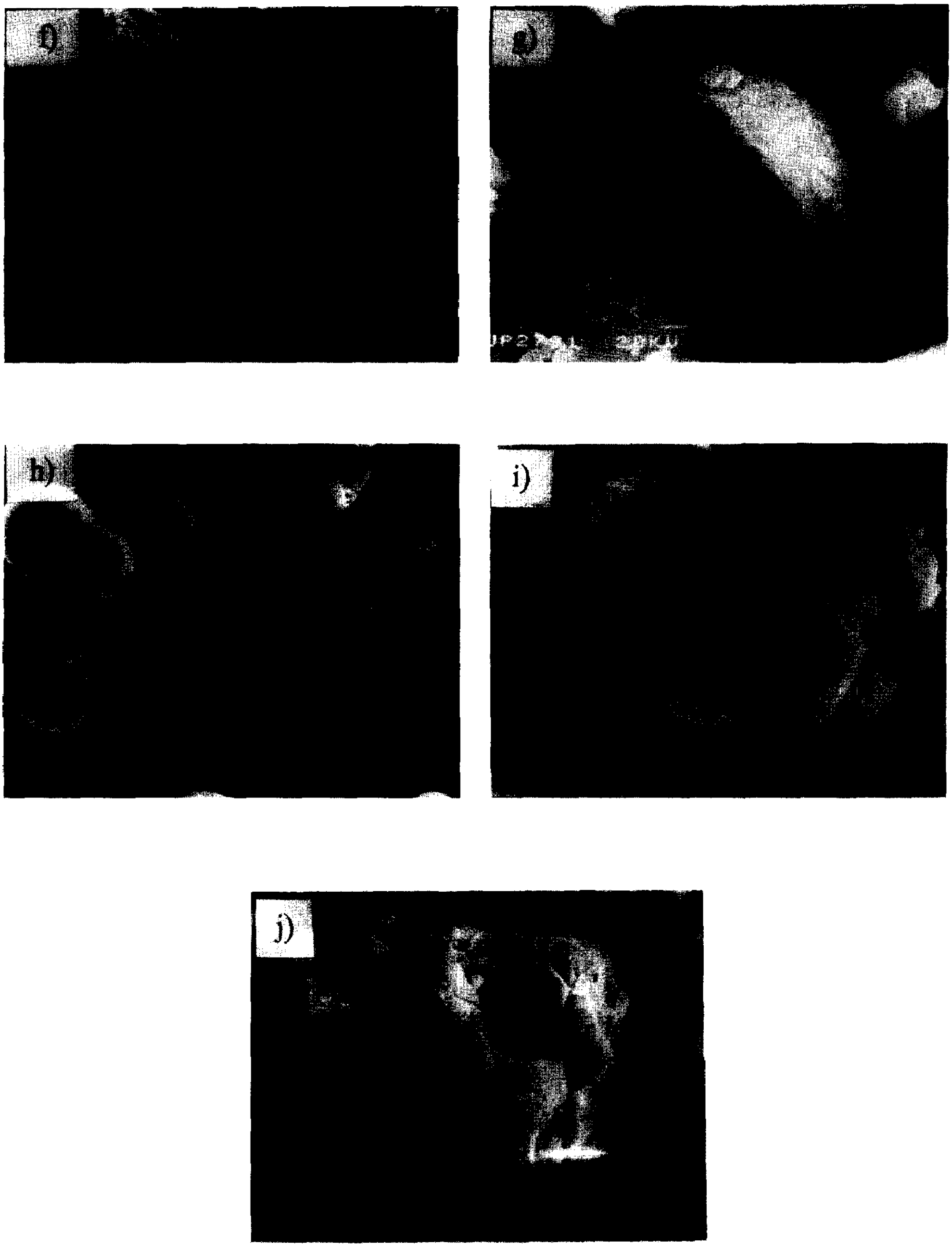

FIG. 1. (cont.)
SEM microphotographs: f–j) solid fragments.

fly ash particles remained unaltered, retaining their optimum spherical conformation; and b) spheres which ranged in size from 20 μm to well over 100 μm were break down yielding two different morphologies: b1) shell-shaped fragments originated from cracking of cenospheres and plerospheres; and b2) solid fragments from solid particles, in part maintaining partially the original morphology (semispheres, eight-part- of spheres, cap-shape particles, etc...) and the other part showing splintered and highly irregular shapes.

Plates 1a to 1e show shell-shaped fragments; plate 1a shows a shell fragment from a vesicular hollow particle; three views of a shell-fragment from a large thin-walled cenosphere are exhibited in plates 1b, 1c and 1d, where little air bubbles into the wall can be observed. Plate 1e show a sliced-in-two cenosphere with a signs of impacts onto surface of the right part. Plates 1f to 1j show solid fragments; microphotographs 1f and 1g contain broken particles from original spherical fly ash particles; in plate 1g little voids are observed into the "new surface" of the particle, indicating that original particle was a "bubblesphere". Microphotographs 1h, 1i and 1j represent typical highly irregular shapes; it can be noticed that particles present small deposits as cracking products and surface irregularities due to brittle cracking pattern.

Specific Gravity of GFA. Grinding process involves crushing particles; so, unburned carbon particles and hollow particles are easily crushed because of their brittleness (9), and, when grinding, an increase of real specific gravity (ρ_R) is observed. However, from the point of view of workability behaviour, bulk or apparent specific gravity (ρ_B) has a more decisive influence on viscosity of fly ash-cement pastes (4). Table 1 summarizes real and bulk specific gravities for original (T0) and GFA (T10, T40 and T60); ρ_B values were measured after compacting a fly ash sample using a flow table (100 drops, one per second). Particle packing factor (PPF) is defined as ρ_B/ρ_R ratio in percentage, and PPF is related to the shape morphology of particles and indicates the possibility of packing. So, a low PPF values reveals high content of particles with poor shapes and, consequently, affecting negatively workability.

Additional information on specific gravity is showed in Table 1: original fly ash was sieved (18) yielding three sized-fractions (sieved fly ash: SFA), named TX (coarsest fraction), TY (medium size fraction) and TZ (finest one). TX shows the lowest real specific gravity value,

TABLE 1

Real (ρ_R) and Bulk (ρ_B) Specific Gravities and Particle Packing Factor (PPF) for Several Processed Fly Ashes

Fly Ash	ρ_R (g/cm^3)	ρ_B (g/cm^3)	PPF=(ρ_B/ρ_R)*100
T0	2.44	1.54	63.1
T10	2.60	1.32	50.7
T40	2.64	1.17	44.3
T60	2.69	1.13	42.0
TX	2.31	1.15	50.0
TY	2.44	1.48	60.7
TZ	2.46	1.28	52.0
TX60	2.71	1.09	40.2
TY60	2.65	1.16	43.8
TZ60	2.62	1.20	45.8

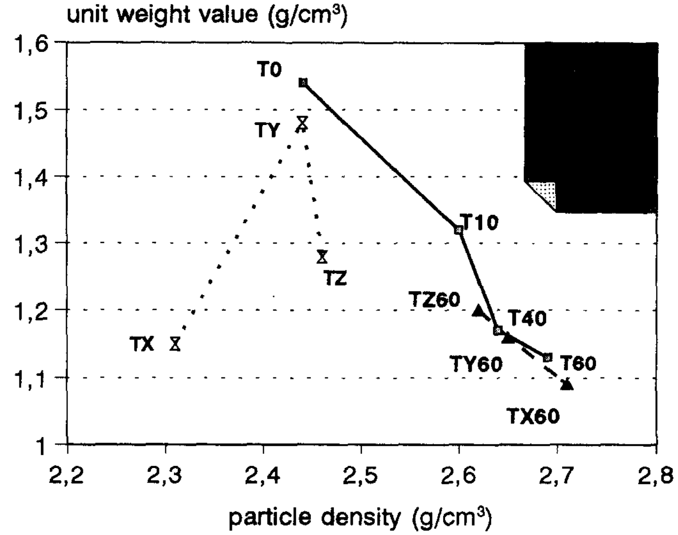

FIG. 2.
Comparison between real (ρ_R) and bulk specific (ρ_B) gravities for ground (GFA), sieved (SFA) and sieved and grinding (SFA+G) fly ash samples.

indicating that is enriched in large cenospheres, whereas highest ρ_R is found for TZ. However, TY presents highest PPF value for SFA samples, indicating that fineness and bulk specific gravity are not directly related, and granulometric distribution has a decisive rol, that is the reason that T0 shows the highest PPF value in the Table 1.

On the other hand, when these SFA samples were ground during 60 minutes (yielding TX60, TY60 and TZ60 respectively) ρ_R is clearly increased; however, highest ρ_B values appears for coarsest fraction TX60, indicating that majority of cenospheres are broken, whereas for TZ60 ρ_R is only 2.62 g/cm^3 suggesting the existence of little hollow particles, cenospheres and "bublespheres", which were not crushed when milling and/or less density for little particles due to high vitreous content. Again, ground sieved fly ashes (SFA+G) yielded PPF values less than 50%, indicating a poor shape morphology. Figure 2 shows correlation between ρ_R and ρ_B for several processed fly ashes.

Workability of Mortars Containing GFA. Flow table spread (FTS) measurements of mortars showed a good correlations with water/(cement + fly ash) ratios in the range 0.5 to 0.45 (8). Now, the procedure is applied for GFA-cement mortars in the same range. Standard control mortars with cement prepared with 200, 208, 216 and 225 mL of water yielded the following FTS values: 107.25, 112.75, 119 and 126.5 mm respectively. FTS values for mortars containing GFA in 15, 30, 45 and 60 % substitution of cement are given in Table 2.

Figures 3a and 3b show linear relationships between FTS values and water volumes for 15 and 60 % fly ash replacing percentages respectively. Generally, an increasing of FTS values with water volume can be observed; original fly ash notably enhances workability respect to "only cement" control mortar. When GFA is used, a clear reduction in FTS values of mortars is observed but, interestingly, being their workability greater than "only cement" one for water volume range studied. A linear correlation between FTS values for

TABLE 2

FTS values (in mm) for mortars containing GFA
(Each section, first value for 200 mL,
last one for 225 mL)

		% of substitution			
		15 %	30 %	45 %	60 %
FLY ASH	T0	113 117.5 127.5 133.5	117.5 131.25 137.25 149.25	126.75 134.75 140.75 152	142.5 148 153.25 164
	T10	111.75 119 125.5 134.25	119 125.75 127.5 142.25	120 129 129.5 146.75	135.75 142 149.25 160.5
	T40	110 116.25 120.25 129.75	112 117 123.75 134.25	119.75 122.75 125.75 138.75	124.5 133 141.5 148.5
	T60	110 118.25 122.5 130	112 118.75 124.75 130.75	113 122.75 129.75 137.5	120.25 133.25 136 141.5

each water volume and the replaced percentage of cement is also found. Figures 4a and 4b show relationships for 200 mL and 225 mL of water respectively. Greatest workability differences of GFA-cement mortars for highest susbtitution percentage are observed in both cases.

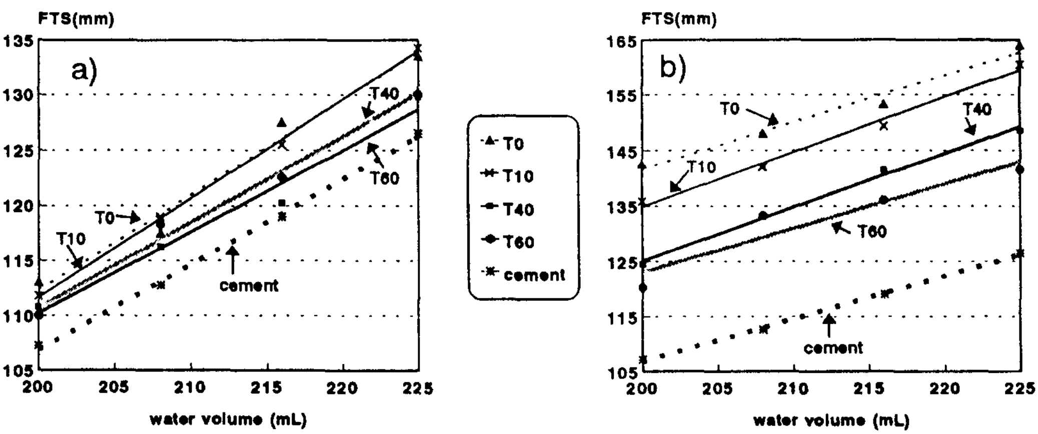

FIG. 3.
Correlations between FTS values and water volume for: a) 15 % substitution percentage; b) 60 % substitution percentage.

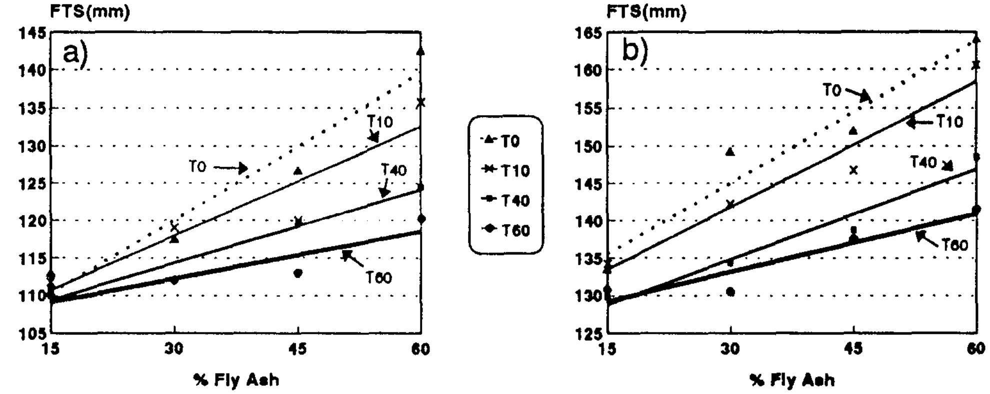

FIG. 4.
Correlations between FTS values for mortars with: a) 200 mL; b) 225 mL of water and the replaced percentage of cement.

So, for a given fly ash, we can estimate the yielded flow per water volume unit as FTS/V; a linear correlation between FTS/V values and the fly ash replacing content is also established (Figure 5). Table 3 summarizes regression data parameters for original fly ash and GFA; additionally, regression data obtained from workability studies on air-classified fly ashes ACFA (8) are also listed (T0*, T1, T2, T3 and T4 fly ashes). T0 and T0* fly ashes were from the same source, but cements used were slightly different.

A value represents the estimate FTS/V for only cement mortars. W value is the slope of the linear regression and it represents the fly ash workability factor for a given fly ash when certain cement and sand are used. But, if different fly ashes, obtained from an original one, may be compared, a new workability parameter W_r (relative workability factor) is defined as W_i/W_o ratio, being W_o the slope for original fly ash and W_i the slope

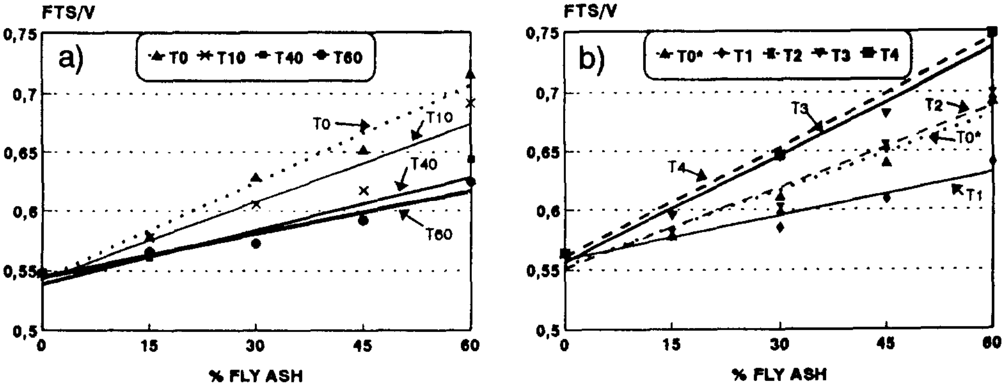

FIG. 5.
Linear dependence of FTS/V ratio with fly ash substitution percentage (only mean values for a given percentage are represented for clarity).

TABLE 3

Linear Regression Data for Dependence of FTS/V Ratio with Percentage (%) Substitution of Cement: FTS/V = A + W*%

Fly Ash		A (mm/mL)	W*1000 (mm/mL)	R-squared	Mean abs. % error
GFA series	T0	0.5428	2.729	0.980	1.18
	T10	0.5423	2.187	0.925	1.60
	T40	0.5387	1.502	0.893	1.65
	T60	0.5445	1.203	0.940	1.03
ACFA series	T0#	0.5533	2.129	0.963	1.41
	T1	0.5582	1.233	0.938	0.98
	T2	0.5510	2.284	0.949	1.55
	T3	0.5564	2.994	0.987	1.02
	T4	0.5608	3.067	0.997	0.63

for the treated one. So, we can observe that when a fly ash is mechanically treated, a diminution of W_r factor is produced, but for air-classified fly ashes W_r values greater than unit can be obtained.

Table 4 summarizes W_r values and fineness data for the studied fly ashes.Certainly, the relative workability factor W_r depends on fly ash fineness parameters. So, good correlations between W_r and specific surface area (S_m) and the inverse of mean particle diameter ($1/d_m$) are observed (Figure 6).

Figure 6a shows the close linear correlation between calculated specific surface area S_m and relative workability factor W_r; the linear correlation for ground fly ashes (GFA) was 0.99, whereas worse correlation was obtained for air-classified fly ashes (ACFA) (r=0.88) Similarly, linear correlations between W_r and $1/d_m$ were obtained (Figure 6b) with appreciably better coefficients of correlation (r=0.998 for GFA and r=0.90 for ACFA).

TABLE 4

Relative Workability Factor (W_r) and Finenes Parameters for GFA and ACFA

Treatment	Fly Ash	W_r	Specific surface area S_m (m^2/Kg)	Mean diameter d_m(μm)	$1/d_m$ (μm^{-1})
GFA	T0	1	398	32.2	0.031
	T10	0.8014	623	13.5	0.074
	T40	0.5504	850	6.9	0.145
	T60	0.4408	893	5.9	0.169
ACFA	T0#	1	419	26.8	0.037
	T1	0.5791	286	53.6	0.019
	T2	0.8369	333	30.6	0.033
	T3	1.4054	410	18.1	0.055
	T4	1.4406	527	11.2	0.089

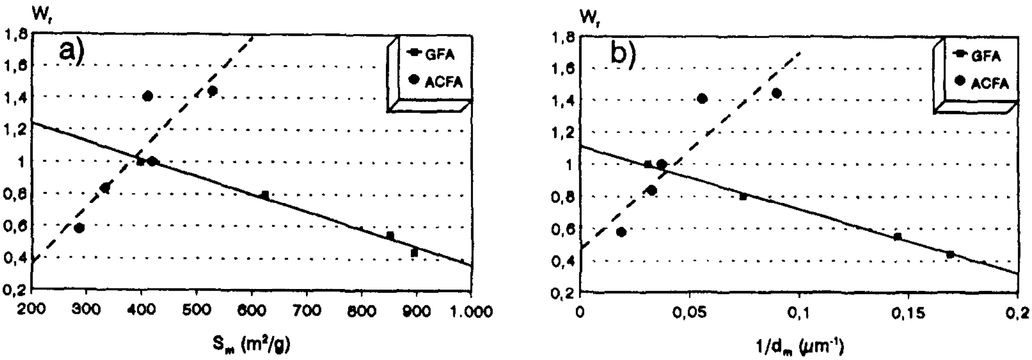

FIG. 6.
Relationships between relative workability factor (W_r) of GFA and ACFA and: a) specific surface area (S_m); b) inverse of mean diameter ($1/d_m$).

Conclusions

Based on the experimental reported in this paper about ground fly ashes (GFA), the following conclusions are made:

1- Grinding of fly ashes mainly break down coarse particles, yielding shell-shaped fragments, from original cenospheres and plerospheres, and solid fragments with or without retaining partially original spherical shape.
2- Increasing of specific gravity of fly ashes is achieved when grinding due to crushing hollow particles, and inversely, bulk specific gravity of fly ashes decreased due to loss of optimum spherical shape morphology.
3- Particle packing factor (PPF), defined as bulk to real specific gravity ratio, revealed higher content of poor shape particles for GFA (PPF less than 50 %).
4- Linear correlations between flow table spread (FTS) values and water volume or fly ash replacing percentage were obtained. In general, a clear diminution of workability was observed when grinding time is increased.
5- GFA-cement mortars yielded greater workability than only cement mortar one for water volume and fly ash replacing percentage ranges studied.
6- Yielded flow per water volume unit FTS/V were estimated, finding a good linear correlation with fly ash replacing percentage, their slope representing a fly ash workability factor W.
7- A relative workability factor W_r was proposed in order to compare the effect of two fly ash processing methods: grinding and air classification. W_r was less than unit for mechanically treated fly ashes, whereas W_r values greater than unit were determined for non-destructive methods.
8- Clear dependence for W_r with the inverse of mean diameter or calculated specific surface were established; W_r decreased with fineness increasing for GFA, contrary to air classified fly ash behavior.

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