

ACI 232.2R-18

Report on the Use of Fly Ash in Concrete

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Report on the Use of Fly Ash in Concrete

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Report on the Use of Fly Ash in Concrete

Reported by ACI Committee 232

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Fly ash is used in concrete and other portland cement-based systems primarily because of its pozzolanic and cementitious properties. These properties contribute to strength gain and are known to improve the performance of fresh and hardened concrete, mortar, and grout. The use of fly ash typically results in more economical concrete construction.

This report gives an overview of the origin and properties of fly ash, its effect on the properties of hydraulic cement concrete, and the selection and use of fly ash in the production of hydraulic cement concrete and concrete products. Information and recommendations concerning the selection and use of Class C and Class F fly ashes conforming to the requirements of ASTM C618 are provided. Topics covered include a detailed description of the composition of fly ash, the physical and chemical effects of fly ash on properties of concrete, guidance on the handling and use of fly ash in concrete construction, use of fly ash in the production of concrete products and specialty concretes, and recommended procedures for quality control. High-volume fly ash concrete is covered in a general way in this report; readers can consult ACI 232.3R for more information.

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Keywords: alkali-aggregate reaction; controlled low-strength material; durability; fly ash; mass concrete; pozzolan; sulfate resistance; sustainability; workability.

CONTENTS

CHAPTER 1—INTRODUCTION, SCOPE, SOURCES, AND SUSTAINABILITY, p. 2

- 1.1—Introduction, p. 2
- 1.2—Scope, p. 3
- 1.3—Source of fly ash, p. 3
- 1.4—Fly ash and sustainability, p. 7

CHAPTER 2—DEFINITIONS, p. 9

CHAPTER 3—FLY ASH COMPOSITION, p. 9

- 3.1—General, p. 9
- 3.2—Chemical composition, p. 10
- 3.3—Crystalline constituents, p. 11
- 3.4—Glassy constituents, p. 13
- 3.5—Physical properties, p. 15
- 3.6—Chemical activity of fly ash in hydraulic cement concrete, p. 17
- 3.7—Future research needs, p. 18

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CHAPTER 4—EFFECTS OF FLY ASH ON CONCRETE, p. 18

- 4.1—Effects on properties of fresh concrete, p. 18
- 4.2—Effects on properties of hardened concrete, p. 20

CHAPTER 5—CONCRETE MIXTURE PROPORTIONING, p. 26

- 5.1—General, p. 26
- 5.2—Considerations in mixture proportioning, p. 27

CHAPTER 6—FLY ASH SPECIFICATIONS, TEST METHODS, AND QUALITY ASSURANCE/QUALITY CONTROL, p. 27

- 6.1—Introduction, p. 27
- 6.2—Chemical requirements, p. 28
- 6.3—Physical requirements, p. 29
- 6.4—General specification provisions, p. 30
- 6.5—Methods of sampling and testing, p. 30
- 6.6—Source quality control, p. 30
- 6.7—Startup, oil, and stack additives, p. 31
- 6.8—Rapid quality control tests, p. 32

CHAPTER 7—FLY ASH IN CONCRETE CONSTRUCTION, p. 32

- 7.1—Ready mixed concrete, p. 32
- 7.2—Concrete pavement, p. 32
- 7.3—Mass concrete, p. 33
- 7.4—Roller-compacted concrete, p. 33
- 7.5—Self-consolidating concrete, p. 33
- 7.6—High-volume fly ash concrete, p. 34
- 7.7—High-performance concrete, p. 34
- 7.8—Long-life structures, p. 34
- 7.9—Bulk handling and storage, p. 35
- 7.10—Batching, p. 36

CHAPTER 8—FLY ASH IN CONCRETE PRODUCTS, p. 36

- 8.1—Concrete masonry units, p. 36
- 8.2—Concrete pipe, p. 37
- 8.3—Precast/prestressed concrete products, p. 37
- 8.4—No-slump extruded hollow core slabs, p. 38
- 8.5—Concrete tile, p. 38
- 8.6—Miscellaneous concrete products, p. 38

CHAPTER 9—OTHER USES OF FLY ASH, p. 38

- 9.1—Grouts and mortar, p. 38
- 9.2—Controlled low-strength material, p. 39
- 9.3—Soil cement, p. 39
- 9.4—Plastering, p. 39
- 9.5—Cellular concrete, p. 39
- 9.6—Shotcrete, p. 39
- 9.7—Waste management, p. 40
- 9.8—Cements, p. 40

CHAPTER 10—REFERENCES, p. 40

- Authored documents, p. 41

APPENDIX A—RAPID QUALITY CONTROL TESTS, p. 54

- A.1—Loss on ignition, p. 54
- A.2—Carbon analysis, p. 54
- A.3—Particle size, p. 54
- A.4—Color, p. 55
- A.5—Density (specific gravity), p. 55
- A.6—Fly ash adsorption, p. 55
- A.7—Organic material, p. 55
- A.8—CaO content, p. 55
- A.9—Presence of hydrocarbons (startup oil), p. 55
- A.10—Presence of ammonia (precipitator additive), p. 55
- A.11—Calorimetry, p. 55

CHAPTER 1—INTRODUCTION, SCOPE, SOURCES, AND SUSTAINABILITY

1.1—Introduction

Fly ash, a material resulting from the combustion of pulverized coal, is widely used as a cementitious and pozzolanic ingredient in concrete and related products. Fly ash is introduced in concrete either as a separately batched material (ASTM C618, Class C or F) or as a component of blended cement (ASTM C595/C595M; ASTM C1157/C1157M; ASTM C1600/C1600M).

Fly ash possesses pozzolanic properties similar to the naturally occurring pozzolans of volcanic or sedimentary origin found in many parts of the world. Two thousand years ago, the Romans mixed volcanic ash with lime, aggregate, and water to produce mortar and concrete (Vitruvius 1960). In modern concrete, fly ash combines with calcium hydroxide ($\text{Ca}(\text{OH})_2$, also known as portlandite, which predominately results from the hydration of portland cement, and with water to form additional cementing product. This process, called the pozzolanic reaction, creates a finer pore structure, which in turn increases the durability of mortar and concrete.

All fly ashes exhibit pozzolanic properties to some extent. However, some fly ashes also display varying degrees of cementitious properties without the addition of $\text{Ca}(\text{OH})_2$ or hydraulic cement. The cementitious nature of the latter type of fly ash is primarily attributed to the presence of reactive constituents such as calcium aluminate and calcium silicate phases, and calcium oxide. The role of fly ash in concrete with hydraulic cement is summarized as:

a) Calcium and alkali hydroxides that are released into solution in the pore structure of the paste by hydrating cement combine with the pozzolanic phases of fly ash, to form additional calcium silicate hydrate (C-S-H) gel (cementing matrix)

b) The heat of hydration helps to initiate the pozzolanic reaction and contributes to the rate of the reaction

When concrete containing fly ash is cured, fly ash reaction products fill spaces originally occupied by mixing water but not filled by the hydration products of the cement, thus reducing the concrete permeability to fluids (Manmohan and Mehta 1981). The slower reaction rate of fly ash, when compared with hydraulic cement, limits the amount of early heat generation and the detrimental effect of early

temperature rise in massive concrete structures. Concrete proportioned with fly ash can develop properties that are not achievable through the use of hydraulic cement alone.

1.1.1 History—Fly ash from coal-burning electric power plants became readily available in the 1930s and, shortly thereafter, the study of fly ash for use in hydraulic cement concrete began (Davis et al. 1937; Stanton 1940). This early research served as the foundation for initial specifications, methods of testing, and use of fly ash. Abdun-Nur (1961) covers much of the early history and technology of using fly ash in construction and includes an annotated bibliography (1934–1959). Since this early work, much research has been performed regarding alkali-silica reaction (ASR) mitigation using fly ash. A recent summary is provided by Thomas et al. (2013).

Initially, fly ash was used as a partial replacement of hydraulic cement, which is typically the most expensive manufactured component of concrete. As fly ash usage increased, researchers recognized that fly ash could impart beneficial properties to concrete. Additional research was done on the reactivity of fly ash with calcium and alkali hydroxides in portland cement paste, and the ability of fly ash to act as a mitigator of deleterious alkali-silica reactions was identified (Davis et al. 1937). Other research has shown that fly ash often improves concrete's resistance to deterioration from sulfates (Dunstan 1976, 1980; Tikalsky et al. 1992; Tikalsky and Carrasquillo 1993). Fly ash also increases the workability of fresh concrete and reduces the peak temperature of hydration in mass concrete. The beneficial aspects of fly ash were especially notable in the construction of large concrete dams (Mielenz 1983). Some major projects, including the Thames Barrier in the UK (Newman and Choo 2003) and the Upper Stillwater Dam in the United States (Poole 1995), incorporated 50 and 65 percent mass replacement of hydraulic cement with fly ash to reduce heat generation and decrease permeability, respectively. The Iraivan Temple, built in Kauai, HI, in 1999, has a foundation composed of high-volume fly ash (HVFA) concrete with an estimated service life of 1000 years (Mehta and Langley 2000). This concept of HVFA concrete was adopted for foundation construction of at least two additional temples in the United States: one located in Chicago, IL, and the other in Houston, TX (Malhotra and Mehta 2012). In addition, numerous projects in the United States have used HVFA concrete for sustainable construction. More information on HVFA usage is available in Chapter 7 and ACI 232.3R.

A new generation of coal-fired power plants were built in the United States during the late 1960s and 70s using efficient coal mills and state-of-the-art pyroprocessing technology. These plants produce fly ash with a smaller average particle size and lower carbon content. Fly ash containing high levels of calcium oxide became available because of the use of western U.S. coal sources, typically subbituminous and lignite. Enhanced economics and improved technologies, both material- and mechanical-based, have led to a greater use of fly ash throughout the ready mixed concrete industry. Extensive research has led to a better understanding of the chemical reactions involved when fly ash is incorporated in concrete.

Fly ash is used in concrete for many reasons (refer to Chapter 4), including improvements in workability of fresh concrete, reduction in temperature rise during initial hydration, improved resistance to sulfates, reduced expansion due to alkali-silica reaction, and contributions to the durability and strength of hardened concrete. In the 1990s and 2000s, some power plants made changes to co-fire coal with biomass and to improve air quality by using scrubbers to reduce sulfur oxide emissions (SO_x), catalytic reduction equipment to reduce nitrous oxide emissions (NO_x), and various systems to reduce mercury emissions. These additional systems have the potential to alter the composition of the fly ash by incorporating such compounds as ammonia, sulfate, sulfite, alkalis, and carbon residues. These changes should be considered when selecting fly ash sources, as additional quality control parameters may be required for acceptance.

1.2—Scope

The scope of this report is to describe the use and characterization of fly ash, its properties, and its impacts on concrete properties. Guidance is provided concerning specifications, quality assurance, and quality control of fly ash itself, as well as that of concrete and related products produced using fly ash.

1.3—Source of fly ash

Due to the increased global use of pulverized coal as fuel for electric power generation, particularly in China and India, fly ash is available in many areas of the world. Approximately 53.4 million tons (48.4 million metric tons) of fly ash are produced annually in the United States (American Coal Ash Association 2015). An estimated 27 percent of that total is used in the production of cement, concrete, and manufactured concrete products.

1.3.1 Production and processing—The ash content of coals by mass may vary from 4 to 5 percent for subbituminous and anthracite coals, to as high as 35 to 40 percent for some lignites. The combustion process, which creates temperatures of approximately 2900°F (1600°C), liquefies the incombustible minerals. Rapid cooling of these liquefied minerals upon leaving the firebox causes them to form spherical particles with a predominantly glassy structure. Many variables can affect the characteristics of these particles. Among these are coal composition, grinding mill efficiency, the combustion environment (for example, temperature and oxygen supply), boiler/burner configuration, mineral additions, processing conditions, and the rate of particle cooling.

Modern coal-fired power plants that burn coal from a uniform source produce very consistent fly ash. Fly ash particles originating from the same plant and coal source will vary in size, chemical composition, mineralogical composition, and density. Particle sizes may run from less than 1 μm to more than 200 μm , and density of individual particles may vary from less than 62.4 lb/ft³ (1 g/cm³) for hollow spheres to more than 187 lb/ft³ (3 g/cm³) for fly ash with a preponderance of solid spheres. The true density of bulk fly ash produced by a single coal-burning plant can vary very dramatically.

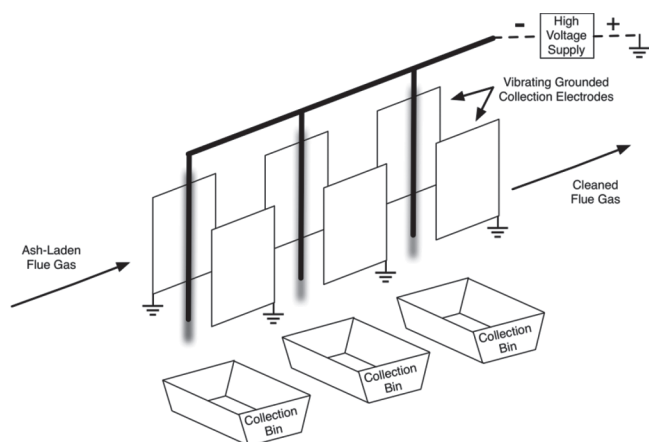


Fig. 1.3.1—Typical gas flow pattern through electrostatic precipitator.

Collection of these particles from the furnace exhaust gases is routinely accomplished by electrostatic or mechanical precipitators or by bag houses. A typical gas flow pattern through an electrostatic precipitator is shown in Fig. 1.3.1.

As fly ash particles are collected in a bag house or mechanical precipitator, they segregate in sequential precipitator hoppers according to their size and density; the larger and heavier particles tend to accumulate closer to the fly-ash-laden gas inlet, whereas the smaller and lighter particles tend to collect farther from the inlet. In electrostatic precipitators, however, the particle size and density trends in sequential hoppers are disrupted due to the influence of the charged collection grids. The fineness, density, and carbon content of fly ash can vary significantly from hopper to hopper in both mechanical and electrostatic precipitators. Hoppers can be selectively emptied and transported to a main silo. Blending occurs as a natural result of pneumatic material handling operations.

1.3.2 Impact of environmental regulations—Nitrous oxide emissions are considered to contribute to the production of ozone levels; along with SO_x , both are considered to contribute to acid rain. Additionally, air regulations are being implemented that further limit fine particulate and mercury emissions.

It has been suggested that some approaches to pollution reduction in coal combustion may modify the cementitious or pozzolanic properties of fly ash. Changes in fly ash glass content and mineralization, combined with changes in particle size distribution and particle morphology, can affect fly ash reactivity. The impact on reactivity can vary from significant to inconsequential, depending on the specific fuel and combustion modification system employed. Post-combustion technologies for reducing NO_x emissions and mercury emissions may also impact fly ash quality. The processes are summarized in the following sections.

1.3.2.1 SO_x reduction technologies—To reduce SO_x emissions, the power-generating industry has adopted a two-pronged approach. The first is a shift toward fuel sources that are lower in sulfur content, and the second is to apply technologies such as flue gas desulfurization (FGD). With regard to low-sulfur coal sources, some coal-fired power

plants have shifted from the use of eastern and central U.S. coal sources in favor of western coal sources, primarily those from the Powder River Basin (Energy Information Administration 2015). Due to low natural gas prices, the increased supply of natural gas due to fracking technology, and the need to reduce CO_2 emissions from power generation, the U.S. is expected to decrease its reliance on coal in future years. In 2013 (Energy Information Administration 2015), U.S. coal production fell below one billion short tons in the United States—3.1 percent lower than 2012—with production from the Western Region representing 53.8 percent of the U.S. total.

FGD methods have been in place for many years as a result of limits placed on SO_x emissions as part of the Clean Air Act (CAA). In general, SO_x is removed from flue gases by a variety of methods that include wet scrubbing using a slurry of sorbent such as limestone or lime, spray-dry scrubbing using similar sorbents, or dry sorbent injection systems (Nolan 2000). Normally, the by-product is a material that is currently unusable for portland cement concrete. However, some FGD materials have been used as a calcium sulfate source for the cement and wallboard industries. The presence of FGD materials in fly ash is detected by testing SO_3 levels.

Other approaches include the increased use of fluidized bed combustors, which result in lower SO_x production but also result in production of fluidized bed combustor ash that currently is not marketed for use in portland cement concrete production. Reducing the excess air in the combustion process also controls formation of SO_x ; however, limits on excess air could lead to increases in unburned fuel, which increases the loss on ignition (LOI) value or, theoretically, could result in incomplete oxidation of mineral species. Effects stemming from the latter concern have not been reported in the literature reviewed.

1.3.2.2 NO_x reduction technologies—The control of NO_x emissions is addressed primarily through the use of low- NO_x burners and a variety of downstream treatment technologies including the use of over-fire air, selective catalytic reduction (SCR), and selective noncatalytic reduction (SNCR).

NO_x forms during the combustion of coal as a result of two primary mechanisms. Thermal NO_x results from the oxidation of nitrogen in air while fuel NO_x results from oxidation of nitrogen in coal. The first source, thermal NO_x , increases exponentially with temperature and is controlled by moderating flame temperature and oxygen concentration at the burner (LaRue et al. 2001). Limiting the oxygen available during the early stages of the combustion process controls fuel NO_x . Technologies that reduce oxygen availability at the flame will effectively reduce NO_x , but as a by-product of this process change, there tends to be an increased amount of unburned fuel that can be found in the flue gases as either carbon monoxide (CO) or as carbon particulate. Even with the addition of over-fire air, higher unburned fuel amounts occur with the same total amount of combustion air (LaRue et al. 2001).

The basic principle of SCR is the reduction of NO_x to N_2 and H_2O by the reaction of NO_x and ammonia (NH_3) over a catalyst bed. SCR catalysts are manufactured using ceramic

materials as a substrate, such as titanium oxide, and active catalytic components are usually either oxides of base metals, zeolites, or various precious metals (Institute of Clean Air Companies 2008). In contrast, SNCR is a chemical process that converts NO_x into molecular N_2 without the use of a catalyst. A reducing agent, typically ammonia, is injected into the flue gases at high temperatures—for example, 1600 to 2100°F (870 to 1150°C) for the conversion of nitrogen oxides into diatomic nitrogen (N_2) and water (H_2O). SNCR is selective in that ammonia reacts primarily with NO_x and not with oxygen or other major components of the flue gas. In both SCR and SNCR, no solid or liquid wastes are generated except for spent catalyst in the case of SCR.

A problem associated with both approaches, especially SNCR, is ammonia slip, where excess ammonia deposits in the fly ash. In some fly ash, ammonium salts have been detected at concentrations ranging from barely measurable to levels exceeding several thousand parts per million (ppm) (Brendel et al. 2001). Low concentrations of ammonia have no impact on concrete properties (Koch and Prenzel 1989); however, a strong ammonia odor can be emitted. Although research has shown that this excess ammonia does not result in decreased concrete performance (Van der Bruggen et al. 1995), it does create a potential work-place hazard, as ammonia gas is released from the concrete mixture when the ammonia-laden fly ash combines with the high-pH pore solution that is created when portland cement is mixed with water (Rathbone et al. 2002). Ammonia absorption is also more concentrated in high-sulfur fly ash through the formation of ammonium salts, and in high-carbon fly ash through adsorption of carbon. The latter problem can be addressed during treatment of fly ash for carbon removal, including carbon burnout (Giampa 2000). A rapid method for determining the ammonia concentration in fly ash as a means for quality control of fly ash used for concrete was provided by Majors et al. (1999). When using fly ash containing ammonia, consideration should be given to material characteristics, applications, environment, and quality control programs in place.

1.3.2.3 Mercury reduction technologies—Technologies to achieve mercury reduction goals clearly pose the most significant potential change to fly ash characteristics. The majority of technologies being used or discussed include various approaches to injecting activated carbon into the flue gas stream to adsorb gaseous mercury. The activated carbon may or may not be commingled with the fly ash, depending on the technology used. Other techniques include capturing the mercury as a result of other pollutant control measures, so-called multi-pollutant control; concrete-friendly amended silicate sorbents; and other methods in development (Ramme and Tharaniyil 2013).

Mercury (Hg) is volatilized from coal during combustion and converted to elemental mercury (Hg^0) vapor, referred to as gaseous phase mercury. As the flue gas cools, the reduced mercury (Hg^0) oxidizes to ionic mercury (Hg^{2+}) and could form mercury compounds that are in a solid phase at flue gas temperatures, or it could occur as mercury that is adsorbed onto the surface of other particles. The presence of chlo-

rine in flue gas favors the formation of mercuric chloride (HgCl_2) at flue gas temperatures (Miksche and Ghorishi 2007). Mercury oxidation reactions, however, are kinetically limited and, therefore, mercury is present in the flue gas as a mixture of species including Hg^0 , Hg^{2+} , adsorbed on other particle surfaces, or as mercury compounds. Reports indicate that most gaseous mercury in bituminous coal-fired boilers is Hg^{2+} , whereas gaseous mercury in subbituminous and lignite-fired boilers tends to be present as Hg^0 (Environmental Protection Agency 2002). The oxidation state of mercury in the flue gas greatly determines the type of control technology that can be used.

In the past, the general approach was to capture mercury as a part of other pollution control strategies used to achieve SO_x , NO_x , or particulate control. Selective catalytic reduction results in mercury oxidation. Once oxidized, the mercury is soluble in wet-scrubber solution and can be captured in the wet scrubber. Note that reduced mercury (Hg^0) is not soluble in the wet-scrubber solution (Environmental Protection Agency 1997). If the mercury is not oxidized in retention or by SCR, then a wet scrubber is unable to efficiently remove gaseous phase mercury. Mercury that has adsorbed onto solid particles, or has formed other solid compounds, can be effectively removed as a result of particulate removal in either an electrostatic precipitator or fabric filter or baghouse. Again, the mercury being oxidized is key, as the oxidized form adsorbs on solids more readily and is also the form that is required to precipitate mercuric compounds (Environmental Protection Agency 1997, 2002).

To achieve higher levels (that is, greater than 90 percent) of mercury reduction, new technologies need to be employed (Hinzy et al. 2013; Wdowin et al. 2014). The most economically feasible technology for existing power plants to meet EPA mercury reduction requirements is by the use of activated carbon injection directly into the flue gas to adsorb gaseous mercury. Activated carbon, most commonly in the form of powdered activated carbon (PAC), is being evaluated for use in power plants throughout the United States. Where the activated carbon is injected in the process has a large impact on whether acceptable fly ash is produced. The simplest and most economical approach is for the activated carbon to be injected prior to the primary particulate control device (PCD), where it will then travel downstream and be commingled and collected with the fly ash in the primary PCD. In this case, the resulting fly ash will contain an increased activated carbon fraction, thereby increasing the fly ash LOI value (Pflughoeft-Hassett et al. 2008).

An alternative option is injection of activated carbon after a primary PCD. The carbon is then removed with the remaining fly ash in a secondary PCD. In this option, the fly ash collected from the primary PCD will not be commingled with the activated carbon. Accomplishing this approach would require capital investment in a secondary PCD, if one is not already being used in the process.

In response to the need to minimize LOI in fly ash, treatment methods have been employed to treat the fly ash resulting after activated carbon injection. Several approaches used to reduce LOI from fly ash in general and

include carbon burnout, triboelectrostatic separation, and activated carbon adsorption neutralization either by chemical treatment or ozonation (Hill and Folliard 2006; Howard et al. 2013). When using thermal treatment methods, it is vital that no mercury is released. Mercury is released from fly ash in the temperature range of 572 to 752°F (300 to 400°C) (Rubel et al. 2003).

1.3.2.4 Carbon dioxide reduction technologies—To date, CO₂ emissions from coal-fired power plants have not been regulated. However, with the release of the EPA Clean Power Plan in 2015, it is inevitable that such controls will be put in place. Therefore, the power industry is investigating methods to reduce CO₂ emissions. Most approaches center on power generation efficiency improvements, carbon capture, and sequestration.

Capture, sequestration, and separation methods for CO₂ emissions will differ much in the same way as mercury capture technologies differ, depending on the flue gas chemistry. Current options include physical and chemical adsorption, distillation at low temperatures, gas separation membranes, mineralization, and biomineralization. Some of these technologies could create new or alter existing inorganic phases in the fly ash.

1.3.3 Beneficiated fly ash—If the quality of some or all of the fly ash produced is less than required by specification or market standards, methods may be used to beneficiate the fly ash. Low-calcium-content ashes that do not harden under water may be used after long-term stockpile or pond storage. Beneficiation and processing, however, are required (McCarthy et al. 2013). Properties that are commonly controlled by beneficiation include fineness and LOI, an indicator of carbon content. The physical and chemical properties of fly ash can vary among individual precipitator or baghouse collection hoppers. This phenomenon can be taken advantage of in some operations to produce a high-quality fly ash. Where the control and piping systems in the power plant allow, fly ash can be selectively drawn from those hoppers that contain the higher-quality fly ash while material of questionable quality can be discarded or directed to other uses.

Air classification systems can be used to reduce the mean particle size of fly ash to meet specification or market requirements. These systems separate particles based on the combination of particle diameter, shape, and apparent density (Wills 1979). Depending on the size, apparent density, and distribution of particles containing carbon, LOI of the processed fly ash can be increased, decreased, or unchanged by this technique. In general, the finer the fly ash, the lower the LOI and the greater the concrete's late-age compressive strength. Increased fineness with spherical-shaped particles also lowers the water demand and increases resistance to sulfate attack in concrete (Electric Power Research Institute 2001).

Numerous investigations have demonstrated that fly ash performance can be enhanced by significantly shifting the particle-size distribution to finer material (Butler 1981; Berry et al. 1989; Obla et al. 2001b). Compared with a fly ash with a mean particle diameter ranging from 15 to 35 µm, processed fly ash can be produced with a mean particle

diameter of 2.5 to 4.0 µm. Particle-size reductions of this magnitude have been achieved by methods of specialized air classification systems (Cornelissen et al. 1995; Hassan and Cabrera 1998) and micronization (Paya et al. 1995; Bouzoubaâ et al. 1997). These processed ultra-fine fly ashes can provide water reductions of 10 to 12 percent in mortar and reduce high-range water reducer demand in concrete (Ferraris et al. 2001). Kruger et al. (2001) and Obla et al. (2001a,b) have demonstrated that ultra-fine fly ashes contribute more toward concrete strength gain and permeability reduction than unsized fly ash and will, when properly proportioned, provide concrete characteristics comparable to highly reactive pozzolans such as silica fume. Concrete durability properties, such as resistance to alkali-silica reaction (Bérubé et al. 1995), sulfate attack (Shashiprakash and Thomas 2001), and concrete permeability (Obla et al. 2000) are enhanced by ultra-fine fly ash.

Commercial technologies now available to reduce the LOI of fly ashes without negative effects to other properties include triboelectric separation (Whitlock 1993) and thermal beneficiation techniques (Cochran and Boyd 1993; Knowles 2009). Triboelectric separation uses electrostatic charge exchange between carbon and mineral particles occurring due to contact during pneumatic conveyance. Bittner and Gasiorowski (1999) reported on a commercial triboelectric process that uses a countercurrent moving belt to facilitate the separation of carbon from fly ash in a high-voltage field. Triboelectric separation systems have generated 500,000 tons (450,000 metric tons) of fly ash per year. Triboelectric operations based on alternate designs have also been demonstrated but not commercialized (Li et al. 1999; Soong et al. 1999).

Thermal beneficiation is another means of reducing fly ash carbon content. Different processes burn the residual carbon in fly ash as a fuel source in an auxiliary fluidized bed combustor or a turbulent reactor, producing a pozzolan meeting the required carbon content. In the case of the turbulent reactor, the residual carbon can be totally removed (Knowles 2009). In the process, heat is recovered and returned to the power plant that originally produced the high-carbon fly ash. One commercially-operating facility has reported processing capabilities of 180,000 tons (163,000 metric tons) per year (Electric Power Research Institute 2001; Frady et al. 1999). In addition to burning carbon, the temperature of these thermal beneficiation processes can remove ammonia from the fly ash (Giampa 2000). Fly ash fuel reburn technology has been in commercial use at some power plants since 1999. High-LOI fly ash from other plants and fly ash recovered from monofill landfills are introduced in a metered proportion to the coal transported to pulverized coal-fueled power plants to recover the energy, and alter the resulting chemical and physical composition of the power plant's conventional fly ash (Ramme and Tharaniyil 2004).

Froth flotation is a method derived from mineral processing that separates carbon from fly ash by introducing the fly ash into a slurry system. The slurry contains frothing chemicals that facilitate the flotation of less-dense carbon particles, whereas the inorganic fraction of fly ash is carried to a collection area. The processed fly ash is dried before use.

(Grosso 2001). Froth flotation can be useful for removing very fine carbon (Electric Power Research Institute 2001).

The potential for a fly ash to impact the air-entrainment level in concrete is not always a simple function of residual carbon, as indexed by LOI values. Hurt et al. (1995) suggested that carbon in fly ash is heterogeneous, ranging from coke-like to lacy in morphology. More recent studies point to the fact that fly ash from different sources can exhibit a varying impact on air entrainment even though LOI values are almost equivalent (Hill et al. 1997, 1998, 1999). Other research has highlighted the important role that total carbon surface area, available surface area, and surface reactivity play in the interaction between fly ash carbon and chemical air-entraining admixtures (Freeman et al. 1997; Gao et al. 1997). Studies indicate that modifying carbon surface properties without significantly reducing carbon mass potentially affects the adsorptive properties of fly ash carbon (Sabanegh et al. 1997; Hill and Majors 2001). Ozonation has been suggested as a means for chemically passivating carbon against chemical interaction with air-entraining admixtures as a means for fly ash beneficiation (Hurt et al. 2000). Some fly ash sources are treated with spray-applied admixtures that adhere to the carbon and lessen its ability to impact the air-entrainment level in concrete. A high-temperature air slide for use in fly ash beneficiation for ammonia removal, mercury removal, or both, from fly ash has also been developed (Ramme and Tharaniyil 2013).

1.4—Fly ash and sustainability

1.4.1 Sustainability considerations in structure design—

Awareness of sustainability has become much more prevalent in concrete construction. Concrete is a widely used and often locally available material. Properly designed and constructed concrete structures can provide the owner or occupant with many years of service. Concrete using fly ash benefits sustainable development by:

- Possibly reducing the portland cement content, thus lowering the CO₂ footprint of a cubic yard of concrete
- Possibly reducing the demand for portland cement or aggregate in concrete mixtures, ultimately reducing use of virgin raw materials and the environmental burdens associated with resource extraction, processing, as well as the transportation associated with the manufacturing of portland cement
- Reducing the need for disposal of this viable industrial by-product to landfill, thereby diverting materials from landfill, reducing potential impacts to groundwater, and encroaching upon valuable open space and biodiversity
- Substantially enhancing concrete durability, thereby increasing the functional service life of buildings and infrastructures, thus lowering the embodied energy from new construction and the energy and environmental impacts from demolition (longer lasting structures are one of the most effective strategies for minimizing environmental and economic impacts)
- Supporting the economy and reducing transportation impacts; in most regions of the world, fly ash is a regionally available material

Table 1.4.1—Considerations for a sustainable and resilient structure design (Brown 2006; AASHTO 2008)

Environmental	Social	Economic
Ecology and biodiversity	Community interaction	Life cycle costs
Landscape	Community liveability	Project management
Storm water impacts	Human health impacts	Financial sustainability
Construction waste management	Historic and cultural preservation	Economic analysis
Material use	Scenic and natural qualities	Safety programs
Energy and carbon	Safety	Land use
Reduce, recycle, and reuse	Equity	Operation and management systems
Reduced energy and emissions	Stakeholder involvement	Bridge management systems
Noise pollution	Transportation impacts	Energy efficiency
Resiliency	Resiliency	Resiliency

f) Requiring less water in manufacture because the concrete typically will have a lower water content, often improving strength and reducing permeability with durability benefits

Sustainability is an evolving term generally associated with the availability and judicious use of finite resources and with decision making that values and considers both present and future generations (World Commission on Environment and Development 1987). The terms “sustainability” and “sustainable” mean to create and maintain conditions, under which humans and nature can exist in productive harmony, that permit fulfilling the social, economic, and other requirements of present and future generations (United States Federal Register 2009).

Functional definitions that align with the three pillars of sustainability, or the triple bottom line (that is, the Three “E”s: environment, economics, and equity) can overlap when the theoretical framework for sustainable decision making is used and when an emphasis can be placed on each of the pillars. However defined, for any process or product to be truly sustainable, it should also have resilience against external disturbances. Van Dam et al. (2015) provides a general discussion of the contributions of fly ash.

Sustainable and resilient design requires an integrated, long-term, and holistic view of all phases of the project: planning, designing, constructing, maintaining, operating, repair/rehabilitation, and final decommissioning and disposal at the end of its service life. The responsibility of a sustainable design team does not lie solely with aesthetical impact and functional performance, but also with key concerns such as integration of context-sensitive solutions; awareness of societal and biodiversity impacts; life cycle costing; climate mitigation/adaptation; and minimizing the impact on the environment, society, and the economy throughout the structure’s life. Table 1.4.1 summarizes numerous key considerations for a sustainable and resilient structure design.

Table 1.4.2—Average environmental impacts and water and energy savings for SCMs in concrete per unit mass of recovered mineral component substituted for cement at a 1:1 level (Environmental Protection Agency 2008)

	Fly ash	Slag cement	Silica fume
	Per metric ton		
Water savings, L	376.3	145.2	–5114
Water savings, in USD	0.20	0.10	–3.20
Energy savings, megajoules	4696	4221	32,915
Energy savings, in USD	129	116	905
Avoided CO ₂ emissions (GHG), g	701,378	668,889	699,876
	Per pound		
Energy savings, in USD	0.059	0.053	0.411
Avoided CO ₂ emissions (GHG), lb	0.318	0.314	0.318
	Per kilogram		
Energy savings, USD	0.129	0.116	0.905
Avoided CO ₂ emissions (GHG), kg	0.71	0.669	0.699

Notes: Impact metrics based on representative concrete products. Negative values represent an incremental increase in impacts relative to the use of portland cement.

1.4.2 Greenhouse gases and fly ash—The most effective means of decreasing both energy consumption and the production of greenhouse gases is to substitute supplementary cementitious materials (SCMs), such as fly ash, for a portion of the portland cement. SCMs incorporated into cement-based building materials are added individually, blended, or interground with portland cement.

The [Environmental Protection Agency \(2008\)](#) has calculated the environmental impact of fly ash, silica fume, and slag cement and expressed the result in energy efficiency savings and corresponding levels of reduced CO₂ emissions. These values are derived from life cycle inventory data and represent the total life cycle savings of using SCMs as a replacement for 1 metric ton of finished portland cement in concrete. Table 1.4.2 summarizes energy savings and CO₂ emissions not occurring from portland cement manufacturing for each of the three common SCMs at the following replacement rates: 30 percent ASTM C618 Class F fly ash, 50 percent slag cement, and 10 percent silica fume. These rates can vary significantly depending on the application and with ternary mixture use (that is, using two SCMs along with portland cement in the same mixture) to achieve the desired properties. For example, very high replacement percentages of cement with Class C fly ash can be appropriate for specific applications. Additional information is available in [ACI 232.3R](#).

1.4.3 Reduction of waste stream materials to landfill—Fly ash is a by-product of coal-fired furnaces at power generation facilities and its use in concrete and concrete product manufacture enables the reduction of landfilled materials. Landfilling is the most common waste management option for fly ash and a majority of the fly ash generated in the United States is disposed of in landfills. Transportation of fly ash to

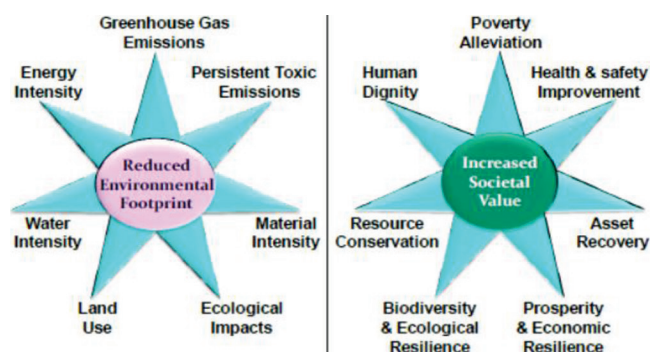


Fig. 1.4.5—Typical categories of sustainability indicators (Fiksel et al. 2013).

a landfill and operation of landfill equipment result in anthropogenic CO₂ emissions from the combustion of fossil fuels in the vehicles used to haul the wastes. Additionally, the diversion of materials from landfills reduces potential impacts to groundwater and encroachment upon valuable open space.

1.4.4 Robustness and durability of fly ash concretes—Fly ash plays a critical role in increasing the longevity of concrete structures. The use of fly ash substantially enhances concrete durability, thereby increasing the functional service life of buildings and infrastructure ([Malhotra and Ramezaniapour 1994](#); [Van Dam et al. 2015](#)). Increases in service life lower the embodied energy from new construction and the energy and environmental impacts from demolition. The design of long-life structures and effective life cycle management of existing structures are one of the most effective strategies for minimizing environmental and economic impacts, as well as ensuring public safety, health, security, serviceability, and life-cycle cost effectiveness ([Lounis and Daigle 2006, 2010](#)).

1.4.5 Measurements of sustainability for construction—Several sustainability indicators are used by varying organizations in the United States and globally. Depending on the perspectives of various stakeholder groups and interested parties, the preferred indicators can vary. Carefully chosen and implemented indicators can assist policymakers ([Singh et al. 2009](#)). Figure 1.4.5 illustrates several common sustainability indicators ([Mitchell et al. 1995](#); [Niemeijer and deGroot 2008](#)).

In the construction industry, sustainable rating systems such as LEED®, BREEAM®, CEEQUAL, and Envision™ have been developed to provide independent assessment standards that evaluate, measure, and improve the performance of buildings, infrastructure, and communities. While each rating system may favor certain strategies over others, there are similar sustainability performance indicators when evaluating building materials. Three performance indicators predominate: reduced net embodied energy and carbon footprint of products, systems, or both (often stated as global warming potential in units of CO₂ equivalents) ([Hart 1997](#); [Kibert 2012](#); [Pezzey 1992](#); [Omer 2008](#)); reduced resource depletion (including increased recycled content) ([Pezzey 1992](#); [Lippiatt 1998](#); [Hill and Bowen 1997](#)); and transparency in reporting environmental impacts ([Kibert 2012](#); [Berardi 2012](#); [Braune et al. 2007](#)).

Table 1.4.5a—Excerpt from example EPD showing potential environmental impacts for fly ash (Danish Technological Institute 2013)

Impact category		Impact per tonne of fly ash			
		Total	Loading at power plant	Depot	Transport
Global warming (GWP)	kg CO ₂ equivalent	3.92	1.31	2.61	5.71×10^{-3}
Ozone depletion (ODP)	kg CFC-11 equivalent	9.88×10^{-10}	3.29×10^{-10}	6.58×10^{-10}	0
Acidification for soil and water (AP)	kg SO ₂ equivalent	7.26×10^{-3}	2.41×10^{-3}	4.82×10^{-3}	2.31×10^{-5}
Eutrophication (EP)	kg PO ₄ equivalent	1.05×10^{-3}	3.48×10^{-4}	6.96×10^{-4}	6.06×10^{-6}
Photochemical ozone creation in tropospheric ozone (POCP)	kg ethene equivalent	5.49×10^{-4}	1.87×10^{-4}	3.73×10^{-4}	-1.10×10^{-5}
Depletion of abiotic resources – elements	kg Sb equivalent	3.29×10^{-7}	1.10×10^{-7}	2.19×10^{-7}	0
Depletion of fossil resources	MJ, net calorific value	43.3	14.4	28.8	0

Table 1.4.5b—Excerpt from example EPD showing resource consumption per declared unit of fly ash (Danish Technological Institute 2013)

Resources		Consumption per tonne of fly ash			
		Total	Loading at power plant	Depot	Transport
Renewable primary energy	MJ	15	4.9	10	0
Nonrenewable primary energy	MJ	43	14	29	0
Renewable secondary energy	MJ, net calorific value	0	0	0	0
Nonrenewable secondary energy	MJ, net calorific value	0	0	0	0
Use of secondary material	kg	0	0	0	0
Water	M3	0.427	0.142	0.285	0

Table 1.4.5c—Excerpt from example EPD showing generation of waste per declared unit of fly ash (Danish Technological Institute 2013)

Waste categories		Waste per tonne of fly ash			
		Total	Loading at power plant	Depot	Transport
Hazardous waste	kg	0	0	0	0
Nonhazardous waste – from excavation of resources	kg	5.71	1.90	3.81	0
Nonhazardous waste – other	kg	7.95×10^{-2}	2.65×10^{-2}	5.30×10^{-2}	0
Radioactive waste	kg	0	0	0	0
Materials for reuse	kg	0	0	0	0
Materials for recycling	kg	0	0	0	0
Materials for energy recovery	kg	0	0	0	0

Environmental product declarations (EPDs), as defined by **ISO 14025** and **ISO 21930**, are currently the method to report environmental impacts in a formalized and comparable structure.

Comparatively, a concrete EPD summary contains the results of a life cycle assessment (LCA) (that is, ecobalance and cradle-to-grave analysis of environmental impacts associated with all the stages of a product's life from raw material extraction through materials processing, manufacture, distribution, use, repair and maintenance, and disposal), conducted according to the ISO 14040 series and based on a specified unit of one cubic meter or one cubic yard of concrete, and a specified design strength and age, with additional options for performance. An example of this type of

declaration is shown in Tables 1.4.5a through 1.4.5d. Use of fly ash in the concrete mixture design would require incorporation of the fly ash EPD into the concrete EPD.

CHAPTER 2—DEFINITIONS

Please refer to the latest version of “ACI Concrete Terminology” for a comprehensive list of definitions.

CHAPTER 3—FLY ASH COMPOSITION

3.1—General

Fly ash particles consist of heterogeneous combinations of amorphous (glassy) and crystalline phases. The largest fraction of fly ash consists of glassy spheres of two types: solid

Table 1.4.5d—Contents of a sample EPD for concrete (Danish Technological Institute 2013)

Mixture performance	Name	Abbreviation	Unit
	28-day compressive strength	CS	psi
Mixture impacts, per m ³	Total primary energy consumption	TPE	MJ
	Concrete water use, batch	CWB	m ³
	Concrete water use, wash	CWW	m ³
	Global warming potential	GWP	kg CO ₂ -eq
	Ozone depletion	ODP	kg CFC-11-eq
	Acidification	AP	kg SO ₂ -eq
	Eutrophication	EP	kg N-eq
	Photochemical ozone creation/smog	POCP	kg O ₃ -eq

particles and hollow particles called cenospheres. These glassy phases usually comprise 60 to 90 percent of the total mass of fly ash, with the remaining fraction of fly ash made up of a variety of crystalline phases. Crystalline phases can exist as independent particles or be commingled with the glass phase as either a surface deposit or inclusion. Fly ash is an inherently complex material to classify and characterize, as the composition and mineralogy depend on numerous factors, including coal type, coal grinding mill efficiency, coal feeding rate, combustion environment (for example, temperature and oxygen supply), type and configuration of boiler/burner, and fly ash collection method.

3.2—Chemical composition

Bulk chemical composition (Table 3.2) has been used by **ASTM C618** to classify fly ash into two types: Classes C and F. The chemical composition data used to determine compliance with ASTM C618 do not directly address the reactivity of the particles, but are used as a quality control or quality assurance tool. Minor variations in the chemical composition of a specific fly ash do not relate directly to the long-term performance of concrete containing that fly ash. Fly ash composition is reported as percent oxides by mass, although the elements analyzed may not always be present in a pure oxide form, and may be incorporated within glassy or other mineral phases. The crystalline and glassy constituents that remain after the combustion of the pulverized coal are a result of materials with high melting points and incombustibility. The amounts of the four principal constituents vary widely. Typical values are SiO₂ (35 to 60 percent), Al₂O₃ (10 to 30 percent), Fe₂O₃ (4 to 20 percent), and CaO (1 to 35 percent). The sum of the first three constituents—SiO₂, Al₂O₃, and Fe₂O₃—need to be equal to or exceed 70 percent for the material to be classified as an ASTM C618 Class F fly ash, whereas their sum need only exceed 50 percent for the material to be classified as an ASTM C618 Class C fly ash. Class C fly ashes typically have a higher CaO content than a Class F fly ash.

The silica and alumina in the glass of fly ash, and Ca(OH)₂ generated with hydration of portland cement, are primary contributors to the pozzolanic reaction in concrete because the amorphous silica and alumina combine with Ca(OH)₂

Table 3.2—Example bulk composition of fly ash with coal sources

	Bituminous	Subbituminous	Northern lignite	Southern lignite
SiO ₂ , percent	45.9	31.3	44.6	52.9
Al ₂ O ₃ , percent	24.2	22.5	15.5	17.9
Fe ₂ O ₃ , percent	4.7	5.0	7.7	9.0
CaO, percent	3.7	28.0	20.9	9.6
SO ₃ , percent	0.4	2.3	1.5	0.9
MgO, percent	0.0	4.3	6.1	1.7
Alkalis, percent*	0.2	1.6	0.9	0.6
Loss on ignition, percent	3	0.3	0.4	0.4
Air permeability fineness, m ² /kg	403	393	329	256
Fineness, percent	18.2	17.0	21.6	23.8
Specific gravity	2.28	2.70	2.54	2.43

*Available alkalis expressed as Na₂O equivalent.

and water to form calcium silicate hydrate (C-S-H) and calcium aluminosilicate hydrates (Lothenbach et al. 2011).

The SiO₂ present in fly ash is due mainly to the clay minerals and quartz in the coal. Anthracite and bituminous (that is, high-rank) coals often contain a relatively greater percentage of clay minerals in their incombustible fraction as compared to subbituminous and lignite (that is, low-rank) coals. Therefore, the fly ash from the high-rank coals is richer in silica. The principal source of alumina (Al₂O₃) in fly ash is the clay in the coal, with some alumina coming from the organic compounds in low-rank coal. The types of clays found in coal belong to three groups of clay minerals: smectites, illites, and kaolinite.

Northern lignites—for example, lignite coal sources in North Dakota, Saskatchewan, and surrounding areas—typically contain smectite. Bituminous coal typically contains only members of the illite group and kaolinite. This difference in types of clay helps explain the lower Al₂O₃ in low-rank coal fly ash. From the alumina/silica ratios of smectite (0.35), illite (0.61), and kaolinite (0.85), it is clear why lignite

fly ashes usually contain 40 percent less analytic Al_2O_3 than bituminous fly ashes (Diamond and Lopez-Flores 1981a,b).

The Fe_2O_3 content of fly ash comes from the presence of iron-containing materials in the coal. The sizes of particles with highest concentrations of iron are typically in the range 30 to 60 μm , whereas the particles with a lower concentration of iron are typically smaller than 15 μm (Abad-Valle et al. 2011; Hower et al. 1999; Zyryanov et al. 2011).

The source of the materials reported as CaO in fly ash is primarily calcium carbonates and calcium sulfates in high-rank coal and from organic calcium compounds in low-rank coals. High-rank coals, such as anthracite and bituminous coal, contain smaller amounts of noncombustible materials, usually showing less than 5 percent CaO in the ash. Low-rank coals can produce fly ash with up to 35 percent CaO, depending on the geochemical character of the basin of coal deposition. The southern lignite coals found in Texas and Louisiana show the least CaO of the low-rank coals (for example, 10 to 15 percent).

The MgO in fly ash is derived from organic constituents and clay minerals, smectite or ferromagnesian minerals, and sometimes dolomite. Magnesium oxide is usually minimal in high-rank coals, but can exceed 7 percent in fly ashes from subbituminous and northern lignites, which are lignite coal sources in North Dakota, Saskatchewan, and surrounding areas. Southern lignites from Texas and Louisiana have MgO contents of less than 2 percent.

The SO_3 in fly ash from high-rank coal sources is primarily a result of pyrite (FeS_2) and, to a lesser degree, gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) present in the coal. The sulfur in low-rank coals comes primarily from organic compounds. The sulfur is released as sulfur dioxide gas (SO_2) and precipitates onto the fly ash or is scrubbed from the flue gases through a reaction with lime and alkali particles.

The alkalis in fly ash from high-rank coal come primarily from clay minerals. Alkalis in low-rank coals come primarily from sodium and potassium-bearing constituents in the coal. Alkali sulfates in northern lignite fly ash result from the combination of sodium and potassium with oxidized pyrite, organic sulfur, and gypsum in the coal. McCarthy et al. (1984, 1988) reported that Na_2O is found in greater amounts than K_2O in lignite and subbituminous fly ash, but the reverse is true of bituminous fly ash. Expressed as Na_2O equivalent (percent $\text{Na}_2\text{O} + 0.658 \times$ percent K_2O), alkali contents are typically less than 5 percent but may be as high as 10 percent in some high-calcium fly ashes.

The carbon content in fly ash is a result of incomplete combustion of the coal and any organic additives injected in the collection process, such as powdered activated carbon when introduced into the flue gas to control mercury (Hinzy et al. 2013). Carbon content is not usually determined directly, but is often assumed to be approximately equal to the LOI; however, LOI will also include any combined water or CO_2 lost by decomposition of hydrates or carbonates that are present in the fly ash. Fly ashes meeting the ASTM C618 specification are required to have less than 6.0 percent LOI. ASTM C618 does provide for the use of Class F fly ash with

up to 12.0 percent LOI, if either acceptable performance records or laboratory test results are made available.

The carbon produced by burning coal in a plant equipped with a low- NO_x burner is produced at somewhat cooler and much more reduced conditions (that is, lower oxygen) compared with traditional burners. The carbon associated with a low- NO_x fly ash is a more activated form than carbon produced using traditional burners. Therefore, low- NO_x carbon has a greater propensity to adsorb liquid chemical admixtures used in concrete, especially the air-entraining admixtures (AEAs). This can result in higher and more variable AEA dose requirements. Studies by Ley et al. (2008) have indicated that modification of the burning process, such as employing low- NO_x burners, may affect the interaction between the produced fly ash and AEA. Because small amounts of low- NO_x carbon can lead to relatively large increases in AEA in concrete, LOI may not be as useful in monitoring fly ash as tests based on measuring the adsorption potential of the fly ash or mortar air content.

Minor elements that may be present in fly ash include varying amounts of titanium, phosphorus, lead, mercury, chromium, and strontium (Flues et al. 2013; Haykiri-Acma et al. 2011; Hower et al. 2013; Li et al. 2012; Shah et al. 2012; Vassilev et al. 2000). Some fly ashes also have trace amounts of organic compounds other than unburned coal. These additional compounds, such as ammonia, are usually from NO_x reduction systems or precipitator conditioning additives and are discussed in 1.3.2.

Table 3.2 gives examples of North American fly ash bulk chemical composition for different coal sources. Other references that provide detailed chemical composition data are also available (Bayat 1998; Berry and Hemmings 1983; Chancey et al. 2010; Das and Yudhbir 2006; Hooton 1986; Hower et al. 1996; Levandowski and Kalkreuth 2009; Du et al. 2013; Liu et al. 2013; McCarthy et al. 1984; Nathan et al. 1999; Pietersen et al. 1992; Pipatmanomai et al. 2009; Sakorafa et al. 1996; Shehata et al. 1999; Sutter et al. 2013b; Tang et al. 2013; Tikalsky et al. 1992; Tishmack 1996; Tsubouchi et al. 2011; Venkateswaran et al. 2003; Williams et al. 2005).

3.3—Crystalline constituents

From the bulk elemental composition of fly ash, a division can be made between the phases in which these chemical compounds exist in fly ash. Developments in the techniques of quantitative X-ray diffraction (XRD) analysis have made it possible to determine the approximate amounts of crystalline phases and amorphous contents in fly ash (Mings et al. 1983; Pitt and Demirel 1983; McCarthy et al. 1988).

Low-calcium fly ashes contain relatively inactive crystalline phases: quartz, mullite, ferrite spinel, and hematite (Diamond and Lopez-Flores 1981a; Sutter et al. 2013a). High-calcium fly ash can contain the previously mentioned phases and may also contain additional crystalline phases such as anhydrite, alkali sulfate, dicalcium silicate, tricalcium aluminate, free calcium oxide, melilite, gehlenite, åkermanite solid solution, merwinite, merwinite-åkermanite, and ye'elimite (McCarthy et al. 1984; Sutter et al. 2013a).

Table 3.3a—Mineralogical phases in fly ash

Mineral name	Chemical composition
Thenardite	(Na,K) ₂ SO ₄
Anhydrite	CaSO ₄
Tricalcium aluminate (C ₃ A)	Ca ₃ Al ₂ O ₆
Dicalcium silicate (C ₂ S)	Ca ₂ SiO ₄
Hematite	Fe ₂ O ₃
Lime	CaO
Melilite	Ca ₂ (Mg,Al)(Al,Si) ₂ O ₇
Merwinite	Ca ₃ Mg(SiO ₄) ₂
Mullite	Al ₆ Si ₂ O ₁₃
Periclase	MgO
Quartz	SiO ₂
Sodalite structures	Na ₈ Al ₈ Si ₆ O ₂₄ SO ₄ Na ₆ Ca ₂ Al ₆ Si ₆ O ₂₄ (SO ₄) ₂ Ca ₈ Al ₁₂ O ₂₄ (SO ₄) ₂
Ferrite spinel	Fe ₃ O ₄
Portlandite	Ca(OH) ₂
Ye'elimite	Ca ₄ Al ₆ (SO ₄)O ₁₂

Some of these additional phases (for example, tricalcium aluminate) found in Class C fly ash are hydraulic, producing cementitious materials in the presence of water, explaining why Class C fly ash exhibits both cementitious and pozzolanic properties. Excessive amounts of the C₃A and CaO compounds can also contribute to rapid set and high water-demand characteristics, which may affect plastic shrinkage. A list of crystalline mineral compounds found in fly ash is given in Table 3.3a.

Alpha quartz, or crystalline silica (SiO₂), is present in all fly ashes. This silica is a result of the quartz content in the raw coal that failed to melt during combustion. Quartz is typically the most intense peak in the XRD pattern from the fly ash.

Mullite (3Al₂O₃·2SiO₂), which is a crystalline aluminosilicate, is found in substantial quantities only in low-calcium fly ashes (Gomes and Francois 2000). Mullite forms within the glass spheres as they solidify around it. Mullite accounts for most of the alumina in fly ash but is not normally chemically reactive in concrete.

In its purest form, magnetite (Fe₃O₄) is the crystalline spinel structure closest to that found in fly ash. A shift in the XRD spacing from that of pure magnetite indicates Mg and Al substitution in the ferrite spinel structure (Gomes et al. 1999; Tevenson and Huber 1986). The ferrite spinel phase found in fly ash is not chemically active. Hematite (Fe₂O₃) can be formed by the oxidation of limonite, siderite, or magnetite and is present in some fly ashes, though it is not chemically active.

Coal fly ashes containing high calcium contents often contain between 1 and 3 percent by mass anhydrite (CaSO₄). The calcium acts as a scrubber for SO₂ in the combustion gases and forms anhydrite. Crystalline CaO (free lime) is present in most high-calcium fly ashes and may be a cause of

autoclave expansion. Lime in the form of Ca(OH)₂ (slaked lime), however, does not contribute to autoclave expansion. Soft-burned CaO hydrates quickly and does not result in unsoundness in concrete. However, hard-burned CaO formed at higher temperatures hydrates slowly after the concrete has hardened. Demirel et al. (1983) hypothesized the carbon dioxide-rich environment of the combustion gases causes a carbonate coating to form on poorly burned CaO particles, creating a diffusion barrier that retards the hydration of the particle and thereby increases the potential for unsoundness. If free lime is present as highly sintered, hard-burned material, there is a potential for long-term deleterious expansion from its hydration. Although there is no direct way to separate soft-burned lime from the sintered lime, McCarthy et al. (1984) noted that when hard-burned lime is present, it is often found in the larger grains of fly ash. If there is sufficient hard-burned CaO to cause unsoundness, it can be detected as excessive autoclave expansion. Ca(OH)₂ is also present in some high-calcium fly ashes that have been exposed to moisture.

Crystalline MgO (periclase) is found in fly ashes with more than 2 percent MgO. Fly ash from low-rank coals can contain periclase contents as high as 80 percent of the MgO content. The periclase in fly ash is not free MgO like that found in some portland cements. Rather, the crystalline MgO in fly ash is similar to the phase of MgO found in slag cement and is nonreactive in water or basic solutions at normal temperatures (Locher 1960).

Phases belonging to the melilite group include:

- Gehlenite Ca₂Al(AlSiO₇)
- Akermanite Ca₂Mg(Si₂O₇)

These phases have been detected in fly ash but are not chemically active in concrete. Each of these phases can have Fe substituted for Mg or Al. Merwinite is a common phase in high-calcium fly ash and in the early stages of the devitrification of Mg-containing glasses. Northern lignites typically have higher MgO contents and lower Al₂O₃ contents than subbituminous-coal fly ash, allowing the merwinite phase to dominate over the calcium aluminate phase in the northern lignite fly ash. Merwinite is nonreactive at normal temperatures.

The presence of calcium aluminate in high-calcium fly ash was confirmed by Diamond (1981a) and others. The intense XRD peaks of this phase overlap those of the merwinite phase, making the quantitative interpretation difficult. McCarthy et al. (1988), however, reported the calcium aluminate phase is the dominant phase in fly ash with subbituminous coal sources, and the merwinite phase is dominant in lignite fly ashes. Neither phase is present in low-calcium fly ash. The cementitious value of calcium aluminate contributes to the self-cementitious property of high-calcium fly ashes. The calcium aluminate phase is extremely reactive in the presence of calcium and sulfate ions in solution.

Phases belonging to the sodalite group, which are formed from melts rich in alkalis and calcium, have a low silica content. Nosean and Hauyne phases have been identified in fly ash by McCarthy et al. (1988). Some researchers have found ye'elimite (Ca₄Al₆(SO₄)O₁₂) in Class C fly ash, the

Table 3.3b—Qualitative XRD test results for 18 separate Class F fly ashes

ID	Sum of oxides, % mass	Major phase(s)	Minor phase(s)	Other phase(s)
FA-A	92.52	α -quartz, mullite	—	—
FA-C	88.99	α -quartz	Mullite, ferrite spinel	Hematite*
FA-E	87.76	α -quartz	Mullite, hematite, anhydrite	Traces of lime, periclase and ferrite spinel*
FA-F	90.62	α -quartz, mullite	Ferrite spinel	Hematite*
FA-G	89.89	α -quartz, mullite	Ferrite spinel, hematite	—
FA-H	91.26	α -quartz, mullite	Lime	Ferrite spinel*
FA-I	89.45	α -quartz	Mullite, ferrite spinel	Trace anhydrite*
FA-J	91.90	α -quartz, mullite, hematite	Ferrite spinel	Trace lime*
FA-K	83.50	α -quartz, mullite, hematite	Anhydrite, ferrite spinel	Trace anorthite, lime*
FA-L	84.80	α -quartz, mullite, hematite, ferrite spinel	Anhydrite, portlandite	Lime
FA-M	81.85	α -quartz	Mullite, ferrite spinel, hematite	Trace anhydrite, trace lime, trace periclase*
FA-N	86.90	α -quartz, mullite, hematite, ferrite spinel	Lime, anhydrite	—
FA-O	79.81	α -quartz	Mullite, lime, periclase, hematite	Trace ferrite spinel,* trace portlandite
FA-P	73.34	α -quartz	Lime, periclase, anhydrite	Trace hematite, trace ferrite spinel, trace mullite,* trace melilite,* trace C ₃ A*
FA-Q	74.34	α -quartz	Mullite, lime, periclase, anhydrite, ferrite spinel	trace melilite,* trace C ₃ A*
FA-R	73.27	α -quartz	Ferrite spinel, hematite, anhydrite, periclase	Trace lime, trace merwinite*
FA-S	70.55	α -quartz	Mullite, anhydrite, lime, periclase, ferrite spinel,	C ₃ A,* C ₄ AF,* trace hematite*
FA-T	77.41	α -quartz	Mullite, anhydrite, hematite, ferrite spinel	Trace periclase,* merwinite,* anorthite*

*Indicates likely but not absolutely confirmed due to low-intensity profiles—for example, trace phases or convoluted profiles.

Note: Sum of the oxides is the sum of the SiO₂, Al₂O₃, and Fe₂O₃, expressed in percent mass (Sutter et al. 2013a).

active constituent of Type K expansive cement. Ye'elimite reacts readily with water, lime, and sulfate to form ettringite (Winburn et al. 2000).

Among the other phases found in fly ash are alkali sulfates and possibly dicalcium silicate. Dicalcium silicate is a crystalline phase that is present in some high-calcium fly ashes. Northern lignite fly ash often contains crystalline alkali sulfates such as thenardite and apththilite.

Tishmack et al. (1999) investigated high-calcium Class C fly ashes derived from Powder River Basin coal, which contain significant amounts of sulfur, calcium, and aluminum, and thus are a potential source of ettringite in concrete. Hydration products of fly ash water pastes contain ettringite, monosulfate, and strätlingite (Bae et al. 2014). Portland cement/fly ash pastes were found to contain calcium hydroxide, ettringite, monosulfate, and smaller amounts of hemicarboaluminate and monocarboaluminate. The portland cement/fly ash pastes generally formed less ettringite than did the control cement pastes, but formed more of the monosulfate phases. Sutter et al. (2013a) performed a qualitative XRD study of 30 different fly ash sources: 18 Class F and 12 Class C. The ash sources in their study represented a broad range of physical and chemical characteristics, as well as geographic representation of the United States. The results

from this study are summarized in Tables 3.3b and 3.3c. Both tables are organized in order of decreasing value of the sum of the oxides—for example, increasing CaO content. As can be seen in these tables, in general, the mineralogy of the coal fly ash samples becomes more complex as the CaO increases. In the same study, four Class F and four Class C fly ash sources were selected for quantitative X-ray diffraction (QXRD) analysis separately using Rietveld analysis and the relative intensity ratio (RIR) method, which is based on the work of Klug and Alexander (1954). The results are shown in Tables 3.3d and 3.3e. When comparing the two analytical approaches, there was generally good agreement for the major phases, although minor phases showed differences between the two analysis methods. To validate application of the Rietveld analysis method for quantitative XRD analysis of fly ashes, Winburn et al. (2000) performed testing with a set of standard mixtures.

3.4—Glassy constituents

Fly ash consists largely of small glassy spheres that form while the burned coal residue cools very rapidly. The composition of these glasses depends on the composition of the pulverized coal and the temperature at which it is burned. Fly ash reactivity is strongly affected by the glass content

Table 3.3c—Qualitative XRD test results for 12 separate Class C fly ashes

ID	Sum of oxides, % mass	Major phase(s)	Minor phase(s)	Other phase(s)
FA-B	67.62	α -quartz	Anhydrite, lime, periclase, ferrite spinel, hematite	Trace mullite, trace thenardite,* trace melilite*
FA-U	65.80	α -quartz	Lime, periclase, ferrite spinel, C ₃ A	Trace mullite, trace melilite
FA-V	63.00	α -quartz	Lime, periclase, ferrite spinel, C ₃ A, melilite	Trace mullite, C ₄ AF,* hematite*
FA-W	62.83	α -quartz	Anhydrite, melilite, periclase, ferrite spinel,	Trace lime, merwinite or C ₃ A,* hematite*
FA-X	61.63	α -quartz	Periclase, thenardite, melilite, ferrite spinel	Trace anhydrite, trace lime, merwinite or C ₃ A,* trace mullite*
FA-Y	62.77	α -quartz	Lime, periclase, C ₃ A, anhydrite, melilite, mullite	Ferrite spinel
FA-Z	61.21	α -quartz	Lime, periclase, anhydrite, melilite, ferrite spinel	merwinite or C ₃ A,* trace mullite, trace hematite, trace ye'elimite*
FA-ZA	55.32	α -quartz, C ₃ A, lime, periclase	Melilite, ferrite spinel, C ₄ AF	Trace thenardite, trace hematite,* trace ye'elimite*
FA-ZB	61.66	α -quartz	C ₃ A, anhydrite, lime, periclase, melilite	Trace mullite, trace ferrite spinel, hematite, trace ye'elimite,* C ₄ AF*
FA-ZC	53.09	α -quartz, C ₃ A, lime, periclase	Anhydrite, melilite, C ₄ AF, ye'elimite	Ferrite spinel, trace hematite*
FA-ZD	54.27	α -quartz, C ₃ A, lime, periclase, anhydrite	Melilite, ferrite spinel, C ₄ AF	Ye'elimite
FA-ZL	61.52	α -quartz	Lime, periclase, ferrite spinel, Na-K sulfate	Trace anhydrite, melilite,* merwinite or C ₃ A,* hematite*

*Indicates likely but not absolutely confirmed due to low-intensity profiles (for example, trace phases) or convoluted profiles.

Note: Sum of the oxides is the sum of the SiO₂, Al₂O₃, and Fe₂O₃, expressed in percent mass (Sutter et al. 2013a).

Table 3.3d—Summary of QXRD test results for the Class F fly ashes

Rietveld method	FA-H	FA-M	FA-O	FA-Q
Quartz	10.8	16.1	13.4	11.5
Mullite	15.8	1.6	3.2	4.0
Hematite	0.5	0.6	1.0	BQL
Magnetite	0.2	0.6	0.2	BQL
Anhydrite	ND	0.4	0.7	0.8
Lime	BQL	BQL	0.6	0.2
Periclase	ND	ND	1.0	1.2
Portlandite	ND	ND	0.6	ND
C ₃ A	ND	ND	ND	2.8
Glass	73	81	79	80
RIR method	FA-H	FA-M	FA-O	FA-Q
Quartz	11.6	16.0	15.5	12.0
Mullite	14.4	3.2	3.8	3.6
Hematite	BQL	0.7	1.1	BQL
Magnetite	BQL	0.5	BQL	BQL
Anhydrite	ND	0.4	0.6	0.6
Lime	BQL	BQL	0.7	0.4
Periclase	ND	ND	0.8	0.6
Portlandite	ND	ND	BQL	ND
C ₃ A	ND	ND	ND	2.0
Glass	74	79	78	81

Notes: Results in percent by mass (Sutter et al. 2013a). BQL: below quantity limit, and ND: not detected.

and glass composition. The major differences in fly ash glass composition lie in the amount of calcium present in the glass. Coal that contains relatively small amounts of calcium—for example, anthracite and bituminous or some lignite coals—results in aluminosilicate glassy fly ash particles. Subbituminous and some lignite coals contain larger amounts of calcium and produce calcium aluminosilicate glassy phases in the fly ash (Roy et al. 1984). This can be seen in the ternary system diagram shown in Fig. 3.4. The normalized average glass composition of high-calcium fly ash falls within the ranges where anorthite to gehlenite are the first phases to crystallize from a melt, whereas the low-calcium fly ashes fall within the regions of the diagram where mullite is the primary crystalline phase. The disordered structure of a glass resembles that of the primary crystallization phase that forms on cooling from the melt. In fly ash, the molten silica is accompanied by other molten oxides. As the melt is quenched, these additional oxides create added disorder in the silica glass network. The greater the disorder and depolymerization of the fly ash glass structure, the less stable the network becomes.

To conceptualize the composition of the glass phase in a fly ash, the mass of crystalline compounds is subtracted from the bulk mass to yield the mass of the glassy portion of the fly ash. Extending this analysis to chemical compounds, the crystalline composition can be stoichiometrically subtracted from the bulk chemical composition to yield an average composition of the glass for any given fly ash. This is important when considering the level of reactivity of a fly ash. Das and Yuthbir (2006) used the pozzolanic potential

Table 3.3e—Summary of QXRD test results for the Class C fly ashes

Rietveld method	FA-U	FA-X	FA-ZA	FA-ZC
Quartz	5.4	3.1	3.1	5.5
Mullite	3.5	ND	ND	ND
Hematite	ND	ND	ND	ND
Magnetite	0.3	0.2	BQL	BQL
Anhydrite	0.7	0.5	2.0	2.6
Lime	0.2	BQL	0.7	1.9
Periclase	1.1	1.3	2.5	4.1
Gehlenite	0.4	1.1	0.5	1.3
C ₃ A	3.6	1.7	4.9	8.2
C ₄ AF	BQL	ND	2.4	2.8
Thenardite	ND	0.7	0.6	BQL
Ye'elimite	ND	ND	BQL	1.6
Glass	85	91	83	72
RIR method	FA-U	FA-X	FA-ZA	FA-ZC
Quartz	5.0	2.4	2.6	5.2
Mullite	2.1	ND	ND	ND
Hematite	ND	ND	ND	ND
Magnetite	BQL	BQL	BQL	BQL
Anhydrite	0.4	0.2	1.5	1.9
Lime	0.4	0.2	0.9	2.3
Periclase	1.0	0.9	2.1	3.7
Gehlenite	BQL	1.8	1.6	3.0
C ₃ A	3.3	1.6	6.1	9.6
C ₄ AF	ND	ND	2.9	3.8
Thenardite	ND	2.2	0.9	ND
Ye'elimite	ND	ND	BQL	1.1
Glass	88	91	82	69

Notes: Results in percent by mass (Sutter et al. 2013a). BQL: below quantity limit, and ND: not detected.

index (10 times the mole ratio of K_2O/Al_2O_3) of Hubbard et al. (1985) to estimate the glass content of fly ash. Fly ash glass content has been successfully determined by XRD (Ibanez et al. 2013; Ward and French 2005). The compositions of glassy and crystalline components of fly ashes have also been investigated using scanning electron microscopy (Kutchko and Kim 2006; Chancey et al. 2010; Aughenbaugh et al. 2013) and a combination of three-dimensional X-ray computer tomography and electron probe microanalysis techniques (Hu et al. 2014).

Additional discussions on the glass phases existing in fly ash can be found in Aughenbaugh et al. (2013), Chatterjee (2011), Hemmings and Berry (1988), Hu et al. (2014), Kutchko and Kim (2006), Pietersen (1993), Valentim et al. (2009), Chancey et al. (2010), and Zhu et al. (2013).

3.5—Physical properties

The shape, size, particle-size distribution, and density of fly ash particles influence the properties of freshly mixed, plastic concrete, and the strength development and other

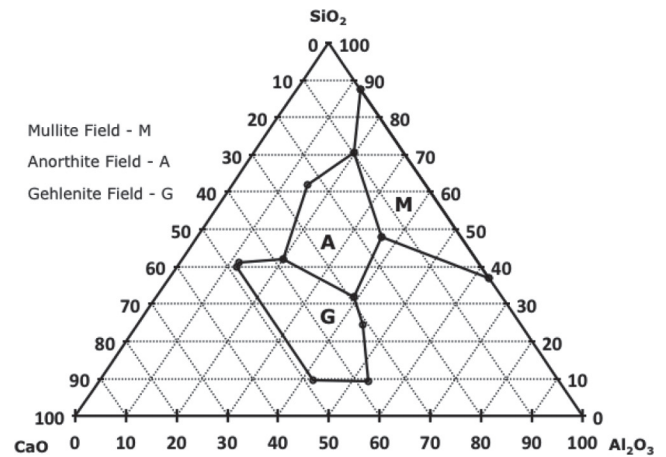


Fig. 3.4—CaO-SiO₂-Al₂O₃ ternary system diagram.

properties of hardened concrete. This is due, in part, to the influence of particle characteristics on the water demand of the concrete mixture. The properties of a fly ash produced at a particular power plant can vary from another fly ash originating from a different source. This is likely to be the case if the power plants are burning different coals or operate using different combustion systems. Plants operating under similar combustion regimes can produce fly ash with very similar chemical and physical characteristics. In addition, fly ashes produced at different power plants, or at one plant using different coal sources or different combustion conditions, may have different colors.

Fly ash color and the amount used can influence the color of the resulting hardened concrete in the same way as changes in cement or fine aggregate color. Fly ash color is not an engineering concern unless aesthetic considerations relating to the concrete require maintaining a uniform color in exposed concrete. A change in the color of a fly ash from a particular source can indicate changes in coal source, carbon content, iron content, or burning conditions and, therefore, changed properties. Refer to Fig. 3.5 for examples of the range in color seen with different fly ash sources.

3.5.1 Particle shape—Particle size and shape characteristics of fly ash depend on the source and uniformity of the coal, the degree of pulverization before burning, the combustion environment (temperature level and oxygen supply), uniformity of combustion, and the type of collection system used (mechanical separators, baghouse filters, or electrostatic precipitators). Lane and Best (1982) reported the shape of fly ash particles is also a function of particle size. The majority of fly ash particles are glassy, solid, or hollow, and spherical in shape as singles or clusters. Examples of fly ash particle shapes are shown in Fig. 3.5.1a and 3.5.1b. Fly ash particles that are hollow are translucent to opaque, slightly to highly porous, and vary in shape from rounded to elongated. The intergrinding of fly ash with portland cement clinker in the production of blended cement has improved its contribution to strength (Diamond 1982). Grinding further reduces particle size, breaks up cenospheres, and separates particles that have surface attachments.

			
CaO = 3.5% LOI = 0.25%	CaO = 5.3% LOI = 0.80%	CaO = 2.3% LOI = 1.22%	CaO = 1.5% LOI = 3.32%
			
CaO = 14.0% LOI = 0.13%	CaO = 13.6% LOI = 0.45%	CaO = 14.3% LOI = 1.19%	CaO = 3.0% LOI = 21.34%
			Examples of Fly Ash Color
CaO = 29.2% LOI = 0.20%	CaO = 24.2% LOI = 0.33%	CaO = 24.8% LOI = 0.50%	

Fig. 3.5—Examples of the range in color seen between different fly ash sources.

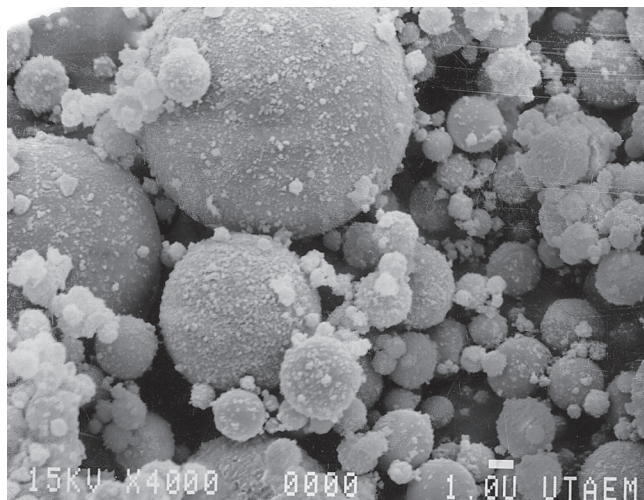


Fig 3.5.1a—Fly ash at 4000× magnification.

3.5.2 Fineness—Individual particles in fly ash may range in size from less than 1 μm to greater than 0.04 in. (1 mm). A coarse fly ash often contains more plerospheres, which are porous particles with surface openings, and are typically filled with smaller particles (Fig. 3.5.1b). Fly ash of 5 to 30 micron particle size is highly reactive compared to coarser fly ash (Malhotra and Mehta 2002). In older plants where

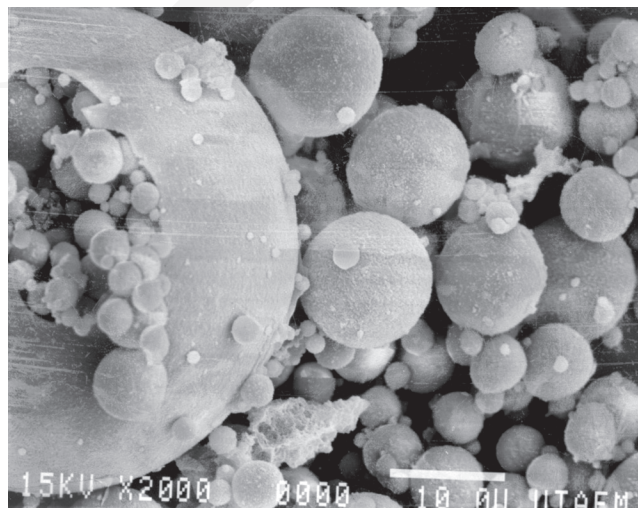


Fig 3.5.1b—Fly ash showing plerospheres at 2000× magnification.

mechanical separators are used, the fly ash is coarser than in more modern plants that use electrostatic precipitators or bag filters. Fly ash meeting ASTM C618 should not have more than 34 percent of the particles retained on the No. 325 (45 μm) sieve. The No. 325 (45 μm) sieve analysis of fly ash from a particular source will normally remain relatively constant,