FINAL TECHNICAL REPORT
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Project Title: **UTILIZATION OF ILLINOIS FLY ASH IN MANUFACTURING CERAMIC TILES**

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ABSTRACT

The overall objective of the project was to utilize fly ash, produced by burning Illinois basin coal which is currently being landfilled (due to lack of resource utilization), as raw material to manufacture value-added ceramic tiles for commercial use.

Among several steps necessary to achieve this objective, the present work focussed on the composition formulation and processing to produce tile bodies with characteristics similar to those of commercial tiles. Two ceramic processing approaches were undertaken to accomplish this objective. In one approach, solid state sintering was performed without any compositional modifications. In the other approach, fly ash particles were sintered using low temperature liquid-forming additives to facilitate sintering.

Two Class F fly ashes were investigated in the current program. Prior to processing, these ashes were characterized for their physical and chemical characteristics. The processing conditions and the temperature range used in the program were well within the practice of the tile industry. The data generated to-date clearly indicate that both fly ashes are sinterable at a very desirable temperature of approximately 1,050°C.

In order to compare the characteristics of the fly ash tile bodies produced in this program with those of commercial tiles, a total of 8 commercial tiles (floor, wall, and patio, both glazed and unglazed) from several manufacturers were also evaluated for two vital properties, tensile strength and water absorption. The results obtained clearly show that the properties of fly ash tile bodies are far superior to those of wall and patio tiles and similar to those of floor tile bodies. The accomplishments made clearly indicate that fly ash is a viable replacement for the raw materials used in manufacturing wall, floor, and patio tiles.

Pages 6, 7, 11-22 contain proprietary information.

EXECUTIVE SUMMARY

The fine particulate material that is electrostatically precipitated or mechanically collected from the stack gases of power plants burning pulverized coal is called fly ash. Annually, about 48 million tons of fly ash are generated in the U.S., of which 3 million tons are generated in Illinois. Approximately 20% of the fly ash generated in the U.S. is utilized by the cement and concrete industry. A small fraction of the fly ash generated in Illinois is utilized by the concrete industry. Any non-concrete utilization of the fly ash being disposed of will not only be environmentally sound and cost effective, but also will create a stable year-round demand. The approach taken in this investigation is unique with respect to the methods of fly ash utilization practiced today.

The ultimate objective of the project was to utilize Illinois fly ash as raw material for manufacturing value-added ceramic tiles for commercial use. Among the several steps necessary to achieve this objective, present work focused on the first step, composition formulation, and processing to produce tile materials with characteristics similar to those of present commercial tiles.

In general, the oxide composition of Illinois fly ash is such that, with or without minor compositional modifications, it can be utilized as raw material for making ceramic tiles in an energy-efficient manner. Furthermore, the fine powder form of fly ash makes the utilization simpler and cost effective, as no further grinding is necessary.

Ceramic tiles are very common construction materials used in households and commercial facilities of all sizes. They are used on floors, walls, and patios, both indoors and outdoors. There are approximately 100 ceramic tile manufacturing plants in the United States, and in 1990, these plants produced approximately 510 million square feet of tiles. The properties of commercial tiles vary depending on the application, and are specified in the American National Standard Specifications for Ceramic Tiles (ANSI 137.1).

Two Class F fly ashes investigated in the current program were characterized for oxide and mineral compositions, density, and particle size distribution (PSD). As fly ash contains a number of oxides, the compositional triangles for both Baldwin and Gibson ashes have been identified through two ternary equilibrium phase diagrams.

In order to make ceramic tiles, fly ash particles need to be formed into a compact mass through sintering. Sintering forms solid bonds between particles when they are heated to appropriate temperatures. In present investigation, two following approaches were taken: (i) sintering of as-received fly ashes with and without minor compositional modification (solid-state sintering) and (ii) liquid-phase sintering of fly ash particles in the presence of additives that form liquid at lower processing temperatures.

Sintering of these two ashes were performed under the following processing conditions:

- a range of forming pressures
- a range of sintering temperatures
- a range of heating and cooling rates
- various times at sintering temperatures
- various liquid-forming agents, and quantities.

In order to optimize the sintering behavior, samples were formed into small discs (approximately 3 cm in diameter and 0.5 cm in thickness), and primary emphasis was given to densification (bulk density) that was measured simply from the weight and dimensions. An increase in bulk density imparts attributes such as increased strength, lower porosity, and reduced water absorption characteristics for the sintered specimens. As true density is the density of the solid material and all the open pores within the fly ash particles are excluded in this density measurement, achieving true density in the sintered bodies is not possible, and may not be necessary for present application.

Solid-State Sintering

Solid-state sintering of both Baldwin and Gibson fly ashes was performed by mixing fly ash with small amounts of water and forming under various pressures with a maximum of 270 MPa.

These discs were then fired in air at temperatures ranging from 900 to 1,100°C, and the soaking times at these temperatures were varied. Approximately twenty-four minutes of sintering appeared to be adequate. The pressures used to compact fly ash powders also influenced the fired bulk density. Both fly ashes were found to be sinterable at a desirable temperature of approximately 1,050°C, and a sintered bulk density of over 90% of the true density was achieved. It was also apparent that the sintering requirements of these two ashes are not quite the same.

It was concluded that both fly ashes can be solid-state sintered to over 90% of true density, and this can be achieved under processing conditions, especially temperature and forming pressure, that are available in commercial tile manufacturing plants.

Liquid-Phase Sintering

In order to perform liquid-phase sintering, various kinds and quantities of liquid-forming agents were used. A pressure of 132 MPa produced green as-pressed discs with reasonable handling characteristics. Firing of the discs along with control specimens (containing no sintering aid) was carried out in air at a temperature of 900 to 1,000°C for 25 min and then slowly cooled to room temperature.

The fly ashes were liquid-phase sintered to a density of up to 97% of true density with a small addition of sintering aid, and the sintering temperatures were 50 to 100°C below that which was necessary for solid-state sintering.

Comparison of Tensile Strength of Fly Ash Tile Bodies with those of Commercial Tiles

In order to compare the characteristics of fly ash tiles produced in the current program with those of commercial tiles, a total of 8 commercial tiles (floor, wall, and patio, both glazed and unglazed) from several manufacturers were evaluated. Tensile strengths were measured on tile bodies of comparable dimensions.

The results indicate that the tensile strength of the fly ash tile bodies are *more than three times* those of commercial wall tiles, *more than five times* those of patio tiles, and *similar* to those of floor tiles.

Comparison of Water Absorption of Various Commercial Tile Bodies with Those Made of Baldwin and Gibson Fly Ashes

Water absorption of tile bodies, both commercial and those made of fly ashes, of comparable dimensions was also measured. According to specifications, water absorption for glazed wall tiles is not to exceed 20% and that for floor tiles varies from impervious to a maximum of 5% depending on the type of tile and its application. Because the water absorption of fly ash tile bodies was *equal or slightly better* than that of all the commercial tiles tested, the ANSI 137.1 specification for water absorption can be met easily.

Deliverables

In order to deliver tile bodies made of fly ash to ICCI, a large die capable of producing specimens with size as large as 10 cm x 10 cm was fabricated. Two early samples of large tile bodies made of Baldwin and Gibson fly ashes are provided along with this report.

The results obtained to-date are very encouraging, and it is apparent that fly ash has potential to be an effective replacement for raw materials used to produce commercial tiles for various applications.

The remainder of this report contains proprietary information and is not available for distribution except to the sponsor(s) of this project.