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RESEARCH AND DEVELOPMENT FOR FLY-ASH UTILIZATION

DP/CPR/86/007

PEOPLE'S REPUBLIC OF CHINA

Technical report: Fly-ash utilization in the construction industry*

Prepared for the Government
of the People's Republic of China
by the United Nations Industrial Development Organization,
acting as executing agency for the United Nations Development Programme

Based on the work of Mr. D. M. Golden,
expert in the utilization of fly-ash in mass amount

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United Nations Industrial Development Organization
Vienna

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Report on UNIDO Coal Ash Utilization Mission to Shanghai, China
by
Dean M. Golden, P.E.

Section I - Introduction

Under the terms of my agreement with UNIDO I delivered a series of lectures on fly ash utilization at the Shanghai Research Institute of Building Sciences (SRIBS) during my nineteen day mission to Shanghai. Prior to my arrival in Shanghai, I had worked out a series of thirteen lecture topics to include all five of the topical areas included in my mission agreement. The topical areas included in the original agreement are as follows:

1. General Condition
   - (a) Total fly ash discharged in USA, general utilization rate for various applications;
   - (b) Organization for fly ash utiliztion research;
   - (c) Main problems encountered in utilization of fly ash and solutions;
   - (d) Development trends for fly ash utilization.
2. Fly ash Recycling Techniques
   - (a) Collection, separation, discharge and storage of fly ash in power plants;
   - (b) Separation and processing of qualified fine ash and conditioned ash;
   - (c) Separation for microspheres;
   - (d) Application of microsphere in plastics and insulative materials.
3. DAL Technology
   - (a) DAL technology and present status;
   - (b) Economic comparison for DAL and other technology;
   - (c) Properties of products extracted by DAL
   - (d) Purpose and market potential for these products
   - (e) Application of spent ash.
4. Roller Compacted Concrete
   - (a) Maximum fly ash admixture and optimum mix design;
   - (b) Design for configuration of road;
   - (c) Construction methods;
   - (d) Economic effect.
5. Other Aspects
   - (a) Fly ash used as filler in asphalt mix;
   - (b) Fly ash brick, maximum fly ash admixture, production method, porosity and technological and economic appraisal;
   - (c) Artificial reef bed. Composition and raw material, properties of blocks and their shape, effect on fishing industry and environment;
   - (d) Fly ash used for coagulant;
   - (e) Desulphurization techniques in power plants.

The objective of my UNIDO sponsored mission to Shanghai was to assist the Shanghai Research Institute of Building Sciences in further enhancement of the utilization potential of coal fly ash. The primary areas of interest are: fly ash separation techniques, microsphere utilization, metal extraction, roller compacted concrete and other miscellaneous applications for ash. The lectures were spaced over seven days covering thirteen distinct subject areas. Visits to a coal fired power plant, a fly ash block factory, and a cement plant, and a year old steel plant were made prior to the lectures so that common areas of use could be noted in the lectures.

Section II - AUDIENCE

In addition to staff members from the Shanghai Research Institute of Building Sciences (SRIBS), the audience included people from a total of thirty-three different organizations. The organizations included industial entitities such as electric utilities, cement companies, building companies, fly ash companies, the power industry bureau, and universities, research organizations for building material, building construction and management, environmental control systems, electric power research, and governmental authorites such as the Shanghai Science and Technology Committee. Sixteen of the organizations represented are located in Shanghai, and seventeen organizations are from outside of Shanghai. People came from six provinces or cities, including Jiangsu, Zhejiang, Hubei, Hunan, Beijing, in addition to Shanghai. Based on the level and number of questions, the level of attention was very high. The simultaneous translation worked very well. The translators were very capable. As background material I had sent a number of the Electric Power Research Institute (EPRI) reports and papers on ash utilization that served as the basis for my lectures.

Section III - Current Ash Utilization Activity in Shanghai

An introduction into the current research program at the Shanghai Research Institute of Building Sciences was given during my first two days in Shanghai (Dec. 1-2, 1987). Various common technical issues were discussed throughout these introductory meetings. A tour of the laboratory facilities of SRIBS was made. The facilities are quite adequate for the type of research undertaken by SRIBS. The laboratory equipment used is very similar to that which would be found in a civil engineering laboratory at a large university in the United States.

Prior to and during my visit to Shanghai I reviewed a number of publications of the Shanghai Research Institute of Building Sciences to provide a basis of comparison in the coal ash types produced in the two countries. This was important because an understanding of the similarities and differences was necessary to properly transfer the results of the applied research from one country to the other.

At the present time, the annual world combustion of coal in electric plants is about 2500 mtce (million metric tonnes coal equivalent), resulting in the production of 250-300 million tonnes of fly ash. It has been predicted that consumption will increase to about 6500-7000 mtce annually in the year 2000., resulting in the collection of some 650-850 million tonnes of fly ash. The current usage of coal by utilities in the United States results in the production of over 65 million tonnes (71 million short tons) of solid combustion by-products each year. In China the total production of coal ash produced by electric power plants is now 43 million metric tons per year. The ash utilization rate for the Peoples Republic of China is estimated to be 22 percent which is similar to the overall rate in the United States, currently estimated to be 28 percent. Given the large coal reserves in China and the industrialization process that is occuring, it is likely the China will become the largest user of coal for electricity production in the early part of the next century. Given the large population and shortage of land for disposal of the ash by-products it is of vital importance for China to develop new ash utilization options to minimize the environmental impact of the industrialization process.

The tables that follow summarize the chemical, mineralogical and engineering property data from the SRIBS reports and from the EPRI reports which were written under my direction.

Chemical Content of 30 Chinese Fly Ashes

Parameter	Range	Average
LOI	1.29 -- 26.46	6.82
SiO_2	41.50 -- 60.76	51.68
Al_2O_3	14.13 -- 31.64	26.90
Fe_2O_3	3.77 -- 13.04	7.89
CaO	1.22 -- 9.58	3.26
MgO	0.22 -- 1.93	0.95
SO_3	0.13 -- 1.03	0.39
K_2O	0.27 -- 2.68	1.19
Na_2O	0.14 -- 0.60	0.29

Source: Fly Ash Quality and its Rapid Assessment, Z. Z. Gu, SRIBS, 1986.

Chemical Content of Fly Ashes from 27 Power Stations in 13 Provinces of China

Parameter	Range	Average
LOI	1.10 -- 26.46	6.45
SiO_2	31.12 -- 60.76	51.30
Al_2O_3	11.88 -- 35.58	27.70
Fe_2O_3	3.77 -- 37.52	8.23
CaO	0.76 -- 9.58	3.02
MgO	0.11 -- 1.93	0.98
SO_3	0.11 -- 1.84	0.43
K_2O	0.27 -- 2.68	1.11
Na_2O	0.10 -- 0.60	0.29
S-SiO_2	1.12 -- 7.20	3.13
S-Al_2O_3	1.09 -- 10.85	3.52
Melting Pt °C	1068 -- 1559	1442

Source: *Rapid Assessment of Activity of Fly Ash Used as Admixture in Cement and Concrete*, Yue M., Wang W., Tang C., and Wang Z., SRIBS,'86.

Mineral Composition of Fly Ashes from 27 Power Stations in 13 Provinces of China

Parameter	Range	Average
α-Quartz	0.90 -- 18.50	8.11
Mullite	2.70 -- 34.10	21.24
Hematite	0 -- 4.70	1.08
Magnetite	0.40 -- 13.80	2.78
Glass	50.20 -- 79.00	60.35
Glassy SiO_2	18.60 -- 45.90	37.10
Glassy Al_2O_3	7.00 -- 22.80	12.57

Source: *Rapid Assessment of Activity of Fly Ash Used as Admixture in Cement and Concrete*, Yue M., Wang W., Tang C., and Wang Z., SRIBS,'86.

Physical Properties of Fly Ashes from 27 Power Stations in 13 Provinces of China

Parameter	Range	Average
Sp. Gravity	1.92 -- 2.85	2.13
Density, Kg/m^3	531 --1261	780
Compactness,%	25.6 -- 47.0	36.5
Surface area, BET	0.8 -- 19.5	3.40
Surface area, Blaine	0.1176 - 0.6531	0.3298
Fineness, >200μm	0.22 -- 32.76	4.17
Fineness, >80μm	0.6 -- 77.8	22.23
Fineness, 80 - 45μm	2.2 -- 32.5	17.99
Fineness, <45μm	13.4 -- 97.3	59.78
Water Demand,%	27.3 -- 66.7	48.0

Source: *Rapid Assessment of Activity of Fly Ash Used as Admixture in Cement and Concrete*, Yue M., Wang W., Tang C., and Wang Z., SRIBS,'86.

Chemical Properties of Sixteen Fly Ashes From Power Stations in Five Regions of the USA

Parameter	Range	Average
Silicon Dioxide,%	30.92 -- 62.76	47.94
Aluminum Oxide,%	12.30 -- 26.95	20.53
Iron Oxide,%	2.84 -- 24.43	10.71
Calcium Oxide,%	1.10 -- 30.53	11.13
Magnesium Oxide,%	0.69 -- 6.69	2.48
Sulfur Trioxide,%	0.31 -- 3.85	1.30
Sodium Oxide,%	0.20 -- 2.04	0.76
Potassium Oxide,%	0.22 -- 3.03	1.62
Available Alkalies,%	0.23 -- 1.54	0.76

Source: *Classification of Fly Ash for Use in Cement and Concrete,* Gumtz G.,White, E , EPRI Report CS-5116, April 1987.

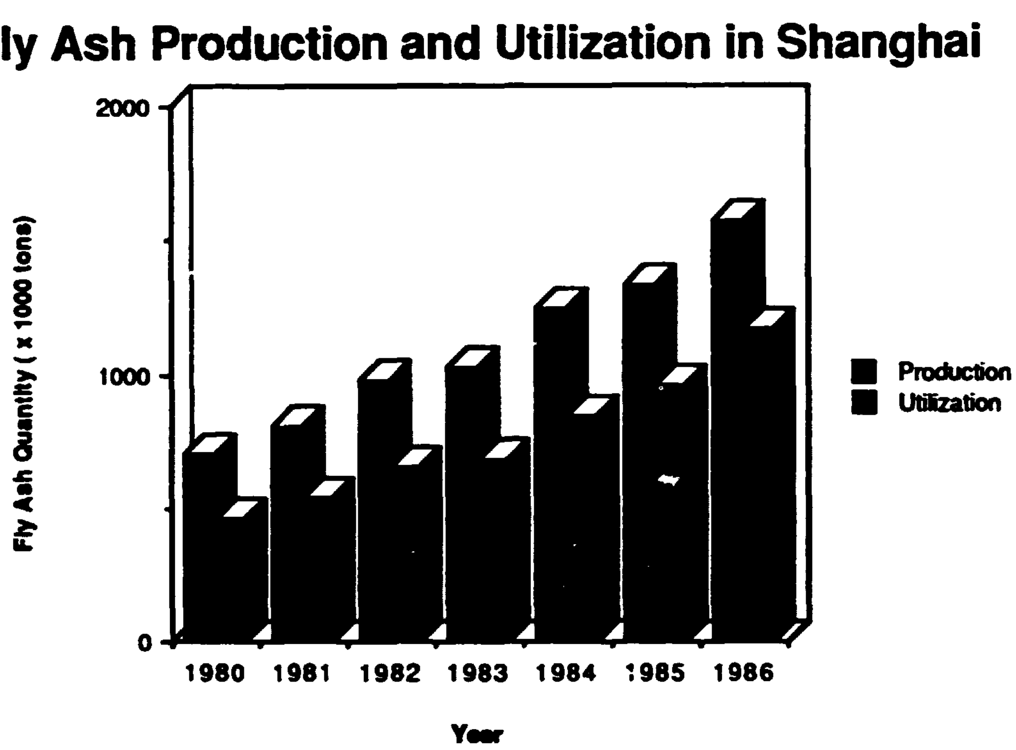
Physical Properties of Sixteen Fly Ashes From Power Stations in Five Regions of the USA

Parameter	Range	Average
Loss on Ignition,%	<0.01 -- 16.60	2.28
Carbon,%	0.02 -- 15.34	1.56
Specific Gravity	2.14 -- 2.69	2.48
Moisture Content,%	0.0 -- 0.38	0.08
% Ret. on 325 mesh	3.55 -- 36.90	17.75
Blaine Fineness	1579 -- 5550	3839
Pozzolanic Act. Index	86 -- 239	113

Source: *Classification of Fly Ash for Use in Cement and Concrete,* Gumtz G.,White, E., EPRI Report CS-5116, April 1987.

There has been a remarkable increase in the ash utilization rates in the Shanghai metropolitan area over the past decade, even with the increased production of fly ash. As noted previously, the utilization rates in the rest of China are not nearly as high. The bar graph on the next page compares the production and utilization quantities during the current decade.

Fly Ash Production and Utilization in Shanghai



Source: *Fly Ash Utilization in Shanghai, China,* Gu, Z.Z., Lu, J.G., Wang, F.Y., EPRI Report No. CS-5362, October 1987.

The three primary ash utilization applications in the Shanghai area are walling material, cement production, and road construction. Fly ash walling material includes brick, block and panel among which block is the largest single field application with annual production of 240,000 cubic meters. During my visit I was given a tour of the fly ash block production plant in Shanghai.

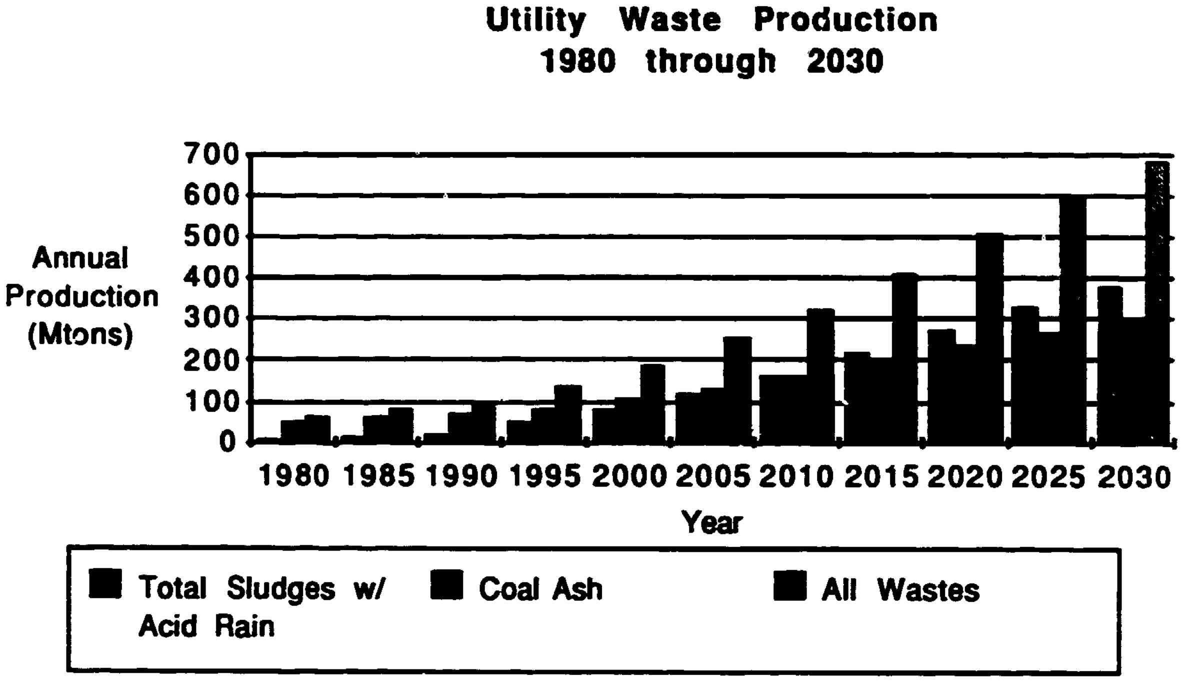
Section IV - LECTURES

Lecture 1: (Dec. 5) Ash Utilization Conditions in the USA

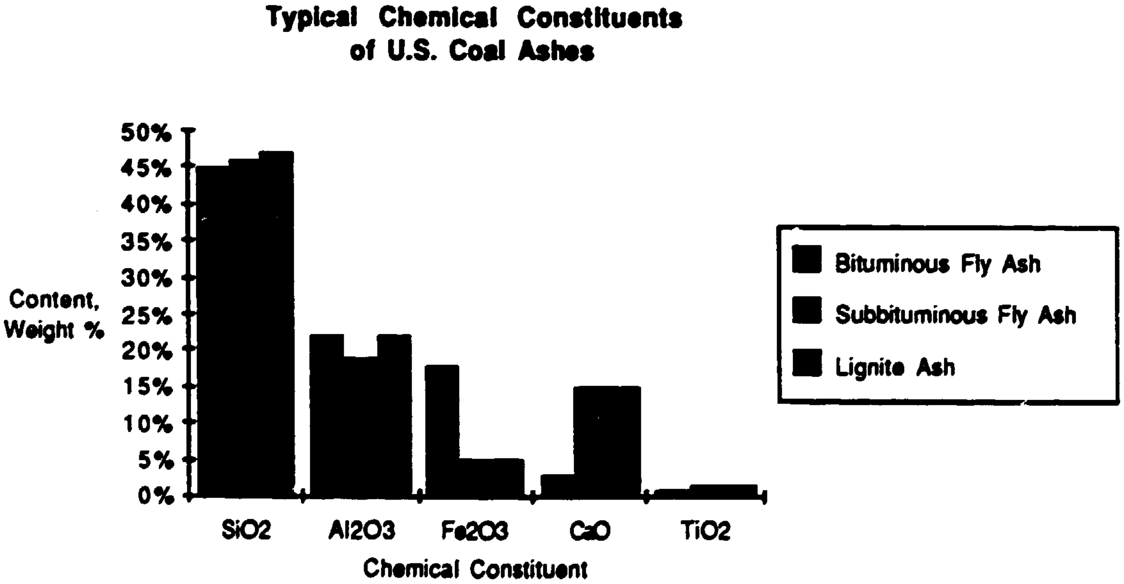
(1) Total fly-ash discharged in USA, general utilization rate for various applications

The emphasis of the environmental regulations in the United States of America has changed during the past decade. The march of enviromental regulations, Clean Air Act (1970), Clean Water Act (1972), Toxic Substances Control Act (1976), Resource Conservation and Recovery Act (1976), Superfund (1980),and SARA (1986) demonstrate the gradual tightening of controls. The air and water regulations result in equipment which pull particulates or compounds from air or water streams and create a solid waste. The problem for the industry and for the society which it serves is coping with this fundamental law of physics -- conservation of matter. It could be stated another way: "What you take out in one place, you have to put back someplace else or find some other use for it".

The largest single "emission" from a coal-fueled power station is the coal ash. Presently coal-fired power generation amounts to about 60% of the electric utility power in the United States by burning approximately 600 million tons of coal annually. This produces about 77 million tons of coal ash (60 million tons of fly ash, 12 million tons of bottom ash, and 5 million tons of boiler slag). The quantities of solid wastes are expected to increase dramatically over the next few decades as shown in the figure on the next page.



The chemical composition in terms of major components of fly ashes from bituminous, subbituminous, and lignite coals burned at US power stations are shown in the figure below. The data indicate that the average silica, alumina, and titania content of the three types of coal ashes are similar. Notable differences in composition are the higher iron oxides content of the subbituminous and lignite coal ashes.



Lecture 2: (Dec. 7) Fly Ash Utilization Research Organizations in the United States

In the United States of America there is not a centralized research organization dedicated entirely to conducting research into utilization of coal ash. In 1967 the American Coal Ash

Association (ACAA) was founded to provide a focal point to utilities and marketer of coal ash. This association sponsors the international ash utilization symposiums every couple of years to bring together researchers from around the world. The ACAA does not have a large enough budget to conduct a research program of its own. The typical annual budget is about $350,000. The Electric Power Research Institute (EPRI) was founded in 1973 to conduct research related to electric utility operations. The USA is somewhat unique amoung industrialized countries, in that it has nearly 3,600 electric utilities. From the 1920's to the late 1960's the cost of electricity decreased every year, do to economies of scale and technological advances. When the costs started to increase, the utility industry was criticised for having a very low expenditure rate for research and development compared to other industries. To prevent duplication among the thousands of utilities, EPRI was formed to centralize the R&D function. Over 500 utilities, representing nearly three-quarters of the generation capacity in the country are members. The annual research budget has exceeded $300 million for each of the last ten years. EPRI is the largest non-governmental research consortium in the world. Research in ash utilization began in 1979 in the Coal Combustion Systems Division at EPRI. The annual budget for waste disposal and utilization research has been between $1 and $2 million per year since 1980. I have managed that research program for its entire history.

EPRI is undertaking a multi-year program to promote the bulk sale of coal ash in high volume applications, primarily in the roadway construction sector. The project is specifically directed to applications that do not require a specific class or quality of fly ash. Some of these uses include:

- Fills
- Embankments
- Backfills
- Landfill Cover
- Soil Amendments
- Subgrade Stabilization
- Pavement Base Courses
- Grouting
- Slurry Walls
- Hydraulic Fills

The principal components of the existing ash utilization project include:

- Documentation of existing ash utilization projects
- Participation in new demonstration projects sponsored by EPRI and host utilities
- Documentation of Federal Highway Administration (FHWA) and other non-EPRI sponsored projects
- Preparation of draft specifications with cooperation of State highway and environmental agencies
- Preparation of a Design Manual for Ash Utilization
- Preparation of a Construction Manual for Highway Ash Utilization
- Development of a generic utility ash marketing program
- Preparation of brochures, films, and slides promoting ash use
- Educational seminars

A major objective of the EPRI ash utilization research project (RP 2422) is to promote the use of fly ash in highway construction applications to contractors, design engineers, and State highway departments. Since many of these groups consider coal ash a new or unconventional construction material, a major effort is underway to document the numerous successful applications of fly ash in general construction.

One of the major accomplishments of this project was the publication in 1986 of CS-4446 which identified over 270 existing projects using fly ash in high volume applications in the United States and Canada.

An important component of the overall program is the development of new demonstration projects, particularily in those areas of the United States where ash has not been widely used but where considerable market potential and utility interest in ash utilization exists. EPRI is supporting the development of candidate "seed" projects in five or six promising demonstration areas proposed by utilities. EPRI expects that through these demonstration projects, potential customers and highway departments will recognize that power plant ash by-products are acceptable building materials and economic substitutes for other natural products now in use. It is expected that the use of fly ash can be increased significantly throughout the United States because of the construction of these projects in areas where state governments and highway departments are receptive to increased by-product utilization.

The demonstration projects will be structured to show the environmental and technological acceptability of ash utilization in road construction in a controlled and monitored segment of a highway. The field demonstrations will also serve as test cases for a draft design manual and specifications developed for highway ash utilization. Near the end of the project, the design manual will be issued as an EPRI final report, and the specifications will be submitted for approval to the state highway agency in the states where the demonstrations are located. The field demonstration projects implemented under this project will be supplemented with information and data obtained from previous highway projects which used coal ash. This will provide information on long term durability and performance which otherwise would not be available in a five year project.

Four field demonstration projects have been approved to date for construction in the 1985-87 period. Additional projects will be approved in 1987 for construction in the 1988 time frame. The Figure below shows the location of the demonstration projects and the type of application.

The Electric Power Research Institute has been actively seeking to increase the utilization of coal ash through an applied research and development project begun in 1979, and has a multi-faceted project underway over the next five years to continue the research in three principal areas. The high volume applications in highway construction uses, medium volume uses in cement and concrete, and the high technology mineral extraction process development make up the triad of ash utilization research at EPRI.

Main problems encountered in utilization of fly ash and solutions

Historically, the use of fly ash in the United States has been primarily directed towards its use in concrete. This is an ideal example of waste product utilization since there are both technical and economical benefits from its use. However, the quantity of ash used in concrete is but a small portion of the volume produced. In fact, if ash was used in all the concrete made in the United States, it would still be only a minor percentage of the volume available. In 1983, only 27 % of all fly ash produced could have been used in cement at a 20 % replacement rate. In fact, only 14 % of all fly ash produced in 1983 was utilized in any form, with the remainder being placed in storage or disposal areas. It is estimated that only one-quarter of the ash produced in the United States will meet the requirements of ASTM C618-83 for concrete-quality fly ash. Therefore one of the major impediments to increased ash use may be the ASTM specification for fly ash as a mineral admixture in Portland cement concrete. This is inevitable given the inherent nature of specifications set up defining compositional or property limits within which a fly ash must fall to be used in certain applications. In general, it has been found that if a fly ash meets the specification ASTM C618-83 an acceptable product can be made with the fly ash. However, there are a sufficient number of exceptions to the general rule to cast doubt on the validity of the specifications and on the characteristics being specified. Information developed in a number of research programs on fly ash over the past twenty years throughout the world (including work under EPRI sponsorship) has led to increasing doubts not only about the ash characteristics being measured, but also about the validity of some of the methods used to measure the characteristics. The test results have shown that fly ash has several distinct functions in concrete. It is (1) a pozzolan, (2) a workability modifier, (3) a fine aggregate, and (4) an adsorbent of air-entraining agents. Additionally, the chemical properties of both fly and bottom ashes make them potentially useful raw materials for the manufacture of Portland cement clinker.

Development trends for fly ash utilization

In the future the ratio of ash generated to coal burned will become larger as electric utilities shift to burning more low-sulfur, high-ash coals because of environmental considerations, and as particulate collection devices used in flue gas cleanup become even more efficient. The use of advanced SO_2 control devices to capture emissions will also increase the quantities of solid by-products. For example a typical AFBC conversion will increase the production of by-products from 120 lbs per 1000 lbs of coal burned to 420 lbs of ash and spent sorbents.

Processing of Fly Ash in Power Plants

The second day of lectures concluded with a number of picture slides showing various types of equipment used in the USA for the separation and handling of the ash within power plants. The lecture reviewed the (1)Collection, separation, discharge and storage of fly ash in power plants; and (2)Separation and processing of qualified fine ash and conditioned ash.

Lecture 3: (Dec. 8) Fly Ash in Cement and Concrete: Roller Compacted Concrete

LECTURE 3 PART 1: FLY ASH IN CEMENT AND CONCRETE

This portion of my lectures described a project funded by the Electric Power Research Institute (EPRI) to develop a fly ash classification model which would aid electric utilities in assessing the suitability of a given fly ash for use in cement/concrete applications. The full utilization potential for fly ash in cement / concrete applications has been held back by the inadequacy of present specifications and methods for testing fly ash. Because of the variability in fly ash from one plant to another and from sample to sample from the same power plant, specifications have been established defining compositional or physical property limits which a fly ash must fall within in order to be used in cement/concrete. This lecture discussed the basis for a model to predict the performance of a given ash in cement / concrete applications based on the composition of fly ash. In this model, fly ash is described by two parameters, maximum efficiency E_m and the lime binding capacity n.

Significant amounts of fly ash already are being used for cement / concrete admixtures in industrialized countries around the world. For example, cement / concrete uses consumed 3.5 million short tons in 1983 in the United States or approximately 40% of all of the ash utilized that year. There is however considerable room for expansion of sales compared to the European usage rates. As the research and development arm of the U.S. electric utility industry, one of the Electric Power Research Institute's (EPRI) goals is to assist utilities in maximizing the utilization of fly ash in cement/concrete applications. Based on a 20% replacement of ash for cement, the potential use in the U.S. market could be 12 million short tons annually. In China the shortage of cement could be alleviated by even higher replacement levels than the 20% level used in the USA.

Limited standards and guidelines already exist, primarily for the cement/concrete industry itself, relating to fly ash usage in cement and concrete. However, ash classification guidelines directed specifically at aiding the electric utility industry in utilizing or marketing their fly ash for cement/concrete applications do not presently exist. The primary goal of the EPRI funded Fly Ash Classification Project is to develop such guidelines. One of the goals of my UNIDO mission to Shanghai was to provide an introduction to the ash classification model developed under the EPRI study.

The inadequacy of present specifications and test methods for fly ash was identified by the EPRI Project Advisory Committee established in 1984 to guide the EPRI ash in cement/concrete products research project. The development of a new fly ash classification was given the highest rating of several research areas which were proposed. For this reason one of the objectives established for the fly ash cement/concrete use project is to integrate new and existing knowledge relating to the effects of using fly ash in cement/concrete products. Existing criteria for determining the suitability of fly ash for cement/concrete use will be validated, and new fly ash classification guidelines developed where existing ones are deemed inadequate for the purposes of the electric utility industry.

A further complication in determining whether a particular fly ash would be suitable for a given application is the fact that different specifying bodies do not agree on specifications. For example, specifications for fly ash to be used as an ingredient in concrete have been established by ASTM, U.S. Bureau of Reclamation, U.S. Army Corps of Engineers, some State Highway Departments, several large municipalities, and some large contractors. The characteristics measured, the methods employed to measure the characteristics, and the specified limits for those characteristics vary. In the broadest sense, the basic objective of the EPRI ash in cement/concrete use project is to fill in some of the more critical gaps in the fundamental knowledge about the characteristics of fly ash and how they affect its behavior. The ultimate aim is to develop a meaningful and reliable method of evaluating fly ash that will allow the use of fly ash in concrete with confidence.

The Fly Ash Classification Project consists of an ash sampling and testing phase to characterize physical and chemical ash properties, followed by concrete testing of selected ashes. The concrete testing was conducted to determine the performance of concrete mixtures incorporating fly ash. The project's primary goal is to relate concrete performance to the physical and chemical characteristics of representative fly ashes. A fly ash classification system, will assist utility or marketing firm personnel in assessing potential fly ash usefulness in cement/concrete applications.

Because of the various coal ash types in the U.S., five regional contractors were selected to perform the ash sampling and testing work in their respective regions. Each contractor obtained fly ash samples from three pre-selected power plants in the region, analyzed the ash for specified chemical and physical properties, and subsequently prepared various concrete mixes containing fly ash and other admixtures and conducted specified concrete performance tests on the mixes.

This data was then compiled by a project coordinating contractor (Baker Engineers) and incorporated along with existing literature data, into the development of predictive models relating fly ash characteristics to resulting cement/concrete properties. The models were based on cement/concrete chemistry as well as empirically observed correlations.

Fly ash was collected for analysis from fifteen power plants from five different regions of the USA. The power plants were selected as ash sources based on the following criteria:

a. Boiler Type - Pulverized Coal.
b. Fly Ash Removal System - ESP or Baghouse
c. Fly Ash Collection System - Dry Conveyance
d. Coal Source - Each plant should have a somewhat differing coal source.

The following two types of ash samples were collected from each ash source:

<u>Base load</u> fly ash samples were taken when the generating unit was operating at base load, i.e., normal plant operating conditions.

<u>Upset condition</u> fly ash samples were taken under rapidly changing or other load conditions (e.g., startup, shutdown, peak load or other operating conditions which would be expected to produce a different, but not necessarily unacceptable quality ash when compared to base load operations).

The ash sampling methods conformed to ASTM C311 procedures with the preferred sampling location at the point of ash delivery to bulk storage. All samples, regardless of collection point, were a composite of a minimum of 13 discrete samples collected at appropriate time intervals. Final composite mixing was accomplished through tumbling or shaking the sample for an extended period or by mixing with a mechanical ash blender.

All ash samples were analyzed for the following chemical and physical properties:

CHEMICAL	PHYSICAL
SiO_2	Specific Gravity
Al_2O_3	325 Sieve Fineness
Fe_2O_3	Blaine Fineness
CaO	• as received
MgO	• ≤ 325 sieve
SO_3	Moisture
Na_2O	Pozzolanic Activity Index
K_2O	• Control #1
Available Alkalies	• Control #2
LOI	Particle Size
Carbon/fixed carbon	Sulfate resistance

Typical concrete mix designs were selected and concrete mixes were prepared in the following three general categories:

CATEGORY A: Concrete mixes with no admixtures.
CATEGORY B: Concrete mixes with water reducing admixture.
CATEGORY C: Concrete mixes with air entraining agent and water reducing admixture.

Categories A and B included control mixes with no fly ash and mixes with 15% and 30% by weight replacement of the Portland cement with fly ash. Category C included control mixes with no fly ash and mixes with 30% by weight replacement of the Portland cement with fly ash.

In each mix category, the control mixes included: (1) control mixes with the same amount of cementitious material as the fly ash mixes, and (2) control mixes with the same amount of Portland cement as the fly ash mixes. Admixture dosage was based on the total volume of cemetitious material (cement and fly ash) at the lower end of the manufacturer's recommended dosage range.

The making, curing and sampling of concrete test specimens conform to ASTM C192 with the following additional restrictions: In an attempt to better simulate field conditions, the concrete was mixed for 3 minutes followed by a 3 minute rest, followed by a 2 minute mixing period (as specified in ASTM C192) and then covered to prevent evaporation. Then "periodic agitation" was applied for 20 more minutes. "Periodic agitation" was defined as agitation for 30 seconds every 5 minutes during the 20 minute period.

CLASSIFICATION MODEL DEVELOPMENT

The lecture descibed the methods used to develop the model and the guidelines which were used as a basis for the predictive model development:

Simplicity. Every nuance of cement/concrete behavior modification due to fly ash addition was not considered, a univerally applicable "global" model was not desired.
Phenomenological basis. "Real world" characteristics of fly ash cement/ concrete admixtures

served as a basis for model development. Observed and observable phenomena were to be reasonably portrayed, relating the precision with which both ash characteristics can be determined and test results can be extrapolated to full-scale applications.

Ease of application. Based upon fly ash and cement properties, application of the model to provide guidance for cement/concrete use should be relatively elementary and straightforward. That is, only simple calculational, nomographic or graphical procedures should be required.

Predicative basis. To use the model, knowledge of basic fly ash, cement and concrete properties would be necessary to permit an assessment of potential applications. Uncertainties in data as well as the model's predictive capabilities will lead to uncertainty regarding applications in some instances. Model verification should provide a basis for assigning such uncertainty.

Indicative guidance. Application of the model with appropriate "basic" fly ash and cement/concrete characteristics need provide only an indication of potential applications, not definitive estimates of concrete mix properties required for these applications. That is, model application is not meant to replace the need for ash-cement/concrete testing for construction. Only guidance for the electric utility industry in terms of potential by-product use is required. Detailed estimation of the complex chemistry of cement and other pozzolans, the relation of cement/pozzolan chemistry and mineralogy to the structural/physical properties of concrete, etc. are beyond the scope of the EPRI funded work. They are best left to the cement/concrete and construction industries themselves.

To attempt to develop a classification model without a thorough review of past literature on the subject would be ludicrous. The EPRI funded study considered the following elements during the technology and literature review phase of the work:

Performance characteristics. Those performance characteristics sufficient to indicate a variety of potential fly ash-cement/concrete applications should be considered.

Previous model development work. Models which have already been developed or postulated relating materials properties to cement/concrete structural and other performance also should be considered and where appropriate, built upon.

Fly ash and cement/concrete testing. Chemical and physical test properties which may relate to relevant performance characteristics identified explicitly; test requirements should be as simple and basic in nature as possible consistent with the premise that the electric utility industry does not desire to become involved with another elaborate test program.

Cement/pozzolan chemistry. Elements of the chemical behavior of cements and pozzolans should be identified as they relate to model development.

Based on the literature review as well as the intent and specific requirements of the overall program, predictive model concepts were formulated. Two distinct model development approaches were pursued.

First, a factor analysis model was developed to relate fly ash characteristics to resulting cement/concrete properties. A factor analysis model has the capability of reducing the independent and/or the dependent parameters into a smaller subset of paramenters while still maintaining the interrelationships between parameters and variables. The intent is to describe the interrelationships with the minimum number of measurements. The factor analysis model transforms input parameter and key variable data so that their variability relates to both pre-hydration and post-hydration parameter values.

To test for independence, whether each of the parameters are related to the same factors or factors which are orthogonal to each other, a statistical test was determined. The distribution of the values of each of the independent variables should be normally distributed. An appropriate method was selected to test for normality. A multiple stepwise regression analysis was then performed on the normalized data. Both standard error and bias were used for the sensitivity analyses.

The resulting independent variables identified used for development of predictive equations and nomographs or other graphical representations which can be used by persons with no prior knowledge of the optimum cement/fly ash mixtures for a given application.

Second, an independent model development effort included development of five empirical formulas for predicting the following fly ash concrete properties: (1) Strength, (2) Setting time, (3) Drying time, (4) Freeze-thaw durability, and (5) Sulfate resistance.

Each formula includes the fly ash quantity and a "constant" which describes the efficiency of the fly ash in comparison to Portland cement.

A relation describing each property was obtained from the literature and appropriately modified to include consideration of fly ash addition. Test data from this project and from the literature was then used to verify the formulas.

Verification, in the above sense, implies that the formula is empirically predictive and that the fly ash "constant" is truly constant for the range of data of interest. The fly ash "constant" should not vary with changes in fly ash quantity or of the concrete mix (e.g., quantities of cement and water).

Universal fly ash "constants" did not result from this work. The formulas that have been developed for concrete with Portland cement as the only binder do not account for all independent variables. The modified formulas used in this study do not account for all possible variables either. "Constants" were identified for limited bounds, however, (for example, a constant may apply for 0-30% fly ash but not for larger percentages). Fly ash "constants" (K) should also be descriptive. For example, a constant "K" of K=0 may indicate no effect on a concrete property while a K>0 indicates a positive effect and K<0 indicates a negative effect. Fly ash "constants" probably will be different for each fly ash.

Fly ash "constants" determined empirically were correlated with ash analysis from this project and from the literature to determine relations between fly ash properties, chemical or physical, and the "constants".

As a result of this EPRI funded study a fly ash concrete model has been developed by the contractor Dunstan, Inc. This model includes factors for concrete strength, setting time, sulfate resistance, drying shrinkage, and freeze-thaw durability. This model is a formula that describes one of the aforementioned properties. To account for fly ash, the formula must include the fly ash quantity and a "constant" which describes the efficiency of the fly ash in comparison to Portland cement. A simple approach is the use of a KF (K times F) value which represents the efficiency or effect of the fly ash in comparison to Portland cement. For example, if K=0.50 and the concrete mix contains 100 pounds of fly ash (F=100), then KF = 50 (0.5 x 100). KF then indicates that 100 pounds of fly ash will act like 50 pounds of cement. Many of the models discussed in the concrete literature use this approach. However, this approach is too simplistic since it does not account for one important factor. For fly ash to react it must be in the presence of lime, a by-product of the cement reaction. If there is no Portland cement there would be no by-product lime, and therefore the fly ash will not react. In this case KF=0. Therefore no matter what the quantity of fly ash K=0. Therefore K is not a constant. In the first example above K=0.50 and in the second K=0. The failure of the KF approach is used as an illustration in the model development description outlined herein. The model derivation is outlined by Dunstan in five steps which follow.

Step 1-- Develop a formula or obtain a formula from the literature which describes the concrete property.

Step 2-- Modify the formula from step 1 to include fly ash or use a formula from the literature which includes a fly ash term. The fly ash term is usually expressed as a "constant".

Step 3a- Use the concrete test data from the regional database developed in the project and from the literature to verify the formula. Verification means the formula is predictive and the fly ash constant is truly a constant. The fly ash constant should be a constant with resect to changes in the fly ash quantity and other changes in the concrete mix such as changes in quantities of cement and water. If the fly ash constant is not "constant" go back the step 1 and start over. The fly ash "constant" will be different for each fly ash. The fly ash K should also be descriptive. For example a constant "K" of K=0 indicates no effect on the concrete property. K>0 indicates a positive effect and K<0 indicates a negative effect.

Step 3b- It does not appear to be possible to find a "true constant". The fly ash effect will not be a constant if all the variables are not included in the formula There are most likely variables that have not been identified. The formulas that have been developed for concrete with Portland cement. as the only binder for Portland cement concete it is unreasonable to expect that the formula or models developed under this research study will account for everything. A constant may be found that is constant within limited bounds. For example, the constant may be reasonably constant for 0 to 30% fly ash substitution, however at larger percentages it is not constant. In these cases the limited constants can be used.

Step 4-- Compare the constant from 3a or 3b above to the test results from the ash analyis results, and to data in the literature. The next question to ask is which ash test or combination of chemical or physical tests, best decribes the constant. When a relationship between the constant and ash testing and ash testing is found proceed to Step 5.

Step 5-- If the model works with the data from the ash analysis data developed under the EPRI project the question to be evaluated is how predictive it is. It must be determined whether or not the model works with data from each of the provinces of China. The most important issue for the eventual acceptance of this or any model is how easy is it to use. If the model can be placed on a nomograph it will increase its usefulness.

By way of illustration of the five steps described above, a model for concrete strength is discussed. The model has been proposed by Dunstan. This is a brief description of the model developed for the compressive strength of fly ash concretes under this EPRI project. The model for compressive strength of fly ash concretes developed under this contract considers all concretes to be normal Portland cement concrete with additional strength produced by the addition of fly ash. The model would have to answer the questions of what is the strength of the concrete both with and without the addition of fly ash. The total strength would be represented by the following relationship:

$$F'_{(c+f)} = (1 + ?)\, F_c \qquad \text{(Equation \#1)}$$

Where F_c is the strength without fly ash and $F'_{(c+f)}$ is the strength with the addition of fly ash.

The question one must ask is what is the fly ash effect represented by the "?" in equation #1? If we assume that fly ashes are pozzolanic, that is the silica and alumina in the fly ash combines with the lime given off by the cement, the "?" is a function of the binding capacity or the binding of lime given off as a by-product reaction of cement hydration. The strength of this reaction or efficiency of this reaction would be a function of the fly ash content and the amount of lime given off by the cement. Consider the following relationship:

$$F'_{(c+f)} = [\,1 + EF / MLC\,]\, F_c \qquad \text{(Equation \#2)}$$

Where F is the weight of fly ash per cubic yard, C is the weight of cement per cubic yard, L is the decimal percent (total) lime given off by the cement reaction, M is the maturity (how much reaction has taken place expressed as a decimal percent), and E is the efficiency of the reaction.

Most references state that the cement reaction gives off about 25% of its weight as lime thus L in the above equation is assumed to be) 0.25. The parameter M is the maturity of the reaction. If the cement is fully hydrated (at long ages) the value of M would be 1. At earlier ages the cement has not fully reacted thus the value of M is less than one. In this model a value of 0.80 is used at 28 days and full hydration is assumed to have taken place by the end of one year. The efficiency parameter E will be discussed in greater depth later.

If the above equation is written in terms of fly ash percent "f" the equation is as follows:

$$F'_{(c+f)} = [\ 1 + Ef\ /\ \{0.25(1 - f)M\}]\ F'_c \qquad \text{(Equation \#3)}$$

Where f is the percent fly ash calculated as $f = \{F/(F+C)\}$

In the above equation F'_c can be any of the many equations that have been developed for concrete strength for concretes containing only Portland cement. The most common referenced equations are those of Feret, Abrams, and Bolomey [2]. The authors prefer the equation by Feret which includes the effect of air content. Using the equation of Feret the following equation is produced.

$$F'_{(c+f)} = [\ 1 + Ef\ /\ \{0.25(1 - f)M\}]\ K\ [\ C_v\ /\ (C_v + W_v + A_v)]^2 \qquad \text{(Eqn. \#4)}$$

Where C_v, W_v, and A_v are the volumes per cubic yard of cement, water, and air, respectively and K is an empirical constant.

The efficiency E in the above equation can be described by the following relationship:

$$E = (1-f)f^n \qquad \text{(Equation \#5)}$$

The n-value is the ratio of the amount of lime given off by the cement to the amount of lime that can be bound by the fly ash (pound per pound). It can be shown chemically that the silica in a pozzolanic material such as fly ash will combine with 1.85 times it's weight of lime-$Ca(OH)_2$ given off by the cement (see calculation of n in equation #7 below). Alumina materials combine with 2.2 times their weight. It can be demonstrated that the optimum percentage of fly ash (for silica reactions) can then be calculated by the following formula:

$$f_{opt} = n/(1 + n) \qquad \text{(Equation \#6)}$$

Where n is the ratio of lime produced to the binding capacity of the fly ash.

The value of n for silica pozzolans can be calculated as follows:

$$n = ML/\ 1.85\ P \qquad \text{(Equation \#7)}$$

Where P is the percent (decimal) of the fly ash that will react at the age in question. Thus a very good pozzolan (100% reactive) would follow a curve similar to n = 0.10.

The question often asked is how reactive is fly ash? What is the n-value for fly ash? The n-value will most likely be different for each ash but should be similar for ashes of similar chemistry and fineness. The n-value can be accurately calculated for one fly ash studied by Mohan and Taylor. Their study was not made with normal Portland cement. Instead they used C_3S the main source of lime in Portland cement. Normal Portland cement is about 50% C_3S therefore the total lime in their study is about twice that of normal Portland cement. In their study, the lime content was 40% with a fly ash reaction of 45%. The calculated n-values are then:

$(0.34/\ (1.85x0.15) = 1.23$ at 28 days, and
$(0.40/\ (1.85x0.45) = 0.48$ at one year

Using 20% lime for normal Portland cement at 28 days and 25% at one year, the corresponding fly ash reactions for these n-values are 8.8% and 28.2% for Navajo Power Plant ash in the EPRI study which indicates that sedigraph fineness of about 3 microns and less have entered into the reaction by 28-days and particles up to 6 microns would react by one year.

If the rate of reaction of the fly ash was the same as Portland cement with the same relative percentages of each reacting the n-value would be a constant. In that case the fly ash reacts at a slower rate at early ages so the n-value is higher at early ages and decreases at longer ages. For comparison, Warris suggested n-values of those used in this model. The n-values in his model are inverse values of those used in this model. Warris suggested 0.8(1/0.8 = 1.25) at 28 days and 2 (1/2 =0.50) as an ultimate value. These values compare very well with 1.23 and 0.48 calculated for the Mohan and Taylor data.

Looking closely at the Mohan and Taylor data provides additional information. At 28 days the fly ash has actually reacted with 12% lime and at one year with 18% lime. If this is a silica reaction the actual weight that has entered into the reaction is (12/1.85) = 6.5% and (18/1.85) = 9.7%. It therefore appears that the n-value calculated above represents ash that is "associated with" the reaction but not all of the ash has actually reacted. Many researchers report that a fly ash reaction is on the surface of the fly ash particle. The fly ash material below the surface does not enter into the reaction except at very long ages. A large fly ash particle would have less surface area in comparison to its total weight than a small particle. Thus the actual amount of material entering into the reaction chemically would be higher for smaller particles than for larger particles. This appears to be true of the Mohan and Taylor data. At 28 days it appears that particles up to 3 microns have raacted (as discussed above) and by one year particles up to 6 microns have reacted. The percentage of actual reaction to that "associated with" the reaction is (6.5/15) =43% at 28 days and (9.7/45) = 21.6% at one year. This relates to about a four fold increase in surface area (the ash particles 3 to 6 micron in size) that has reacted at one year compared to 28 days. A corresponding weight increase for solid spheres (3 to 6 microns) would be an 8 fold increase. The Mohan and Taylor data indicates a 3 fold increase of 15 to 45%. It is a well known fact that fly ash is commonly described as a hollow sphere rather than a solid sphere which would explain the three fold increase rather than an eight fold increase for solid spheres.

$n = 10 - [2.6 \mathrm{Log}\,(B_f)]$ (Equation #8)

Where Log is logarithm and B_f is Blaine fineness (cm^2/g)

However one must consider how the amount of reacted material and the amount of lime bound relates to the concrete strength. Equation #5 has been modified to the following form:

$E = E_m(1 - f)f^n$

Where E_m is the maximum efficiency of the reaction.

The maximum efficiency of the reaction depends on whether the reaction is a lime-alumina reaction or a lime-silica reaction. The lime-alumina reaction produces about 10% the strength of a lime-silica reaction. If the alumina reactions and silica reactions are in the same ratio as the alumina and silica reactions in cement, the value of E_m should be 1.0. If the E_m value is higher than 1.0 the strength has been produced by a higher percentage of silica reactions than 1.0 the efficiency represents a reaction which is higher in alumina components than in the control cement.

Substituting equation #9 into equation #4 produces the final equation for fly ash concrete strength which is as follows:

$F_{(c+f)} = [\,1 + E_m\, f^{(n+1)} / (0.25M)]\, K\, [C_v /(C_v + W_v + A_v)]^2$ (Equation #10)

Equation #10 has been used to produce graphs. These figures are based around the ashes investigated in this model development study.

The parameters E_m and n are both descriptive. The parameter n has defined as lime binding capacity. The parameter n probably also includes other reactions such as alkali reactions. Possible reactions include, lime-silica, lime-alumina, alkali-silica, alkali-alumina, and sulfate reactions. In a purely pozzolanic material the total strength (sum of all of the reactions) is related to E_m (maximum efficiency). In materials such as blast-furnace slag, and Class C fly ashes that contain a high proportion of calcium E_m also includes all of the strength reactions of the material that take place. Many of these reactions do not require by-products of the cement reaction such as lime and alkalies. Many Class C ashes produce considerable strength without Portland cement, thus the lime-fixation constant n may be low and the efficiency index-E could be high. These two parameters that will give a very good description of the secondary material irrespective of whether it is a fly ash, silica fume, or blast-furnace slag.

The lime-binding capacity parameter n may also relate to another concrete property--durability. Sulfate resistance is related to the amount of lime consumed by the secondary material. If a high amount of lime is given off by the cement is used by the fly ash, the concrete will have good sulfate resistance. In similar fashion, if a high amount of the alkali is used by the fly ash, the potential for alkali-aggregate reaction is reduced. Therefore a high value for the parameter n indicates improvements in concrete durability (sulfate resistance and resistance to alkali-aggregate reaction).

Hopefully a good relationship exists between these parameters and the ash testing data from the EPRI project. As of the date of writing this data has not been thoroughly evaluated. The parameter n will be related to the chemistry and the fineness (physical characteristics) of the fly ash. When the relationship for n is established, E_m can be calculated from the physical testing (pozzolanic index with cement).

The above comments are the basics of the model that is described in further depth in the final report published by EPRI in April 1987 (CS-5116).

There are a number of additional models being developed under this EPRI project which time did not permit me to descibe at the SRIBS lectures. This includes models for setting time, freeze/thaw resistance, and dry shrinkage. Of these models only the setting time model is expected to require a new formula. The important parameters (E_m and n) for the freeze/thaw resistance and shrinkage phenomena have already been defined. The setting time model is being developed around the OIF (Omega Index Factor) proposed by Dodson. This factor relates setting time to the cement content and the water to cement ratio. The model for freeze/thaw resistance will most likely state that resistance of fly ash concrete is comparable to Portland cement concrete with the same strength and entrained air content. The model for drying shrinkage wil use an equivalent cement approach. The equation for concrete strength can be used to calculate how much cement is required to obtain an equivalent strength as the cement plus fly ash quantity. If the equivalent cement quantity is known it is expected that drying shrinkage will be similar to normal Portland cement concrete.

This lecture summarized a comprehensive project funded by the Electric Power Research Institute to develop a series of fly ash classification models for predicting the performance of fly ash in cement/concrete applications. This material was covered only in summary form in my lectures. I left a copy of the EPRI report which reviews the model developed by Dunstan under this project to predict the performance of fly ash based on two fundamental parameters, maximum efficiency E_m and the lime binding capacity n. These parameters E_m and n serve in many models--sulfate resistance, alkali-aggregate reaction, freeze/thaw resistance, drying shrinkage, and possibly setting time. There is research underway at EPRI to further improve these predictive models.

LECTURE 3 PART 2: ROLLER COMPACTED CONCRETE

The afternoon of the third lecture day was to review roller compacted fly ash use in the USA and to discuss the recent developments in RCC pavement. The primary use of RCC containing fly ash in the USA has been used in dam construction. The Upper Stillwater Dam in Utah, USA became the worlds largest RCC dam, when completed in August 1987. Its total volume of 1.22 million cubic meters of concrete of which 1.07 million cubic meters will be RCC. In that project, fly ash was used in structural concrete (23% by weight), and in RCC (70% by weight.) Over 204,000 metric tons of Type F fly ash were used in the Upper Stillwater Dam. RCC dam construction combines strength and impermeability of concrete dams with the rapid placing rates of embankment construction. It is possible to make no slump concrete containing fly ash by mixing it in a conventional drum or shaft-driven paddle mixers, transporting it by end-dump trucks, spreading it into horizontal layers (0.3 to 0.9m) by a bulldozer, and compacting it to a solid mass by a smooth-drum vibratory roller. The strength gain of RCC is governed by the water to cementitious materials ratio. Pozzolan mixes have lower early strength followed by higher long-term strength, RCC mixes act the same. Dams using RCC concrete are designed for 1-year strength criteria. The cost of fly ash compared to cement is significant factor in its use in RCC dam construction. The Upper Stillwater Dam bid prices were $47/ton for ash and $85/ton for cement.

The use of fly ash is a key element in RCC construction due to the benefits for both fresh and hardened concrete.

- Increased bond potential because of longer setting times.
- Reduced temperature rise in mass concrete.
- Increased long-term strength.
- Reduced cost.

The use of RCC pavement is growing rapidly in the USA and Canada. The first known RCC test pavement was in 1975 at the US Army Corps of Engineers test station in Mississippi. This 12 ft by 105 ft street section is still performing well. In 1984 a 63,900 sq yd hardstand for tracked military vehicles was installed in Texas. A demonstration road was built in Washington state on a military base in late 1984. A city street was constructed of RCC in Portland , Oregon in 1985. Docks, freight yards, coal yards, and truck stop parking areas have been constructed during the past two years. Most RCC pavements have been placed on granular base courses instead of directly on a subgrade. The mix is placed with conventional paving machines, typically in 7 to 8 inch lifts. There are substantial cost savings compared to convential asphalt materials, typically ranging from 25 to 40%. A typical RCC pavement mix uses Type I or II portland cement in the range of 480 to 625 lbs per cu. yd. Class F fly ash has been used in most RCC pavement projects to replace 20 to 30% of the cement. Almost all RCC pavements have used a nominal maximum size aggregate of 3/4 in or 5/8 in. Larger aggregate size tends to produce a mixture with significant segregation. RCC pavement placement methods have been established by the US Army Corps of Engineers. Their specifications require two aggregate sizes, split on the No. 4 screen size. The minus 200 material should be between 6 and 10%. Air entraining agents have not been used successfully. Continuous-type plants have been used rather than batch-type plants.

Mixing RCC Pavement has not been found to be particularly difficult. Twin-shaft pugmills are necessary because their scrubbing action is far superior to the tumbling action of a drum-type mixer. Corps of Engineer specifications require adjustable paddles in mixer for better control in mixing. Corps of Engineer specifications require cement and fly ash be fed by vane feeders or similar positive metering devices, and that each aggregate bin feed into a variable speed belt or gate controlled remotely. The amount of water going into the mixer most be continuously controlled, since placing and compaction are sensitive to small (0.1 or 0.2%) variations in water content. Prior to 1985 American-made asphalt paving machines were used to lay the RCC material. Since 1985 heavy duty pavers from Germany with tamping screeds and vibrating screed have been made specifically for RCC pavement. The German paver compacts the RCC course to 94 - 95% modified Proctor density, leaving less compaction to be performed by the vibratory rollers. The disadvantage is the close-spaced tearing cracks in the top surface which can be only partially closed with proper use of rubber-tired roller. Corps of Engineer specifications require that grade be contolled with electronic controls operating from a stringline. A 12 ft straightedge must be used continually to assure pavement smoothness. Grade conformance and slab thickness must be checked regularly. A Nuclear density meter should be used to determine the in place density of the compacted RCC. A control density reading should be taken 2 inches from the bottom of the course. Minimum field density for pavement is 98.5% o lane interiors and fresh joints, and 96.5% on cold joints. Density percentages are % of the maximum wet density attained in modified Proctor test (ASTM D1557). To date, most RCC pavements have been designed without transverse contraction joints. Pavement is allowed to crack at 40 to 70 ft spacings. The primary compaction equipment used for RCC pavement is the heavy drum steel wheel vibratory roller weighing at least 150 lbs per lineal inch of drum, and vibrating at least 1,500 cycles per minute. The Corps of Engineer specifications require at least four passes of the double drum roller to produce a density of 98.5%.

There were a considerable number of questions from the audience on RCC pavement design and placement techniques.

Lecture 4: (Dec. 9) Fly Ash Resource Recovery

It is clear that there is a continuing and intense international research interest in the recovery of resources from coal ash. Resource recovery from fly ash is inherently attractive because of the prospects for avoiding disposal costs and generating revenue from metal and mineral sales, and conserving the world's strategic mineral resources. Of equal importance is the reduced environmental concerns from reduced volumes in disposal facilities. Coal ash represents a source that is already mined and crushed, and available for the recovery of its constitutent metals. By using the mineral content of coal ash, not only is the environmental problem of ash disposal eliminated or greatly reduced, but also the dependence of the China on imported ores for strategic use and other metals can be lessened. Judging from the amount of attendance and the level of attention during this lecture their is a high interest in China as well.

The chemical composition of coal ash is dependent on the composition of the soil strata from which the coal was mined, and therefore, is a function of the geology and hydrogeology of the surrounding strata. Generally speaking, it can be stated that coal ash has a chemical composition similar to clay, which has also been evaluated as a non-bauxitic alumina source. Table 1 shows typical levels of the major ash/clay constituents.

TABLE 1. Typical Levels of Major Ash/Clay Constituents

	Fly Ash	Bottom Ash	Kaolin Clay
SiO2	40 to 60 %	50 to 60 %	40 to 50 %
Al2O3	20 to 35 %	15 to 25 %	35 to 40 %
Fe2O3	3 to 6 %	4 to 9 %	0.5 to 2 %
CaO	3 to 20 %	4 to 15 %	<0.1 %

Silica, alumina, and iron oxide represent about 90 % of the total. Because of the high temperature at which fly ash is produced in a high efficiency boiler, the ash consists of glassy particles (generally spherical) of complex silicates of these three elements. In addition to the constituents contaioned in Table 1, coal ash contains other elements in trace quantities. If the sensitivity of the analytical method were high enough, virtually all the naturally occuring elements on the periodic table might be found.

The literature is full of data giving the concentrations of the many trace elements in coal ash. The data show the substantial variations in concentration between samples. Table 2 lists the typical ranges for the more common trace elements.

TABLE 2. Concentrations of the Trace Elements In Coal Ash

Range (mg/kg)	Elements
100 to 1000	B.Ba,Cu,Mn,Sr
10 to 100	As,Cr,La,Mo,Ni,Pb,Th,U,Zn
1 to 10	Cd, Ga, Sb, Se,Ti,V
<1	Hg

Under the existing economic conditions, the resource components of coal ash which may be considered for their intrinsic value can be summarized as follows:

- Carbon
- Magnetite
- Cenospheres
- Alumina, iron oxide, and other metal values

The recovery of metals from coal ash is not a new idea. Literature citations go back fifty or more years. The technical literature is full of different methods of recovery of the metal values from coal ash. Of the elements present in coal ash, iron and aluminum have been examined most extensively for recovery. Of the minor and trace elements present, extraction of gallium, germanium, uranium, and molybdenum have been investigated most frequently. The recovery of the elemental constituents from coal ash can be accomplished using either physical or chemical processes. The chief roadblock to ash metal recovery process development is the economics of recovering minerals from such a "lean ore body" as coal ash.

Physical processes for recovery of the iron-rich fraction of ash from bituminous coal ash by magnetic separation is commercially viable. A commercial facility is in operation at a West Penn Power plant near Pittsburgh, Pennsylvania, USA producing a fly ash heavy medium material specifically for use in coal beneficiation. The market price for the magnetic ash product is about $75/ton.

Chemical processes for the recovery of the major constituents from coal ash that have been formulated or at least tested on a laboratory scale can be divided into four general categories:

- Hydrothermal leaching
- Sinter-leach
- Gas-solid reaction
- Direct reduction

Hydrothermal leaching processes involve contacting the solid ore with an aqueous solution to selectively dissolve the minerals to achieve separation. After the solution is isolated from the insoluble residue, it is treated to recover the minerals in pure form by solvent extraction, ion exchange, crystallization, precipitation, or similar methods. Leaching processes for the recovery of alumina and other minerals from coal ash can be placed in one of two categories, alkaline or acidic, depending upon the nature of the solution which initially contacts the fresh coal ash. Hydrothermal leaching of fly ash with alkaline solutions has been examined by several research organizations. The process would appear to be most viable on a high pH ash.

In sinter-leach processes, a high-temperature solid-state reaction is used to modify the aluminosilicate matrix and make it more amenable to leaching. Various processes then use either an alkaline or acidic leach to extract the aluminum from the sinter residue.

Gas-solid reaction processes involve reacting coal with a gaseous reagent(s) to convert the metals values present into volatile compounds and permit their recovery. Typically one reagent is needed to reduce the aluminosilicate -- or serve as an oxygen sink -- while another reagent is needed to form the volatile species.

Direct reduction processes produce a metallic alloy by high-temperature or electric-arc reduction. High-temperature reduction of coal ash (1700°-1750°C) with charcoal produces a silicoaluminum alloy. Electric-arc reduction of coal ash can produce a ferrosilicon alloy of a silicoaluminum alloy from which aluminum can be obtained by extraction with zinc.

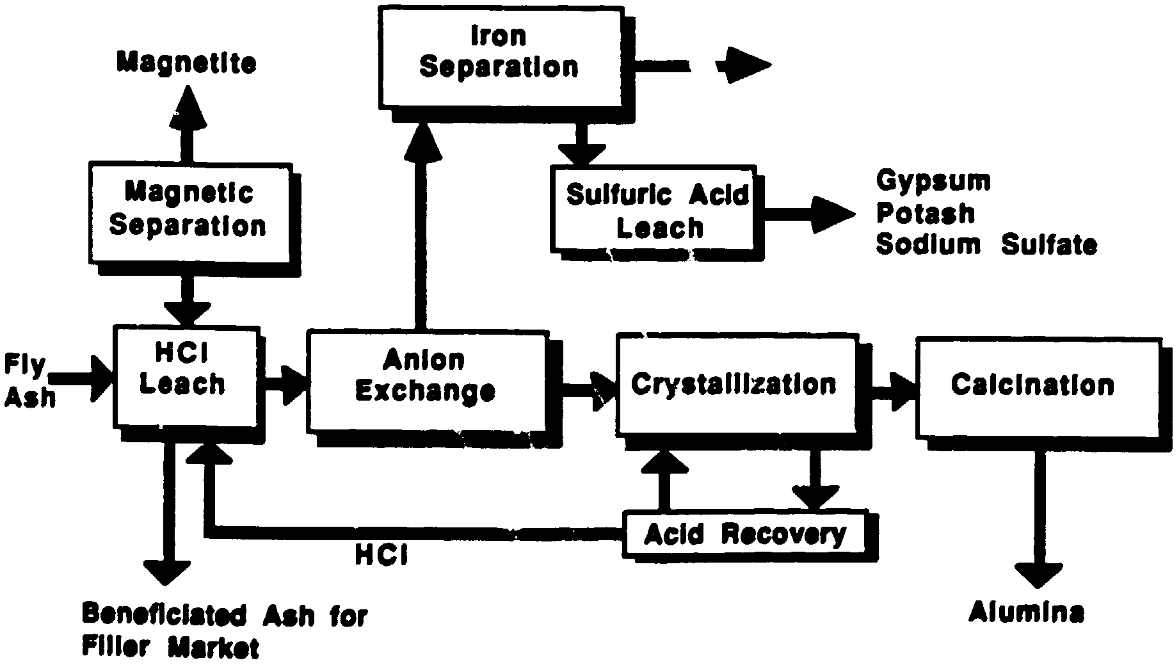
The coal ash metal recovery processes that appear to hold the most promise at the present time are the EPRI/ORNL Direct Acid Leach (DAL) process, and two sinter-leaching processes, the Calsinter (ORNL) and the Lime-Soda Sinter (Ames Laboratory).

The large scale implementation of technologies for mineral recovery form fly ash will require processing primarily for Aluminum recovery as well as for utilization of all of the process residues. That is the beauty of the DAL process, since the combination of subprocesses has been design so as not to create any new wastes which would require disposal. Commmercialization of the technology will require a cooperative effort between those wishing to dispose of or sell the fly ash (electric utilities) and those seeking a nonbauxite source of alumina and other metals, and fillers.

The direct acid leaching (DAL) process was first studied as a means of treatment for fly ash to remove the leachable heavy metals. What differentiates the DAL process evaluated under EPRI sponsorship at Oak Ridge National Laboratory is that it is designed to convert the entire ash resource at a given facility into a variety of products, without regard to obtaining high extraction percentages of individual metals. During 1983-84, a conceptual commercial scale design and financial estimate were developed for a plant to remove metal oxides and other salts from coal ash using the direct acid leaching process. The financial estimates projected a rate of return on the direct acid leaching process (DAL) of approximately 20 to 30 %.

In the direct acid leach process, fly ash is leached at about 100°C for two hours in hydrochloric acid to remove the metals. The resultant chloride leachate contains most of the metals, including trace amounts of strategic metals such as chromium, cobalt, and manganese, but the major components of potential value are aluminum and iron. The leachate is then passed through a series of anion exchange columns to produce a partially purified solution of aluminum chloride and a very high purity iron chloride. Final purification of the aluminum chloride is by hydrogen chloride gas - sparged crystallization in two stages. The figure below is a simplified process flow diagram.

SIMPLIFIED DAL PROCESS DIAGRAM



The technique of producing relatively pure compounds by precipitating chlorides from solution with an excess of HCl has been proven on a commercial scale. Purified salts like $AlCl_3 \cdot 6H_2O$ can be prepared by a saturated solution of HCl, which causes the salt to precipitate. In the DAL ash metal recovery process, the purification and recovery of an aluminum chloride hexahydrate product is made. Waste liquor from the multi-stage crystallization system goes into a gypsum reactor where it is reacted with 98 % H_2SO_4. Gypsum is crystallized and HCl gas generated by this reaction. The aluminum chloride hexahydrate crystal slurry from the crystallizer is converted to alumina by calcination of the hexahydrate and absorption of the off-gases, which provides the HCl for recycle.

EPRI became involved in fly ash metal recovery research in 1979 primarily as a method of treatment of ash prior to disposal. At that time, there was great uncertainty as to how coal ash would be regulated under the federal government's Resource Conservation and Recovery Act. The EPRI contractor, Oak Ridge National Laboratory, investigated several potential processes and identified the two most promising recovery processes. The project report included process flowcharts, preliminary designs for a demonstration plant, cost estimates, and expected by-product revenues from recovered resources. EPRI contractor Raymond Kaiser Engineers did a detailed engineering, cost, and financial evaluation for a conceptual commercial plant to process fly ash into marketable metal oxides by the direct HCl acid leach process during 1983. During 1984-1985, a fly ash resource evaluation and product market assessment are being conducted. During 1985 preliminary design of a five ton a day pilot plant was completed.

An engineering, cost, and financial evaluation study was performed for EPRI by Raymond Kaiser Engineers during 1983-1984. The study was carried out for a conceptual commercial size plant to process 1,180,000 short tons of ash (dry basis) per year into marketable metal oxides. The hypothetical plant site in the study was adjacent to the Tennesseee Valley Authority (TVA) Kingston Power Plant. The cost estimates included capital, operation, and maintenance costs. The revenue projections were made based on several selling prices for the principal products.

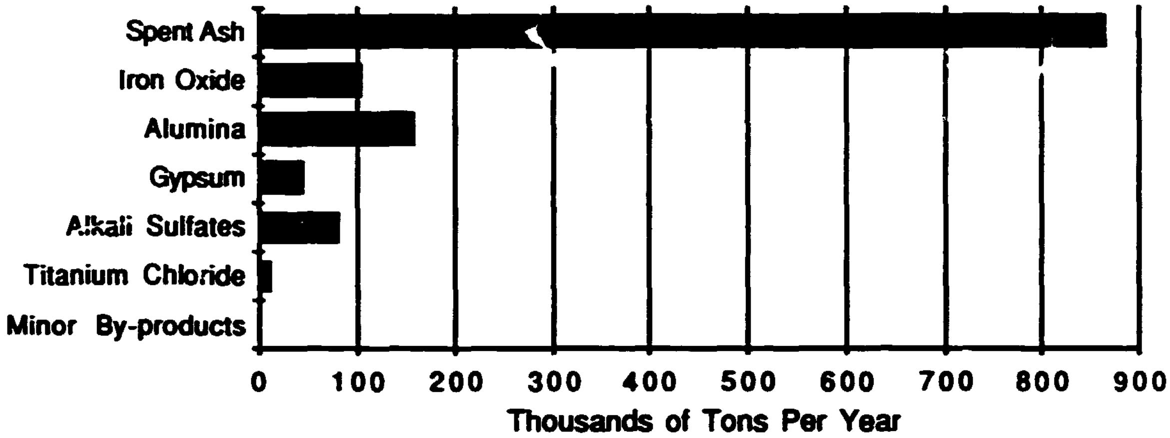
The estimated capital cost of the commercial scale plant located next to the Kingston Power Plant is $270,000,000. In addition to the by-products produced, the co-generation plant will produce an excess of 7,189 kW of electric power with an estimated value of $2,144,000 per year.

It is interesting to note that the process produces more hydrochloric acid than it consumes, due to the vapor recovery system. The additional chlorides are introduced into the process by the chlorine which is used as an oxidizer. The major cost item in raw materials is the 96,000 short tons per year of sulfuric acid. It is conceivable that the DAL process could be run in conjunction with a regenerable FGD scrubber system which produces sulfuric acid as a by-product. This would provide economic benefits to both processes, by avoiding limestone purchases and FGD waste disposal costs.

One of the primary objectives in the EPRI sponsored DAL process development is that there be no waste by-products created requiring disposal. The process has been modified from the earlier concept by the addition of a sulfuric acid loop to produce gypsum and alkali sulfates, rather than create a chloride waste. In all, over a dozen by-products are possible from the DAL process. A summary of the potential products from the hypothetical commercial scale facility is shown below for a full-scale plant.

The spent ash material produced in the leaching process is a unique product. This spent ash material has properties which, with proper processing and marketing programs, could become an appreciable revenue source for the process. One of the principal advantages of the spent ash is that it will be uniform in quality. There are many potential applications for spent ash because it is an inert, stable, opaque aluminum silicate particle with enhanced surface characteristics. In all, over a dozen by-products are possible from the DAL process.

Full-Scale DAL Plant Production
Thousands of Tons



The spent ash product will be dried and classified, so that five different size fractions can be supplied into different markets. The finer size fractions would go to pigment and filler markets, and the coarse size for pumice pigments, cements, and aggregates. Based on the laboratory studies, the spent ash will have superior properties such as high surface area (8 to 12 times original ash), high abrasion resistance, and low density. The potential market for spent ash in fillers and extenders applications is estimated a 12,000,000 short tons per year in the United States. The average price for these products is estimated at $85 per short ton. A study completed in 1986 (CS-4765) showed these spent ash particles make ideal fillers in nylon and polyprolene. Additional research is needed to assess the technical and economic feasibility of these ash fillers in other plastics.

The alumina production rate from the commercial scale plant studied is about 158,000 short tons per year, which is less than 2 % of current U.S. consumption.

The ion exchange system included in the DAL process will allow production of a very high purity iron chloride. This is converted into an agglomerated iron oxide particle. The target markets for this product are finished pigments, magnetics, and catalysts in specialty chemical markets.

The gypsum produced in the DAL process should be viewed as a chloride recovery and waste disposal operation. The crude gypsum produced would contain impurities, such as magnesium, strontium, and barium, which could limit marketability and may need to be recovered separately. The critical requirements in gypsum markets are purity, particle size, moisture content, and specific impurities (such as chlorides).

The remaining alkali sulfates are the mixed salts of sodium, potassium, and aluminum. The estimated chemical analysis is as follows:

$Al(SO_4)_3$	9.7 %
K_2SO_4	42.4
$MgSO_4$	24.4
$Mn(SO_4)_2$	0.7
Na_2SO_4	6.5
$Ti(SO_4)_2$	16.3
Total	100.0 %

The alkali salts are the variable product in the series of products recovered in the DAL process. The heavy metals would also end up in this product unless they are recovered earlier in the process. These salts would appear to have value as fertilizer and as salt cake. The titanium dioxide could be a valuable by-product.

Synthetic magnetite appears to be a relatively small market since the present market as a coal cleaning media is relatively limited. Some ashes have considerably higher magnetic ash contents. Magnetic ash is a relatively low purity, low value product compared to higher value iron oxide products. Since magnetic ash is mostly silica with some iron oxide, magnetic ash could possibly be a good ferrosilicon raw material.

As noted previously fly ash contains many trace metals. Given the large size of the presumed commercial plant, even the trace amounts can add up to hundreds of tons per year. In many cases the technology for recovery of these products is known, but application to ores with trace amounts is not economical.

For ash resources with a high carbon content, a carbon recovery circuit can be added in front of the DAL process. In this system the difference in the physical characteristics (size and density) of the ash and carbon particles could be utilized to separate the carbon fraction of the ash. The carbon fraction can be further purified using wet flotation techniques to produce a carbon concentrate which then would be dried and ground if necessary. The thermal history of the ash is likely to produce a carbon with sufficient surface activity so that the potential exists for entering such traditional activated carbon markets as: municipal and industrial waste treatment, dry cleaning, sugar and syrup industries, motor vehicles, and air cleaning equipment. If so, the price per ton can reach several hundred dollars.

Although the percentage of cenospheres in ash varies widely, it generally represents only a fraction of a percent of the total ash. Nevertheless, it can be easily collected in ash ponds with flotation collection devices. The literature indicates a diverse market potential for ash cenospheres in industrial applications. The primary applications are as mineral fillers and as lightweight refractory materials. The high strength, low weight, chemical inertness, spherical shape, and particle size distribution of cenospheres allow them to serve as low-cost substitute for manufactured glass microspheres. A great many applications are possible when binders such as organic resins or cement are used. In these applications the cenospheres act as both an extender and a material conferring desirable engineering properties on the finished products. When used as lightweight refractory materials, cenospheres can be used as a refractory aggregate that can be added to refractory cements or sintered without any binders. The market price for cenospheres is also several hundred dollars per ton.

It should be noted that the DAL process is still in the early stages of commercialization. It is expected that if suitable funding arrangements can be made, the pilot plant could be built in 1987 and operated through 1989. If the pilot plant proves as successful as the bench scale unit has, a larger semi- commercial scale unit would be planned for construction in 1990 to 1992. I would guess that a semi-commercial unit would cost $8-9 Miliion.

This UNIDO report has briefly reviewed the lecture presented on the DAL technology and the target markets for the DAL process by-products. As part of the ongoing EPRI sponsored research, product samples were being produced in a bench scale DAL unit at Oak Ridge National Laboratory with ash supplied by two utilities. The samples were then evaluated by test laboratories recommended by potential ash product users. The goal of the testing was to determine whether or not the by-products meet the specifications for use by the target industries. The results of this testing will be available later this year.

The overall goal of EPRI in sponsoring the DAL process technology development is to produce high quality, attractively priced products with a reliable, long term availablility in environmentally acceptable operations and in grades specifically processed to meet customer needs.

In summarizing the so-called high technology applications for coal ash it is important to realize that ash possesses a number of unique characteristics which should enhance its marketability as a filler or extender:

- Ash can be produced and brought to market without the penalty of high-energy related costs required for the mining, processing, and size-reduction (grinding) of conventional materials.

- Ash is stockpiled in large quantities (800 million short tons since the end of World War II), ensuring the dependable supply of a process feedstock.

- The wide geographic distribution of coal-fired power plants provides an advantage to ash resources over existing mineral resources because of proximity to manufacturing centers.

- The fly ash particles are predominantly spherical in shape with a size distribution extending from <0.5 to >200 µm, and it shares with only a few other minerals the unique property of an almost perfectly spherica' shape in the submicron sizes. This property contributes to improved packing and rheological characteristics.

- The high compressive strength and good thermal stability make fly ash suitable for applications requiring high production temperatures or when specifications call for high temperature resistance in the finished products.

The need to re-evaluate mineral resources from a purely nationalistic or even world-wide perspective has lead to numerous paper studies on the utilization of by-products as a source material for a variety of products. Fly ash represents a possible source of fillers and additives to the oil-drilling, plastics and paint industries, a source of activated carvon for various purification problems, and a source of magnetite for heavy media cleaning of metallurgical and thermal coals. Cenopheres, the very light componenet of fly ash, is becoming a valuable ingredient to certain oil-drilling cements, insulating resins, and in general as a filler that is lightweight and inert. Other spherical components of fly ash, fractionated to narrow specifications, are beginning too be used in the manufacture of resins and paints as a filler with high loading factor. Magnetite has been successfully removed from fly ash by both wet and dry processes and is reported to have been successfully used for heavy media washing of coal. Carbon has been removed by both wet and dry processes but there is nor reported currently-operating process in North America. Since the carbon has been shown to be "activated" there is a large and growing market available at a reasonably high return. It is recommended therefore that the Shanghai Research Institute of Building Sciences set a high priority to further studies to beneficiate fly ash using both wet and dry processes. A theoretical cost-benefit analysis of the fly ash fractions indicates a six to tenfold increase in the fly ash market value based on the selling proce. Since there are no methods of separation available for study and factors such as transportation and plant equipment are not known, it is impossible to determine the net proceeds from beneficiation. Fly ash beneficiation could provide the material currently being imported, therefore providing needed foreign exchange.

Lecture 5: (Dec. 10) Fly Ash Fillers in Metals and Plastics

The morning lecture covered solidification processing of metal matrix - fly ash particle composites. Although the technology involved in fully utilizing the Direct Acid Leaching process to recover the many resources within fly ash could hinder its development within China for a few years, another possibility is solidification processing of the fly ash into metal matrices. The processing involved in preparing the fly ash for use in metals is minimal. Recent research in India, the Soviet Union and the United States of America has suggested the possibility of dispersing ceramic particles in molten alloys prior to solidification to obtaining metal matrix - ceramic particle composites. To date the research has focused on other ceramic materials rather than coal fly ash. Considerable research and development is required to get adequatee dispersions of fly ash in metal matrices by casting techniques to obtain sufficient enhancement in properties and reduction in the cost of components to generate industrial interest in this new family of materials. Fly ash represents a unique waste by-product resource of hollow microspheres of silica and alumina which are otherwise quite expensive for incorporation into low cost industrial composites. Some of the unique features of dispersing the hollow microspheres of fly ash in composites are as follows:

1. One can produce composite materials with tailored stiffness, density, bouyancy, damping, electrical and thermal conductivity by varying the wall thickness of the hollow fly ash particles, or by modifying the surfaces with various coatings.

2. The thermal diffusivity of hollow particles can be varied by changing the wall thickness while keeping the surface characteristics constant. This will permit separating the influence of interfacial enercy, thermal diffusivity, and density on particle entry and entrapment during solidification.

3. The hollow ash microspheres can be incorporated in the metal matrices using only solidification techniques since the powder metallurgy methods would rupture the hollow particles.

4. The density of the hollow microspheres can be tailored to be equal to melts, and the inner surface of punctured micorspheres tailored to develop interparticle repulsion leading to better dispersions.

5. The residual stresses in the matrix with hollow microspheres could be very different than solid ceramic particles.

6. The fracture characteristics of the composites containing microspheres could be advantageous and tailored by changing the wall thickness much more easily than solid spheres. The fracture, or flexing and buckling of hollow spheres can provide a mechanism of energy absorption and arrest cracks.

7. The inner cavity of punctured hollow microspheres can be filled with substances which can respond to external fields thereby permitting control on the motion of the particles.

8. The tribological properties of the composites containing hollow ash microspheres could be much better than composites containing solid particles. At the surface the hollow microspheres could act as oil reservoirs and supply fluid lubrication.

9. The stiffness (in bending) of metals containing hollow ash microspheres will be much higher in a manner similar to high-stiffness obtained with foamed materials.

10. The sound transmission and dapmping porperties of composites containing hollow ash microspheres can be tailored more readily than composites containing solid ceramic particles.

11. The fine perfectly spherical fly ash particles can improve flow characteristics of molten metals and could be used as dispersants of fibers in hybrid composites containing both particles and fibers. The shrinkage and porosity of cast allowys with microspheres is likely to be much lower.

Over the past decade extensive experimental observations have been made on solidification behavior of molten alloys in presence of suspended solid ceramic particles and fibers, in terms of their effects on nucleation, growth, particle pusing, micro and macro-segregation, and formation of shrinkage and proosity. These observations along with studies of thermodynamics and kinetics of segragation of alloying elements at the melt ceramic interfaces, formation of reaction products, allow researchers to make some predictions on the kind of solidification structures that can be obtained during the freezing of metal-ceramic particle composites. Several studies on surface precipitated phases and dislocation structures at the metal ceramic fiber interfaces in composites, and their influence on properties are now available in literature. These provide adequate initial basis for tailoring the interfaces in aluminum-fly ash particle composites, even though much more work remains to be done in this area. The scientific background of the concept includes thermodynamics and kinetics of introduction of ceramic particles in molten alloys, their movement in the melt and with respect to moving solid-liquid interfaces during solidification. It has been established that if the matrix particle interfacial energy is low, the particles can enter the melt and uniform dispersions can be obtained in the liquid, and in the liquid, and inthe solid after freezing of the composite. Mass and thermal diffusivity paramentsrs have been derived which successfully predict whether solid particles will be rejected or entrapped by the moving solid-liquid interface. This theoretical work gives a basis for tailoring the solidification conditions and the interfacial energies to obtain uniform dispersion of particles in the solid which forms on freezing of suspensions. The changes in thermal and hydrodynamic properties of the melts as a result of dispersions of solid ceramic particles inluding changes in thermal conductivity, temperature gradients, viscosity, fluidity, convections have been worked out and experimentally confirmed in certain cases.

Cast metal - fly ash composite materials represent a low cost, high performance, tailor made substitute material for a variety of automotive and electromechanical applications such as pistons, cylinder liners, bearings, and current collectors, which will result in a savings of material and energy. According to Rohatgi, et al (Ref. 1 - International Metals Reviews, 1986, Vol. 31, No. 3), various hard or soft ceramic particles or short fibres of zircon, graphite, alumina, mica, shell char, and silicon carbide in several shapes and sizes (including micospheres) have been dispersed in severl molten alloys before solidification to synthesize cast metal - ceramic composites. These cast composites represent one of the most inexpensive tribological materials for automotive and electromechanical applications and have been successfully tested as bearings, pistons, cylinder liners, and current collectors, resulting in savings of materials and energy.

LECTURE 5 PART 2: ENGINEERING PROPERTIES OF PLASTIC COMPOSITES

When improvements in the engineering properties of a plastic composite are required, many factors must be considered in the selection of an appropriate filler in a particular matrix. Some of these properties include:

- Particle size distribution
- Bulk and particle density
- Color, opaqueness, & transparency
- Thermal behavior
- Electrical properties
- Surface area
- Particle packing
- Anti-stripping behavior
- Chemical composition (incl. pH)
- Particle shape
- Inertness
- Dispersion, suspension in matrix
- Surface properties (incl. zeta potential, and adsorption)
- Forming aids
- Cost

Consistency of properties is generally an over-riding concern when assessing these characteristics, since in some applications the filler make up 50% of the final product.

In general, a user will select fillers on the basis of their ability to modify one or more of the following characteristics in the end product: (1) Cost, (2) Physical properties, (3) Flow, (4) Density, (5) Particle size distribution, (6) Color or brightness, (7) Hardness, brittleness and strength, (8) Fire resistance, and (9) Heat or electrical conductivity.

During my lecture I described the various advantages of fly ash microspheres as fillers in plastics, as follows:

- The inclusion of fillers or extenders in product formulations reduces the demand for expensive matrix compenents such as resins in plastics, solvents and extender pigments in coatings, elastomers in rubbers, and bitumen in asphalts; this reduced demand for "real" material improves the cost-effectiveness of the product in the marketplace provided the filler is of low cost relative to the replaced components.

- By careful selection of appropriate filler(s), the engineering properties of a raw material matrix may be modified, for example, to increase strength, to improve casting or moulding characteristics, to reduce thermal expansion or to control density or thermal conductivity.

Fine particles of spherical form have been used as fillers in plastic composites in increasing quantities over the past few years. Spherical fillers bring tie following advantages to composite production in comparison to filler of more irregular shapes:

- Low surface area/volume and, thus, low resin and oil absorption.
- Ease of wetting and dispersion.
- Uniform stress distribution and isotropic physical properties of composites.
- The ability to process plastics with high filler loadings.
- Reduced warpage of injection molded parts.
- Reduced wear of fabrication and mixing equipment.

The conclusion of the fifth lecture covered the research project sponsored by EPRI to evaluate the fly ash residue from the DAL process to assess its performance in two common plastics. The contractor for this research was Ontario Research Foundation.

The objectives of the ORF research were as follows:

(1) To demonstrate how pre-treatment of a selected raw fly ash may be used to upgrade it as a feed material for the DAL-process.

(2) To investigate the extractability of metal values from the selected ash in relation to the pre-DAL benefication process; and to confirm the scale-up of the DAL-process to a pilot plant level.

(3) To demonstrate how beneficiation of the DAL-residue may be used to provide filler grade products.

(4) To evaluate the technical performance of the beneficiated DAL-residues as fillers in polymer composites.

(5) To evaluate the potential marketability of DAL-residues in the fillers industry.

My lecture described the plastic filler tests used and the results of the testing. The test methodology was as follows:

For comparative testing, three widely used commercial fillers were selected: (1) talc, a reinforcing filler widely used in automotive parts manufacturing; (2) ground calcite, a low-cost general purpose filler; and (3) glass spheres, a typical spherical filler.

- Tests were selected on the basis of their ability to reveal any major technical advantages or disadvantages affecting the use of DAL-fillers in composites:
- Melt index - this test reveals the effect of filler type and loading on the flow of a molten plastic composition and indicates its probable performance during injection-molding
- Impact strength
- Tensile strength and elongation under tensile stress.

- Flexural and tensile modulus, a measurement of stiffness.
- Color of finished composite.
- Morphology of fracture surfaces as seen by scanning electron microscopy as an an indication of bond quality and failure mechanism.

Lecture 6: (Dec. 11) Fly Ash Backfills and Other Power Plant By-Products

The lecture on fly ash backfills was primarily a slide show containing examples of successful backfill projects in the United States and Canada. Historically, the use of fly ash as a fill material has been the direct result of its light unit weight, high strength, low compressibility, and relative low cost when compared to natural material. The light unit weight is particularly advantageous in situations where filling is necessary on relatively weak and compressible subsoils. When natural earthen materials are used which commonly have unit weights of 100-130 lbs per cubic foot, excessive settlement on a compressible subsoil is often the result. the average range of compacted dry unit weights for fly ash is 70 to 95 lbs per cubic foot, which represents a considerable reduction in the surcharge placed on these in-place materials. Anytime there is a lack of suitable borrow material near a construction site, fly ash from a nearby power plant can represent an economical source of borrow material.

Fly ash or cement-fly ash mixtures can be used as a replacement for well-graded sandy gravel or other backfill materials in bridge abutment backfills. The advantage of fly ash in these type of applications is its low compressibility and lower unit weight which reduce lateral loads on the abutment. Other commonly cited advantages include lower material cost and a source of low moisture content borrow for use during periods of wet or freezing weather. Portland cement- fly ash mixes can be used in lieu of earthen material for pipe backfills. The advantages of this application are the high strength and low compressibility of the material and ease of use.

LECTURE 6 PART 2 - UTILIZATION OF OTHER POWER PLANT BY-PRODUCTS

The objective of one of the research projects at the Electric Power Research Institute in the USA has been to identify and evaluate the potential for utilizing the solid waste by-products from coal plants using advanced SO_2 control processes. The work already in progress has been divided into four discrete subtasks with specific output objectives as follows:

- Evaluation of current utilization practices -- the objective of this subtask has been to identify and characterize existing utilization practices.
- Screening of utilization practices for new wastes -- the objective of this subtask is to identify new utilization practices relative to the characteristics of advanced SO_2 control process wastes and itentify data deficiencies.
- Identification of new alternatives -- the objective of this subtask has been to identify new utilization alternatives which may be feasible because of differences in advanced wastes characteristics as opposed to conventional wastes.
- Assessment of utilization practices -- the objective of this subtask is to rank utilization alternatives for advanced SO_2 control process wastes on the basis of technical and economic feasibilities.

The initial evaluation of utilization alternatives was completed and summarized in an interim report issued by EPRI in June 1987. The basis for the evaluation was threefold: (1) a review of previously published literature on conventional and advaced SO_2 control by-product utilizaation: (2) a technological evaluation of potential by-product utilization applications based on literature information; and (3) a preliminary market assessment based on comparisons with conventional combustion by-products' marketability. The results of the evaluation were used to prioritize alternatives for further analysis and identify testing needs to support that analysis. The initial feasibility screening was conducted with the use of an evaluation matrix containing such technical factors as physical properties, handling characteristics, chemical composition, environmental effects, and processing requirements. Market considerations were not included in this phase of the analysis. The table on the next page lists the high potential options by technology.

The second phase of the evaluation focused on the assessment of probable markets for the by-products using data from previous studies on the marketability of conventionaol coal combustion by-products. Key considerations include market volume, expected revenues, competing products, seasonal use restriction, geographic restrictions, and product addeptance. When the technical and marketing assessments were integrated, they yielded a set of priority alternative as summarized in the Task 4 Interim Report, issued in June 1987 as EPRI Report CS-5269.

Advanced SO_2 By-Product Utilization Testing

Utilization testing of all high and moderate potential alternatives is nearly complete. Utilization test results available to date are summarized as follows:

- Using standard test mixtures, all by-products, except those from calcium spray dryers, produced acceptable stabilized soil.
- Based on standard test mixtures, only AFBC residue produced an acceptable road base material.
- Sludge stabilization testing is complete, but further work is required to define minimum strength requirements.
- At 5% substitution levels, fine aggregate by-products, except AFBC, produced acceptable quality asphalt.
- Using standard test mixtures, only AFBC and calcium spray dryer residues produced grout with the required minimum compressive strength.
- All attempts to produce fired brick and ceramic products were unsuccessful.
- All by-products produced acceptable quality mineral wool.
- Synthetic and lightweight aggregate production and related testing are still in progress.
- Calcium spray dryer and limestone furnace injection residues met ASTM C618 criteria for use in concrete; at 30% cement replacement levels, these by-products yielded concretes with 90-day compressive strengths of over 6,000 psi.

Future EPRI Plans

The present plans are to complete the waste utilization laboratory work in 1987. Recommendations will be formulated regarding additional utilization development work, including demonstration projects. There appears to be industry support in the USA for additional funding of research to develop the by-product potential of the new wastes created by the clean coal technologies. To not do so would be counterproductive in the long run. I believe for example that the greatest impediment to the utility industry implementation of AFBC technology over the next decade will be the issues related to the disposal of the waste by-products. Many of the plants that would otherwise be likely candidates for AFBC are retrofit facilities that are located in urban areas or confined sites. At the Oak Creek Plant for example the present unit will produce 120 lbs of ash for every 1,000 lbs of coal burned. When AFBC is installed the waste production will go to 420 lbs per 1,000 lbs of coal consumed. This dramatic increase in disposal quantities even when compared to conventional wet FGD systems puts this technology at a competitive disadvantage compared to other SO_2 control technologies. It is imperative for EPRI, the US Department of Energy, equipment manufacturers, and other organizations to develop the utilization options for these by-products to mitigate the disadvantage.

Lecture 7: (Dec. 12) Desulphurization Technologies and the Artificial Fishing Reef Project

LECTURE 7 PART 1: Environmental Aspects of Power Plants and FGD Technologies

Power plant sulfur dioxide emissions and waste disposal are the two most prevalent environmental issues facing electric utilites building or operationing coal-fired power plants today. Concern over the impacts of sulfur dioxide emissions has led to the development of a number of flue gas desulfurization (FGD) techologies. The original generation of FGD control equipment consisted of wet processes to wash the flue gas, and resulted in wet scrubber sludge. The second generation of FGD systems achieve the desulfurization through injection of calcium or sodium based sorbents at various stages in combustion or flue gas systems. Unlike the more traditional wet scrubber FGD systems, these new processes generally produce a dry product. The disposal of these wastes as practiced in the USA is by either landfilling, ponding or mine disposal. Landfilling and mine disposal options are used to dispose of solids and dewatered sludges, while ponding is used for disposal of slurries or for interim storage prior to treatment. In reviewing the environmental issues related to the disposal of the combustion products, an understanding of the properties of the by-product wastes is essential.

About half of the coal ash produced in the USA is currently handled in wet systems and disposed of in ponds. The national trend, however, is toward dry collection and landfilling. A number of factors have been responsible for this change, including environmental laws protecting groundwater quality, as well as economic reasons. Mine disposal of these wastes is not very widespread. There are less than ten full-scale mine disposal systems in operation.

The disposal characteristics of wet FGD scrubber sludge will, to a great extent vary with the proportion of sulfate to sulfite. This is due to the differing particle morphology. The important physical properties of FGD sludge are as follows:

- Moisture Content
- Solids Content
- Dry Bulk Density
- Permeability
- Compression Index
- Angle of Internal Friction
- Cohesion
- Unconfined Compressive Strength

The FGD sludges which are predominantly calcium sulfate dewater more easily and to a greater degree than the sulfite forms. A preponderance of sulfate will be beneficial to the handling and physical characteristics of the product after disposal. The factors affecting FGD sludge charateristics are as follows:

- Extent of Oxidation
- Fly Ash -- Amount and Type
- Coal -- Type and Source
- Reagent -- Type, Durability, Purity
- Water -- Quantity, Purity

The potential interactions of the environment with land-disposed utility wastes are numerous. Some of the trace elements discussed in Lecture 4 (Metal Recovery), present in the coal ash, are phytotoxic, other are toxic to fish and other aquatic organisms, and others have adverse effects on humans and animals. The land disposal environmental issues discussed during my lecture included:

- Effects on Local Air Quality
- Effects on Soils and Vegetation
- Phytotoxicity
- Effects on Groundwater
- Effects on Surface Waters
- Disposal Site Washout

No discussion of the potential environmental pathways and effects of utility disposal operation would be complete without a review of the natural mitigation mechanisms and engineered controls available for assuring the safe disposal of utility coal combustion waste by-products. The natural mitigating mechanisms I reviewed are listed below:

- Soil Buffering Capacity
- Attenuation of Trace Metals in Some Soil Types and Organic Matter
- Low Soil Permeability

The mobility of metals in soil is strongly dependent on soil-specific characteristics. For example the soil can exert a buffering influence over the fly ash leachate by raising or lowering the pH. The solubility of most trace metals (the notable exceptions being As and Se) tend to decrease with increased pH. In general, trace metals are less mobile in alkaline soils because the metals will precipitate and/or adsorb onto hydrous iron and aluminum oxides. Clay content, and the presence of organic matter in soil can also strongly affect attenuation of trace elements. Clay serves two functions by adsorption of metal ions from leachate, as well as retardation of water movement due to small pore size and low permeability. Organic matter in soil can chelate metals. For example the mobility of cadmium, lead, and nickel in soils is limited, since the clay-organo fractions of the soil have a high affinity for these heavy metals.

In addition to the existence of natural mitigating measures, engineered disposal systems can control the migration of leachate from disposal sites. The measures I reviewed in my lecture were as follows:

- Liner Systems
- Leachate Collection Systems
- Fixation / Stabilization of Waste
- Site Grading
- Waste Cover
- Surface Water Control

These engineering control methods are described in detail in two EPRI disposal manuals that I forwarded to SRIBS upon my return to the USA. The manuals are intended for the use in designing new disposal facilities.

ADVANCED SO_2 CONTROL PROCESSES

1. Atmospheric Fluidized Bed Combustion (AFBC)

AFBC technology involves the combustion of coal in a bed of fluidized sorbent such as limestone. Crushed coal and limestone are mixed and held in suspension by an upflow of combustion air. Currently there are about 2,000 small-scale AFBC boilers in China, although they normally do not combine the coal and limestone for SO_2 control. In AFBC boilers designed for SO_2 control, the coal burns in the bed and in the freeboard above the bed while the limestone is calcined to form lime. By introducing the sorbent to the combustion chamber, SO_2 and other acid gases react with and are absorbed by the sorbent, thereby reducing emissions. Fluidizing the coal and sorbent creates turbulent mixing, causing the entire mass of solids to behave as a pseudo-fluid. This turbulent mixing enhances heat generation and transfer in a manner similar to conventional pulverized-coal fired boilers but as lower combustion temperatures and with essentially no loss in efficiency. The lower combustion temperatures in AFBC boilers are well below the level for thermally induced formation of nitrogen oxides (another cause for concern regarding electric utility emissions). Lower combustion temperatures also minimize the slagging, fouling, and other fireside problems associated with conventional pulverized and stoker-fired units. During combustion, a portion of the bed material is continuously withdrawn to maintain proper bed conditions. A fraction of the bed material (i.e. the fly ash, reaction products, and unreacted limestone or lime) are elutriated and carried by the combustion gases through cyclones where a substantial fractin of the particulate matter is collected and recyhcled to the bed. The remaining particulate matter is carried by the flue gas to a fabric filter or electrostatic precipitator where it is collected for utilization or disposal. AFBC technology may permit electric utilities to burn lower cost, higher sulfur coals while meeting emission requirements without additional, expensive, downstream emission controls. The principal impediment to this technology appears to be the realitively large volumes of wastes produced compared to other emission control technologies. My lecture (No. 6) discussed the utilization research activities for AFBC by-products to mitigate this concern.

2. Calcium Spray Drying

In spray drying, a lime solution/slurry is sprayed as a fine mist into flue gas in a reaction vsssel where the mist reacts with the SO_2 and dries to a fine particulate. The particulate laden flue gas then passes to a fabric filter (bag house) or electrostatic precipitator where reaction products and fly ash are collected.

Spray drying has several advantages over wet scrubbing FGD systems. It produces a dry waste and, therefore eliminates the complexity and operating problems associated with the large volumes of liquid wastes produced in wet FGD systems. The dry waste product can be utilized or landfilled directly without dewatering and/or ponding. The major disadvantage of this process compared to conventional FGD is the significantly higher unit cost of the alkaline absorbent material. Spray drying requires a highly reactive absorbent like lime to attaine high SO_2 removal efficiencies. Prior to my trip to Shanghai, I met with Mr. Ye Yisen, Vice Secretary General of Chinese Association of Environmental Protection Industry, from Beijing. He indicated that calcium spray drying technology is the technology that is being considered most likely for implementation in China.

3. Sorbent Furnace Addition

Sorbent furnace addition accomplishes SO_2 control by injection by injection of a pulverized calcium-based material such as limestone or hydrated lime directly into the furnace of a pulverized coal fired boiler. The sorbent rapidly decomposes at high temperatures releasing either carbon dioxide (in the case of limestone) or water (in the case of lime). The sorbent forms a porous micro-structure upon decomposition, having a much increased and more exposed surface area than the original sorbent material. The resulting lime particles are highly reactive, and chemically combine with SO_2 and oxygen to form, mostly, solid calcium sulfate. This calcium sulfate, together with the fly ash and unreacted lime is collected as dry particulates in a fabric filter or electrostatic precipitator. The limestone furnace injection process is similar in overall SO_2 removal chemistry to that for fluidized bed combustion, althoughthe two processes operate under distinctly different temperatures, residence times, and combustion conditions.

4. Dry Sodium Sorbent Injection

In dry sodium sorbent injection, sodium compounds, such ash naturally occurring forms of sodium carbonate and bicarbonate (nahcolite and trona) are pulverized and introduced in the flue gas ahead of a particulate collection device. The SO_2 in the flue gas reactes with the sorbent, and the reaction products, and together with the fly ash, are carried by the gas stream to a fabric filter or electrostatic precipitator where they are removed and then stored for subsequent management.

5. Advanced Coal Cleaning

The processes I discussed above are effective in controlling SO_2 emissions during and after coal combustion. An alternative in some situations is to use a precombustion process, such ash coal cleaning, to reduce a fuel's sulfur content. Coal cleaning removes some of the mineral impurities present in coal, such as mined rock and pyritic material. Coal cleaning processes are designed typically to yield a fuel with lower ash and sulfur contents and a higher unit heating value. Higher-ash content, higher-sulfur raw coals become more attractive via this route since flue gas emissions can be lowered. The technology is like the others in that it generates a solid waste by-product which must be managed properly. Typically, conventional coal cleaning operations have been situated remote from the power plants with refuse management done by the mine operators. With its advent, advanced physical cleaning technology can reasonably be anticipated to be located at power plant sites. In such cases the management of coal cleaning residuals could become the responsibility of the electric utilities.

LECTURE 7 PART 2: The Coal Waste Artificial Fishing Reef Project

My last lecture topic discussed the results of an EPRI project which investigated the technical feasibility and environmental acceptability of using coal waste blocks as artificial fishing reefs. In highly urbanized areas like Europe, China, and the Northeast United States, the land available for disposal of wastes is very limited. One alternative is the disposal at sea of coal waste in the form of stabilized blocks of fly ash, cement and FGD scrubber sludge. EPRI, through the coal waste artificial reef program (C-WARP) evaluated the feasibility and environmental effects of stabilized coal waste blocks in an ocean reef constructed off Long Island, New York in 1980. The program constructed a 500-ton reef, consisting of over 15,000 blocks of coal by-products. The block production process would if commercialized be fully automated, from raw material supply, through block molding, curing, and cubing using conventional concrete block-making technology. The ocean disposal system consisted of four phases: (1) collecting blocks from the storage yard and transporting them to a barge loader, (2) loading the blocks onto the barge, (3) transporting the blocks to the ocean disposal site and depositing them in a reef, and (4) monitoring the disposal area.

From an environmental standpoint the disposal of stabilized coal waste blocks at sea appears to be acceptable. Diverse data from laboratory and field investigations at the reef site of the physical, chemical and biological interactions of the two mixes of coal waste blocks used in the program have all suggested that in the form of solid blocks, the material is compatible with the marine environment. Block elemental composition was determined using a variety of techniques including x-ray fluourescence, neutron activation, and atomic absorption spectrophotometry. Mineral phases and mineralogical changes were determined employing scanning electron microscopy, x-ray diffraction, and light microscopy. Long-term leaching studies yielded leaching rates for major block components.

The stablished coal waste blocks have proved to be suitable substrates for the settlement and growth of marine organisms which are characteristics of artificial reefs. Analyses of colonizing organisms collected from reef blocks, which had been in the sea over four years, found no evidence of elevated levels of block compounds, including trace elements, in the biomass. Bioassays of coal waste eluriates in seawater had no significant effects upon developing fish eggs and larvae nor upon growth of cultures or marine plant cells. The physical integrity of the blocks has been maintained and the material compressive strength has increased. Block densities were also found to increase after immersion in seawater. Leaching of major components decreased with time and trace elements appear to remain absorbed in the blocks. Tests on the organisms colonizing the reef indicate that toxic materials are not being absorbed into organisms.

Copies of EPRI reports on this program have been sent to SRIBS upon my return to EPRI. There may be a possible use for this technology in Shanghai, since the block production facility is already in place. Perhaps broken or off-spec blocks could be barged to off-shore locations in the East China Sea.

SECTION V: Discussion on Cooperation (Dec. 14)

The next to last day of my visit I met with the Director of SRIBS, Dr. Wang, to discuss possible areas of future cooperation between EPRI and their organization. It is obvious that there is a common area of interest in ash utilization research. There would be benefits to both organizations for technical cooperation between organizations. After all China is second only to the United States in the amount of coal utilized in power production. The amount of ash generated in China last year was 43 million metric tons. The rate of increase in coal fired generation capacity over the next several years as a result of the industrialization of China will no doubt cause China to exceed the USA in coal capacity early in the next century. SRIBS recognizes EPRI's leadership in the USA in ash utilization research, and has expressed the willingness to develop closer ties in the months and years ahead. This technical cooperation can be at several levels. I have aggreed to provide copies of EPRI reports related to ash utilization to them in exchange for copies of the technical articles that they write in English. They provided me a set of about 15 papers that they have already written in English. Their younger engineers typically study English now in college, so there is an increasing level of proficiency among thier staff. However, the language barrier still exists, since the full technical reports and data analysis are still written in Chinese (Mandarin dialect).

The next level of cooperation could be to have a bilingual SRIBS engineer spend a year here at EPRI much like others have from industrial organizations from Japanese utilities. The SRIBS Director indicated that they have been doing this successfully with other organizations around the world. One of these engineers worked at Ontario Hydro (16 month duration) and served as one of my translators for my lectures. Another is currently in Oslo, Norway studying silica fume utilization. Others have worked or studied in France and the USA. I am not sure what impact the new US immigration law would have on having a foreign national work at EPRI. The engineers in China that I met with are well versed technically, having completed a rigorous engineering curriculum in school and possess hands-on experience in ash utilization concepts. Bringing one of the engineers to EPRI for a year would allow for a more meaningful technical exchange of data and study results because the language barrier would be overcome.

The third level of technical cooperation could take the form of actual research contracts issued to SRIBS to conduct specific studies for EPRI. Obviously their labor rates, make the work cost effective. The language barrier could be overcome by implementing this level of cooperation while an "exchange" project manager is in residence here at EPRI. Two areas of possible research are in the area of plastic filler and metal filler mix design.

SECTION VI: RECOMMENDATIONS

It is evident from the current scale of the research program at SRIBS as well as at other organizations within China which sent representatives to the lecture series, that the utilization of industrial by-products is recognized and is considered very important. However, given the budgetary limitations on research into by-product utilization in China as well as in western countries in North America and Europe, it is important to develop cooperative ties to minimize duplication of effort and exchange technical knowledge. This could be accomplished through cooperative activities such as the three levels that I described above.

Throughout the lecture series there were a number of questions or comments which suggest the need for follow-up with more specialized presentations by other experts or areas for additional research. The recommendations below are based on my perceptions of the level of interest and unresolved questions.

- The prediction model for fly ash concrete strength (Lecture 3) generated considerable interest. This model is presently being expanded to include other important parameters like sulfate resistance, alkali-aggregate reaction, freeze/thaw resistance, drying shrinkage, and set time. The EPRI project will be completed in April 1988. The EPRI contractor for this work, Ed Dunstan, would be an ideal candidate for a future UNIDO short-course at SRIBS.

- The use of RCC concrete in pavement (Lecture 3 Part 2) evoked considerable interest from the audience. It is recommended that a specialist in this reuse option from an organization such as the US Bureau of Reclamation be considered for a follow-up short-course.

- Lecture 4 on ash resource recovery generated considerable questions and a high level of interest. The high level of technology and exotic materials of construction required would make this alternative less attractive for China in the near future. On the other hand, ash beneficiation methods using size classification, magnetic separation in both wet and dry ashes would be useful to the development of products for activated carbon, magnetite, and filler markets. A recommended lecturer for this type of short-course would be Dr. Ray Hemmings of the Ontario Research Foundation, in Mississauga, Ontario, Canada.

- Composite materials made from cast metal and fly ash filler represent a potentially large market for fly ash. SRIBS indicated that they have some research underway in this area. It might be useful in the future to have Prof. Rohatgi, of the University of Wisconsin at Milwaukee, provide a series of lectures on the ash cast composite materials. He is the recognized world expert on this technology, and is currently under contract to EPRI to do developmental work in this area.

- EPRI research in ash fillers in plastics to date has been limited to nylon and polypropylene. Future R & D needs to be done on a wide variety of other plastics, like polyelefins, styrenes, TFE, polyesters, silicones, urethanes, and vinyls. This is a possible area of future co-operation between EPRI and SRIBS and a possible subject area for future UNIDO sponsored ash utilization short-courses.

- The half day lecture 7 on desulphurization technologies generated considerable interest from those members of the audience with utility companies. An utilization assessment of SO_2 control by-products should begin prior to the introduction of scrubbers. The utilization potential should be factored into the selection of the appropriate technology. This subject is a possible area of future short-courses. The Chinese Association of Environmental Protection Industry in Beijing, might be interested in participating in such a short-course. A possible lecturer from EPRI would be Stuart Dalton, who is Program Manager for Desulphurization Processes. The contact person at the CAEPI would be Ye Yisen, Vice Secretary General, who is with the Ministry of Urban and Rural and Environmental Protection, in Beijing.

- The flowable ash backfill lecture also generated interest. A follow-up lecture by an expect in this technique would be beneficial. Robert Collins, a consultant in Springfield, Pennslyvania is the acknowledged American expert in this area.

The three primary ash utilization applications in the Shanghai area are walling material, cement production, and road construction. Fly ash walling material includes brick, block and panel among which block is the largest single field application with annual production of 240,000 cubic meters. During my visit I was given a tour of the fly ash block production plant in Shanghai. The SRIBS personn[illegible]gnize that if they are to maintain the high levels of ash utilization (now about 75% in th[illegible]ai area) over the next few years that they will have to develop other markets as well. For this reason they have developed other markets for ash in ceramic tiles, ornamental brick, and rubber products. The new SRIBS headquarters building has the ornamental brick on the exterior. They are just beginning research on plastic filler and metal filler applications. I think one of the ways that SRIBS has been able to stay in the forefront of research into ash utilization is that over the last five years they have had at least four UNIDO experts on ash utilization from organizations all over the world come in and lecture. This "cross pollination" funded by the the United Nations has helped them in formulating a research program that has proven to be very effective in increasing the ash utilization in the Shanghai area even when the ash production rates have increased dramatically with new plants coming on line. I understand that there are four additional UNIDO experts on various aspects of ash utilization who will be visiting SRIBS during 1988. I strongly endorse the continued support of this kind of program.