Pergamon

Cement and Concrete Research, Vol. 25, No. 2, pp. 395-407, 1995

0008-8846/95 $9.50+.00

0008-8846(95)00025-9

CHARACTERIZATION AND POZZOLANIC PROPERTIES OF SILICA FUME STORED IN AN OPEN POND

David Bonen[1] and Kamal H. Khayat
Department of Civil Engineering
University of Sherbrooke
Sherbrooke, QC Canada J1K 2R1

(Refereed)
(Received March 25; in final form November 1, 1994)

ABSTRACT

A wide range of properties (e.g., physical, chemical, mineralogical, microstructural, and pozzolanic) of silica fume are reported. This silica fume was derived from silicon metal production and stored in ponds under water for about a year. Its properties are compared to those of the silica fume from the daily production. The characteristics of the ponded silica fume are high SiO_2 >95%, LOI <2.6%, and total trace element content of about 3000 ppm. It crystalline matter is composed of quartz, cristobalite, silicon metal, silicon carbide, and wood chips. This silica fume has undergone some degree of diagenetic process that yielded aggregation and degraded its pozzolanic reactivity as measured by the rate of consumption of $Ca(OH)_2$. In turn, the silica fume from the daily production is characterized by its high pozzolanic activity. Nevertheless, the compressive strength of these two types of silica fume is about the same. This is in accord with other findings illustrating that strength gain results from both the filler effect and the pozzolanic properties.

Introduction

As part of a research project, the properties of silica fume that was dumped in an open pond were evaluated. At the time of production this silica fume was not sold either due to overproduction, or concern was raised as to its possible interference with other chemical admixtures, such as, air-entraining agents. The "finger prints" of this silica fume were "bubbling" effects as gas was generated during immersion in water. Further details about this gas were not available. As little information on the properties of this kind of silica fume exists, a detailed characterization was carried out.

The rejected silica fume was dumped in open artificial ponds that were lined with double-layer thick plastic sheets to avoid contamination of both the ground water and the dumped silica

[1]Present address: Department of civil Engineering, Northwestern University, 2145 Sheridan RD., Evanston, IL 60208, U.S.A.

fume. This research investigation was carried out on the silica fume stored in the last of a series of ponds. This pond contained about 4400 tons of silica fume, most of it was continuously submerged in water and only minor quantities exposed to air were on the slopes of the pond. The height of the water level above the silica fume bed varied from about 1.5 to 4 m and averaged to 2.5 m. It has been found that the silica fume in the pond neither possessed a uniform thickness, nor the same degree of consolidation. Consolidation probably took place due to some amount of aggregation, compaction, dissolution and precipitation.

In order to retrieve the submerged silica fume, a rectangular grid was set, and the silica fume was sampled at intersections of "rows" and "columns" of this grid by employing a modified Shelby sampler device operated by two persons from a boat. In all, a total of 29 specimens was taken: 25 specimens submerged in water, and four from the sides of the pond that were above the water level. In addition, five more samples from the daily production of the plant were utilized in this research, in which four were in the form of finely divided powder, and one was in a granulated (pelletizied) form. The latter was prepared in a commercial granulating dish by adding water to the powder silica fume in order to facilitate it handling. Granulated silica fume is also ground with cement clinker to produce blended silica fume cement.

The objectives of this paper are to characterize the microstructure and pozzolanic properties of the ponded silica fume, and to report on a possible alteration due to a continuous storage under water.

Analytical Procedure and Method

Table 1 summarizes the analytical procedures and instrumentation used in this investigation. Free-water content was determined on representative silica fume specimens by repeatedly quartering each sample down to about 100 g, and drying it out in an oven at 100°C for 24 hours. Loss-on-ignition (LOI) measurements were carried out by placing about 10 g of the oven-dried silica fume in a furnace at 1000°C for 4 hours. In addition, the weight loss of the samples between 100 and 1000°C was recorded by TGA in an oxygen-free environment, thus eliminating oxidation. The TGA weight loss is presented on the basis of the initial weight of the samples.

Particle size distribution was measured by a sedimentation technique. For disintegrating aggregated materials, approximately 45 g of dry silica fume was placed in 990 mL of distilled water containing 10 mL of deflocculating agent and suspended in a blender for 15 seconds. The suspension allowed to rest for about 2 days, and its density change was measured at set intervals by a calibrated hydrometer. The equivalent diameters and percentages of each of the size groups were calculated according to Stoke's Law.

Specimens for scanning electron microscopy examination were prepared by dispersing a small amount of oven-dried silica fume in methanol, and mounting it on microscopical graphite grids. Since the graphite grid is conductive, no further coating was needed.

Pozzolanic Reactivity

The pozzolanic activity was determined by measuring the rate of consumption of $Ca(OH)_2$ that reacted with silica fume. Pastes composed of 26 g of analytical grade $Ca(OH)_2$ powder, 12 g of silica fume, 25 g of double distilled water, and 0.76 g of naphthalene-based superplasticizer were mixed manually for 10 minutes. The water was added to the superplasticizer and followed

Table 1: Analytical Procedure

Character	Technique/Instrumentation
Free-water	100°C oven dried
$LOI_{100\text{-}1000}$ total Volatile hydrocarbons	1000°C furnace TGA (Seiko SSC/5200)
Particle size distribution	Sedimentation
Chemical composition (all elements) Carbon and sulfur	ICP (ARL 3560 AES) Leco (CS244)
Mineralogical composition	XRD (Rigaku)
Uptake rate of $Ca(OH)_2$	TGA & XRD
Microstructure	SEM (Jeol JSM 840A)
Compressive strength	Testing machine

by the addition of the preblended dry ingredients. The pastes were cast in polyethylene cylinder molds, sealed off with sheets of polyethylene film, and moist cured for 1, 3, 7, and 28 days at room temperature. At the end of the curing periods, the pastes were manually ground in an agate mortar and pestle in the presence of acetone for 10 minutes, then dried out by filtration. The consumption rate of $Ca(OH)_2$ was determined by both XRD and TGA.

Compressive strength development

Mortar cubes were cured in two ways:
(a) Accelerated curing at 38°C, at variable water-to-cement ratio (w/c).
(b) Standard curing in lime-saturated water at room temperature and at a constant w/c.

1. Mixture proportioning

An ASTM Type I portland cement was used throughout. Mortars were proportioned in compliance with ASTM C109: sand (Ottawa) to cementitious material ratio of 2.75:1, and w/c of the portland cement control 0.494. The specific gravities of the cement and silica fume as measured by a liquid displacement technique were 3.14 and 2.25, respectively. For mortars subjected to accelerated curing, silica fume replaced 28% of the cement by mass (35% by volume) and the water content was adjusted so that the obtained flow spread was 100 to 115% of that of the control mixture. On average, the w/c ratio of the silica fume mortars was about 0.65 (Table 7). For the water-cured mortars, the silica fume dosage was 8% by mass (about 11% by volume) of the cement, and a constant w/c of 0.53 was employed.

2. Mixing, Curing, and Testing

The mixing procedure employed complied with ASTM C109. The silica fume, either in slurry form (the pond specimens) or as powder (for specimens from the daily production) was introduced into the mixing water, and mixed at low speed for one minute before introducing the cement. The flow spread of the mixtures prepared for accelerated curing was then measured, and if needed, water was added for securing a flow spread corresponding to 100 to 115% of that of the control mixtures. Once the desired water content was obtained, the mixtures were remixed for 15 seconds at high speed and cast into 50.8 mm cubes for compressive strength measurements.

Following casting, the cubes were covered and kept for 24 hours at room temperature, then demolded, and cured for 27 days in two ways: (a) accelerated curing: storage in sealed containers at 38°C, and (b) standard curing: storage in lime-saturated water at 23°C. At the end of the 28-day curing period, 3 to 6 cubes were tested for compressive strength determination.

Results

Some scatter in all measured properties was found. However, due to the voluminous raw data, the results are presented in average terms of two categories; the silica fume from the pond and that from the daily production.* Results of two other subcategories, namely the samples from the pond slopes (above the water), and the granulated sample from the daily production appear when their characteristics differ from those of the main categories.

1. Free-Water and LOI Contents

The free-water content of the silica fume specimens submerged in the pond ranged from a low of 36.4 to a high of 45.8% and averaged 40.7%. Interestingly, the corresponding values for the specimens from the slopes of the pond were relatively high (27.2% on average), and probably derived from frequent rain precipitation. This high free-water content illustrates the capability of the silica fume to hold water which is absorbed to the high surface area of this finely divided material.

In contrast, the silica fume from the daily production is characterized by a very low free-water content, typically less than 1%. The high free-water content of about 17% of the granulated sample is due to water addition during the granulation process.

Loss on ignition measurements yielded two sets of data: a total weight loss measured in a furnace, and volatile hydrocarbon content that was measured by TGA (Table 2). The differences in these two sets of data derive from differences in the environmental conditions in which the analyses were carried out. The furnace measurements were done in ordinary atmosphere, so all the carbon (mainly derived from wood chips, electrodes, and other furnace load) could completely be oxidized and volatilized as CO_2 gas, whereas the TGA analyses were conducted in non-oxidizing environment by flushing the samples with nitrogen. The recorded weight loss in the TGA (referred to as volatile hydrocarbon) is mainly attributed to the volatilization of hydrocarbons. But, as carbon could not be oxidized, some residual carbon was likely to be remained. Therefore, the differences between these two sets of data lie in the content of the residual fixed carbon which is not measured in the TGA analysis.

Accordingly, the total LOI content of the pond samples varied from a low of 1.7 to a high of 2.6% and averaged about 2.2%, whereas their volatile hydrocarbon content varied from 1.3 to 1.8% with an average of 1.5%. The corresponding values for the specimens from the daily production were somewhat lower with LOI content ranging from about 1.3% to 2.0%, and volatile hydrocarbon content from 1.1 to 1.4%. The total LOI content recorded here is fairly low and lies in the lower range characterizing silica fume from silicon metal production.[1]

TGA thermograms show that the specimens from the daily production have somewhat different curves than those from the pond. The former specimens are characterized by the

*The detailed data are available upon request addressed to the first author.

presence of a conspicuous shoulder at a temperature interval from about 500 to 730°C, which accounts for about one-quarter of the total weight loss. This shoulder is completely absent in the pond specimens whose thermograms are characterized by a monotonic linear weight loss. This indicates that the hydrocarbons of the pond specimens have been altered, and up to about one-fourth of them were lost during the disposal period. This conclusion conforms with the data presented in Table 2. The pond specimens have lower volatile hydrocarbons-to-carbon ratio than the corresponding ratio of the daily production specimens.

Table 2: Average LOI, Volatile Hydrocarbons (TGA), and Carbon Contents (wt. %)

	LOI (total)	Volatile Hydrocarbons	Carbon
Pond	2.2	1.5	1.7
Daily Production	1.6	1.2	1.1

2. Particle Size Distribution

Silica fume is known as a finely divided material with a BET specific surface area on the order of 20 000 m^2/kg and a mean grain size of 0.2-0.3 μm. However, since the silica fume from the pond appears to have undergone some compaction and aggregation, concern was raised as to the ability of breaking down this aggregation. Thus, prior to the analyses of particle size distribution, some shear stress was applied and the materials suspended in a blender. The results therefore, are not necessarily a measure for the surface area of the materials, but their fineness after aggregation disintegration, which could be the case prior to any potential use of the silica fume from the pond.

In all fraction sizes, the silica fume from the slopes of the pond appear as the coarsest one. This is probably related to sequences of wetting and drying. The fineness of the pond specimens submerged in water lies between those of the powder and the granulated samples from the daily production. It may be concluded therefore, that (a) the applied shear stress needed for dispersing the silica fume from the pond is on the same order as that needed for the dispersion of the granulated silica fume from the daily production, and (b) potential differences in strength development between these two categories cannot be attributed to variations in grain size distribution.

Table 3: Particle Size Distribution

Fraction[1]	Pond		Plant	
	Submerged[2]	Slopes	Powder	Granulated
< 70 μm	97.3	89.3	98.5	95
< 22 μm	94.8	85.0	97.5	91
< 6 μm	91.7	78.8	97.0	85
< 3 μm	89.7	75.3	95.5	81
< 1 μm	86.8	72.0	95.0	77

[1] Mass fraction smaller than the specified size, [2] Submerged in water.

3. Chemical and Mineralogical Composition

Table 4 shows the average and coefficient of variation (CV) of the major element

Table 4: Average Major Element Composition (wt. %)

Oxide	Daily Production			Pond		
	Average	Range	CV%	Average	Range	CV%
SiO_2	97.22	96.7-97.5	0.28	95.87	94.3-96.8	0.53
Al_2O_3	0.25	0.22-0.30	13.6	0.31	0.20-0.54	24.4
Fe_2O_3	0.17	0.13-0.23	26.8	0.61	0.31-1.03	31.7
CaO	0.27	0.24-0.31	9.2	0.40	0.28-0.57	20.6
MgO	0.46	0.37-0.58	17.6	0.50	0.40-0.62	11.7
MnO	0.03	0.02-0.04	29.9	0.06	0.03-0.10	27.3
TiO_2	0.003	0.002-0.003	16.0	0.004	0.002-0.017	71.3
Na_2O	0.13	0.12-0.15	10.2	0.11	0.08-0.16	14.7
P_2O_5	0.17	0.14-0.20	16.52	0.20	0.13-0.26	21.7
S	0.08	0.01-0.11	49.8	0.04	0.001-0.08	68.1
LOI	1.6	1.3-2.0	21.6	2.2	2.0-2.6	9.9

composition of the silica fume. The silica fume from both categories is characterized by a high SiO_2 content averaging 97.2% for the silica fume from the daily production and 95.9% for the pond specimens with small CV values. Except for SiO_2, only LOI and carbon contents (Table 2) are greater than 1 percent. Otherwise, the contents of the rest of the elements constitute a fraction of percentage each.

Interestingly, Pistilli et al.[2] analyzed 32 samples of silica fume powder from the daily production of the same plant on 32 consecutive days. Except for sulfur, the variations in the composition of the rest of the elements were greater than the corresponding CV values reported here for both, the specimens from the daily production and those from the pond. The silica fume analyzed at that time had distinctly lower SiO_2 content of 92.1% on average and higher LOI of 3.2%. The differences in the SiO_2 content appear to be related to a change in the raw feed composition. Similarly, it is doubtful whether the silica fume from the pond was manufactured from the same raw feed as that of the daily production. This conclusion is also supported by the trace element distribution. Nevertheless, the small variations in the SiO_2 content likely bear little effect on the potential pozzolanic properties of these materials.

Table 5 summarizes the average trace element composition. It is evident that the silica fume from the pond contains more impurities. The trace element content of the specimens from the daily production totals to 1900 ppm compared to 3000 ppm for the pond specimens. Much of this difference derives from the greater content of zinc (2094 vs. 1300 ppm) and lead (515 vs. 273) in the latter. In addition, the pond specimens are mainly enriched in copper (88 vs. 42) strontium (65 vs. 46), tin (55 vs. 26), and chromium (32 vs. 3), whereas, it is relatively depleted in bismuth (18 vs. 42), antimony (36 vs. 58), and vanadium (4 vs. 15). The higher scatter that is observed in the pond specimens is probably related to the number of samples analyzed, and the duration of time in which the samples were produced. The silica fume from the daily production was sampled during about two-week period, whereas that from the pond was dispersed throughout several months, and probably manufactured from several raw material shipments.

Silica fume always contains several percent of coarser materials on a micrometer to a millimeter scale. This oversized fraction is usually composed of wood chips, Si metal, SiC, quartz, and other silica polymorphs.[3] Thus, the oversized fraction of the silica fume retained on

Table 5: Average Trace Element Composition (ppm)

Oxide	Daily Production			Pond		
	Average	Range	CV%	Average	Range	CV%
ZnO	1300	1110-1440	10.4	2094	1590-2600	14.1
PbO_2	273	90-510	76.7	515	350-670	15.3
CuO	42	35-55	21.4	88	55-160	26.1
SnO_2	26	10-35	29.5	55	35-85	26.7
Cr_2O_3	3	<1-7	77.8	32	9-60	41.7
NiO	4	3-5	24.9	11	4-20	38.5
As_2O_3	41	30-50	12.2	38	25-55	25.0
BaO_2	23	20-25	8.6	32	20-50	21.8
CdO	1	1		1	1-2	32.8
La_2O_3	<1	tr.-1		1	tr.-1	18.0
MoO_3	<2	tr.-3	55.9	1	tr.-3	66.5
Sb_2O_3	58	55-60	4.9	36	13-105	64.9
Sc_2O_3	2	2		1	tr.-3	39.5
SrO	46	44-50	8.7	65	45-100	18.6
Bi_2O_3	42	35-50	12.0	18	2-50	86.2
CoO	12	11-13	9.3	4	<1-13	113
V_2O_5	15	14-16	7.2	4	<1-14	118
ZrO_2	9	9		3	<1-11	103
Total	1900			3000		

80 μm sieve was X-ray diffracted in the range of 10 to 75° 2θ (Cu Kα λ=1.5418 A was used). A conspicuous hump reaching a maximum at about 21° 2θ indicated the presence of the expected siliceous glass. The other recorded peaks corresponded to quartz, cristobalite, silicon metal, and silicon carbide. In a few runs, other small unidentified peaks were detected.

On average, the intensities of the crystalline phases of the silica fume from the daily production and that from the pond yielded similar intensities. For a given facility, the coarse fraction of the silica fume (other than wood chips) depends on the operational production conditions, such as, temperature, atmosphere, efficiency of phase separation, and cooling rate. Since no compositional differences have been observed, it may be concluded that the silica fume of these two categories were manufactured under similar operational conditions.

4. Microstructure

Figure 1 provides a characteristic TEM view of a silica fume powder specimen from the daily production. Most of the particles are rounded; some are subrounded. The particle size ranges from about 0.1 to 0.5 μm in diameter; however, some particles are extremely small: about 15 nm and less. In general, the particles are dispersed, or loosely packed in aggregates connected by narrow bridges or contact points. Tiny particles are commonly attached to larger particles. Some of these tiny particles are transparent and composed of hollow, extremely thin-shell grains (Figure 2). The granulated specimen contains more aggregated materials than the powder specimens, although its aggregates are porous and do not seem to be firmly bonded (Figure 3).

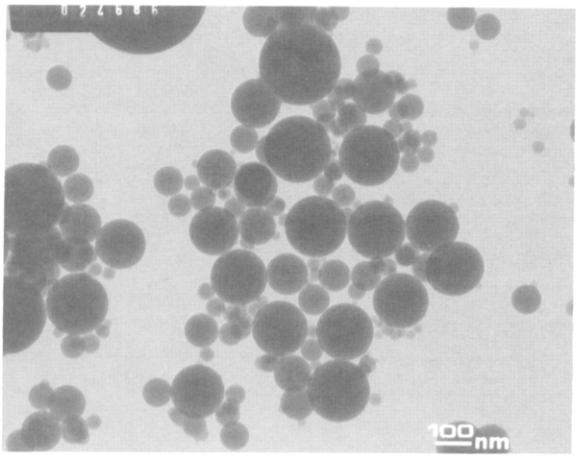

Figure 1: TEM micrograph of a powder silica fume from the daily production. The spherical silica grain particles range form about 15 to 400 nm and are loosely packed.

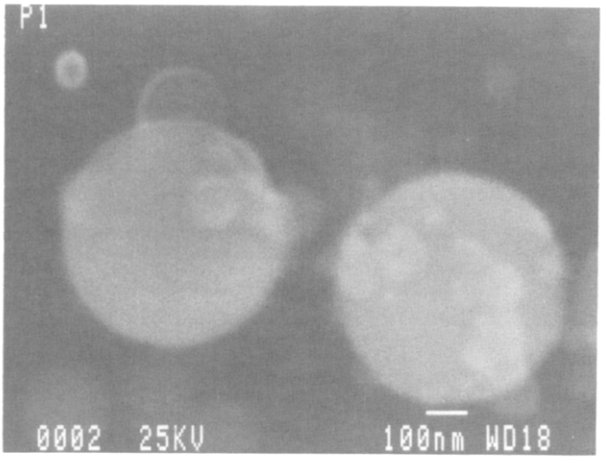

Figure 2: SEM micrograph of a powder silica fume from the daily production showing transparent, extremely thin-shell hollow silica fume grains.

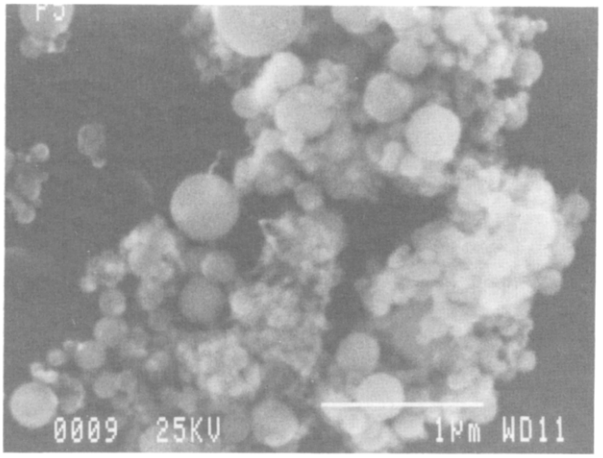

Figure 3: SEM micrograph of the granulated silica fume from the daily production form porous aggregate

The silica fume from the pond has similar particle size distribution as that from the daily production (Figure 4). However, it contains a greater proportion of aggregated materials, and these aggregates have somewhat different morphology. They are larger, more compact, and less porous implying a stronger particle cohesion due to a formation of a greater number of contact points. Some of these aggregates appear as amorphic massive lumps with various aspect ratios ranging up to several tenths of a micrometer in their larger dimension. These lumps are probably tightly packed (Figure 5). Fused-type aggregates were also encountered (Figure 6). Their rounded grains are packed by some sort of fused or cemented materials (Figure 7). The better bonding of the aggregates from the pond is probably related to a diagenetic process involving compaction, and some extent of dissolution and precipitation during the relative long disposal period under water.

5. Pozzolanic Reactivity and Strength Development

Table 6 summarizes the consumption rate of $Ca(OH)_2$ in silica fume-$Ca(OH)_2$ pastes as calculated from TGA. These results are in a complete agreement with the monotonous decrease of the $Ca(OH)_2$ peak over time measured by XRD. The silica fume from the daily production reacted with greater amount of $Ca(OH)_2$ than the pond specimens. Quantitatively, the powder silica fume consumed on the average 23, 41, 55, and 70% of the original $Ca(OH)_2$ presented in the pastes at 1, 3, 7, and 28 days, respectively, whereas the average corresponding values for the silica fume from the pond were 20, 29, 35, and 44% for 1, 3, 7, and 28 days, respectively. It is evident, therefore, that the silica fume form the daily production is more reactive than that from the pond. Interestingly, the granulated sample was found less reactive than the powder specimens, but still displayed greater reactivity than any of the pond specimens.

The evaluation of the pozzolanic activity of silica fume (as well as other pozzolanic materials) is often based on a comparison of the 28-day compressive strength of mortar samples made with and without silica fume. The two reference methods for determining the

Table 6: Mean Percentages of $Ca(OH)_2$ Retained in Lime-Silica fume Pastes [TGA (wt. %)]

Days	Daily Production			Pond	
	Powder Av.	Range	Granulated	Average	Range
2	77	74-79	81	81	74-86
3	59	52-66	66	71	67-75
7	45	44-46	61	65	62-71
28	30	29-31	47	56	51-62

pozzolanic activity index in North America are ASTM C 1240 (1993), and the corresponding Canadian method CAN/CSA-A23.5-M86 (1986). In the former test procedure, silica fume replaces 10% of the cement by mass, whereas in the latter the amount replaced is variable and depends on the specific gravities of the pozzolanic materials and the cement. For a common range of specific gravity of silica fume of 2.2 to 2.3 g/cc,[4] this means a replacement of about 35% (by volume) of the original cement. In practice, however, silica fume rarely exceeds 15% of the cement by mass and is commonly 8% or less. In this view, and for a further comparison in the future to other pozzolanic materials prepared according to ASTM C 311 test procedure, two levels of replacement were chosen, 11 and 35% by volume (8 and 28% by mass).

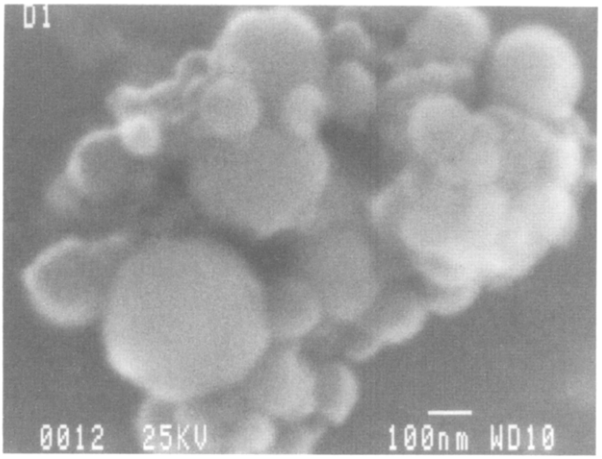

Figure 4: SEM micrograph of the silica fume from the pond. The silica fume grains cluster to form a packed aggregate.

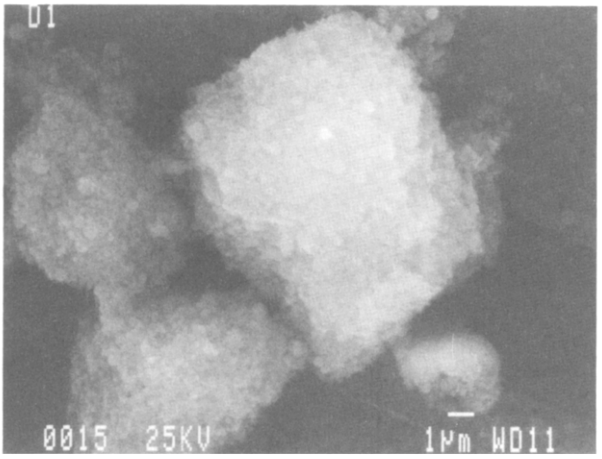

Figure 5: SEM micrograph showing tightly packed large aggregated materials that were formed in the silica fume from the pond.

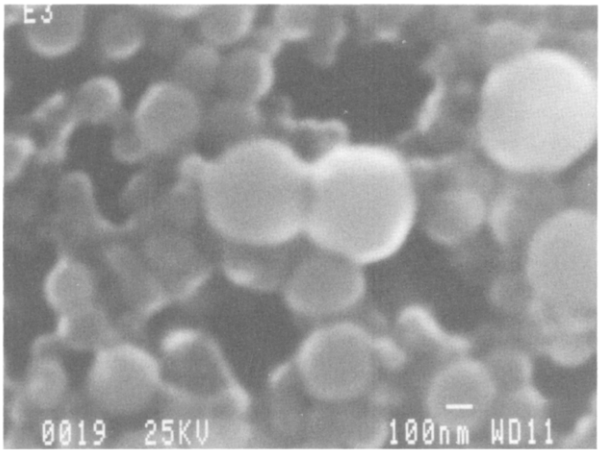

Figure 6: SEM micrograph of the silica fume from the pond presenting fused-type aggregate.

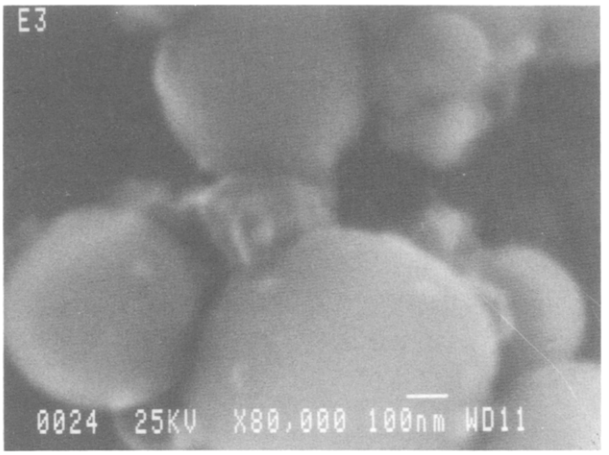

Figure 7: SEM micrograph of silica fume from the pond showing cemented-type aggregate.

Table 7: Mean 28-day Compressive Strength (MPa), Pozzolanic Index, and Water Content.

	SF vol. %	w:(c+sf)	Compressive Strength			Pozzolanic Index %	Water req. %
			MPa	Range	CV %		
Accelerated Curing - 38° C							
Cement (control)	0	0.494	25.8	-	3.0	100	100
Daily production	35	0.663	33.6	31.6-35.4	1.7	130	134
Pond	35	0.650	34.2	31.2-36.6	2.9	133	132
Water Curing - 23° C							
Cement (control)	0	0.494	24.8	-	4.3	100	100
Daily production	35	0.655	32.5	31.7-33.2	3.0	131	133
Water Curing at 23° C at Constant w/c Ratio							
Daily production	11	0.530	38.0		3.5	100	100
Pond	11	0.530	38.0	36.5-39.7	1.4	100	100

Table 7 presents the compressive strength values of the silica fume mortars and their pozzolanic indices. The latter is defined as the ratio between the compressive strength of the silica fume mortar over that of the control, times 100. Thus, the higher the pozzolanic index, the greater the pozzolanic characteristics. Similarly, the water requirement is defined as the ratio between the mixing water of the silica fume mortars and that of the control.

Relation Between the Pozzolanic Reactivity and Compressive Strength

Table 7 shows that all the mortars that contained 35% silica fume by volume possess remarkable pozzolanic characteristics, and their 28-day pozzolanic indices scatter around 130% irrespective of the curing procedure and the type of silica fume used. It may also be argued that since the control sample has the lowest w/c, then for equal water to cementitious material ratio, the pozzolanic index of the silica fume cubes should be even higher than that registered in Table 7. Interestingly, no difference between the compressive strength of the mortars containing silica fume from the daily production and those containing silica fume from the pond was recorded. The mean pozzolanic index of the former averaged 130% , whereas the corresponding average of the latter was 133%. Similar results were obtained for mortars prepared at constant w/c of 0.53

and cured at 23°C. The pozzolanic indices of the pond mortars scattered around that of the powder sample from the daily production (which served as control), and averaged 100%. These results do not conform with the clear differences in the pozzolanic reactivity of these two categories recorded by both TGA and XRD.

It should be noted that the pozzolanic index cited above is not a direct measurement for the pozzolanic activity by itself, because the compressive strength depends on several other factors. Part of the difference in the pozzolanic activity evaluated by the consumption rate of $Ca(OH)_2$ (Table 6) can be attributed to differences in morphology. It has been shown that the specimens from the daily production contain less aggregated materials, and their aggregates are loosely packed and more fragile. On the other hand, the pond specimens contain more aggregated materials and large lumps, which are less porous, more tightly packed, and occasionally fused. It may be argued, therefore, that the shear stress applied during the manual mixing for preparing the silica fume-$Ca(OH)_2$ pastes was adequate for breaking down existing loose aggregates present in the silica fume powder, but not sufficiently vigorous for comminuting the rigid aggregates present in the pond specimens. Thus, the greater pozzolanic reactivity (TGA and XRD data) of the powder specimens can be related to their higher effective surface area. Conversely, the machine mixing procedure used for mortar preparation yielded a greater shear stress that dispersed the silica fume particles more evenly irrespective of their origin.

This also explains the differences in the pozzolanic reactivity (as measured by TGA and XRD) between the powder and the granulated sample from the daily production, as the latter appears to have the coarsest particle size distribution (Table 3). But, this explanation fails to account for the differences in pozzolanic reactivity between the granulated and pond specimens (Table 6). It appears, therefore, that the long disposal period under water degraded to some extent the pozzolanic characteristics of the pond silica fume. Thus, the compressive strength development observed is not necessary a measure for the degree of pozzolanic reactivity or the amount of the C-S-H produced. Rather, strength gain depends on the beneficial "filler" effect of incorporating this ultrafine materials that densifies the interfacial zone developed between aggregate and paste, and refines the pore size distribution of the entire paste. In fact, since the same strength gain was measured, it is suggested that the contribution of the filler effect to the compressive strength is greater than that of the pozzolanic reaction. This conclusion agrees with a recent study that has shown that addition of non pozzolanic, ultrafine material, such as, carbon black, gives similar strength gain to that of silica fume.[5]

Conclusions

Based on the results presented in this paper, the following conclusions can be drawn:

1. Silica fume that has been stored under water for about a year has undergone some diagenetic processes involving a loss of hydrocarbons and aggregation and compaction which degraded its pozzolanic characteristic as measured by lime consumption.
2. Proper dispersion of existing aggregated materials plays an important role on increasing the pozzolanic reaction.
3. Aggregation of the silica fume does not apparently present any practical problem, provided that a suitable shear stress is applied during mixing.
4. Results show that the pozzolanic index of high-level silica fume mortars prepared at variable w/ and cured at a moderate temperature can be adequately evaluated by using low-level silica fume mortars, constant w/c ratio, and a curing at room temperature.
5. The strength gain of mortars containing silica fume is attributed to both the filler effect, and the pozzolanic reactivity.

References

1. V.M. Malhotra, V.S. Ramachandran, R.F. Feldman, and P-C Aitcin, "Condensed Silica Fume in Concrete," CRC Press, Inc., 221 pp. Boca Raton, 1987.
2. M.F. Pistilli, G. Rau and R. Cechner, Cem. Concr. Agg. 6[1]33-37(1984).
3. D. Bonen and S. Diamond, in Proc. 14th Inter. Confer. on Cement Microscopy. International Cement Microscopy Association, Duncanville, TX. pp. 103-113, 1992.
4. M.F. Pistilli, R. Wintersteen, and R.Cechner, Cem. Concr. Agg. 6[2] 120-124(1984).
5. A. Goldman and A. Bentur, Cem. Concr. Res. Vol. 23[4]962-972(1993).