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Fly Ashes in Cements and Concretes: Technical Needs and Opportunities

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, *Secretary*
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, *Director*

ABSTRACT

Following a brief review of the nature of fly ashes and their levels of production and use in various countries, an estimate is made of the potentially achievable level of use of fly ash in cement and concrete in the United States. The estimate assumes that 20 percent of the mass of all the portland cement used in the United States could be replaced by the same mass of fly ash; it ignores possible competition from granulated blast-furnace slag as a finely-divided mineral admixture for concrete. It appears that about 16 million tonnes (18 million tons) per year of fly ash could be consumed in cement and concrete, provided there were sufficient ash of suitable quality and a general understanding of the technical requirements for satisfactory fly ash use. Present standards which affect the use of fly ashes are discussed. Steps which could be taken to improve knowledge of factors affecting fly ash performance in cement and concrete, and hence to improve standard test methods and specifications, are outlined.

EXECUTIVE SUMMARY

This report represents, in part, the response of the National Bureau of Standards (NBS) to the mandate of Subtitle E of the Resource Conservation and Recovery Act (P.L. 94-580). Specifically, the report addresses technical needs and opportunities in the use of fly ash in cements and concretes. The Department of Commerce, through NBS, is required to provide guidelines for specifications for recovered materials, stimulate the development of markets for recovered materials, promote proven technology, and provide information necessary to allow governmental agencies to procure products with the highest feasible percentage of recovered material.

Fly ashes are finely-divided, predominantly inorganic byproducts collected from the exhaust gases from coal-burning furnaces. Although they are generally wasted they are a potentially important resource. While fly ashes have a number of uses, mostly in civil engineering, only about 13 percent (seven million tonnes) of that produced in the U.S. in 1979 was used. The remainder was disposed of by placing in stockpiles. The total amount of fly ashes stored in stockpiles is now about 300 million tonnes. The present report addresses opportunities for increased use of fly ash as an ingredient of cement and concrete (but not as a raw material for the manufacture of portland cement clinker) and outlines research needed to aid exploitation of the opportunities.

The high contents of silica and alumina in fly ashes, together with the finely-divided state and glassy nature of the ashes, frequently impart pozzolanic properties which make them useful as partial replacements for portland cements in concrete or as ingredients of blended cements.

According to Manz, about 280 million tonnes of coal ashes (fly ash and bottom ash) were produced in the world in 1977. The largest producers were, in decreasing order, the Soviet Union, the United States, Czechoslovakia, Poland, West Germany, and East Germany. Although the United States was the largest user of coal ash, and presumably fly ash, it ranked only ninth in percentage use.

Of ten major uses for fly ashes, three of the uses -- cement replacement, addition to cement, and aerated concrete blocks -- exploit pozzolanic properties of fly ashes by using them as finely-divided mineral admixtures in concrete. These three appear to to be the highest value uses since they involve partial replacement of portland cmeent which now sells for between $50 and $70 per ton. All three are successfully practiced in the United States, though not at the highest possible level.

The report estimates the tonnnages of fly ashes which could, conservatively, be used in each of several applications in concrete if it were available in suitable quality at a suitable price. In each case we have assumed that, on the average, 20 percent of the mass of portland cement could be replaced by an equal mass of good quality fly ash. This seems justified because there are few, if any, applications where a replacement of 20 percent of the portland cement by the same mass of good quality fly ash would cause unacceptable changes

of performance. Indeed, there will often be significant benefits from such use in concrete including:

1. lower cost of the concrete
2. improved workability
3. lower rate of heat liberation during hardening
4. improved durability in sulfate environments and with alkali-reactive aggregates
5. higher long-term strength

The calculations suggest that about 16 million tonnes (18 million tons) of fly ash could be used. This is about twice the present level of use.

Requirements for fly ashes for use as mineral admixture in concrete are given in ASTM C 618, Standard Specification for Fly Ash and Raw or Calcined Natural Pozzolan for Use in Portland Cement Concrete, and those for fly ashes for use in blended cements are given in ASTM C 595, Standard Specification for Blended Hydraulic Cements. Both standards rest on a weak foundation of knowledge about fly ashes and their technical performance in concretes and both need improvement.

Apart from cost savings and performance benefits, the use of fly ash in cements and concretes contributes to energy conservation because fly ash requires less energy to produce than cement, and to environmental protection because fly ash disposed of in dumps can cause environmental problems from dust and water pollution. Factors, either real or imaginary, which may account for the present relatively low levels of use of fly ash in cement and concrete are:

- Lack of knowledge and experience in fly ash use and difficulty in assessing the risks associated with its use,

- Increased costs due to requirements for storage and handling of fly ash,

- Lack of uniformity of fly ash composition and performance,

- Lack of adequate performance-based criteria for selection of fly ashes for use in concrete,

- Inability to predict performance of fly ashes in concrete,

- Uncertainty about the position to be taken by regulatory agencies, specifically the EPA, on whether fly ashes should be classified as hazardous wastes, and

- Uncertainty about the strength of possible competition from ground, granulated blast-furnace slag as a finely-divided mineral admixture.

We believe these factors to be the ones most often cited by cement and concrete manufacturers as reasons for not using fly ash in their products. The list includes facctors which may deter users of concrete from specifying the use of cements and concretes containing fly ash.

The major technical barriers to the use of fly ash in cement and concrete are associated with the lack of understanding of the nature of fly ashes and the lack of adequate standards which could help users recognize ashes likely to achieve desired levels of performance in concrete. The benefits to be obtained from reducing these barriers can be a combination of reduced waste for disposal, energy conservation through reduced use of virgin materials and, in many cases, a better product. In overview, in order of priority, the main research needs can be summarized as:

- Development of methods for characterizing fly ashes in terms of important characteristics controlling performance. These should include particle size distribution, mineral (phase) distribution by particle size, and surface characteristics.

- Development of methods for using the measured characteristics for predicting performance of fly ashes in concretes containing various cements and chemical admixtures.

- Development of standards based on methods for predicting performance from ash characteristics.

- Development of methods for beneficiating marginal ashes to improve their uniformity and performance.

The first three of these would assist development of improved standards for fly ash-containing cements and concretes. It is recommended that research in these areas be carried out and that it be performed in consultation with the relevant standards committees.

Table of Contents

1. INTRODUCTION

This report represents, in part, the response of the National Bureau of Standards (NBS) to the mandate of Subtitle E of the Resource Conservation and Recovery Act (P.L. 94-580). Specifically, the report addresses technical needs and opportunities in the use of fly ash in cements and concretes. The Department of Commerce, through NBS, is required to provide guidelines for specifications for recovered materials, stimulate the development of markets for recovered materials, promote proven technology, and provide information necessary to allow governmental agencies to procure products with the highest feasible percentage of recovered material.

Various research areas that need to be addressed in order that larger quantities of fly ash can be used in cement and concrete are identified. The results of such research will aid standards setting organizations, industry and governmental agencies in encouraging the increased use of fly ash and increasing its viability as an article of commerce.

Fly ashes are finely-divided, predominantly inorganic byproducts collected from the exhaust gases from coal-burning furnaces. Although they are generally wasted, they are a potentially important resource. Ashes which are collected from the bottom of the furnace are called bottom ashes. The United States produced 75 million tons (66 million tonnes) of coal ashes in 1979 [1], mostly from power plants, and the production is increasing each year as coal replaces oil as an energy source. Of this quantity, 57.5 million tons (51 million tonnes) were fly ashes. While fly ashes have a number of uses, mostly in concrete and other civil engineering applications, only about 13 percent (seven million tonnes[a]) of that produced in the U.S. in 1979 was used in concrete [3]. The remainder was disposed of by placing in stockpiles. The total amount of fly ashes stored in stockpiles is now about 300 million tonnes [4]. Since the stockpiling of fly ashes in ways which will not cause environmental problems is costly [5], it is important to identify potential uses and potentially attainable levels of use. The present report aims to identify opportunities for increased use of fly ash as an ingredient of cement and concrete (but not as a raw material for the manufacture of portland cement clinker) and to outline research needed to aid exploitation of the opportunities. The report reviews information on the nature of fly ashes (chapter 2), the U.S. and world productions and uses of coal ashes (fly ash and bottom ash) (chapter 3), the potential for increased use of fly ashes in cement and concretes (chapter 4), requirements for fly ashes for use in cement and concrete (chapter 5), and possible technical barriers to increased use of fly ashes in cements and concretes (chapter 6). Institutional barriers are only discussed briefly. Finally, a research program to aid exploitation of the opportunities is outlined (chapter 7).

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[a] Because the main source of data [2] for this report used the tonne as the unit of mass, the tonne is used throughout this report. Its relationship to other units is as follows:

$$1 \text{ tonne} \equiv 1000 \text{ kg} = 1.1225 \text{ tons} = 2245 \text{ lb}$$

2. THE NATURE OF FLY ASHES

Fly ashes are byproducts from the combustion of powdered coal. No two fly ashes are alike because the composition and nature of an ash reflects a) the composition of the coal, b) the fineness to which the coal is ground in preparation for combustion, and c) the details of the design and operation of the combustion and dust collection system.

The oxide compositional ranges of some fly ashes from U.S. coals [6] are given in table 1. These overall compositions are averages for all the particles in an ash. The individual particles may have quite different compositions and may themselves be heterogeneous [7]. Possible constituents of ash particles and the ranges of their quantities are listed in table 2. The constituents are residues from the inorganic minerals in the coal, together with some unburned carbon and other organic matter of low volatility from the coal, and sometimes traces of additives used to aid ash collection by electrostatic precipitation. The mineralogical compositions of the inorganic residues can, at least qualitatively, be explained in terms of the effects of the combustion temperatures on the grains of the inorganic minerals in the coal [8]. For example, a grain of a crystalline aluminosilicate such as a feldspar in a coal particle may melt in the flame and give rise to a spherical, glassy particle of approximately the same oxide composition after the rapid cooling on being carried out of the combustion chamber. Similarly, a grain of calcite, $CaCO_3$, is likely to be decarbonated in the combustion chamber without melting and end up as a nonspherical, crystalline grain of calcium oxide, CaO. Fly ashes should therefore be thought of as heterogeneous mixtures of predominantly spherical grains of different individual compositions and different degrees of crystallinity which will depend on their compositions and thermal histories. In addition, the different minerals must be expected to have different size distributions and may tend to be in particles of different shapes [26,27]. Overall, the constituents of fly ashes may be categorized as in table 2. The mineralogical compositions of some fly ashes are given in table 3.

While table 3 indicates the variety of more easily recognizable minerals in ash particles [8, 9], it does not show that there may be some contamination of the particle surfaces as indicated in table 2. Such contamination may be residual organic matter from the coal, or it may be from additives sprayed into the exit gas stream to aid electrostatic precipitation of the ash. Because there is some circumstantial evidence that contaminants on the particle surfaces can affect fly ash performance in concrete, it is important to consider the possibility of their presence.

The high contents of silica (SiO_2) and alumina (Al_2O_3) in fly ashes frequently make them potential raw materials for pyroprocesses such as the manufacture of portland cement clinker [10], clay brick [11], and lightweight aggregates [12]; they also help to impart pozzolanic activity which makes many ashes suitable for use in concrete [13]. Pozzolanic activity is the property whereby some inorganic materials react with calcium hydroxide and water to form cementitious compounds. While the finely-divided state and glassy nature of the ashes is usually beneficial in pozzolanic applications, the variability of the ashes may sometimes be a problem.

Table 1. Compositional Ranges of Some Ashes from Various Types of U.S. Coals

	Anthracite	Bituminous	Subbituminous	Lignite
% SiO_2	47-68	7-68	17-58	6-45
% Al_2O_3	25-43	4-39	4-35	6-23
% Fe_2O_3	2-10	2-44	3-19	1-18
% CaO	0-4	1-36	2-45	15-44
% MgO	0-1	0-4	0.5-8	3-12
% Na_2O	-	0-3	-	0-11
% K_2O	-	0-4	-	0-2
% SO_3	0-1	0-32	3-16	6-30
% TiO_2	1-2	0-4	0-2	0-1
% P_2O_5	0-4	0-3	0-3	0-1

After Sondreal, Kube and Elder [6].

Table 2. Categories of Constituents of Fly Ashes

Type of Constituent	Typical Range (%)
Mineral particles, glassy	50-90
Mineral particles, crystalline	10-50
Carbon	0-12
Other organic matter of low volatility[a]	0-1
Additives used to aid ash collection[a]	0-0.1

[a] Probably as surface layers on more refractory particles.

Table 3. Mineralogical (Phase) Compositions of Some Fly Ashes

Phase	Idealized Composition	U.K. Ashes[a]	Yugoslav Ash[b]
% Glass[c]	-	71-88	53.6
% Mullite	$3Al_2O_3 \cdot SiO_2$	6-14	19.0
% Quartz	SiO_2	2-9	3.2
% Corundum	Al_2O_3	-	1.0
% Magnetite	Fe_3O_4	1-3	6.5
% Hematite	Fe_2O_3	1-3	2.0
% Goethite	(Al,Fe,Mn)O(OH)	-	2.5
% Wustite	FeO	-	1.5
% Pyrite	FeS_2	-	trace
% Free lime	CaO	-	2.7
% Calcite	$CaCO_3$	-	1.0
% Anhydrite	$CaSO_4$	-	2.1
% Periclase	MgO	-	1.9
% Carbon	C	0-3	3.0

[a] Watt and Thorne, 1965 [8].

[b] Brzakovic, 1970 [9].

[c] Non-crystalline material of variable composition; consists predominantly of SiO_2 and Al_2O_3 with lesser quantities of other oxides.

3. WORLD-WIDE PRODUCTION AND USE OF FLY ASHES

According to Manz [2], about 280 million tonnes of coal ashes (fly ash and bottom ash) were produced in the world in 1977. The largest producers were, in decreasing order, the Soviet Union, the United States, Czechoslovakia, Poland, West Germany, and East Germany. Little is known about the use of coal ashes in the Soviet Union and Czechoslovakia, but information is available on the tonnages used in many other countries. Leading users of coal ashes are listed in table 4 in decreasing order of quantity used. The table also gives information for each country on the coal ash production in 1977, the percentage of the ash which was used, and the major uses. Although the United States was the largest user of coal ash listed in the table, it ranked only ninth in percentage use.

Data on the world use of coal ashes, by category of use, is given in table 5 in order of decreasing quantity used. The four largest volume uses are all in civil engineering or construction applications. They are as fill material, cement replacement in concrete, addition to cement to form blended cements, and road stabilizer. All of these uses are found in the United States. Cement replacement and addition to cement are similar applications of coal ashes since both depend on the pozzolanic properties of the ashes. However, the degree of quality control which can be exercised is usually greater when fly ash is added to the cement; this could sometimes be an important consideration to the user.

Table 4. Coal Ash Production and Use by Country in 1977[a]

Country	Use, 10^6 tonnes/yr	Production, 10^6 tonnes/yr	Percent Used	Major Uses
U.S.A.	9.07	61.0	13	fill material, cement replacement, blast grit and roofing, ice control
Poland	5.51	15.0	37	road stabilizer, aerated concrete blocks, addition to cement
England	5.06	11.9	39	fill material, cement replacement, aerated concrete blocks
West Germany	3.41	15.0	23[b]	asphalt filler, cement replacement, blast grit and roofing
East Germany	2.48	15.0	18	addition to cement, lightweight aggregate, brick, and ceramics
France	2.00	4.8	42	addition to cement, road stabilizer, cement raw material
Canada	0.71	2.6	27	fill material, cement replacement
Spain	0.65	5.0	13	addition to cement
Australia	0.58	5.4	11	addition to cement
Scotland	0.56	2.0	30	fill material, cement replacement
Japan	0.45	2.0	23	addition to cement, cement replacement
Hungary	0.425	5.0	9	addition to cement, aerated concrete blocks

[a] The figures are taken from Manz [2]; they are for coal ash with no distinction being made between fly ash and bottom ash.

[b] The percentage use of hard coal ash was much higher than the use of soft coal ash.

Table 5. World Use of Coal Ash in 1977[a]

Use	10^6 tonnes/yr	Leading Users[b]
Fill material	5.8	England, USA, Canada
Cement replacement	5.6	USA, England, E. Germany
Addition to cement	4.1	France, Spain, W. Germany Poland, Australia, USA
Road stablizer	3.1	Poland, England, USA
Aerated concrete blocks	1.8	Poland, England
Blast grit and roofing	1.6	USA, W. Germany
Asphalt filler	1.2	W. Germany, USA
Cement raw material	1.0	USA, France
Lightweight aggregate	0.8	England, USA, E. Germany
Bricks and ceramics	0.6	W. Germany, Ukraine, E. Germany
Other	9.6	USA, Poland, E. Germany W. Germany
	35.2[c]	

[a] The figures are from Manz [2].

[b] In order of decreasing quantity used.

[c] For comparison, total ash production is 278 x 10^6 tonnes/yr.

4. POTENTIAL FOR INCREASED USE OF FLY ASHES IN CEMENTS AND CONCRETES IN THE UNITED STATES

Ten major uses for fly ashes, and coal ash in general, are indicated in table 5. All relate to construction. Three of the uses -- cement replacement, addition to cement, and aerated concrete blocks -- exploit pozzolanic properties of fly ashes by using them as finely-divided mineral admixtures in concrete. Four of the uses -- fill material, road stabilizer, blast grit and roofing, and asphalt filler -- exploit the particulate nature of fly ashes but do not take advantage of their pozzolanic nature. The other three uses -- cement raw material, lightweight aggregate, and bricks and ceramics -- use fly ashes as replacements for clay and other aluminosilicates in high temperature manufacturing processes. The uses which exploit the pozzolanic properties of fly ashes as ingredients of cements and concretes are the main subject of this report. They appear to be the highest value uses in construction since they involve partial replacement of portland cement which now sells for $50 to $70 per ton [14].

The approximate quantities of cements and mineral admixtures presently produced and used in the United States are listed in table 6. The quantities produced and used are in balance except for fly ashes and blast-furnace slags. The fly ashes and blast-furnace slags are byproducts which have potential for being used, either directly or after suitable processing, as additions to portland cement to make blended cements or as mineral admixtures which can partially replace or supplement portland cements in concrete. In either case, they are used as part of the cementing material in concrete. At the risk of over-simplification, it may be stated that fly ashes and granulated blast-furnace slags are probable competitors for use as part of the cementing material in concrete [15]. However, we shall not consider this probable competition further until we have discussed the potential for growth in the use of fly ashes in cement and concrete in the absence of such competition.

In discussing the potential use of fly ash as part of the cementing material in concrete, it will be helpful to know the distribution of cement among end uses. The distribution is indicated in different ways in tables 7 and 8. In table 7, percentages of cement shipments going to the main categories of purchasers in 1976 [16] are indicated. Assuming the percentage distribution to have remained the same since 1976, the tonnages going to each type of purchaser in 1979 are also given. Table 8 shows estimated percentages [24] of all the cement used in the United States which go into major end uses in cast-in-place and precast concrete. We have included in both tables a column giving our estimates of the tonnages of fly ashes which could, conservatively, be used in each application if it were available in suitable quality at an attractive price. In each case, we have assumed that, on the average, 20 percent of the mass of portland cement could be replaced by an equal mass of good quality fly ash. This seems justified because there are few, if any, applications where a replacement of 20 percent of the portland cement by the same mass of fly ash would cause unacceptable changes of performance. In almost all cases, changes in mixture properties could keep the performance of fly ash-containing concrete in an acceptable range and bring one or more of several benefits:

Table 6. Approximate Quantities of Cements and Mineral Admixtures Produced and Used in Concrete in the United States in 1979

Type of Material	Quantity Used 10^6 tonnes/yr	Quantity Produced 10^6 tonnes/yr	Reference
Portland cements	80	80	14
Portland-pozzolan cements	2[a]	2[a]	--
Portland-blast-furnace slag cements	0.5[a]	0.5[a]	--
Natural pozzolans	0.5[a]	0.5[a]	--
Fly ashes	8	51	1
Granulated blast-furnace slags	0.2[a]	1	23
Blast-furnace slags	0.2[a]	23	23

[a] Estimates to indicate the orders of magnitude.

Table 7. Cement Shipments by Type of Purchaser and Actual and Potential Fly Ash Use

Type of Purchaser	Percent of Total Cement Use[b]	Estimated Cement Use, 10^6 tonnes[a]	Ash Purchased, 10^6 tonnes[c]	Potential Ash Use, 10^6 tonnes[d]
Ready-mixed concrete	65	52	} 2	10
Concrete products	14	11		2.2
Building material dealers	8	6	-	1.2
Highway contractors	8	6	1	1.2
Other contractors	4	3	-	0.8
Government agencies (Federal and other)	0.5	0.5	-	0.1
Miscellaneous	1.5	1.5	-	0.3
	101	80	3	15.8

[a] Calculated from percent of total cement use assuming an annual production of 80 million tonnes.

[b] Percentages are for 1976 [16].

[c] Figures in this column are rough estimates for 1978 based on reference 28; the figure for use in ready-mixed concrete and concrete products is probably too low by at least a factor of two [3].

[d] Can be in cement or used as mineral admixture for concrete; assumes 20 percent replacement of portland cement and no use of granulated blast-furnace slag.

Table 8. End Uses for Cement and Potential for Replacement of Cement by Fly Ash or Other Mineral Admixture

End Use	Percent of All Cement Used (est.)[a]	Cement Use, 10^6 tonnes/yr (est.)	Estimated Replacement, 10^6 tonnes/yr[b]
a. Cast-in-Place Concrete			
Foundation	36	29	5.8
Slabs	14	11	2.2
Highways	13	10	2.0
Columns	7	6	1.2
Masonry mortar	4	3	0.6
Canal linings	2.5	2	0.4
Tunnel linings	2	1.5	0.3
Other cast-in-place	8	6	1.2
b. Precast Concrete			
Concrete block	4	3	0.6
Pipe	2	1.5	0.3
Panels	2	1.5	0.3
Beams	2	1.5	0.3
Fiber-reinforced products	2	1.5	0.3
Other precast	1.5	1.0	0.2
	100	78.5	15.7

[a] Based on Table 7 of reference 24 with figures adjusted to give a total of 100 percent.

[b] Assuming 20 percent of the mass of portland cement is replaced by mineral admixture on a 1-for-1 mass basis.

1. lower cost concrete
2. improved workability
3. lower rate of heat liberation during hardening
4. improved durability in sulfate environments and with alkali-reactive aggregates
5. higher long-term strength

One example of a possibly unacceptable change in a critical aspect of performance due to fly ash use might be a reduction in early age strength resulting from replacement of 20 percent of a high early strength portland cement (ASTM Type III). If no other changes in mixture proportions were made, the replacement would almost certainly result in a reduction of the early strength for which the cement was selected. In practice, the early age strength reduction resulting from fly ash use could be lessened by redesign of the mixture, including the possible use of chemical admixtures. In any case, since ASTM Type III cements account for less than five percent of all portland cements manufactured, this consideration does not have a significant effect on the estimates of fly ash use made in this report.

On the basis of a 1-for-1 replacement of 20 percent of the portland cement now used, almost 16 million tonnes of suitable fly ash could be used. This is about twice the present level of use. What constitutes a suitable fly ash will be discussed in chapters 5 and 6.

5. REQUIREMENTS FOR FLY ASHES USED IN CEMENTS AND CONCRETES

It is probable that almost all fly ashes can be used in cements and concretes at some level without significant loss of performance. Nevertheless, in the present limited state of knowledge about relationships between fly ash characteristics and fly ash performance in concrete, it is necessary to restrict the fly ashes permitted for use. The restrictions on fly ashes for use in concrete are specified in standards of the ASTM (American Society for Testing and Materials). The relevant specifications are ASTM C 618, Standard Specification for Fly Ash and Raw or Calcined Natural Pozzolan for Use in Portland Cement Concrete [17], and those for fly ashes for use in blended cements are given in ASTM C 595, Standard Specification for Blended Hydraulic Cements [18]. These and other important ASTM standards relevant to fly ash use in cement and concrete are listed in table 9.

Two classes of fly ash, Classes F and C, are referenced in ASTM C 618 (see table 10). The chemical and physical requirements for Class F and Class C fly ashes are summarized in table 11. The requirements for fly ashes and other pozzolans in the manufacture of pozzolan-containing blended cements meeting ASTM C 595 are sufficiently similar to those defined in ASTM C 618 that it is not necessary to give separate discussions about them in this report. In fact, it has recently been suggested that the ASTM C 595 specification be changed to indicate that fly ashes and other pozzolans for use in blended cements should meet the requirements of ASTM C 618.

In considering possible improvements to the standards for fly ashes, and the needs for research to provide the technical basis for improved standards, it will be helpful to consider how the categories of fly ash constituents listed in table 2 affect the performance of cements and concretes containing fly ashes. These comments are based predominantly on experience with use of bituminous coal ashes.

<u>Glassy particles</u> (i.e., non-crystalline particles) in fly ashes are probably the most important pozzolanic constituent [19]. Their quantity is probably closely related to the quantity of (SiO_2 + Al_2O_3 + Fe_2O_3) in the ash. The reactions of the glassy particles with the calcium hydroxide liberated by the reactions of portland cements with water give cementitious calcium silicate hydrates similar to those produced by portland cements. Although the mechanisms of the reactions and the factors determining the kinetics are not well understood, there is no reason to suspect that the glassy particles cause any difficulties in concrete, apart from a possible reduction in the rate of strength gain at early ages.

<u>Crystalline particles</u> in fly ashes may be almost inert in portland cement concretes (e.g., mullite or corundum) or quite reactive (e.g., free lime or high-lime minerals). The relatively inert crystalline particles may be considered as finely-divided sand grains in their effects on concrete properties, but it is not yet possible to make an equally general statement about the reactive crystalline particles. For the most part, reactive crystalline particles appear to occur in higher quantities in high-lime fly ashes from subbituminous

Table 9. ASTM Standard Specifications and Test Methods for Cements, Pozzolans, and Cement-Aggregate Combinations

ASTM No.	Title
Cements	
C 114	Chemical analysis of hydraulic cement
C 150	Specification for portland cement
C 595	Specification for blended hydraulic cement
Pozzolans	
C 311	Sampling and testing fly ash or natural pozzolans for use as a mineral admixture in portland cement concrete
C 618	Specification for fly ash and raw or calcined natural pozzolan for use in portland cement concrete
Cement-Aggregate Combinations	
C 227	Test for potential alkali reactivity of cement-aggregate combinations (mortar-bar method)
C 342	Test for potential volume change of cement-aggregate combinations
C 441	Test for effectiveness of mineral admixtures in preventing excessive expansion of concrete due to the alkali-aggregate reaction

Table 10. Classes of Fly Ash and Other Pozzolans in ASTM Standard C 618 on Fly Ash and Raw or Calcined Pozzolan for Use as a Mineral Admixture in Portland Cement Concrete

Class	Description
N	Raw or calcined natural pozzolan, e.g., some diatomaceous earths; opaline cherts and shales; tuff and volcanic ashes or pumicites; and some clays or shales requiring calcination
F	Fly ash with pozzolanic properties normally produced from anthracite or bituminous coal
C	Fly ash with pozzolanic and cementitious properties normally produced from lignite or subbituminous coal. May have a lime content higher than 10 percent

Table 11. Summary of Chemical and Physical Requirements Defined in ASTM C 618 for Fly Ashes for Use in Portland Cement Concrete

	Class F	Class C
A. Chemical Requirements		
(SiO_2 + Al_2O_3 + Fe_2O_3), min, %	70.0	50.0
SO_3, max, %	5.0	5.0
Moisture content, max, %	3.0	3.0
Loss on ignition, max, %	12.0	6.0
Optional Chemical Requirements		
MgO, max, %	5.0	5.0
Available alkalies, as Na_2O, max, %	1.50	1.50
B. Physical Requirements		
Fineness:		
Amount retained when wet-sieved on No. 325 (45 μm) sieve, max, %	34	34
Pozzolanic activity index:		
With portland cement, at 28 d, min, % of control	75	75
With lime, at 7 d, min, psi (kPa)	800 (5500)	800 (5500)
Water requirement, max, % of control	105	105
Soundness:		
Autoclave expansion or contraction, max, %	0.8	0.8
Uniformity:		
Specific gravity, variation from avg, max, %	5	5
Percent retained on No. 325 (45 μm) sieve, max variation from avg. of previous 10, percentage points	5	5
C. Optional Physical Requirements:		
Multiple factor, product of LOI and fineness, max, %	255	-
Increase of drying shrinkage of mortar bars at 28 d, max, %	0.03	0.03
Uniformity requirements:		
When air-entraining concrete is specified, quantity of AE agent required to produce air content of 18.0 vol % shall not vary from the average of previous 10 by more than, %	20	20
Reactivity with cement alkalies:		
Mortar expansion at 14 d, max, %	0.020	0.020

coals and lignites which occur west of the Mississippi River. These subbituminous coals have only recently attained major importance as fuels in the United States. Because of their shorter history of use, less is known about the exact natures of the subbituminous coal and lignite fly ashes and of their effects on concrete performance than is the case for bituminous coal ashes. All that is known is that the high-lime ashes tend to show a greater range in their contributions to strength development of concrete than lower lime ashes with predominantly glassy particles. Indeed, some of the high-lime ashes can develop significant strength when mixed with water, even in the absence of portland cement.

Unburned carbon in fly ashes is usually the largest contributor to the loss on ignition. The carbon can be considered inert as far as cementing reactions are concerned. However, the carbon in fly ash has a high specific surface area and it appears to be able to adsorb significant quantities of organic chemical admixtures, such as water-reducing agents, set retarders, and air-entraining agents [20] which might be added to the mixing water to improve the performance of the concrete. Chemical admixtures are frequently used in concretes and it can cause problems if their performances become unpredictable. Experience suggests that fly ashes with less than three or four percent of carbon do not cause serious problems in the performance of organic chemical admixtures. It is possible that significantly larger quantities of carbon can be tolerated as long as there is awareness of its possible effects.

Organic matter of low volatility, other than carbon, can sometimes be extracted from fly ashes with solvents such as chloroform [21]. There is no standard method for its determination. It has been suggested that such organic matter, though present in only small quantities [26], may interfere with the performance of organic chemical admixtures. This area needs further investigation.

Additives used to aid ash collection in electronic precipitation [22] are relatively volatile, ionizable compounds and as ammonium and alkali metal salts. When used to aid fly ash collection, they may be deposited on the surfaces of ash particles in quantities which depend on the operating conditions of the power plant. Little has been published about their effects on concrete performance though there have been reports that detectable quantities of ammonia vapor have been liberated during mixing when some fly ashes have been used in concrete. If this proves to be a real and a recurring phenomenon, it must be taken into account in selecting fly ashes for use in concrete. Also, an investigation should be made of other possible effects of traces of the additives such as on the performance of chemical admixtures.

With the preceding discussion as background, further consideration can now be given to the factors which may hinder acceptance of fly ashes as ingredients of concrete.

6. BARRIERS TO INCREASED USE OF FLY ASHES IN CEMENTS AND CONCRETES

Just over 50 million tonnes of fly ash were generated in the United States in 1979 [1]. If, as was suggested in chapter 4, about 16 million tonnes of suitable fly ashes could be used in concrete at current levels of cement and concrete production, it is important to try to understand the reasons for the much lower actual level of use (e.g., seven million tonnes in 1979).

As mentioned in chapter 4, fly ash use in concrete can bring cost savings and performance benefits. This is substantiated by the fact that the U.S. Army Corps of Engineers, the largest Federal construction agency, uses large quantities of fly ash in concrete. On the other hand, few, if any, smaller construction organizations have levels of fly ash use approaching those of the Corps of Engineers. Other benefits are conservation of energy because fly ash requires less energy to produce than cement, and protection of the environmental because fly ash disposed of in dumps may cause environmental problems due to wind-blown dust and leaching of constituents into ground water. Factors, either real or imaginary, which may account for the present relatively low levels of use of fly ash are:

- Lack of knowledge and experience in fly ash use and difficulty in assessing the risks associated with its use,
- Increased costs due to requirements for storage and handling of fly ash,
- Lack of uniformity of fly ash composition and performance,
- Lack of adequate performance-based criteria for selection of fly ashes for use in concrete,
- Inability to predict performance of fly ashes in concrete,
- Uncertainty about the position to be taken by regulatory agencies, specifically the EPA, on whether fly ashes should be classified as hazardous wastes,
- Uncertainty about the strength of possible competition from ground granulated blast-furnace slag as a finely-divided mineral admixture

We believe these factors to be the ones most often cited by cement and concrete manufacturers as reasons for not using fly ash in their products. The list includes factors which may deter users of concrete from specifying the use of cements and concretes containing fly ash. In this connection, it is to be expected that, in the absence of definitive information, some of the possible concerns would be exploited by vendors of competing materials. Some comments on the individual factors follow.

a. Lack of Knowledge and Experience in the Use of Fly Ashes in Cements and Concretes

The general knowledge of the characteristics and performance of fly ashes in cements and concretes is still at a low level compared with knowledge about portland cements. Nevertheless, at least two cement companies have made portland-pozzolan cements (ASTM Type IP) successfully for several years and the U.S. Army Corps of Engineers, the U.S. Bureau of Reclamation (now the Water and Power Resources Service), and the TVA (Tennessee Valley Authority) have long records of successful use of fly ash in their concrete work. In spite of these uses, the knowledge gained by these users has not yet been applied extensively by other cement manufacturers and civil engineering and construction organizations. The reasons for this are complex and probably include the fact that other organizations tend to have somewhat different types of projects (e.g., less mass concrete), less well-developed testing capability, and less purchasing power than the governmental agencies. On the other hand, there are now several companies which market fly ashes and take responsibility for their quality and provide technical information about their use.

A number of possible actions could assist in breaking down the knowledge barrier. These include publication of research results giving a sound basis for understanding the behavior of fly ashes on the basis of their characteristics, and the organization of training courses for the persons in manufacturing and construction organizations who must make decisions about the use of fly ash. Training could be done by trade associations and by universities and colleges which educate persons to enter the cement and concrete fields.

Research which could help overcome the barriers mentioned in this section is discussed in chapter 7.

b. Requirements for Storage and Handling of Fly Ash

If a fly ash is to be included among the concrete ingredients by concrete manufacturers, they must provide equipment, such as bins or silos, for storing and handling the material. This might require a capital investment that is difficult to justify as long as the benefits of fly ash use are not easily assessable. In any case, the handling of an additional material can put a burden on concrete manufacturers which they may want to avoid unless there is a clear benefit to be obtained.

c. Lack of Uniformity of Fly Ashes

The characteristics of fly ash from a single source may be uniform, or quite variable, depending on factors such as the source of the coal, the age of the plant, and operating conditions and procedures. As a class, fly ashes are significantly more variable than portland cements because their compositions are determined by nature, rather than by synthesis. Doubts about uniformity of composition raise doubts about uniformity of performance. One of the variables is the quantity of unburned carbon in the ash. This can influence the effect of added fly ash on the behavior of concrete containing air-entraining and other chemical admixtures. More quantitative knowledge is needed about factors affecting the performance of fly ashes in concrete so that the effects of changes in fly ash characteristics can be predicted.

d. Lack of Adequate Performance-Based Criteria for Selection of Fly Ashes for Use in Cement and Concrete

As mentioned in chapter 5, the present ASTM standard for fly ash and raw or calcined natural pozzolans for use in concrete (ASTM C 618) provides specifications with compositional and performance requirements for two classes of fly ash -- ashes normally produced from anthracite and bituminous coal (Class F ashes) and fly ashes normally produced from lignites and subbituminous coal (Class C ashes). To the extent that ashes complying with the specifications may differ significantly in performance, the specifications may be considered to be too broad. For example, because the strength tests used as a measure of ash reactivity are not very severe, it is possible for ashes of relatively low reactivity to pass the tests. In addition to not being very discriminating, the activity tests suffer because they take too long to carry out. Ultimately, it would be desirable to have a method for predicting the performance of a fly ash in a concrete with a cement of known composition using measured characteristics of the ash as a guide. There is a need for the development of improved standards by organizations such as ASTM, ACI (American Concrete Institute) and AASHTO (the American Association of State Highway and Transportation Officials). The standards should emphasize performance rather than composition and recognize that different levels of performance and uniformity may be needed for different purposes. By emphasizing performance, the standards would remove barriers based on composition and ensure that any suitable material could be used.

e. Regulatory Agency Position with Regard to Fly Ash

Under the Resource Recovery and Conservation Act of 1976, the Environmental Protection Agency (EPA) has the responsibility for deciding which waste materials should be considered hazardous and those for which use should be encouraged through use of Federal purchasing power. It appears that final decisions have not yet been made for fly ashes. It is possible that cement and concrete manufacturers might be reluctant to commit themselves to the use of fly ash until the EPA positions are finalized. Other barriers result from state agencies, such as some state highway departments, failing to mention fly ash as an acceptable ingredient in concrete purchased by them.

f. Possible Competition from Ground, Granulated Blast-Furnace Slag

In some industrialized countries, fly ashes and other pozzolans are the preferred mineral admixtures for concrete, while granulated blast-furnace slags are preferred in others. Factors affecting the patterns of use include the amounts of fly ashes and blast-furnace slags available, their compositions, investments by the producers and others in exploiting the byproducts, and the strength of opposing interests. In the United States, fly ashes are already available to producers of cement and concrete and a smaller but significant amount of ground, granulated blast-furnace slag is expected to be available by 1982 [23]. If the slag is favored as a mineral admixture for concrete and as an ingredient of blended cements, it will to some extent reduce the amount of fly ash which can be used. We believe that both ground, granulated blast-furnace slag and fly ashes will grow in importance in U.S. cement and concrete technology and that good performance-based specifications will foster the use of both. We also believe that the ultimate balance achieved will cause the amount of fly ash used in cement and concrete to be somewhat less than the amount indicated in tables 7 and 8 and chapter 4. A detailed discussion of the potential for the use of slag and fly ash in concrete is given in a report which was prepared for the Department of Energy by Price, Troop and Gershman [29].

7. TECHNICAL RESEARCH NEEDS FOR FACILITATING THE USE OF FLY ASHES IN CEMENTS AND CONCRETES

The major technical barriers to the use of fly ash in cement and concrete are associated with the lack of understanding of the nature of fly ashes and the lack of adequate standards which could help users recognize ashes likely to achieve desired levels of performance in concrete. The benefits to be obtained from reducing these barriers can be a combination of reduced waste for disposal, energy conservation through reduced use of virgin materials and, in many cases, a better product. In overview, in order of priority, the main research needs can be summarized as:

- Development of methods for characterizing fly ashes in terms of important characteristics controlling performance. These should include particle size distribution, mineral (phase) distribution by particle size, and surface characteristics.

- Development of methods for using the measured characteristics for predicting performance of fly ashes in concretes containing various cements and chemical admixtures.

- Development of standards based on methods for predicting performance from ash characteristics.

- Development of methods for beneficiating marginal ashes to improve their uniformity and performance.

These four areas are consistent with more general recommendations on cement and concrete research needs given in the recent National Materials Advisory Board report on the Status of Cement and Concrete R and D in the United States [24]. Each of the four areas will be discussed in turn. The first three would assist the development of improved standards. It would therefore be important for research in these areas to be carried out in collaboration or consultation with the relevant committees of ASTM and ACI and, possibly, a proposed new committee of RILEM (the International Union of Materials and Structures Research and Testing Laboratories) on engineering properties of fly ash. The relevant ASTM and ACI committees are ASTM C 1 on Cements, ASTM C 9 on Concrete and Cement Aggregates, ACI 201 on Durability of Concretes, ACI 212 on Admixtures, and ACI 225 on Hydraulic Cements.

a. Development of Methods for Characterization of Fly Ashes

It must be assumed that if fly ashes could be characterized adequately, their characteristics could be used in predicting performance under various conditions of use. The methods of characterization that appear most likely to be important are:

- Determination of particle size distribution;

- Phase identification and determination of distributions of phases by particle size and within particles, o Detection and analysis of surface phases.

The technology for these methods of characterization is available but has not yet been applied systematically in the ways proposed here.

The information obtained from application of such methods should be used in seeking correlations with performance of fly ashes in concrete which could become the basis for performance-based standard specifications and test methods.

b. Development of Methods for Determining or Predicting Performance of Fly Ashes in Concretes

The rates of reactions and the performances of fly ashes in concretes depend on a number of factors, including the characteristics of the cements used, and the temperature. Test methods which will indicate performance reliably are needed and, because of the complexity of the chemistry of the cement-fly ash-water system, it would be valuable if mathematical models suitable for predicting the courses of reactions under various likely conditions of use could be developed. These models would complement those now being developed for portland cements [25]. They should take into account characteristics such as the mineralogy and particle size distributions of fly ashes, the cement characteristics and the conditions of use.

c. Development of Standards for Fly Ashes for Use in Cements and Concretes

Information on methods for characterizing ashes and for predicting their performance in concretes should be used as a technical basis for new standard performance tests and specifications for fly ashes for use in concrete. The standards, which should be developed in collaboration with relevant ASTM, ACI and, possibly, RILEM committees, should include test methods for ash reactivity, for sulfate resistance of cement-ash combinations, for suitability of cement-ash combinations for use with alkali-reactive aggregates, and for cement-ash combinations for use in concretes containing chemical admixtures. The tests should be more rapid than those which now exist or are being proposed for acceptance in ASTM, and they should be relatively simple to carry out.

d. Development of Methods for Beneficiating Fly Ashes for Use in Cements and Concretes

The most useful ingredients of fly ashes for use in cement and concrete appear to be the predominantly glassy particles [19]. The carbon and crystalline inorganic phases are frequently either detrimental or relatively inert constituents of fly ash; some ashes would be more suitable for use in concrete if these constituents could be removed. Methods for removing the carbon and crystalline material from ashes on a large scale basis should be investigated. Possible methods might include burning off of carbon and other organic matter, magnetic separation of the iron-containing phases, air separation of the larger and denser particles, and froth flotation.

8. SUMMARY AND CONCLUSIONS

The purpose of this report has been to review the present uses of fly ashes in the United States and to indicate technical research needs for supporting their increased use in cement and concrete. From the consideration of present uses and the present state of knowledge about factors affecting the performance of concretes containing fly ashes, it is concluded that the present level of use of about seven million tonnes per year could probably be increased to about 16 million tonnes per year. This increase may be possible without the development of any new technology. However, the rate of increase of fly ash use should be enhanced by research addressing the following questions:

1. How may fly ashes be best characterized to aid their selection for use in concretes for different applications?

2. How may methods for predicting the performance of fly ashes in concretes be developed?

3. How may standards based on (1) and (2) be established most expeditiously?

4. Can viable processes be developed for beneficiating fly ashes to be used in concretes?

The research needed to answer these questions is outlined in this report. It is recommended that the research needed to answer questions (1) through (3) be carried out in collaboration with ASTM committees C 1 on Cements and C 9 on Concretes and Concrete Aggregates; and with ACI committees 201 on Durability of Concretes, 212 on Admixtures, and 225 on Hydraulic Cements; and on an international basis through the new RILEM committee on Engineering Uses of Fly Ash.

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15. SUPPLEMENTARY NOTES

[ ] Document describes a computer program; SF-185, FIPS Software Summary, is attached.

16. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)

Following a brief review of the nature of fly ashes and their levels of production and use in various countries, an estimate is made of the potentially achievable level of use of fly ash in cement and concrete in the United States. The estimate assumes that 20 percent of the mass of all the portland cement used in the United States could be replaced by the same mass of fly ash; it ignores possible competition from granulated blast-furnace slag as a finely-divided mineral admixture for concrete. It appears that about 16 million tonnes (18 million tons) per year of fly ash could be consumed in cement and concrete, provided there were sufficient ash of suitable quality and a general understanding of the technical requirements for satisfactory fly ash use. Present standards which affect the use of fly ashes are discussed. Steps which could be taken to improve knowledge of factors affecting fly ash performance in cement and concrete, and hence to improve standard test methods and specifications, are outlined.

17. KEY WORDS (six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)

Cement; concrete; fly ash.

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