

**Session I:**

**COAL COMBUSTION BY-PRODUCTS  
CHARACTERIZATION**

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# LEACHING TESTS: COMMONLY USED METHODS, EXAMPLES OF APPLICATIONS TO COAL COMBUSTION BY-PRODUCTS, AND NEEDS FOR THE NEXT GENERATION

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## Abstract

There are a variety of tests available for characterizing the leaching behavior of waste materials. These vary in test type, leaching aspects that are addressed, and the particular use for which the test is designed. Types of leaching tests include agitated extraction tests, serial batch tests, flow-through tests, flow-around tests, etc. These tests address different aspects of leaching, such as the physical mechanisms involved, chemical interactions between the waste and the leaching fluid, the kinetics of leaching, leaching as a function of time, etc. Some of the uses of leaching tests include regulatory purposes, scientific research, and environmental assessment. This paper discusses some of the commonly used leaching methods, results of specific application of some of these to coal combustion by-products, and what is needed for the next generation of test methods to more accurately assess the release of contaminants into the environment.

## Introduction

There are a variety of leaching tests available for application to waste materials. A leaching test involves contacting the waste material with a liquid to determine which constituents will be leached by the liquid and released to the environment. Leaching methods vary in test type, leaching aspects addressed, and the particular use for which the test is designed. Types of leaching tests include agitated extraction tests, serial batch tests, flow-through tests, flow-around tests, etc. These tests address different aspects of leaching, such as the physical mechanisms involved, chemical interactions between the waste and the leaching fluid, the kinetics of leaching, leaching as a function of time, etc. Some of the uses of leaching tests include regulatory purposes, environmental impact assessment, scientific studies, and waste management. Commonly used leaching procedures in the United States include those developed by the U.S. Environmental Protection Agency (EPA) and the American Society for Testing and Materials (ASTM). In addition, there are a number of leaching methods commonly being used in Canada and the European countries.

The purpose of this paper is to discuss (1) some of the commonly used leaching methods, (2) results of specific application of some of these to coal combustion by-products, and (3) what is needed for the next generation of test methods to more accurately assess the release of contaminants into the environment.

## Commonly Used Leaching Procedures

A number of leaching procedures have been developed in the United States over the last two decades. The U.S. EPA and ASTM Committee D-34 on Waste Management have been primarily responsible for these method development activities. A brief summary of EPA and ASTM leaching methods is given below. State regulatory agencies have adopted variations of these methods; however, discussion of the state methods is beyond the scope of this paper.

### EPA Methods

*Toxicity Characteristic Leaching Procedure (TCLP) (U.S. EPA Method 1311):* The TCLP (U.S. EPA 1990c) is designed to simulate the leaching a waste will undergo if disposed in an unlined sanitary landfill. It is based on a co-disposal scenario of 95% municipal waste and 5% industrial waste. The method is an agitated extraction test using leaching fluid that is a function of the alkalinity of the solid

phase of the waste. Either a sodium acetate buffer solution having a pH of  $4.93 \pm 0.05$  or an acetic acid solution having a pH of  $2.88 \pm 0.05$  is used. The procedure requires particle size reduction to less than 9.5 mm. The TCLP is designed to determine the mobility of 40 Toxicity Characteristic (TC) constituents in liquid, solid, and multiphase wastes. The TC constituents include both inorganic and organic species. Leachability of volatile organic compounds is determined using a zero-headspace extractor and the sodium acetate buffer solution.

The TCLP was developed in 1984 under the Hazardous and Solid Waste Amendments to the Resource Conservation and Recovery Act (RCRA) (1984) and is the U.S. EPA regulatory method for classifying wastes as hazardous or nonhazardous based on toxicity. If the TCLP extract contains any one of the TC constituents in an amount equal to or greater than the concentrations specified in 40 CFR 261.24 (1992), the waste possesses the characteristic of toxicity and is a hazardous waste.

*Synthetic Precipitation Leaching Procedure (SPLP) (U.S. EPA Method 1312):* The SPLP (U.S. EPA 1990a), which is an agitated extraction method, was developed in 1988 by the U.S. EPA for use in evaluating the impact that contaminated soils may have on groundwater. The procedure calls for use of simulated acid rain or reagent water as the extraction fluid, depending on the constituents of interest. The extraction fluid used to determine the leachability of soil from a site that is east of the Mississippi River is a solution of sulfuric and nitric acids in reagent water having a pH of  $4.2 \pm 0.05$ . This extraction fluid is also used to extract waste or wastewater using the procedure. The extraction fluid used to determine the leachability of soil from a site that is west of the Mississippi River is a solution of sulfuric and nitric acids in reagent water having a pH of  $5.0 \pm 0.05$ . When the leachability of volatile organic compounds or cyanide is being evaluated, reagent water is used as the extraction fluid. The procedure requires particle size reduction to less than 9.5 mm, and as with the TCLP, extraction for volatile constituents is performed in a zero-headspace extractor.

*Multiple Extraction Procedure (MEP) (U.S. EPA Method 1320):* The MEP (U.S. EPA 1990b) is designed to simulate the leaching a waste will undergo from repeated precipitation of acid rain on an improperly designed sanitary landfill. The repetitive extractions are to reveal the highest concentration of each constituent that is likely to leach in a natural landfill (U.S. EPA 1990b).

The MEP is a serial batch test in which the first extraction is performed according to the Extraction Procedure (EP) Toxicity Test (U.S. EPA 1980). The EP is the U.S. EPA regulatory method that was replaced by the TCLP for determining the characteristic of toxicity. It is a leaching procedure that involves monitoring the pH of the waste in reagent water and using an acetic acid solution to maintain the pH of the slurry at  $5.0 \pm 0.2$ . After the first extraction in the MEP is completed, the remaining solid is re-extracted nine times (or more) using synthetic acid rain as the leaching fluid. If the concentration of any constituent of concern increases from the 7th or 8th extraction to the 9th extraction, the procedure is repeated until the concentrations decrease. The synthetic acid rain leaching fluid is prepared by adding the appropriate amount of a 60/40 weight percent sulfuric acid and nitric acid mixture to distilled, deionized water to give a pH of  $3.0 \pm 0.2$ . The method is applicable to liquid, solid, and multiphase materials.

#### ASTM Standard Methods

*ASTM Method D-3987, Standard Test Method for Shake Extraction of Solid Waste with Water (ASTM 1995a):* This method is a procedure for rapidly generating a leachate from solid waste that can be used to estimate the mobility of inorganic constituents from the waste under the specified test conditions (ASTM 1995a). The final pH of the leachate is to reflect the interaction of the leaching fluid with the buffering capacity of the waste. In the method, it is stated that the procedure is not intended to produce a leachate representative of leachate generated in the field, and the method does not simulate site-specific leaching conditions (ASTM 1995a).

ASTM Method D-3987 is an agitated extraction method that uses reagent water as the leaching fluid. The procedure involves an 18-hour contact time between a solid waste and reagent water with rotary agitation. The method calls for testing a representative sample of the waste, and as a result, it does not require particle size reduction. The method has been tested to determine its applicability to inorganic constituents, but it has not been tested for application to organic constituents.

*ASTM Method D-4793, Standard Test Method for Sequential Batch Extraction of Waste with Water (ASTM 1995b):* This method is a procedure for obtaining serial leachates of a waste that can be used to estimate the mobility of inorganic constituents from the waste under the specified test conditions (ASTM 1995b). The final pH of the leachate is intended to reflect the interaction of the leaching fluid with the buffering capacity of the waste (ASTM 1995b). Similar to ASTM Method D-3987, this method also contains statements that it is not intended to produce leachates representative of leachate generated in the field, and that it does not simulate site-specific conditions (ASTM 1995b).

ASTM Method D-4793 is a serial batch test that uses reagent water as the leaching fluid. The method has been tested for its applicability to inorganic constituents only and can be used to test any waste containing at least five percent solids. The leaching steps are repeated so that ten leachates are generated.

*ASTM Method D-5284, Standard Test Method for Sequential Batch Extraction of Waste with Acidic Extraction Fluid (ASTM 1995e):* This method is a modification of ASTM Method D-4793. It calls for use of a leaching fluid having a pH that reflects the pH of acidic precipitation in the geographic region in which the waste being tested is to be disposed (ASTM 1995e). The leaching fluid is prepared by mixing a 60/40 weight percent mixture of sulfuric acid/nitric acid with reagent water. This method has only been tested for its use in leaching inorganic constituents. The information given in the method concerning its intent and limitations is the same as described above for ASTM Method D-4793.

*ASTM Method D-5233, Standard Test Method for Single Batch Extraction Method for Wastes (ASTM 1995d):* This is an agitated extraction test that is very similar to TCLP. The major difference between the two procedures is that ASTM Method D-5233 does not require particle size reduction. The method states that it is applicable for leaching samples of treated or untreated solid wastes or sludges, or solidified waste samples, to provide an indication of the leaching potential (ASTM 1995d). This method is based on the same disposal scenario as TCLP, a sanitary landfill with co-disposal of 95% municipal waste and 5% industrial waste. According to the method, interpretation and use of the test results are limited by the assumption of a single co-disposal scenario and by differences between the extraction fluid used in the method and the real landfill leachate (ASTM 1995d).

*ASTM Method D-4874, Standard Test Method for Leaching Solid Waste in a Column Apparatus (ASTM 1995c):* This is a column method using reagent water in a continuous up-flow mode to generate aqueous leachate from waste materials. The current published version of the method (ASTM 1995c) is applicable for evaluating the leachability of inorganic constituents. However, a revised method will be published in 1996 (ASTM 1996) that can be used to evaluate the leachability of semivolatile and nonvolatile organic compounds, as well as inorganic constituents. The column method is intended to provide aqueous leaching in a dynamic partitioning manner (ASTM 1995c). It is written so that many of the specific column operating conditions can be selected by the user to meet their specific objectives (ASTM 1995c). The method states that analysis of the column effluent can provide information on the leaching characteristics of the waste under the testing conditions used (ASTM 1995c). It is also stated that the method is not intended to produce results to be used as the sole basis for (1) the engineering design of a landfill disposal site or (2) classification of wastes based on leaching characteristics (ASTM 1995c).

There are also a number of leaching methods that are being used in Canada and the European countries. Some of the commonly used international procedures are described below.

## Canadian Methods

*Leachate Extraction Procedure (LEP):* The LEP (Ministry of the Environment 1985) is an agitated extraction test that is the regulatory leaching test used in the province of Ontario, Canada. In addition, this method is identical to the regulatory leaching procedure used by the Canadian provinces of British Columbia, Alberta, and Manitoba (Environment Canada 1990). The test is very similar to the EP Toxicity Test. As previously mentioned, the EP involves monitoring the pH of the waste in reagent water and using acetic acid solution to maintain the pH of the slurry at  $5.0 \pm 0.2$ . The EP is based on a single co-disposal scenario of industrial waste in a municipal landfill (U.S. EPA 1980).

*Quebec R.s.Q. (Q.R.s.Q.):* The Q.R.s.Q. (Ministere de L' Environnement 1985) is an agitated extraction test that is the regulatory leaching method used in the province of Quebec, Canada. It is very similar to the TCLP, with some procedural differences (Environment Canada 1990).

## European Methods

*Availability Test (NEN 7341):* The availability test (Netherlands Normalization Institute 1993a) is an agitated extraction method that was developed in the Netherlands. It is based on leaching a finely ground sample under two controlled pH conditions, pH = 4 and pH = 7. The purpose of the test is to indicate the quantity of an element that may be leached from a material under environmentally extreme conditions, e.g. in the very long term, after disintegration of the material, when the material is fully oxidized, and with complete loss of acid neutralization capacity (de Groot and Hoede 1994).

*Column Test (NEN 7343):* The NEN 7343 column test (Netherlands Normalization Institute 1993b) is believed to simulate the leaching behavior of a waste material in the short, medium, and long term (van der Sloot 1995). This is done by relating contaminant release, expressed as mg/Kg leached, to the liquid-to-solid ratio (van der Sloot 1995). The time scale relation is obtained from the height of the column and rate of infiltration (van der Sloot 1995). The procedure involves passing demineralized water (pH = 4) upward through ground material (95% < 4 mm). Seven consecutive leachate fractions are collected, corresponding to a liquid-to-solid ratio range of 0.1 to 10 L/Kg (de Groot and Hoede 1994). van der Sloot (1995) states that very slow changes in mineral composition are not addressed by this test; and NEN 7343 test conditions do not correlate on a one-to-one basis with field conditions because several variables, such as temperature, channeling, aging, and degree and length of contact, must be considered.

*French Leach Test (Agence Francaise de Normulisation 1987):* This is an agitated extraction test that is a French Ministry of the Environment standard regulatory method for determining the soluble fraction of a solid waste in an aqueous solution (Environment Canada 1990). It requires particle size reduction and involves mechanical stirring of the sample with water in a 10:1 liquid-to-solid ratio for 16 hours. A second and third extraction are performed, and the cumulative extracted soluble fraction for each constituent can be determined.

*Germun Leach Test (DIN 38414 S4) (Institut fur Normung 1984):* This is the leaching test that is generally used in Germany (Rankers and Hohberg 1991). It is an agitated extraction test involving table shaking of the extraction slurry for 24 hours. The liquid-to-solid ratio is 10: 1. The method is applicable to solids, pastes, and sludges (van der Sloot 1995). Advantages of the method are its rapidity, extremely good reproducibility, and simplicity. However, the method is criticized for its use of a 10: 1 liquid-to-solid ratio, which is rarely reached under landfill conditions (Rankers and Hohberg 199 1).

*TVA (TVA 1988):* This agitated extraction test is the standard regulatory method used in Switzerland. The method calls for 100 to 200 grams of sample to be extracted using carbon dioxide-saturated water at a liquid-to-solid ratio of 10:1 for 24 hours. The continuous carbon dioxide injection is considered to represent a time-scale reduction (van der Sloot 1995).

## **Examples of Applications of Leaching Tests to Coal Combustion By-Products**

Several studies have been conducted in which coal combustion by-products have been tested by a variety of leaching methods. Listed below are just a few examples of studies involving leaching of coal combustion by-products using some of the specific leaching methods described above.

### TCLP Applied to Utility Wastes

Following proposal of the TCLP to replace the EP for hazardous-waste classification, the Electric Power Research Institute (EPRI) funded a study to evaluate application of the TCLP to utility wastes (Jackson and Sorini 1987). This study involved testing 41 electric utility wastes produced by conventional and advanced sulfur dioxide control technologies to obtain information on the results of testing these materials using TCLP test conditions and to compare TCLP and EP results. The study evaluated three aspects of the TCLP: (1) the effect of increasing the filter pore size from 0.45- $\mu\text{m}$  in the EP to 0.6-0.8- $\mu\text{m}$  in the TCLP; (2) the effect of changing the composition of the leaching fluid on the concentrations of the inorganic analytes in the leachates; and (3) the concentrations of organic compounds in the TCLP leachates of the coal combustion by-products. The results of the study show the following (Sorini and Jackson 1988): (1) the change in filter pore size does not affect the composition of the coal combustion waste leachates; (2) the TCLP is more aggressive than the EP towards leaching silver, arsenic, and chromium from the wastes tested, while the EP is more aggressive towards leaching barium, and selenium is equally well leached by both TCLP and EP; and (3) the concentrations of the TC organic compounds leached from the coal combustion by-products were very low, mostly in the low parts-per-billion range.

EPRI also sponsored a round-robin study to compare the results of the TCLP with those generated by the EP (Mason and Carlile 1986). In this study, three laboratories extracted seven utility wastes and determined the concentrations of 14 constituents in the resulting leachates. Data generated in the study show that the reproducibility of the TCLP and EP methods varied between constituent, type of utility waste, and method. The concentrations of the analytes of interest were generally higher in the TCLP leachates than in the EP leachates.

### Leachability of Lead from Fly Ash Using the TCLP Sodium Acetate Buffer Solution as the Leaching Fluid

A study was conducted to evaluate the effects of pH and extraction fluid composition on the leachability of lead and arsenic from fly ash (Sorini and Brown 1993). One of the testing conditions that was evaluated was TCLP extraction of the fly ash using the sodium acetate buffer solution. Results of this study definitely show that acetate complexation of lead species prevents precipitation of lead minerals, and as a result, increases extractable lead concentrations. This is an example of where the co-disposal scenario represented by TCLP, which does not represent a typical disposal scenario for fly ash, results in over estimation of the amount of lead that would be leached from the material.

### Development of ASTM Method D-3987, Standard Test Method for Shake Extraction of Solid Waste with Water, and Its Application to Fossil Energy Wastes

During the time of EPA's proposal to classify wastes as hazardous or nonhazardous based on the EP test method, a research team became involved with ASTM and worked on development of ASTM Method D-3987 (Jackson and Moore 1984). Since fossil energy wastes are normally disposed in sites where they are the only waste type, the co-disposal scenario represented by the EP (9.5% municipal waste co-disposed with 5% industrial waste in a sanitary landfill) did not seem appropriate for these types of materials. As a result, ASTM Method D-3987, which uses reagent water as the leaching fluid and is believed to be more representative of the leaching environment found at fossil energy waste disposal sites (Jackson and Moore 1984) was developed.

Jackson and Moore (1984) describe a study in which 18 fossil energy wastes were tested using the ASTM and EP methods. The results of the study showed that most of the wastes tested gave very low concentrations of the eight RCRA metals in their leachates, and that none of the concentrations of these metals in the ASTM or EP leachates exceeded the regulatory levels.

There are some differences between the original ASTM Method D-3987 (Jackson and Moore 1984) and the current version of the method (ASTM 1995a). However, both versions require use of reagent water as the leaching fluid, and both allow the final pH of the leachate to reflect the interaction of the leaching fluid with the waste.

#### Evaluation of NEN Methods 7341 and 7343 Applied to Fly Ash

According to Janssen-Jurkovicova et al. (1994), NEN 7341 (availability test) and NEN 7343 (column test) are laboratory leachability tests that were introduced by the Dutch Standardization Institute for evaluating the leaching behavior of granular materials and /or fly ash. A study they conducted evaluated the correlation between leaching behavior of fly ash simulated by the NEN 7343 column method and leaching of fly ash occurring in a field lysimeter. The results of the study show discrepancy between data generated by applying the column test to fly ash and data generated by analyzing the lysimeter leachate. The reason given in the paper for this discrepancy is that the test conditions (e.g. time, pH, Eh) under which the column method is performed vary considerably from natural conditions, and this results in different leaching behavior of the material. The study also showed that the concentration of an element leached by standard tests can be much higher than expected based on the results of the availability test (NEN 7341). As a result, it was concluded that the availability test does not accurately predict the maximum elemental concentrations available for leaching from fly ash. The findings of the study also indicate that results of the column method do not provide sufficient information to relate the liquid-to-solid ratio used in the method to a time scale under field conditions. The authors of the paper describing this study propose development of a model to predict field leaching based on column leachate data.

#### **Future Direction of Leachability/Mobility Methods Development**

The applications of leaching methods to coal combustion by-products described above provide data that are specific to the particular application. However, these data provide little information for accurately assessing the release of contaminants from the materials into the environment. As pointed out in the descriptions of the leaching methods and the examples of their application to coal combustion by-products, these methods have deficiencies, such as modeling a single disposal scenario, not being intended to produce leachate representative of leachate generated in the field, specifying test conditions that do not correlate with field conditions, etc. As a result, there is a need for a new generation of test methods that address the deficiencies listed above. The U.S. EPA Science Advisory Board (SAB) has recommended that development of a leaching method to characterize the leachability/mobility of wastes involve the following: (1) a better understanding of the mechanisms controlling leaching, (2) multiple tests to address different disposal scenarios, (3) improved models to complement the leaching tests, and (4) field validation of leaching tests and predictive models (U.S. EPA SAB 1991).

Recently, there has been a move towards developing leaching methods that involve multiple procedures. Because a single leaching test cannot address all leaching aspects, van der Sloot (1995) describes combining existing procedures to address the particular leaching parameters of concern. He refers to this as a "concise test."

ASTM Task Group D-34.02.01 on Waste Leaching Techniques has also been working on developing a draft method for determining the leachability/mobility of wastes. Proposed aspects of this method are use of multiple procedures, such as a serial batch test and column test, and design of the method for site-specific applications, so that test parameters, e.g. leaching fluid composition and pH, soil type, etc., can be adjusted to accommodate specific disposal/management scenarios. In addition, at a recent task group meeting, a method for characterizing leachability by evaluating fundamental parameters, such as

constituent solubility, availability, and diffusivity, was discussed (Garrabrants et al. 1995). The method involves applying a number of characterization procedures, including leaching tests and physical tests, to the material being evaluated.

With particular relation to coal combustion by-products, Kosson et al. (1996) describe an approach to estimate constituent release from disposal and utilization of municipal waste combustion residues. The approach described varies greatly from use of a single test to predict leaching behavior. It involves use of portions of the method described above (Garrabrants et al. 1995) to measure fundamental leaching parameters, mathematical modeling to extrapolate laboratory results to field scenarios, and field verification.

In addition, Hassett et al. (1992) describe a study in which a solid scrubber residue was characterized by (1) qualitative screening to identify the elements present, (2) quantitative determination of total concentrations of selected elements, (3) determination of mineral phases present, and (4) application of selected leaching procedures. The leaching methods that were used were TCLP, a synthetic groundwater leaching procedure (SGLP), and a long-term leaching procedure.

The SGLP is a generic agitated extraction procedure that was developed to (1) simulate natural groundwater conditions with respect to groundwater chemistry, (2) be flexible enough to be site specific, and (3) be directly comparable to the TCLP (Hassett 1987). The steps in the method are the same as those performed in the TCLP, except synthetic groundwater is used in place of the specified acetic acid solution or sodium acetate buffer solution. Since the test is generic, any composition of leaching fluid can be used to simulate the groundwater chemistry of any location (Hassett 1987).

The long-term leaching procedure was performed to evaluate mineralogical changes occurring in the waste during long-term contact with water. Samples were analyzed after 18 hours, 48 hours, 1 week, 4 weeks, and 12 weeks of contact.

In the study, results of the various characterizations performed were used in combination to make conclusions concerning environmental impact of the material. For example, solubility trends in the leachate were related to formation of soluble and insoluble mineral phases. One of the conclusions of the study concerning the material tested is that the leachability of most of the RCRA metals in the scrubber residue is very low. A more general overall conclusion of the study is that characterization of waste materials for disposal must include more than a single leaching procedure. Chemical, physical, and mineralogical characteristics of the material must be considered, as well as site characteristics and interactions of the leachate and disposal environment (Hassett et al. 1992).

Based on the above discussion, it appears that a new generation of test methods will involve multiple procedures to address the various aspects of leachability and that these will be more waste- and site-specific oriented. Ideally, what is needed for the next generation of test methods are research methods, which can be longer and more complicated, condensed into shorter, more simple methods that can be used as good predictive tools for regulatory purposes and management practices.

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## **BULK CHEMICAL AND MINERAL CHARACTERISTICS OF COAL COMBUSTION BY-PRODUCTS (CCB)**

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### **Introduction**

Coal ash is a versatile material that can be used as a construction material as well as in agricultural applications. It can be used successfully in mineland reclamation as a substitute for soil in structural fills, capping material, liming material, and topsoil. Although coal ash can be used like soil, its characteristics and properties are very different. There are very few standard tests for coal ash, most tests are modified versions of cement or soil tests. There is no broad classification system for coal ash that covers all of the materials available. The classification system established by ASTM C 618 (Class F and Class C) is very limited and does not describe the different types of CCB, their mineralogy, or its range of properties. Many of the applications mentioned above do not require the material specifications defined by ASTM C 618. When evaluating the types of CCB available for mineland reclamation, it is very important that the basic characteristics of the CCB be understood and related to the mechanical or physical property necessary to successfully utilize the material in an engineering or agricultural application.

### **Elements in Coal and Coal Ash**

Coal is a highly variable, heterogeneous, fossilized material that formed from ancient plant material exposed to elevated temperatures and pressures after burial. In addition to the major elements of carbon, oxygen, hydrogen, nitrogen, and sulfur, coal also contains varying levels of trace elements such as sodium, mercury, chlorine, etc. Coal occurs in association with various types of inorganic minerals such as aluminosilicates (clay minerals), carbonates (calcite and dolomite), sulfides (pyrite), chlorides, and silica (quartz). Some elements such as sulfur occur in both the organic and inorganic coal fractions. The inorganic minerals, deposited along with the plant material, are inherent and make up 5 to 10% of the coal. It is principally these incombustible materials that form the ash that remains after combustion of the coal.

The chemical composition of coal ash is typically made up of silicon, calcium, aluminum, iron, magnesium, and sulfur oxides, along with carbon and various trace elements. These elements are found in the ash because of their high melting points and the short time the ash particles actually remain in the furnace during combustion. The mineral quartz ( $\text{SiO}_2$ ) survives the combustion process and remains as quartz in the coal ash. Other minerals decompose, depending on the temperature, and form new minerals. The clay minerals lose water and may melt, forming aluminosilicate crystalline and noncrystalline (glassy) materials. Elements such as Fe, Ca, and Mg combine with oxygen in the air to form oxide minerals, such as magnetite ( $\text{Fe}_3\text{O}_4$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), lime (CaO), and periclase (MgO).

### **Coal Combustion Technologies**

Conventional coal combustion involves pulverizing the raw coal into small particles and injecting it into a furnace ("boiler") where it is burned in a long luminous flame at temperatures of  $1500^\circ\text{C}$  or greater. Temperatures this high melt 70 to 90 percent of the mineral phases leaving fused mineral impurities we call coal ash or coal combustion by-products (CCB). Approximately one fifth of the ash particles fall to the bottom of the furnace and are collected as bottom ash. The rest of the fused matter is transported to a low temperature zone ( $200^\circ\text{C}$ ) in about 4 seconds. Here it solidifies into crystalline and non-crystalline glassy phases that are carried out of the furnace with the flue gas. We refer to this material as pulverized coal fly ash (PCFA) or simply "fly ash".

Coal combustion technologies are changing in order to burn coal more cleanly. The so called "Clean Coal Technologies" work at several stages of the combustion process; pre-combustion, combustion, and post-combustion. Pre-combustion involves cleaning the coal prior to combustion. Physical coal cleaning removes mineral particles, especially pyrite, to reduce ash content and SO<sub>2</sub> emissions. Combustion and post-combustion technologies remove the SO<sub>2</sub> and NO<sub>x</sub> gases, and/or particulate matter during combustion, or from the gases leaving the furnace. It is the combustion and post-combustion technologies that have a major influence on the chemical and mineralogical composition of the resulting CCB.

One new type of combustion technology is called Fluidized Bed Combustion (FBC). This technology involves mixing limestone with the crushed coal to act as a sorbent for SO<sub>2</sub> gas formed in the furnace. The mixture of coal and limestone are suspended on jets of air inside the furnace. The turbulent mixing of coal and sorbent causes the solids to behave as a pseudo-fluid allowing the coal to be burned at lower temperatures (800-900°C). As much as 90% of the SO<sub>2</sub> can be removed in this manner. The lower temperatures minimize melting resulting in CCB that do not contain the spherical glassy material typical of the fly ash formed during pulverized coal combustion. The amount of limestone used depends on the sulfur content of the coal, but generally it requires 1/3 to 1/2 ton of limestone for each ton of coal burned. This causes a significant increase in the volume of by-products formed from combustion.

Post-combustion cleaning technologies clean the down stream flue gases after they leave the boiler or combustion zone. They are used in conjunction with pulverized coal combustion furnaces and are often referred to as "scrubbers." There are two approaches to post-combustion cleaning technologies, Flue Gas Desulfurization (wet and spray dryer FGD) and Duct Injection. In wet FGD, the flue gases are bubbled through a tank containing hydrated lime slurry or some other alkaline sorbent, reacting with calcium and forming a wet sludge precipitate. In spray dryer FGD the hydrated lime slurry is sprayed directly into the hot flue gases, reacting with the SO<sub>2</sub> and forming a dry particulate material. Duct Injection is a similar process to spray dryer FGD except the sorbent is added directly into the existing furnace ductwork instead of a separate chamber.

### **Mineralogical Composition of CCB**

The bulk chemical and mineralogical characteristics of CCB and their hydration behavior depend mainly on the rank of coal burned, the calcium content of the coal, and the combustion process or scrubbing technology (if applied) (McCarthy, 1988; McCarthy et al., 1990; McCarthy et al., 1993). It has been shown (McCarthy et al., 1990) that the amount of silicon, aluminum, and iron in a coal ash varies proportionately with the amount of calcium in the coal ash and the calcium content of the ash is related to the source coal (Table 1). Bulk chemical analyses are routinely done for CCB that are sold for use in the concrete industry. Mineralogical characterization is not done routinely and the few laboratories that do this type of work are usually involved in research.

#### Pulverized Coal Fly Ash

The various minerals that have been identified with x-ray diffraction analysis of pulverized coal fly ash (and bottom ash) include: quartz (SiO<sub>2</sub>); mullite (Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>); tricalcium aluminate (Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>); melilite (Ca<sub>2</sub>(Mg,Al)(Al,Si)<sub>2</sub>O<sub>7</sub>); merwinite (Ca<sub>3</sub>Mg(SiO<sub>4</sub>)<sub>2</sub>); C<sub>3</sub>S (Ca<sub>3</sub>SiO<sub>5</sub>); C<sub>2</sub>S (Ca<sub>2</sub>SiO<sub>3</sub>); ferrite spine1 ((Mg,Fe)(Fe,Al)<sub>2</sub>O<sub>4</sub>); magnetite (Fe<sub>3</sub>O<sub>4</sub>); hematite (Fe<sub>2</sub>O<sub>3</sub>); lime (CaO); anhydrite (CaSO<sub>4</sub>); periclase (MgO); and alkali sulfates ((Na,K)<sub>2</sub>SO<sub>4</sub>). The predominant minerals in ash resulting from combustion of eastern and midwestern coals are quartz, mullite, hematite, and magnetite. This ash (as will be discussed later) is referred to as ASTM Class F fly ash. These minerals are also present in ash derived from western subbituminous coal along with the minerals periclase, lime, anhydrite, and tricalcium aluminate. This ash is referred to as ASTM Class C fly ash. It is these later minerals that give Class C fly ash its unique hydration behavior (Solem and McCarthy, 1992).

Quartz (SiO<sub>2</sub>) in the fly ash originates from silt and clay in the coal that is not fluxed by other inorganic materials. The amount of quartz does not correlate with chemical SiO<sub>2</sub> content. CCB from eastern and midwestern bituminous coal contain 40 to 50% chemical SiO<sub>2</sub> and 5-10% quartz. Quartz does not significantly react with water when the ash is hydrated.

Mullite (Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>) is the principal aluminum containing phase in Class F fly ash. It crystallizes directly from the molten ash or by devitrification of the glass after cooling. Its formation requires temperatures above 1000 °C. Mullite is twice as abundant in low-calcium ashes as in high-calcium. This is due to differences in the composition of the clays found in the coal. The higher aluminum/silicon ratio in kaolinite (0.85) and mica-illite (0.61) produce an ash richer in mullite compared to ash from coals containing smectitic clays (0.35). Coals that contain more calcium also produce ashes having less mullite because some of the aluminum combines with the calcium forming tricalcium aluminate (C<sub>3</sub>A, Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>). Like quartz, mullite is nonreactive when the ash grains contact water.

Ferrite spinel ((Mg,Fe)(Fe,Al)<sub>2</sub>O<sub>4</sub>), magnetite (Fe<sub>3</sub>O<sub>4</sub>), and hematite (Fe<sub>2</sub>O<sub>3</sub>) are the principal iron containing phases in PCFA. These minerals, present in almost all fly ashes, form from the oxidation of iron sulfides and other iron bearing minerals associated with the coal. Approximately one-third to one-half of the iron present in PCFA is speciated as crystalline iron oxides. These minerals are largely inert during reactions with water. The rest of the chemical Fe<sub>2</sub>O<sub>3</sub> is contained in the glass and will be available under high pHs.

Calcite and dolomite in the coal provide most of the calcium and magnesium in the PCFAs, but the another source is from organic exchange sites and organic complexes in the coal. Crystalline lime (CaO) has been identified in all high-calcium, most intermediate-calcium and some low-calcium PCFAs. Even in high-calcium PCFAs, only a small percentage of the chemical CaO crystallizes to form lime. In high- and intermediate-calcium ashes, the calcium also speciates as C<sub>3</sub>A (Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>), melilite (Ca<sub>2</sub>(Mg,Al)(Al,Si)<sub>2</sub>O<sub>7</sub>), and anhydrite (CaSO<sub>4</sub>). The hydration of lime to portlandite in high-calcium PCFAs is one mechanism for providing a source of the high pH necessary to activate cementitious reactions.

Tricalcium aluminate (Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>) occurs in most intermediate- and all high-calcium PCFAs, along with what is speculated to be a calcium aluminate-rich glass. C<sub>3</sub>A is soluble and participates, along with the calcium aluminate-rich glass component, in the formation of the hydration product ettringite.

Periclase (MgO), the principal magnesian mineral in high- and intermediate-calcium PCFAs, contains about half of the MgO present. Magnesium also speciates as melilite (Ca<sub>2</sub>(Mg,Al)-(Al,Si)<sub>2</sub>O<sub>7</sub>) and merrillite (Ca<sub>3</sub>Mg(SiO<sub>4</sub>)<sub>2</sub>), neither of which participate in cementitious reactions. The high temperatures of conventional coal combustion form periclase that is dead-burned (i.e., does not react significantly with water).

In the presence of higher CaO, MgO, Na<sub>2</sub>O, and K<sub>2</sub>O content, some of the SO<sub>2</sub> that results from the oxidation of pyrite during combustion of the coal can be captured as water soluble alkali and alkaline earth sulfate salts. This partial “self-scrubbing” effect has made these more alkaline low-rank coals useful for reducing SO<sub>2</sub> emissions without installing scrubbers.

Anhydrite (CaSO<sub>4</sub>) results from the reaction of calcium, oxygen and SO<sub>2</sub> in the gases leaving the furnace. Anhydrite is characteristic of high calcium PCFAs, but has also been observed in intermediate- and low-calcium ashes. In the lower alkali ashes, anhydrite accounts for most of the chemical S<sub>03</sub>. but in high alkali ashes, SO<sub>3</sub> is also speciated as alkali sulfates, ((Na,K)<sub>2</sub>SO<sub>4</sub>). Anhydrite in PCFAs is soluble and will react with water to form gypsum, ettringite, or monosulfate (Solem and McCarthy, 1992).

The particle density of PCFA grains varies with the type of minerals present. Iron oxide-rich grains have a density of 4.5-5.1, quartz 2.65, mullite 3.03, carbon 1.2-2.0, and alumino-silicate glass 2.5-2.7 g/cm<sup>3</sup> (Hemmings and Berry, 1988). Therefore, the average bulk density of PCFA reflects the proportions of solid and hollow particles, their size, the mineralogy of the crystalline components, the nature of the glass, and the amount of unburned coal. These characteristics also affect the reactivity of the PCFA when hydrated.

These materials have been used successfully in the U.S. in a number of applications including structural fill and as a topsoil replacement in direct revegetation of coal ash landfills.

#### Flue Gas Desulfurization By-Products

Flue gas desulfurization (FGD) by-products consist of PCFA grains entrained with reacted and unreacted sorbent. The average particle size ranges from 20-40 micrometers. The fly ash in the FGD sludge is similar in particle size, particle density and morphology to those of conventional PCFAs, but the sorbent reaction products have lower bulk densities due to differences in their chemical and mineralogical characteristics. FGD by-products contain higher concentrations of calcium and sulfur, and lower concentrations silicon, aluminum, and iron than PCFAs. The principal sorbent reaction products are hannebachite (calcium sulfite hemihydrate) or gypsum (calcium sulfate dihydrate). Unused sorbent remains as hydrated lime (portlandite) in the by-products.

These materials have been used successfully in Indiana as both structural fill, low permeability caps, underground mine stabilization (injected as a slurry), as well as a replacement for agricultural lime.

#### Fluidized Bed Combustion Residues

The bottom ash collected from FBC technology is sometimes referred to as spent bed, however, the fine material captured in the baghouse is still referred to as “fly ash”. The chemical and physical characteristics of FBC by-products vary from one power plant to another depending on several factors including; the composition of the “bed” material used to fluidize the coal particles (originally limestone, sand or shale), the amount of sulfur in the coal, reactivity of the sorbent, and when the bed is removed. There are times when the load in the boiler becomes too high and portions of the bed are removed from the furnace. This influences the amount of char or partially burned coal present in the spent bed. By-products formed in plants using inert beds (such as sand and shale) also contain variable amounts of x-ray amorphous phases that are produced by the thermal decomposition of the clay minerals in the bed materials. These amorphous materials form a “hump” on the X-ray diffraction scan that resembles the glasses in PCFAs but is not composed of melted material.

The main crystalline phases in the FBC by-products are anhydrite (reacted sorbent), lime (unreacted sorbent), quartz, and iron oxides. Anhydrite forms from the calcination of limestone into lime followed by its reaction with SO<sub>2</sub> and oxygen. The amount of reacted and unreacted sorbent depends on the scrubbing efficiency and the amount of limestone added. Power plants that burn high-calcium, low-sulfur subbituminous coal have to use more sorbent to attain the high percent sulfur removal required by regulations. These ashes contain more lime (due to increased use of limestone) and less anhydrite (due to lower sulfur content of the coal). High sulfur coal also contains more iron resulting in higher concentrations of iron oxide (hematite and magnetite) in the ash. Other crystalline phases present include calcite, periclase, and feldspars.

FBC by-products, bottom ash and fly ash, contain the same type of minerals but often in different proportions. The bottom ash is often enriched in anhydrite and lime while the fly ash is richer in silicon and iron oxides. FBC fly ash is a fine grained homogeneous material that consists of angular particles smaller than 45 micrometers. It is generally irregularly shaped and does not have the fused, spherical characteristics of PCFAs, due to the lower combustion temperatures of FBC. The spent bed

residues are generally larger than 75 micrometers, with a narrower range of particle size than fly ash particles.

Most published studies that included characterization of FBC residuals looked at materials from only one or two different plants. According to a 1991 Electric Power Research Institute database, there were more than 115 plants in the United States operating more than 161 FBC boilers. This technology can be used for small-scale and large-scale power generation. FBC technology can utilize many different types of alternative fuels, such as petroleum coke, bituminous coal mine wastes (GOB), wood chips, agriculture waste, process wastes, refuse derived fuel, and municipal sewage sludge. There is as yet no literature that describes the effects that alternative fuels have on the nature of the FBC residues or their variability due to changing sources of coal. The use of these materials as a soil amendment has been described in a manual by Stout et al. (1988).

These materials have been used successfully at Purdue as structural fill and in the manufacture of synthetic topsoil.

### **Classification of Coal Combustion By-products**

There is currently only one commonly used classification system for CCB defined by ASTM C 618. It distinguishes two classes of pulverized coal fly ash, Class C and Class F (Helmuth, 1987). Materials are first tested according to ASTM C 311, "Standard Test Methods for Sampling and Testing Fly Ash or Natural Pozzoloans for use as a Mineral Admixture in Portland-Cement Concrete", and then classified according to ASTM C 618, "Standard specification for Fly Ash and Raw or Calcined Natural Pozzolan for Use as a Mineral Admixture in Portland Cement Concrete". ASTM C 311 describes the methods for bulk chemical analyses. The chemical and physical requirements specified in ASTM C618 are given in Table 2.

The distinction between Class F and Class C fly ash is based on the sum of the total silicon, aluminum, and iron ( $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ ) in the ash. When the sum is greater than 70% an ash is classified as Class F. When the sum is between 50% and 70% the ash is classified as Class C. A bituminous coal will always produce a Class F ash, but a subbituminous or lignite coal can form either a Class C ash or a Class F fly ash depending on the calcium content of the coal. Ash that results from blending of bituminous and subbituminous coal have not been studied in any detail.

In every state, where CCB specifications have been developed by the Department of Transportation, it is required that coal ash materials must pass C-618 specifications. There are several utilization practices, such as high-volume structural fills and control density fills (flowable fills), where ASTM C618 requirements are irrelevant. In these engineering applications, CCB can be used successfully even though they do not meet ASTM C618 specifications. ASTM C 618 specifications were developed for a specific application, the use of fly ash as a mineral admixture in Portland cement concrete. Their use as a generic material specification for all CCB used in any engineering applications has eliminated a significant volume of CCB from potential utilization.

ASTM C 618 does not adequately characterize the physicochemical properties of fly ash. It only requires a bulk chemical analysis that indicates the total amount of certain elements such as calcium, silicon, aluminum, iron present in the fly ash. It does not provide any indication of the minerals or compounds in which these elements are speciated. Minerals have varying solubility and they control the rate and amount of an ion available in solution, which controls the formation of various hydration products.

### **Physical Characterization of CCB**

Modified soil and cement or concrete testing procedures can be used to characterize the physical properties of CCB that relate to its use as a construction material. However, in many cases, the results of

the tests must be reevaluated in order to understand what the tests are actually measuring, since coal ash is a fundamentally different material than either soil or Portland cement.

The physical characteristics (such as particle size) depend mainly on where the residues are collected (baghouse or bottom ash). Both bottom ash and fly ash usually have a fairly uniform particle size gradation but when commingled can result in a somewhat wider particle size distribution. CCB do not contain clay and therefore exhibit no plasticity. Index tests which measure plasticity and are used to classify soil for engineered applications are not appropriate for CCB. The physical properties most relevant to engineering applications are the particle size, gradation, and hydration behavior. Hydration behavior will depend on whether the material is chemically reactive with water. The compaction behavior and water demands, strength, density, permeability, and leachability will depend very strongly on hydration behavior.

Eastern and midwestern bituminous coals (unscrubbed) produce low sulfur, low-calcium ashes that are unreactive when mixed with water. These materials can be easily compacted provided sufficient water is applied. A simple rule of thumb to follow is that if the ash is dusty, you don't have enough water. Most of this type of coal ash will drain freely and therefore if too much water is applied the situation remedies itself fairly quickly. These CCB will tolerate a relatively wide range of water contents and still produce a well compacted material. They can be placed in thicker lift sizes than most soils and will compact more densely using less compaction energy.

Western subbituminous coal produces ash that typically contains more than 20% CaO and exhibits a self-cementing behavior when mixed with water. Much of the fly ash produced is used by the cement industry and not usually available free of charge. CCB that result from FBC and FGD technology typically contain large amounts of calcium and sulfur in the mineral forms of lime and anhydrite (in FBC) and gypsum or hennrichite (in FGD). The engineering behavior of these CCB is largely controlled by the dominant type of mineral(s) present. Some have cementitious properties, and others do not. The strength gain in these materials will depend on the amount of lime present. The presence of anhydrite and lime causes considerable expansive behaviors due to the formation of ettringite and gypsum (Solem and McCarthy, 1992).

### **Summary**

Coal ash is a versatile material that can be used as a construction material as well as a liming agent or topsoil substitute. These applications are the most relevant to mineland reclamation. The only existing standard specification for coal ash is ASTM C 618. It defines the chemical and physical requirements for fly ash that permits its use as a mineral admixture in portland cement concrete. The bulk chemical and mineralogical characteristics of CCB and their hydration behavior depend mainly on the rank of coal burned, the calcium content of the coal, and the combustion process or scrubbing technology. Modified soil and cement or concrete testing procedures can be used to characterize the physical properties of CCB that relate to its use as a construction material; however, in many cases, the results of the tests must be reevaluated in order to understand what the tests are actually measuring. It is possible to mix various types of CCB, modify them by adding other materials, or use them to modify existing materials at a site in order to achieve successful results in almost any application.

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Table 1. Calcium Content in Coal Ash is Related to the Coal Source

Source Coal	CaO (in ash)
Eastern and Midwestern bituminous	1-6%
Colorado bituminous	4-8%
Utah and Alberta subbituminous	6-12%
Texas Lignite	7-15%
Saskatchewan lignite	10-15%
North Dakota lignite	18-25%
Montana and Wyoming subbituminous	22-32%

Table 2. Chemical and Physical Requirements Defined by ASTM C 618.

Chemical Requirements:	Class F	Class C
SiO <sub>2</sub> + Al <sub>2</sub> O <sub>3</sub> + Fe <sub>2</sub> O <sub>3</sub> , min. %	70.0	50.0
SO <sub>3</sub> , max. %	5.0	5.0
Moisture content, max. %	3.0	3.0
Loss on ignition (LOI), max. %	6.0	6.0
Available alkalies, as Na <sub>2</sub> O, max. %	1.5	1.5
Physical Requirements:	Class F	Class C
Fineness, max. %	34	34
<i>Strength Activity Index:</i>		
-with portland cement, 7 days, min. %	75	75
-with portland cement, 28 days, min. %	75	75
-with lime, 7 days, min. psi	800	800
Water Requirements, max. % of control	105	105
Soundness; autoclave expansion or contraction, max. %	0.8	0.8
<i>Uniformity Requirements:</i>		
Specific Gravity, max. % variation from average.	5.00	5.00
Percent retained on #325 sieve, max. % variation from average	5	5

## ENGINEERING AND REGULATORY ISSUES FOR COAL COMBUSTION BY-PRODUCT CHARACTERIZATION AND UTILIZATION

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### Introduction

The term coal combustion by-products (CCBs) generally refers to the typical high-volume residues that inevitably result from coal use in energy production. These high-volume residues are fly ash, bottom ash, boiler slag, and FGD (flue gas desulfurization) byproducts. Many of these CCBs have properties advantageous for engineering, construction, and manufacturing applications (Baker, 1984; Helmuth, 1987; University of North Dakota Mining and Mineral Resources Research Institute [UNDMMRI], 1988). The first university research study on coal fly ash was reported in the Proceedings of the American Concrete Institute (ACI) in 1937, where the term "fly ash" first appeared in the literature. In 1946, the Chicago Fly Ash Company was formed to market coal fly ash as a construction material for manufacturing concrete pipe (Faber and Babcock, 1987). The first large-scale use of coal fly ash was by the U.S. Bureau of Reclamation in the construction of the Hungry Horse Dam in Montana in 1949 (Faber and Babcock, 1987). Six other dams were constructed during the 1950s using coal fly ash concrete. Initial markets opened up by the Chicago Fly Ash Company were as a cement replacement and as an enhancer of the qualities of concrete to meet the new postwar demands. The technology used to establish these markets came from the U.S. Bureau of Reclamation and Army Corps of Engineers experience using natural pozzolans in concrete for dam construction. Pozzolanic technology dates back to Roman times, some 2000 years ago, in the building of the aqueducts and the coliseums.

Currently, CCBs are an underutilized industrial by-product in the United States. Approximately 20% of all U.S. by-products produced in 1991, including FGD material, were utilized (American Coal Ash Association [ACAA], 1991). Large potential markets for these byproducts have not yet been exploited. Many of the applications referred to above are in the initial stages of commercial development. Although extensive research, development, and promotional effort have been expended, much more work is needed to achieve full commercial potential. Research, development, and demonstration are continuing under the auspices of numerous institutions across the United States. The ACAA, Edison Electric Institute and the Utility Solid Waste Activities Group, the Electric Power Research Institute (EPRI), and many individual utilities and marketing organizations are devoting their best efforts to promote CCB use, especially in engineering and construction applications.

### Engineering Applications for CCBs

Historically, the principal use of coal by-products has been in concrete, and this still holds true today. However, many other utilization applications are discussed in the literature, including controlled low-strength materials, highway road base and subgrade, soil amendments for agricultural uses, waste stabilization, extenders in plastics and paints, and the manufacture of products such as cement, insulating materials, lightweight building block, brick, and other construction materials.

The use of CCBs in concrete and concrete products is primarily in the incorporation of fly ash as a partial replacement for portland cement in concrete. This use generally has the following beneficial effects:

- . Reduced water requirements
- . Increased ultimate strength
- . Improved workability
- . Extended setting time
- . Reduced heat release
- . Lower permeability
- . Improved durability
- . Increased resistance to chemical degradation

Mechanisms proposed to explain the improved microstructure and density obtained by using fly ash are 1) the packing of finer fly ash particles into interstices, 2) an increased binder-water ratio resulting from this packing, and 3) the pozzolanic reaction of calcium hydroxide and fly ash occurring over a period of weeks or months. Analytical characterization of fly ash cement during curing shows that a variety of calcium-silicon-hydrate gels are responsible for the gain in both strength and density notably occurring from about 28 days (Pietersen and others, 1991). These reactions are restricted by low alkalinity, low temperatures, or high water-solid ratios in pozzolanic systems. Reaction mechanisms overall are similar for either pozzolanic or cementitious fly ash, except that cementitious fly ash provides both cementitious calcium and silicon reactants, whereas pozzolanic fly ash supplies primarily silicon, which reacts more slowly with the calcium hydroxide released from the portland cement.

Fly ash used in this application generally must meet one of the commonly applied standard specifications developed by ASTM (American Society for Testing and Materials) or AASHTO (American Association of State Highway and Transportation Officials). These specifications contain both chemical and physical requirements for fly ash to be used as a mineral admixture in concrete. Cementitious fly ash generally falls into the ASTM and AASHTO specification for Class C, and pozzolanic fly ash generally falls into the ASTM and AASHTO specification for Class F. These are material specifications and not performance-based standards, so they do not always accurately predict performance.

There are varying reports of the effects of the use of fly ash in concrete relative to its performance in several situations. Freeze-thaw durability of fly ash concrete can be ensured by the use of practices that ensure good freeze-thaw performance for other concretes (Tyson, 1991; Dunstan, 1991). Freeze-thaw performance is reported to improve with the addition of Class C fly ash up to a 50% replacement level (von Fay and others, 1993). Overall, the resistance of fly ash concrete to salt scaling due to deicing in cold climates appears to be similar to that of conventional concrete, and improved resistance correlates with strength and reduced permeability (Soroushian and Hsu, 1991). Unwanted alkali-silica reactions are the cause of detrimental expansion in concrete both during and after curing. This problem occurs in various forms when free alkalies (usually reported in terms of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$ ) are introduced with any of the raw materials and is common when portland cement is used with high-silica aggregate. As commonly used, the term alkali-silica reaction appears to represent a class of related problems. These problems are variously reported to be remedied or aggravated by the addition of specification-grade fly ashes, suggesting that mineralogical properties not currently considered may be important. In general, the addition of fly ash, and particularly Class F ash, is considered to be beneficial because of the ability of finely divided silicious particles to tie up alkalies and free lime by pozzolanic reactions (Butler and Ellis, 1991; Smith, R. L., 1993). The concrete expansion encountered during curing when using certain Class C fly ashes has been reported to depend on the amount of free lime introduced with the ash, which has been correlated with reduced furnace temperatures and a less vitrified (fused) ash (Kruger and Kruger, 1993). High concentrations of sodium in certain western coals, occurring in an organically associated form, are an identifiable cause of alkali-silica reaction.

Manufacture of artificial aggregates from fly ash or other CCBs can be accomplished by either sintering processes (Puccio and Nuzzo, 1993) or hydrothermal and cold-bonding processes. Sintering processes are well-established technology and have operated successfully for decades, but lower-temperature processes under development have attracted more recent attention because of their lower

energy requirement and greater cost-effectiveness. Commercial production of lightweight aggregate for building block based on low-temperature processing has commenced recently in Florida using the Aardelite process (Smith, C.L., 1993; Hay and Dunstan, 1991) and in Virginia using the Agglite process (Courts, 1991).

Synthetic aggregate was experimentally produced from lime-based spray dryer FGD byproducts in the early 1980s by pelletizing at pressures of 5 to 20 tons per square inch followed by extended 10- to 60-day curing under controlled moisture conditions (Donnelly and others, 1986). Strength properties were adequate for confined applications such as road base. Pellets produced from Coolside, LIMB (limestone injection modified burner), and fluidized-bed combustion (FBC) by-products in the Ohio Coal Development Office demonstration project have passed the ASTM abrasion test for use as road base aggregate (Hopkins and others, 1993). Commercial production of lightweight aggregate suitable for use in lightweight building block commenced in Florida in 1992 using bituminous coal fly ash and FGD scrubber sludge as raw materials in the low-temperature Poz-0-Lite process (Smith, C.L., 1993).

A novel method for producing lightweight aggregate by agglomerating fly ash or sand in foamed cement has been developed in Germany (Gorsline, 1986). The properties of the aggregate can be controlled to meet a range of specifications on size, strength, density, and porosity. The cellular structure imparts high strength in relation to weight and reduces the amount of cement required.

Autoclaved cellular concrete used in building blocks, roof slabs, and other cast products represents an important market for fly ash in Europe. In this process (Pytlík and Saxena, 1991; Payne and Car-roll, 1991), fly ash is combined with cement, lime, sand, and aluminum powder and mixed with hot water. The reaction of aluminum and lime generates hydrogen gas which forms an aerated cellular structure. Curing in high-pressure steam autoclaves produces a physically chemically stable product.

The use of fly ash alone or together with lime or cement in self-hardening road base is an evolving technology which is receiving increased attention. New information in this area includes the results of laboratory testing and extended field monitoring, use of reclaimed pond ash, incorporation of FGD by-products, and use of the ash-based aggregates discussed previously.

Monitoring of a 1500-ft test section using cement-fly ash base for a Michigan 4-lane highway has indicated quite satisfactory performance since its construction in 1987 (Gray and others, 1991). Some heaving and cracking occurred in winter months due to frost effects. Laboratory leachate concentrations for heavy metals using ASTM and RCRA (Resource Conservation and Recovery Act) (U.S. Environmental Protection Agency [EPA]) procedures approached drinking water standards. Replacement of lime with Class C fly ash in the subbase for a Kansas racetrack reduced the cost by one-half; swelling potential was reduced compared to lime stabilization, but strength was reduced at soil temperatures below 40°F (Ferguson and Zey, 1991). Laboratory evaluation of fly ash stabilization of caliche, a red-brown calcareous material used for roadways in South Texas, indicated that both Class C and Class F ashes were more effective than lime for reducing plasticity and that Class C fly ash also significantly increased strength (Keshawar and others, 1991). Laboratory and field tests on the use of artificial aggregate produced from fly ash in asphalt paving, both as road base and in the asphalt mix, indicated that bitumen is absorbed in the pores of the aggregate, producing good bonding but a relatively dry and stiff mix; replacement of commonly used gravel with fly ash aggregate did not result in higher leaching of any of the heavy elements analyzed (Mulder and Houtepen, 1991).

Reclaimed pond ash containing fly ash and bottom ash from Canadian lignite has been used to stabilize road base for asphalt paving (Culley and Smail, 1986). The wet pond ash, when compacted in 1-in. layers using standard equipment and handling procedures, had good structural bearing, but the unconfined surface suffered rapid surface abrasion when dry. Adequate bonding between the ash subbase and asphalt paving was achieved by blade-mixing the first layer of asphalt into the underlying ash. Road surface condition was adequate over time where appropriate construction techniques were used. Recent laboratory testing on strength development for a reclaimed high-calcium fly ash used along with kiln dust

to stabilize road base materials indicated strengths in the range of 200 to 1000 psi (Bergeson and Overmohle, 1991).

Fly ash has been successfully used in combination with lime sludge to stabilize unstable sand in Florida road base projects (Jones, 1986). A base prepared by mixing lime and fly ash with in situ sand hardened sufficiently after several weeks to allow heavy truck traffic. By-products from coolside, LIMB, and FBC sulfur control technologies are currently being evaluated for use in road base. Laboratory tests on the compaction, swelling, shear strength, permeability, and leaching properties of the coolside FGD by-product indicate a good potential for use in road base applications, but final assessment awaits the performance of field trials and engineering analysis (Hopkins and others, 1993).

Controlled low-strength material (CLSM) is a fill formulated from a pozzolanic material such as fly ash along with small amounts of cement, a natural filler such as sand, and water. CLSM is also commonly called flowable fill, flowable mortar, or controlled-density fill (CDF). CLSMs have been investigated for numerous applications, including subbase for paving and foundations; backfills for trenches, culverts, and bridge abutments; and fillings for abandoned tanks, sewers, and mine shafts. Starting in the late 1960s, the Detroit Edison Company working with the Kuhlman Corporation pioneered the development of flowable fill formulations using fly ash, which they call K-Krete. Advantages of CLSM over compacted soil include delivery in ready-mix trucks, placement without tamping or compacting, strength development supporting equipment within 24 hours, and the opportunity to formulate mixes having an ultimate strength well in excess of that of compacted fill. Significant savings in time and related cost can be achieved in designing rapid turnaround projects for high-traffic road applications. For example, bridge replacement in the Mississippi Basin in the aftermath of the 1993 flood could be accomplished more quickly and economically by substituting large culverts imbedded in fly ash fill for damaged abutment-type bridges, where applicable. This type of bridge replacement has been reported to save as much as 75% of the cost of conventional construction (Buss, 1990).

The properties of CLSM vary widely depending on the class of fly ash used and the mixing proportions. Nonspecification fly ashes, relative to requirements established for fly ash as a mineral admixture in concrete, can be quite satisfactory for flowable mortar. Compressive strengths within a nominal range of 50 to 1500 psi can be tailored to fit the requirements of the application, including the possible requirement for reexcavating. In flowable tills, Class F fly ash serves primarily as aggregate, and large amounts can be used. Recent research on the mechanical properties of formulations using Class F fly ash (Maher and Balaguru, 1991) indicates that satisfactory 28-day strengths in the range of 198 to 1726 psi were obtained for mixes containing up to about 40% fly ash along with sand and 3% to 7% portland cement; strength development continued up to and possibly beyond 180 days, at which time a 7% cement mixture testing at 172 psi at 28 days had reached a strength of 3000 psi. Class C fly ash is itself a cementing agent, and 1500-psi 28-day strength is achieved using only about 3% portland cement and 5% high-calcium ash (Naik and others, 1991). The amount of Class C fly ash that can be used in CLSM is limited by the desired strength, where higher proportions of fly ash alone, without cement, will produce compressive strengths exceeding low-strength concrete.

The use of CLSM to correct acid mine drainage and subsidence in old underground coal mines is a well-demonstrated technology that could be more widely applied (Ryan, 1979). The bonding material in the grouting used is typically fly ash and cement in a 10:1 ratio, although ash alone can be used in less critical applications. Subsidence can be prevented either by backfilling the entire mine void with a low-strength grout or by establishing stronger grout/gravel columns at appropriate intervals to support the mine roof. Acid mine drainage and underground burning in spoil piles can be remediated by similar grouting methods engineered to isolate, fill, and/or extinguish affected areas in a mine. Advantages of using flowable mortars are minimum disturbance (no excavation), engineering flexibility, and low cost. Fly ash is returned to the in from whence it came, while at the same time remedying in related environmental problems,

## Manufacturing Applications for CCBs

A large number of potential uses for CCBs in industrial applications have been investigated. Commercial practice is limited in the United States, but is more common in some other countries. These applications include the following:

- Gypsum
- Brick
- Fillers in paint, plastics, and metal
- Mineral wool
- Ceramics

## Land Applications for CCBs

It is important to point out land application of CCBs as a special case of CCB utilization. Land application includes use as soil amendments and in mining applications. Soluble forms of calcium, magnesium, sulfur, and certain necessary trace metals such as boron, molybdenum, zinc, selenium, and copper that are present in CCBs can be used to provide needed plant nutrients. No significant amounts of the primary nutrient elements-nitrogen, phosphorus, and potassium-are found in CCBs, but wood ash is rich in potassium and phosphorus. By-product gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) can be used to improve the tilth of clayey soils and mitigate the toxicity of exchangeable aluminum in acid soils. Calcium contributes to soil aggregation by displacing sodium on clay minerals and providing microscopic cementation. Concerns relating to the agricultural use of CCBs involve the presence of soluble salts and trace concentrations of toxic metals which may be present in CCBs.

### Soil Amendment Applications

A review of past work on the effect of CCBs on plant growth (Clark and others, 1993) indicates that little agricultural utilization is occurring and that information is limited. Scrubber sludge impoundments have been successfully vegetated using wheat grass, tall fescue, sweet clover, millet, cottonwoods, and red cedars. Scrubber sludges have been successfully used as a source of boron and selenium trace nutrients. FBC bed residues are variously reported to increase maize and soybean yields and to provide a necessary source of calcium for apples. Research has been conducted by the Tennessee Valley Authority (TVA) on the inclusion of lime/limestone scrubbing waste into fertilizer formulations (Santhanam and others, 1981). Research is being conducted on the agricultural use of wood-fueled power plant ash from generating units in California (Wheelabrator Shasta Energy Company, 1991; Meyer and others, 1992).

In controlled greenhouse tests on several different CCBs (Clark and others, 1993) the addition of FBC residues to an acid soil of known severe aluminum toxicity served to double the yield of maize at an optimum add rate of 2% to 3% in the soil mix, but yields decreased at higher use rates. The effect of fly ash addition varied with coal type, with a bituminous Class F fly ash showing its highest growth enhancement at 3% addition. whereas lignitic Class C fly ash continued to increase yields at rates up to 25% of the soil mix. FGD by-products generally provided less growth enhancement, and optimum results were obtained at very low rates of 1% or less of the soil mix, possibly owing to detrimental effects of sulfite contained in these by-products. The use of an FGD sludge that had been processed to convert sulfate to gypsum enhanced growth rates at add rates up to 75% of the soil mix, consistent with the known beneficial effect of gypsum application to acid soil.

A major study on land application of FGD and pressurized fluidized-bed combustion (PFBC) by-products (Beeghly and others, 1993) is in progress in Ohio, sponsored by the Ohio Coal Development Office, the U.S. Department of Energy (DOE), EPRI, Ohio Edison, American Electric Power, Dravo Lime Company, and Ohio State University. By-products from fifteen sources are being investigated, representing four major clean coal technologies, including furnace injection FGD (LIMB), duct injection FGD, spray dryer FGD, and fluidized-bed combustion (atmospheric FBC and PFBC). These by-products

are characterized by high alkalinity expressed as calcium carbonate equivalents of 25%-70%; sulfur contents of 2.4%-10.3%; fly ash contents of 10%-32%; and, with the exception of FBC bed material, a high surface area and fineness. Selected by-products, alone or in combination with sewage sludge, were mixed with acid soils and mine spoils and tested in greenhouse growth studies. Interactions of different materials gave somewhat different results. For example, growth of tall fescue was enhanced in overburden spoil, but was suppressed in acid underclay. Sulfite-bearing material did not harm seed germination. LIMB by-product was successfully composted with sewage sludge. The conclusion reached from the greenhouse tests was that the by-products tested, when used appropriately, are suitable substitutes for traditional soil-liming materials for acid soils. Field tests are under way to demonstrate the practicality of this use.

The commercial N-Viro Soil Process (Burnham, 1993) combines agricultural use with waste stabilization by composting CCBs, or cement/lime kiln dust as originally used, with municipal wastewater treatment sludge. The soil conditioner produced has a low nutrient value (1 % N, P, K); a high lime equivalency of 25%-60%; good storage, handling, and spreading properties; and acceptable odor. The product is being produced from sludges produced in several municipalities and is used in agriculture and in cover for landfill. The key to the success of this process is that pathogenic microorganisms are destroyed by the alkalinity and heat associated with the addition of CCBs and possibly quicklime (CaO), followed by temperature-controlled composting and air drying. Leachability tests at various pH levels have indicated that the heavy metals are below EPA toxicity limits.

The efficacy of using CCBs in agricultural applications cannot be generalized since it is evident in comparing case studies that success is varied and depends on the suitability of the amendment to the soil and use conditions. For example, composting coal fly ash with field-collected waste vegetation was found to have no detrimental effect on bean germination in clayey and sandy soil, but reduced germination in a high-humus soil (Varallo, 1993). Alkaline treatment is appropriate for eastern acidic soil, but not for many midwestern soils that are already alkaline in nature. Novel applications in specialized areas may provide some of the more immediate commercial opportunities. FBC bed residues have been used at high rates of over 100 tons per acre as a mulching agent applied directly to cap the soil surface in orchards and raised-bed tomato rows (Korcak, 1993). Coal bottom ash has been demonstrated as an acceptable root medium for growing flowers in a hydroponic nutriculture system (Bearce and others, 1993). Widespread acceptance of CCBs in agriculture still has performance and economic barriers to overcome, but opportunities exist today where the properties of a utility's by-products meet the needs of a local market,

Some concerns still exist about the environmental safety of using CCBs in agriculture, despite findings that leachable concentrations of toxic metals are very low (Beeghly and others, 1993; Burnham, 1993; Bennett and others, 1981). While results vary somewhat for different by-products and soil types, the general finding reported is that leachates are nontoxic relative to the eight RCRA toxic metals (arsenic, barium, cadmium, chromium, lead, mercury, selenium, and silver) and often approach the more stringent primary standard for drinking water. The mobility of metals depends on the mineral matrix and on pH, and solubility is generally reduced at the high pH levels associated with alkaline CCBs. Certain beneficial trace plant nutrients present in coal fly ash, such as boron, selenium, and molybdenum, are assimilated in animal tissues (Lisk, 1981), and selenium deficiency in farm animals has been shown to be correctable by feeding the animals fly ash-grown crops. In coordinated tests on farm crops and animals (Bennett and others, 1981), there has been little evidence of detrimental effects on the food chain. One reason for caution is that standard tests for determining the leachability of trace metals, primarily the EPA toxicity characteristic leaching procedure (TCLP) tests for acid solubility, do not accurately represent all utilization environments, and they may either evidence problems that do not exist or miss worst-case problems that would occur in practice. At the current stage of understanding, states will tend to regulate ash reuse on farmland as solid waste management, requiring case-by-case permitting (e.g., California [Marshack, 1992]).

## Mining Applications

There are several scenarios in which CCBs may be utilized in a mined setting:

- The use of CCBs for abatement of acid mine drainage or for treatment of acid mine spoils (Schueck and others, 1993; Ackman and others, 1993; Stehouwer and others, 1993).
- The use of CCBs in reclamation activities or highwall mining (Bergeson and Lapke, 1993; Paul and others, 1993; Robl and Sartaine, 1993).

The placement of ash, as a low-strength structural material, in an underground mine for reclamation and prevention of subsidence (Chugh, 1993, Butler and others, 1995).

These three options represent most, but not all, scenarios under which CCBs would be returned to the environment in a mined setting. Mine applications have previously been considered disposal but, because of the relatively benign nature of CCBs, should more appropriately be considered reuse for reclamation of mined land because of the benefits derived in these applications.

Solid residues from the combustion of low-rank coals, which generates leachate at extremely high pH, tend to form the mineral ettringite. The alkaline nature of some CCBs (including duct injection residues/FBC residues and low-rank coal fly ash) can be capitalized on for abatement of acid mine drainage and spoils (Schueck and others, 1993; Ackman and others, 1993; Stehouwer and others, 1993). Ettringite has the capacity to chemically fix elements such as arsenic, boron, chromium, molybdenum, selenium, and vanadium that exist as oxyanions in aqueous solution. Thus ash that generally leaches low concentrations of most potentially problematic trace elements tends to form stable minerals of some of the most highly problematic of the trace elements known to concentrate in ash from coal combustion (Hassett and others, 1991). Although CCBs are generally benign with respect to leaching significant concentrations of potentially problematic elements, proper and environmentally sound testing should be conducted (Hassett, 1991; Hassett, 1994). This testing should be done using long-term as well as short-term leaching to determine the total mass of trace elements that may potentially be mobilized and the trends of analyte chemistry evolution (Hassett, 1987; Hassett and Pflughoeft-Hassett, 1993). Although leachate chemistry of most trace elements is accumulation of analyte to an equilibrium concentration that increases to a plateau, some of the oxyanionic trace elements can actually increase to a plateau quickly and then exhibit a trend of decreasing solution concentration. This is important to understand, since it is the long term that is usually important in assessment of potential for environmental impact.

A field demonstration at Center, North Dakota, where scrubber sludge was placed into a mined area was performed by the Energy & Environmental Research Center (EERC). The only observed impact was caused by the disturbance of the environment at the time of mining. An increase in total dissolved solids, mostly from sodium sulfate, was seen, but this rapidly returned to background levels (Beaver and others, 1987).

The primary conclusion that can be drawn is that return of ash to the mined settings is a sound high-volume use of this versatile engineering material. Not only can land be reclaimed, but in the case of underground mines, the setting can be stabilized to prevent future subsidence. Treatment of acid mine drainage and spoils has high potential, especially for high-volume alkaline residues from advanced coal processes. Impacts from trace elements, the primary concern, have been minimal or unmeasurable in almost all instances where monitoring has been carried out. There have been examples where groundwater quality has been shown to actually improve from the placement of CCBs in the environment (Ackman and others, 1993; Paul and others, 1993).

## Coal Combustion By-Product Classification

Alternative classification standards are needed for a growing list of CCBs to address engineering and environmental performance in a manner that will provide public assurance of safety and effectiveness, while not impeding utilization. It is widely believed, as evidenced by this and other

studies, that the present ASTM and EPA test protocols and specifications and related state standards do not adequately predict performance under actual use conditions. Important advances are being made in applicable analytical methods, but their application is hindered by misplaced reliance on existing empirical standards. For example, specifications developed for the use of fly ash in concrete are sometimes used by default in applications involving flowable fill or road base. New, automated analytical methods for statistically characterizing individual particles contained in bulk by-product samples by size, surface composition, and mineral type give a true indication of the chemical and mineralogical diversity in CCBs (Folke Dahl and others, 1993). Leaching tests for determining the mobility of RCRA elements under conditions simulating actual field conditions are being compared with the EPA TCLP acid-leaching procedures (Hassett and Pflughoeft-Hassett, 1993). These and other advanced characterization methods provide powerful tools for understanding the behavior of various by-products in diverse applications. Advanced analytical characterization can be systematically correlated with practical performance experience to provide an entirely new basis for classifying CCBs for optimum beneficial use. A scientifically based classification offering broad coverage of different by-product types and applications would assist greatly over time in providing the added assurance of safety and effectiveness needed to break down overly conservative practices currently prevailing under federal and state regulations.

### **Federal Regulations Applying to Coal Combustion By-Products**

The Resource Conservation and Recovery Act of 1976 and the 1980 Solid Waste Disposal Act Amendments provide for comprehensive cradle-to-grave regulation of solid waste generation, collection, transportation, separation, recovery, and disposal (Jagiella, 1993; Findley and Farber, 1992; Butler and Binion, 1993). Subtitle C of RCRA and its implementing regulations impose specific federal requirements on materials deemed to be "hazardous," either because of being listed by EPA as hazardous or by reason of having hazardous or toxic characteristics. Subtitle D of RCRA delegates regulation of nonhazardous solid wastes to the individual states. In its original form, RCRA did not specify whether CCBs fell under Subtitle C or D. The 1980 amendments temporarily excluded CCBs from Subtitle C regulation pending an EPA study report addressing appropriate classification. In the interim, CCBs were subject to regulation under state laws pertaining to solid wastes.

On August 2, 1993, EPA presented its final regulatory decision on fly ash, bottom ash, boiler slag, and flue gas emission control waste (40 CFR Part 261), stating that, effective September 2, 1993, these materials are not regulated as hazardous wastes under Subtitle C and officially placing them under Subtitle D as solid wastes under the jurisdiction of individual states. Further evaluation will be made by EPA of hazardous or toxic properties of industrial solid wastes, but at this time, CCBs are expected to remain under state regulation where little positive change is expected regarding beneficial use.

An important barrier issue originating in RCRA legislation is the indiscriminate designation of CCBs as solid wastes, whether they are recovered for use or disposed of in a landfill. In the absence of special state exemptions from solid waste regulations for beneficial use, which exist in only a few states, the "waste" designation can trigger case-by-case approval and permitting procedures that discourage CCB use because of unreasonable cost and delay. The remedies for this barrier problem include both the elimination of the "waste" designation and the creation of appropriate exemptions from regulation based on environmentally sound regulatory classifications for various classes of by-product use.

While RCRA is the principal federal law affecting the regulation of CCBs, a larger statutory framework of federal laws that are more or less integrated with state and local statutes may ultimately have to be considered. It is not within the scope of this study to unravel this potential regulatory maze. However, other federal statutes that potentially apply to CCB use or disposal in particular circumstances, as well as to virgin raw materials and derived products, include the Clean Water Act of 1972, the Safe Drinking Water Act of 1974, the Toxic Substances Control Act of 1976, and the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA, the Superfund Act). All of these statutes deal with the control of toxic substances and ultimately rely on environmental testing

and risk assessment to establish regulatory criteria. The final answer to regulatory questions constituting barriers to beneficial use, therefore, lies in obtaining adequate environmental data to demonstrate environmental safety, a process which is well advanced for CCBs but requires systematic compilation and refinement to provide the basis for regulatory classification.

### **State Regulation of Coal Combustion By-Products**

Limited information has been gathered and reported that defines and discusses state regulations pertaining to CCB utilization and barriers to utilization. State regulations have been summarized in a survey of use and disposal provisions (Jagiella, 1993). Hudson and others (1982) discussed barriers to CCB utilization in Maryland, Alabama, Illinois, New York, Pennsylvania, Ohio, Texas, Virginia, and West Virginia. Changes in regulations and practices relating to CCB utilization in these states have been noted in more recent works. The Texas Coal Ash Utilization Group (TCAUG) (1992) addressed regulatory issues in Texas in a recent report. The *Pennsylvania Bulletin* (1992) discussed current regulatory issues in Pennsylvania. These summaries point out that most state regulation of CCBs is designed to regulate disposal. Very few states have regulations regarding utilization of CCBs, either allowing or disallowing use. Common uses include concrete paving by state highway departments. A summary of state department of transportation CCB utilization specifications was prepared by the EERC (Docket, 1994) for the Coal Ash Resources Research Consortium. This report also summarizes the percentages of fly ash typically allowed as a mineral admixture and other utilization applications accepted by the state departments of transportation where specified, although these are not common.

### **Legal Barriers**

It is important to include a brief discussion of the key legal barrier to CCB utilization, which is the potential for environmental liability. Other issues involving commercial law and patents pose limited constraints of much less significance. The most serious environmental issue centers on the wide divergence in the legal and regulatory treatment of the beneficial use of CCBs under state laws. Whereas EPA confirmed in a ruling on August 2, 1993, that CCBs (fly ash, bottom ash, boiler slag, and FGD material) are not hazardous materials under RCRA Subtitle C; the delegation of regulatory authority under RCRA Subtitle D for solid waste allows various states to regulate the use and disposal of CCBs by very different standards (EPA, 1993). Some states restrictively control CCBs as a de facto hazardous material, while other states treat recycled ash as an unregulated construction material (Jagiella, 1993). Some states regulate CCBs on a case-by-case basis. In recent years, several states have adopted statutes prohibiting the importation of solid wastes. Although these statutes have been regularly overruled as restraint of trade, their temporary status has impeded ash sales in some instances.

The principal federal statute affecting the regulation of solid waste, and therefore related beneficial use, is RCRA. Other federal environmental statutes that may affect barriers to CCB utilization are the Clean Water Act and CERCLA. A 1988 summary of state statutes compiled by the Utility Solid Waste Activities Group (USWAG) identified 43 states that exempt CCBs from hazardous waste regulations; seven states-Kentucky, Tennessee, Oklahoma, Washington, New Jersey, Maine, and California-require testing to determine whether the ash would be regulated as a solid waste or a hazardous material and one state, Ohio, exempts CCBs from both solid and hazardous waste regulations (Wald and others, 1983).

Legal review is needed to clarify the grounds and remedies that apply to environmental liability. As a general consideration, statutory liability under environmental law is not based on fault and imposes strict responsibility without regard to negligence. Tort law, on the other hand, applies where a dangerous condition can be traced back to the point of manufacture of a product, which is not a condition that commonly applies to CCB utilization. The commonly held opinion, that semantic reclassification of CCBs as a product rather than a solid waste would by itself simplify regulatory liability, appears to have little legal validity since the intent of the statutes would not change and their wording could be readily adapted. Also, compliance with one statute would not remove jeopardy on others: therefore, compliance

with state regulations under delegated RCRA authority does not prevent liability under the Clean Water Act or CERCLA. The CERCLA statute appears to be the broadest statute covering hazardous materials that present "substantial danger to public health or welfare or the environment," and it incorporates by reference any substance designated as hazardous or toxic in the Clean Water Act or RCRA (Findley and Farber, 1992). CERCLA places strict liability for remediation and restitution on the party responsible for the hazardous material without regard to negligence. However, it is very significant to note that petroleum and natural gas are specifically exempted from liability under CERCLA. This type of exemption from liability establishes a precedent that could appropriately be considered in legislation for CCBs, owing to its importance as the largest- volume recyclable material in the United States and the record of environmental testing that indicates CCBs are not hazardous substances, pollutants, or contaminants.

Other legally recognized remedies for environmental liability, apart from statutory exemption, involve demonstration of compliance with a regulatory authority based on recognized technical specifications and environmental criteria. Improved regulatory classification of CCBs for use in various classes of applications would help to reduce environmental liability by providing background and specificity for legally defending particular utilization practices. By controlling the end use of CCBs, utilities and marketers can limit their liability by providing material only for those uses that are demonstrated to be environmentally safe (Hudson and others, 1982). More effectively, exemption from regulatory control as solid wastes under RCRA could be provided for pre-approved classes of by-product use. Although such federal deregulation of pre-approved products may be politically difficult, it would permit approved CCBs to move into unrestricted interstate commerce. Federal regulatory clarification and improved specifications would, at a minimum, provide leadership and direction for state regulators.

Some difficulties may exist in applying commercial or contract law to the sale of CCBs because of the current lack of both technical specifications and environmental criteria applying to some uses. Suggestions have been advanced for developing a uniform commercial code for by-product transactions that would incorporate specifications to assist buyers and sellers in writing clear and enforceable contracts. Legal research is needed to establish the usefulness of this approach. As better specifications are incorporated, quality control in the production of CCBs becomes a more significant factor in meeting legal responsibility (Hudson and others, 1982).

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## LUNCHEON REMARKS

Kathrine L. Henry, Acting Director  
Office Of Surface Mining  
U.S. Department Of The Interior

It is indeed a pleasure to be here today at Southern Illinois University for three **days** of discussion and information sharing on an important environmental topic. I am glad that so many people from so many parts of the country are participating, from all levels of government, and from industry, universities, and the general public. This is an excellent venue for communicating problems, solutions, and concerns related to coal combustion by-products associated with surface mining. I especially want to thank our colleagues from Southern Illinois University for hosting this forum, and I'd like to recognize the other sponsors, as well:

- . The Illinois Clean Coal Institute
- . Illinois Office of Mines and Minerals
- . Indiana Division of Reclamation
- . Texas Utilities, Inc.

Most of us think of what's left after burning coal as simply "ash." But as with so many environmental aspects of coal mining and coal use, it turns out there's much more to it than that. As a lawyer, I promise not to come before this audience as a technical expert, because my area of expertise is in the law, not chemistry, soil science, agronomy, hydrology, or any of the related fields we depend on for scientific solutions to environmental problems related to mining and using coal. But I would like to summarize where we are in dealing with issues related to coal combustion by-products, and how we got there.

In May of 1994, OSM solicited recommendations for technical studies and applied research topics from the states, industry, and public interest groups. A wide variety of them identified the field of coal combustion by-products as a priority topic for consideration. In September of 1995, OSM began assessing interest in a national interactive forum on coal combustion by-products associated with coal mining. Then in February of 1996, a steering committee was formed to organize and develop such a forum. That brings us to today. The meetings we are now participating in are the result of the combined efforts of numerous interested parties to make this timely and much needed discussion possible.

Back when the Surface Mining Control and Reclamation Act (SMCRA) was passed in 1977, planning for any significant utilization or disposal of coal combustion by-products at surface coal mines did not seem like much of a concern. OSM regulations provide guidance for protecting surface and ground water quality. They also require specific plans for the disposal of coal cleaning wastes and non-coal wastes normally associated with on-site repair shop facilities. Neither SMCRA nor the OSM regulations, however, specifically address the use or disposal of the by-products of electric power generation at surface coal mines, even though a truly systematic plan for producing and using coal would logically take into consideration what to do with the final waste products, things like ash, for instance. After all, according to the First Law of Ecology, *Everything Has To Go Somewhere*.

Since those early days of SMCRA, dramatic changes have taken place. More people are starting to take a more full-systems, full-circle look. U.S. coal consumption is now approximately one billion tons per year. It's increasing at a rate of 2 percent per year. Eighty percent of that coal is used as fuel for the generation of electrical power. Major environmental legislation passed during the 1970s and more recently in 1990, has led to significantly increased removal of contaminants associated with coal fired electric power generation. That is, power plants practically everywhere are requiring cleaner coal, and clean air regulations are requiring more emission control. It's working. Sulfur emissions produced by burning coal have *declined* by 11 percent for the entire U.S. over the period 1970 through 1992, while at the same time coal consumption *increased* by 70 percent.

Where did all that “stuff” go? Controls on particulates and acid forming chemicals from electric utility stack gases plus the addition of more stringent controls on water discharges and waste disposal have provided the nation with effective solutions for improving air and water quality at electrical power plants, no doubt about it. But those very solutions to the problem of air and water contaminants associated with coal fired power plants have generated new challenges for the coal mining industry and for the state and federal programs charged with environmental regulation of coal mining activities. A big part of the challenge comes from the production of large volumes of fly ash, bottom ash, boiler slag, spent bed material from fluidized bed combustion, and flue gas desulfurization residues, collectively referred to as coal combustion by-products. That’s why I no longer think of it simply as “ash.”

More important to the discussions at this forum is the idea of taking the system full-circle, that is, looking at the potential for coal mines to be disposal sites for excess solid residues, and a potential market for products derived partly or wholly from coal combustion by-products. Logically, it makes perfect sense.

To give an idea of the immense volume of coal combustion by-products being generated, our late lamented sister agency, the U.S. Bureau of Mines, provided an estimate. They calculated that a typical power plant burning 100 rail car loads of high sulfur coal (3.5 percent sulfur) will produce 30 car loads of coal combustion by-products. Total annual production of such residues in the U.S. has risen from approximately 68 million tons, at the time of the passage of SMCRA in 1977, to over 100 million tons today. The total could exceed 170 million tons by the year 2000. That’s a 240 percent increase over a period of just 23 years. As I said before, Everything Has To Go *Somewhere*.

Although a major emphasis of this administration and the electric utility industry has been to recycle coal combustion by-products into economically viable commodities, the recycling of coal combustion by-products has remained steady at around 25 percent over the last decade. Potential uses on the mine site have included:

- Injection as fill into old underground mines to combat or reduce subsidence.
- Use as a soil amendment to neutralize acidic spoil and thereby reduce acid mine drainage.
- And as a an ingredient in synthetic concrete substitutes for traditional underground mine timbers.

OSM supports those efforts to recycle coal combustion by-products into commercial items for use on or off the mine site. Despite everything that’s been done to create economically viable products for those residues, however, only about one-quarter of them are used in that way. The other 75 percent of the coal combustion by-products still has to be stockpiled or disposed of, *somewhere*. Interest in coal mines as potential disposal facilities or markets for new products produced from coal combustion by-products has gone up with the dramatic cost increases and mounting difficulties involved in handling those residues on site at coal fired power plants. Typical costs for disposal of solid residues produced by coal combustion in a new disposal facility are at least two to three times what it costs to dispose of them at existing sites, So why not keep on using the existing sites? Well, many existing disposal facilities are just about full. When they reach their design capacity, the next step is to replace them with new disposal facilities that, unfortunately, cost lots more.

In 1993, the Environmental Protection Agency issued its final regulatory determination that coal combustion by-products are deemed non-hazardous and are to be regulated by the individual states under Sub-title D of the Resource Conservation and Recovery Act, when disposed of as solid waste. As a result, the states have been challenged to develop appropriate strategies for integrating the concerns of state solid waste programs with SMCRA programs regarding disposal on permitted coal mine sites.

When the use or disposal of coal combustion by-products happens at surface coal mines, state coal mining regulators are involved to the extent that SMCRA requires:

- The mine operator to ensure that all toxic materials are treated, buried, and compacted, or otherwise disposed of, in a manner designed to prevent contamination of ground or surface water.
- Making sure the proposed land use does not present any actual or probable threat of water pollution.
- And ensuring the permit application contains a detailed description of the measures to be taken during mining and reclamation to assure the protection of the quality and quantity of surface and ground water systems, both on- and off-site, from adverse effects of the mining and reclamation process, also to assure the rights of present users of such water are protected.

Any disposal of coal combustion by-products at mine sites must be in accordance with those standards, and with applicable solid waste disposal requirements. The states differ in their regulatory requirements for disposal of coal combustion by-products as solid waste. Trace element concentrations in coal combustion by-products vary according to where the coal came from. Chemical and physical characteristics differ by region, as do mine site conditions. Accordingly, regulatory programs to allow use or disposal must be designed to handle those differences. At OSM, we are supportive of state efforts to develop appropriate methods and criteria. We will do what we can to help, on request.

Currently, the debate over use or disposal of coal combustion by-products at coal mines centers on the potential for the materials to release toxins back into the environment. We recognize that improved knowledge of the risks and benefits associated with disposal and use of coal combustion by-products, is badly needed, as is a greater acceptance of that knowledge by regulators and the public. The more we know, the more options we have. So I look forward to the discussions that will take place at this forum. I am optimistic that constructive dialogues such as those held here will lead to better understanding of the benefits and risks involved with using coal combustion by-products associated with coal mining.

Thank you for inviting me here today. I commend all the forum participants for being part of this valuable information exchange. The public and the coalfield residents can only benefit from the information that is shared and the knowledge that is gained at this event. I thank you for applying your minds to the task and I wish you success in your efforts on behalf of the coalfield environment.