## FINAL TECHNICAL REPORT

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Project Title: ENGINEERING DEVELOPMENT OF FGD-GYPSUM CONVERSION TO FERTILIZER AND PCC

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## **ABSTRACT**

Utility plants burning high-sulfur coal using the wet flue gas desulfurization (FGD) technologies, in addition to installation and operating expenses, face increasingly expensive landfill disposal for the gypsum they produce. Due partially to the diminution of sulfur compounds from air deposition, there is a growing demand for sulfur in the sulfate form as a plant nutrient. Successful production of fertilizer from FGD-gypsum would economically favor FGD technology and provide the agriculture community with sulfur and nitrogen fertilizer for the soil.

Since 1994, a chemical process which converts FGD-gypsum to ammonium sulfate fertilizer and fine calcium carbonate (PCC) has been developed at the bench scale at the ISGS. The goal of this project for the process development during the 1998-2000 period was to obtain critical information and additional industrial support to build a pilot plant for process demonstration. During this first year, the following tasks were completed: a scaled-up batch unit construction and operation; scaled-up batch unit production and products characterization; a market survey; and the process design and its economic estimation; a survey of utilities that use the FGD system with Illinois coals; and the process feasibility tests for the use of FGD-gypsum samples from producing plants in Illinois.

During the second year, the engineering package was to be refined for the design of a pilot plant to be built and operated at a site in Illinois. In preparation for this task four candidate sites and companies were considered for the location and operation of the pilot plant. Further discussions on the location and design of the pilot plant were not implemented because of the uncertainty of the second-year funding. Also, confirming process conditions for crystallization of the ammonium sulfate product in a pilot-scale operation, a goal of the second year's investigation, was not pursued.

The cost estimates showed that by allowing \$16/ton for the return of the PCC back to the utility FGD unit, a 21% return on investment in the gypsum conversion process can be realized. The results of trace elements analysis showed that the PCC produced had 90% less trace elements when compared with its source limestone suggesting that the PCC could be used for other higher value applications. Also, separation by filtration showed that a portion of the PCC could be recovered with a high brightness and might be marketable in the paper or paint industry where the cost for PCC can be as high as \$300/ton.

## Pages 1 to 31 contain proprietary information.

## **EXECUTIVE SUMMARY**

Wet flue gas desulfurization (FGD) processes, that use limestone as a scrubber for  $SO_2$  and produce gypsum as a by-product, have a considerable level of commercial development and demonstrated operational experience. These pollution-control technologies will remain preferred choices for Phase-II compliance if successful commercial utilization of FGD by-products is developed to offset the cost of equipment installation and operation and decrease or eliminate by-product disposal cost. A FGD system installed on a 687-MW plant burning 2.5% sulfur coal, with a desulfurization efficiency of 95%, and a load factor of 75%, generates about 30 tons of gypsum per hour. In the future, the cost of disposal of the gypsum produced may increase to between \$10 to \$30/ton. From both environmental and economical standpoints, it is desirable to use this by-product as a feed material to produce salable products.

Ammonium sulfate is a valuable nutrient source of both nitrogen and sulfur for growing plants. There is an increasing demand for sulfur in the sulfate form as a plant nutrient because of decreased sulfur compounds in atmospheric deposition. More stringent regulations to reduce sulfur dioxide emissions by the year 2000 (from 2.5 lbs SO<sub>2</sub>/10<sup>6</sup> Btu to 1.2 lbs) suggest a growing demand for sulfur nutrients in the soil by the first decade of the 21st century. Also, the trend toward using high-nitrogen content fertilizers has pressed incidental sulfur compounds out of traditional fertilizer. The current market for ammonium sulfate in the United States is about two million tons per year. It is anticipated that 5 to 10 million tons of new ammonium sulfate production may be required in fertilizer markets annually as a result of the acid-rain control program. The fertilizer industry is seeking a greatly increased source of such product to supply sulfur in nitrogen-phosphorus-potassium (N-P-K) fertilizer blends. The average sale price per ton of ammonium sulfate has increased each year. For example, from 1993, to 1994, to 1995, the average price per ton has increased from \$157, to \$170, to \$182 respectively. By 1998 the average price was \$187 per ton.

Since 1994, the Illinois State geological Survey (ISGS) Division of the Department of Natural Resources has been developing a chemical process which converts FGD-gypsum to ammonium sulfate fertilizer and precipitated calcium carbonate (PCC). During phase I of the project (1994-95), process development on gypsum from the Abbott Power plant focused on an economical process to produce ammonium sulfate fertilizer. The results from prefeasibility cost estimates based on a conceptual design of the process, without considering the high-value commercial application of the PCC produced, indicated that there was a healthy profit margin of 25% for converting FGD-gypsum to ammonium sulfate if a granule size of (1.2 to 3.3 millimeters) ammonium sulfate product could be produced.

During phase II of the project (1996-97), various specifications for commercial applications of calcium carbonate, both ground and precipitated, were reviewed for determining the quality of PCC produced from Abbott gypsum and gypsum from City Water Light and Power's Dallman Generating Station (CWLP). The results from both literature and laboratory studies indicated that the particle size of PCC is within the Grade II size classification and is suitable for applications in paints, adhesives, and sealants. The influence

of impurities in the starting gypsum on the quality of the PCC produced was also established. The results suggested that in order to reduce impurities such as iron and organic carbon in the PCC product, either limestone with low iron and low-organic carbon content should be used during FGD process, or purification procedures for the starting FGD-gypsum (front end purification) or product PCC (back end purification) are needed to produce PCC that is white enough for higher-quality commercial applications.

During phase III (1997-98), the chemical properties of ammonium sulfate made from FGD-gypsum were evaluated and used to estimate its effects on soil fertility. Purification procedures and modified processes to control the purity of the starting FGD-gypsum were initiated. Also, the preparation of a basic engineering package including a process flow diagram to aid in the design and construction of a pilot plant was initiated. The results of product evaluation indicated that the impurities in the ammonium sulfate produced would not impose any practical limitations on its use at application levels employed by farmers. Purification steps were developed for the starting gypsum and the brightness of the PCC products was improved.

The overall goal of this two-year phase IV of the project was to obtain critical information and additional industrial support to build a pilot plant for process demonstration. The specific objectives were to build and conduct a scaled-up batch operation unit (SBU) to produce ammonium sulfate slurry for small-pilot scale crystallization tests, to complete and update the basic engineering package for designing and building a pilot plant, and to obtain/analyze additional FGD-gypsum samples from combustion of Illinois coals to evaluate whether the process under optimization could successfully apply to FGD-gypsum of various sources. Confirming process conditions necessary to produce a suitably sized ammonium sulfate product will increase the process profit margin which is critical to the overall success of process commercialization.

During this first year, the following tasks were completed: SBU unit construction and operation; SBU production and products characterization; a market survey; and the process design and its economic estimation; a survey of utilities that use the FGD system with Illinois coals; and the process feasibility tests for the use of FGD-gypsum samples from producing plants in Illinois.

The SBU was designed based on the most current needs for product and by-product evaluation. Construction and the shake-down testing of the SBU were successfully completed. Two production tests were conducted. The results of one test indicated that about 12% of the PCC produced was of high brightness, and might be marketable in the paper or paint industry where the cost for PCC can be as high as \$300/ton.

The concentrations of trace elements were tracked from the source limestone used in the scrubber to the gypsum, then to the PCC and ammonium sulfate produced from the gypsum conversion. If no build-up of contaminants occurs, the PCC would be a viable product to be recycled back to the scrubber or could be used for a higher value application. If no inordinate amounts of heavy elements occur in the ammonium sulfate, it would be viable as a high-quality fertilizer. The results of the trace element study indicated that most measurable contaminants in the PCC decreased by more than 90% when compared with the source limestone. In addition, this process produces a high-purity ammonium sulfate fertilizer that can be applied to the land indefinitely without risk of buildup of any toxic elements.

The engineering package was to be refined during the second year for the design of a pilot plant to be built and operated at a site in Illinois. In preparation for this task, four candidate sites and companies were considered for the location and operation of the pilot plant. These companies are City Water Light and Power, Dallman Generation Plant, Springfield, Illinois; Agrium, Henry, Illinois; Archer, Daniels, and Midland, Decatur, Illinois; and Allerton Fertilizer Company, Allerton, Illinois. Since the second-year's funding was not considered, no further discussions on the location and design of the pilot plant were pursued. Also, confirming process conditions for crystallization of the ammonium sulfate product in a pilot-scale operation, a goal of the second year's investigation, could not be performed.

Market surveys were conducted for ammonium sulfate fertilizer in both solid and liquid form and for the PCC. The results indicated that the sale price of solid ammonium sulfate fertilizer increased significantly from 1974 at \$110/ton to 1998 at \$187/ton. The basic engineering package was developed to a level that the economics of the process could be updated. The economics of the process were updated for a plant to produce152,000 tons of ammonium sulfate fertilizer per year. The results of this study indicated that a profit of up to 25% can be made while producing and selling crystallized ammonium sulfate at the current market price of \$187 per ton, and a rational sale price of PCC for \$30 per ton . While allowing a purchase price of \$5 per ton for the FGD gypsum used, a profit of 23% can be realized. If the PCC produced is returned back to the FGD unit for \$16 per ton (the price that the utility would pay for its limestone), the profit would be 21%. For a similar plant that produces and sells liquid ammonium sulfate, a profit of up to 19% can be realized.

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